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Co-Chairs
Zh. Alferov
L. Esaki

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Phones: (812) 247 2617, 247 9932
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E-mail: vgrig@eo.ioffe.rssi.ru

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Spin relaxation of charge carriers in quantum dots

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Inverted electron–hole alignment in InAs/GaAs self assembled quantum dots
AIXTRON Young Scientist Award

In 1999, the Symposium Programme Committee and the Board of AIXTRON AG (Germany) established a special award to honour a young scientist who will present at the Symposium the best paper in the field of solid state nanostructures. The award comprises a diploma and a $500 reward sponsored by AIXTRON.

The 1999 awardee was selected by the Award Committee from six nominees proposed by the Programme Committee after consideration of extended abstracts of their contributions.

Alexey R. Kovsh
of the Ioffe Institute became the first recipient of AIXTRON Award for the presentation of the paper:

3.3 W injection heterolaser based on self-organized quantum dots


In this paper, authors realized, using more dense InAlAs quantum dots as centers for stimulated nucleation of InGaAs QDs, a very dense array of QDs and thereby achieved the record output power reported for QD lasers.

Alexey Kovsh was born in Leningrad (now St Petersburg) in 1973. After finishing secondary school he entered the Chair of Optoelectronics of St Petersburg State Electrotechnical University — an educational unit founded by and having close connections with the Ioffe Institute. Since this time he linked his professional activity with the Ioffe Institute.

In 1996, A. Kovsh received with honour his M.S. in optoelectronics from the Electrotechnical University and in 1999 his Ph.D. in semiconductor physics from the Ioffe Institute. A part of his dissertation was devoted to a new thermodynamic approach to molecular beam epitaxy, based on non-equilibrium consideration of growth processes. In another part of his work, the MBE growth and laser application of semiconductor quantum dots were considered. Several tricky ideas put forward in his dissertation led to a significant improvement of the QD laser technology.

Alexey Kovsh has co-authored more than 120 professional publications. In addition to reports at several conferences, he gave talks at the Institute of Electronic Materials Technology in Warsaw (1998), University of California, San Diego (1999) and held six seminars in different cities of Germany on invitation of the Technical University of Munich (1999).
Collective behaviour of the interwell excitons in biased GaAs/AlGaAs double quantum wells

V. B. Timofeev†, A. V. Larionov†, J. M. Hvam‡ and C. B. Soerensen‡
† Institute of Solid State Physics, 142432 Chernogolovka, Russia
‡ Microelectronics Center, DK 2800 Lyngby, Denmark

Photoluminescence (PL) and photoluminescence excitation spectra of the interwell excitons of GaAs/AlGaAs double quantum wells (n-i-n structures) with a thin AlAs barrier (four monolayers) have been investigated under applied electrical bias. In the limit of a rather weak excitation power and low enough temperatures interwell excitons are strongly localized on the random potential fluctuations and exhibit in photoluminescence spectra an inhomogeneously broaden line (FWHM of 3–4 meV) related with a laterally fluctuating potential relief. Under resonant photoexcitation of 1s HH-intrawell excitons by circularly polarized light we have observed a significant narrowing of PL interwell exciton line (down to 1 meV) on excitation power. Simultaneously we have found a superlinear growth of PL intensity and a sharp increase of the circular polarization degree of the corresponding interwell exciton line. Besides of optical orientation of interwell exciton by resonant circularly polarized resonant excitation we have found a sharp increase of the optical dipole moment alignment of the interwell excitons by linearly polarized resonant photoexcitation. All above described phenomena are very sensitive to the temperature and are observed only at $T < 5 \text{ K}$. Same behavior of the interwell radiative recombination, — narrowing of the interwell exciton line, its critical behavior on temperature and significant increase of the corresponding radiative decay rate, — we have observed under picosecond laser photoexcitation and by consequent analysis of the time evolution of PL spectra.

Described phenomena are associated with the creation of a collective excitonic phase. Dielectric excitonic nature of this new phase is confirmed by its diamagnetic behavior and by Zeeman splittings in magnetic field parallel to the structure growth direction (Faraday geometry). Space coherence of collective excitonic phase is investigated and discussed.
InGaAsN/GaAs heterostructures for long-wavelength light-emitting devices

H. Riechert, A. Yu. Egorov†‡, D. Livshits†, B. Borchert and S. Illek
Infineon Technologies AG, Corporate Research, D-81730 München, Germany
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ guest scientist with Infineon Corporate Research

Abstract. We report on the growth and properties of InGaAsN/GaAs heterostructures and on their applications for lasers emitting at \( \lambda \approx 1.3 \mu m \). Structures are grown by molecular beam epitaxy using an RF plasma-source. Broad area and ridge waveguide laser structures based on such QWs exhibit performance that can compete with those of 1.3 \( \mu m \) InGaAsP lasers. In particular, we have achieved 300 K operation of broad area lasers at 1.3 \( \mu m \) with threshold current density down to 400 A/cm\(^2\) and 650 A/cm\(^2\) for single and triple QW structures. Similar structures with heatsinking at 10\(^{°}\)C yield maximum output powers of 2.4 W (cw) and 4 W (pulsed). Ridge waveguide lasers have thresholds down to 16 mA and show cw operation up to 100\(^{°}\)C with a \( T_0 \) of up to 110 K.

1. Introduction

InGaAsN has recently been proposed as a novel material for near-infrared lasers on GaAs and pioneering laser results were reported by Kondow et al [1]. When replacing the wells in InGaAs/GaAs QWs, the quaternary GaInNAs alloy allows both strain compensation and a significant decrease of the ground state transition energy. Also, an increased conduction band offset has been predicted, which should greatly improve high temperature laser performance.

Most attractively, the realisation of 1.3 \( \mu m \) GaInNAs vertical cavity surface emitting lasers (VCSELs) on GaAs is expected to be possible by adopting the well-established fabrication techniques for short-wavelength VCSELs based on GaAs.

In this paper we will review essential aspects of InGaAsN growth by MBE, discuss properties of InGaAsN/GaAs QWs and report state-of-the-art results on lasers which we have realised in this material system.

2. Material growth

QW test structures and laser structures were grown by solid source MBE on (001) GaAs substrates. An RF-coupled plasma source was used to generate reactive nitrogen from \( N_2 \). Growth proceeds much like that of InGaAs, in particular, we use As-stable conditions. It is found that at growth temperatures below 520\(^{°}\)C, all reactive nitrogen is completely incorporated. Thus, the N-content is only determined by the N-flux.

The experiments on growing InGaAsN heterostructures show that one of the more significant growth factors is the growth temperature [2]. The brightest and narrowest PL spectra are obtained for growth below 450\(^{°}\)C. As reported before [3], we find that InGaAsN heterostructures are very susceptible to post-growth heat treatment. Only by annealing at temperatures around 700 to 750\(^{°}\)C do we obtain a luminescence efficiency which is sufficient for high quality laser material. However, during this process, the PL peak shifts by up to
50 meV towards higher energies for strained InGaAsN QWs. Compared to this, GaAsN and InGaAsN lattice-matched to GaAs show a peak shift of less than 15 meV after the same annealing procedure. It should be noted that strained N-free InGaAs QWs do not show any significant shift in PL energy. We therefore interpret these results by a nitrogen-induced out-diffusion of In from the quantum well region.

Thermally annealed, 7 nm thick InGaAsN/GaAs QWs with In- and N-contents of about 35% and 1.7%, respectively, allow to achieve 300 K luminescence at 1.3 µm. Increasing either the N-content or the QW thickness leads to a significant reduction of luminescence intensity, such that the realisation of devices emitting at still longer wavelengths appears to be difficult.

An analysis of the PL and PLE of InGaAsN/GaAs QWs of different thicknesses in [4] has shown that (i) the confinement energy for electrons in these structures is about 400 meV due to the fact that the conduction band discontinuity amounts to 80% the band gap difference and (ii) that there is clear evidence for a strongly increased electron mass compared to InGaAs with the same In-content, as has been predicted in [5]. Both facts promise an excellent high temperature performance of lasers based on InGaAsN/GaAs QWs.

3. Device application

Laser structures were grown in the optimal temperature range and annealed during the growth of the upper cladding layer. The layer sequence was chosen to be similar to those in Ref. [1]. In case of 3 QW lasers, three In$_{0.35}$Ga$_{0.65}$As$_{0.98}$N$_{0.017}$ QWs of 6–7 nm thickness, separated by 20 nm barrier layers, are symmetrically inserted into a 300 nm thick, undoped GaAs cavity. Best laser results were obtained by using quaternary InGaAsN barriers with the same N-content, which were lattice-matched to GaAs. The p- and n-type cladding layers consist of 1.5 µm thick Al$_{0.3}$Ga$_{0.7}$As, doped with Be and Si to $4 \times 10^{17}$ and $5 \times 10^{17}$ cm$^{-3}$, respectively. The 0.6 µm thick p-type GaAs contact layer is doped to $(1-5) \times 10^{19}$ cm$^{-3}$ in the top 200 nm. Broad area lasers were fabricated by metallisation and subsequent wet-chemical etching of the p-contact layer.

3.1. Broad area lasers

For the assessment of material quality, broad area lasers (fabricated by the shallow mesa stripe technology, width 100 µm) were characterized. The emission wavelength of all lasers referred to here is around 1.29 µm. The threshold current density of 3 QW lasers is found to decrease from 1 kA/cm$^2$ to less than 0.7 kA/cm$^2$ for cavity lengths increasing from 400 µm to 1.2 mm. The lowest threshold for $L = 800$ µm is 650 A/cm$^2$. Using heatsinking at 10°C, cw operation could be demonstrated for these lasers with record output powers of 2.4 W [6], which is by far the highest value ever reported so far for any wavelength in the GaInNAs material system.

Further evaluation of these lasers leads to estimated values of 81% for the internal quantum efficiency and of 10 cm$^{-1}$ for the internal waveguide losses. By using these values and assuming in a first approximation, that the radiative and non-radiative (Auger) recombination coefficients for the GaInNAs QWs are roughly the same as for 1.3 µm InGaAsP QWs (which remains to be verified in future work) we attempted to extract the gain parameters of the GaInNAs QWs [7]. The constants $g_0$ and $N_0$ refer to the empirical gain-carrier-density relationship $g = g_0 \ln(N/N_0)$, which is used to describe the gain saturation in QWs. Values of 2800 cm$^{-1}$ for $g_0$ and $2.4 \times 10^{18}$ cm$^{-3}$ for $N_0$ are obtained for our InGaAsN lasers. Both values are significantly higher than those for 1.3 µm
InGaAsP QWs where the corresponding values for $g_0$ and $N_{tr}$ are found to be $1545 \text{ cm}^{-1}$ and $1.45 \times 10^{18} \text{ cm}^{-3}$, respectively. The higher values for $g_0$ and $N_{tr}$ in GaInNAs are considered to be a consequence of the heavier electron mass in this material [4, 5] ($m_e$ around $0.1 m_0$, $m_0$ is the free electron mass).

3.2. Ridge waveguide (RWG) lasers

Narrow-stripe RWG lasers were processed by using Ar ion dry etching technique for the ridge formation. Stripe widths of 3.5 $\mu$m were realised, passivated with RF-sputtered SiNx. After conventional p- and n-contact formation chips were mounted epi-side up on copper heatsinks for detailed characterization.

The pulsed light-current characteristics of 350 $\mu$m long as-cleaved RWG laser diodes at room temperature show threshold currents as low as 21 mA as well as efficiencies of 0.25 W/A per facet. To the authors’ knowledge both values represent improvement factors of $>2$ and 1.5, respectively, as compared to previously published results [8]. At 90°C the threshold current increases above 50 mA but even at 100°C lasing operation could be maintained. As above, the emission wavelength is around 1290 nm at room temperature.

![Fig. 1. Temperature dependence of the threshold currents of as-cleaved ridge-waveguide lasers of two different lengths.](image)

The temperature dependence of the threshold current is shown in Fig. 1 for 350 $\mu$m and 700 $\mu$m long as-cleaved devices. The corresponding values for $T_0$ are 80–90 K and around 110 K, respectively. These values compare favorably to those of 1.3 $\mu$m InGaAsP RWG lasers, where values around 70 K are typical.

The performance of 2 QW devices with a one-side highly reflection (HR)-coating (reflectivity $\sim 75\%$) was also investigated [9]. Threshold currents of only 16 mA and differential quantum efficiencies of 0.35 W/A were measured at 25°C, while at 80°C the corresponding values are 33 mA and 0.25 W/A, respectively.

First measurements of differential gain of InGaAsN QW lasers were performed and yield values similar to InGaAsP lasers ($dg/dN = (5 \pm 1) \times 10^{-16} \text{ cm}^2$). This comparison indicates that also InGaAsN-based 1.3 $\mu$m LDs will be suitable for use as transmitters in high speed transmission systems.

Also, first lifetime tests at accelerated aging conditions (operation at 80°C with a current of 100 mA cw, corresponding to a current density of 6 kA/cm²) show no noticeable degradation of the threshold current after more than 700 h.
4. Conclusion

In summary, we have demonstrated low threshold current density CW operation of MBE-grown InGaAsN lasers at wavelengths of about 1.3 µm. Their performance is comparable to InGaAsP lasers emitting at the same wavelength, but they have the advantage of a significantly enhanced $T_0$. The combination of the active region used in present work with GaAs/AlAs DBR-mirrors is expected to lead to novel vertical cavity lasers for optical fiber communication systems.

Acknowledgement

We gratefully acknowledge the collaboration of D. Bernklau, M. Komainda and M. Schuster. A. Yu. E. is supported by an Alexander-von-Humboldt fellowship. Our work was partly funded by the EU under BriteEuram BRPR-CT98-0721 (OPTIVAN).

References

Theory of threshold characteristics of quantum dot lasers

L. V. Asryan and R. A. Suris
Ioffe Physico-Technical Institute, St Petersburg, Russia
e-mail: asryan@theory.ioffe.ru, http://www/Dep,TM/asryan.html

Abstract. A theory of the gain and threshold current of a semiconductor quantum dot (QD) laser has been developed which takes account of the line broadening caused by fluctuations in QD size. Expressions for the threshold current versus the surface density of QDs, QD size dispersion and total losses have been obtained in explicit form. Optimization of the structure has been carried out, aimed at minimizing the threshold current density. The characteristic temperature of QD laser has been calculated considering carrier recombination in the optical confinement layer and violation of the charge neutrality in QDs.

Quantum dot (QD) lasers are of particular interest because of the following expected advantages over the conventional quantum well lasers: the narrower gain spectra, significantly lower threshold currents and the weaker temperature dependence of the latter [1]. As a consequence of quantum confinement in all the three dimensions, the energy spectra of carriers in QDs are discrete. For this reason, structures with QDs have generated much interest as a new class of artificially structured materials with tunable (through varying the composition and size) energies of discrete atomic-like states that are ideal for use in laser structures.

Here, we briefly review the theory of the threshold current of a QD laser, developed in [2–6].

Equilibrium or nonequilibrium filling of carrier levels in QDs has been shown to be realized depending on temperature $T$, QD sizes and conduction and valence band offsets at the QD–optical confinement layer (OCL) heteroboundary $\Delta E_{c,v}$ [2, 3].

If the characteristic times of thermally excited escapes of an electron and hole from a QD are small compared with the radiative lifetime in QDs, $\tau_{QD}$, redistribution of carriers from one QD to another occurs, and quasi-equilibrium distributions are established with the corresponding quasi-Fermi levels. As a consequence of such a redistribution, the level occupancies (and numbers of carriers) in various QDs will differ.

The condition for the equilibrium filling of QDs may be written as $T > T_g$ where

$$T_g = \max \left( \frac{\Delta E_c - \varepsilon_n}{\ln(\sigma_n v_n N_c \tau_{QD})}, \frac{\Delta E_v - \varepsilon_p}{\ln(\sigma_p v_p N_v \tau_{QD})} \right).$$

(1)

Here $\varepsilon_{n,p}$ are the quantized energy levels of an electron and hole in a mean-sized QD (measured from the corresponding band edges), $\sigma_{n,p}$ the cross sections of electron and hole capture into a QD, $v_{n,p}$ the thermal velocities, and $N_{c,v}^{OCL}$ the conduction- and valence-band effective densities of states for the OCL material.

The peak modal gain appearing in the threshold condition is

$$g = \frac{\xi}{4} \left( \frac{\lambda_0}{\sqrt{\varepsilon}} \right)^2 \frac{1}{\tau_{QD}} \frac{\hbar}{(\Delta \varepsilon)_{\text{inhom}}} \frac{\Gamma}{a} N_S (f_n + f_p - 1).$$

(2)
where \( f_{n,p} \) are the electron and hole level occupancies averaged over the QD ensemble, \( \xi \) a numerical constant appearing in QD-size distribution function (\( \xi = 1/\pi \) and \( \xi = 1/\sqrt{2\pi} \) for the Lorentzian and Gaussian functions, respectively), \( \lambda_0 \) the wavelength at the maximum gain, \( a \) the mean size of QDs, \( \Gamma \) the optical confinement factor in a QD layer (along the transverse direction in the waveguide), \( N_S \) the surface density of QDs and \( (\Delta \epsilon)_{\text{inhom}} \), the inhomogeneous line broadening due to the QD-size dispersion.

The current density is

\[
j = eN_S f_n f_p + e\beta B n_1 p_1 \frac{f_n f_p}{(1 - f_n)(1 - f_p)} \tag{3}\]

where \( b \) is the OCL thickness, and \( B \) is the radiative constant for the OCL,

\[
n_1 = N_c^{\text{OCL}} \exp\left(-\frac{\Delta E_c - \epsilon_n}{T}\right), \quad p_1 = N_c^{\text{OCL}} \exp\left(-\frac{\Delta E_v - \epsilon_p}{T}\right). \tag{4}\]

If the characteristic times of thermally excited escapes of carriers from a QD are large compared with the radiative lifetime in QDs (relatively low temperatures, \( T < T_g \)), the redistribution of carriers from one QD to another and establishment of quasi-Fermi levels for the conduction and valence bands do not occur; in this case, nonequilibrium filling of QDs is realized. Having no time to leave a QD, the carriers recombine there. Since the initial numbers of carriers injected into various QDs are the same, the QD level occupancies are also the same. The contribution of each QD to the lasing is the same. In this case, too, the peak modal gain is given by (2) wherein the level occupancies common to all QDs appear. The current density is given by

\[
j = eN_S \frac{f_n f_p}{\tau_{\text{QD}}} + eB n_1 p_1 \frac{f_n^2 f_p^2}{\sigma_n \sigma_p n_v p_v \tau_{\text{QD}}^2 (1 - f_n)(1 - f_p)} \tag{5}\]

In (3) and (5), the first and second terms are the current densities associated with the spontaneous radiative recombination in QDs and in the OCL, respectively.

With (2) and the threshold condition \( g = \beta \) where \( \beta \) is the total loss coefficient, the population inversion in QDs required for lasing may be written as

\[
f_n + f_p - 1 = \frac{N_S^{\text{min}}}{N_S} \tag{6}\]

where \( N_S^{\text{min}} \) is the minimum tolerable surface density of QDs required to attain lasing at given loss \( \beta \) and inhomogeneous line broadening \( (\Delta \epsilon)_{\text{inhom}} \) [2]–[4, 6]:

\[
N_S^{\text{min}} = \frac{4}{\xi} \left(\frac{\sqrt{\pi}}{\lambda_0}\right)^2 \frac{\tau_{\text{QD}} (\Delta \epsilon)_{\text{inhom}}}{\hbar} \frac{a}{\Gamma}. \tag{7}\]

The mean level occupancies in QDs are related to each other by (6). The second equation relating \( f_n \) to \( f_p \) should be derived from the solution of the corresponding self-consistent problem for the electrostatic field distribution across the junction and depends on the QD laser design [4].

The dependence of \( j_{th} \) on \( N_S \) is nonmonotonic (Fig. 1(a)). In the case of equilibrium filling of QDs, whatever the specific type of the second equation relating \( f_n \) to \( f_p \) is, the
minimum threshold current density has been shown to be [3, 4]

\[ j_{th}^{\text{min}} = \left[ \left( \frac{eN_{S}^{\text{min}}}{\tau_{QD}} \right)^{1/2} + (ebn_{1}p_{1})^{1/2} \right]^{2}. \]  

(8)

- **Fig. 1.** Threshold current density versus (a) the normalized surface density of QDs, (b) RMS of relative QD size fluctuations and (c) cavity length.

In the special case of a symmetric structure \((f_n = f_p)\),

\[ f_{n,p} = \frac{1}{2} \left( 1 + \frac{N_{S}^{\text{min}}}{N_S} \right). \]  

(9)

The electron and hole level occupancies at the lasing threshold may be expressed as a function of the root mean square (RMS) of relative QD size fluctuations \(\delta\) or the cavity length.
length $L$ as follows:

$$f_{n,p} = \frac{1}{2} \left( 1 + \frac{\delta}{\delta_{\text{max}}} \right), \quad f_{n,p} = \frac{1}{2} \left( 1 + \frac{L_{\text{min}}}{L} \right). \quad (10)$$

All other parameters of the structure being constant, $\delta_{\text{max}}$ and $L_{\text{min}}$ are the maximum tolerable RMS of relative QD size fluctuations and the minimum tolerable cavity length at which lasing is possible. For such $\delta$ or $L$, the surface density of QDs is equal to its minimum tolerable value $N_{s_{\text{min}}}$. 

As $N_{s} \rightarrow N_{s_{\text{min}}}$, or $\delta \rightarrow \delta_{\text{max}}$, or $L \rightarrow L_{\text{min}}$, the mean electron and hole level occupancies in QDs tend to unity ($f_{n,p} \rightarrow 1$), which demands infinitely high free-carrier densities in the OCL. As a result, $j_{th}$ increases infinitely (Figs. 1(a)–1(c)).

As $\delta \rightarrow 0$, or $L \rightarrow \infty$ ($\beta \rightarrow 0$), $j_{th}$ decreases and approaches the transparency current density (Figs. 1(b) and 1(c)).

Ideally, the $j_{th}$ of a QD laser must be temperature-independent and the characteristic temperature, $T_{0} = (\partial \ln j_{th}/\partial T)^{-1}$, must be infinitely high [1]. This would be so indeed if the overall injection current went entirely into the radiative recombination in QDs and the charge neutrality in QDs were the case [5, 6]. In fact, because of the presence of free carriers in the OCL, a fraction of the injection current is wasted therein. This fraction goes into the recombination processes in the OCL (the second term in (3) and (5)).

In the case of nonequilibrium filling of QDs ($T < T_{g}$), the threshold current is essentially temperature-independent. More precisely, there is a weak temperature dependence of $j_{th}$ due to the temperature dependence of the cross sections of carrier capture into a QD $\sigma_{n,p}$, thermal velocities $v_{n,p}$ and radiative constant $B$ (see (5)).

In the case of equilibrium filling of QDs ($T > T_{g}$), the current component associated with the recombination in QDs (the first term in (3)), $j_{QD}$, would be temperature-dependent. Examination of the problem shows [4]–[6] that the electron and hole level occupancies in QDs at the lasing threshold, $f_{n}$ and $f_{p}$, become temperature-dependent if the violation of the charge neutrality in QDs is taken into account properly. Thus, correct consideration of the QD charge reveals the $T$-dependence of $j_{QD}$.

The characteristic temperature of a QD laser, $T_{0}$, can be represented as [5, 6]

$$\frac{1}{T_{0}} = \frac{j_{QD}}{j_{QD} + j_{OCL}} \frac{1}{T_{0}^{QD}} + \frac{j_{OCL}}{j_{QD} + j_{OCL}} \frac{1}{T_{0}^{OCL}} \quad (11)$$

where $T_{0}^{QD}$ and $T_{0}^{OCL}$ are defined similarly to $T_{0}$ for the functions $j_{QD}(T)$ and $j_{OCL}(T)$, respectively: $1/T_{0}^{QD} = \partial \ln j_{QD}/\partial T$ and $1/T_{0}^{OCL} = \partial \ln j_{OCL}/\partial T$.

Hence, the reciprocal of $T_{0}$ is a sum of the reciprocals of $T_{0}^{QD}$ and $T_{0}^{OCL}$, each weighted by the relative contribution of the respective component of $j_{th}$.

The $T$-dependences of $f_{n,p}$ are much weaker compared to that of the exponential in (4). Consequently, $j_{QD}$ increases with $T$ much more slowly than $j_{OCL}$ does (Fig. 2). Hence, $T_{0}^{QD}$ is much greater than $T_{0}^{OCL}$. Nevertheless, as it can be seen from (11), $1/T_{0}$ is controlled not only by $1/T_{0}^{QD}$ and $1/T_{0}^{OCL}$, but by the relative contributions of the threshold current density components, $j_{QD}/j_{th}$ and $j_{OCL}/j_{th}$, as well. For this reason, under temperature
Fig. 2. Threshold current density and its components versus the temperature for $N_S = 7.7 \times 10^{10} \text{cm}^{-2}$. The inset shows $j_{\text{QD}}(T)$ and $j_{\text{OCL}}(T)$ on an enlarged (along the vertical axis) scale. The broken line depicts $j_{\text{QD}}$ calculated assuming the charge neutrality in QDs.

Fig. 3. Characteristic temperature $T_0$ versus (a) the normalized surface density of QDs, (b) RMS of relative QD size fluctuations $\delta$ (for $\beta = 10 \text{cm}^{-1}$, bottom axis) and the total loss $\beta$ (for $\delta = 0.05$, top axis) for $N_S = 1.3 \times 10^{11} \text{cm}^{-2}$ and (c) temperature for $N_S = 7.7 \times 10^{10} \text{cm}^{-2}$. The broken curves depict $T_0$ calculated assuming the charge neutrality in QDs.
conditions wherein \( j_{th} \) is controlled by \( j_{QD} \) (Fig. 2), the contribution of the first term in the right-hand side of (11) is every bit as important as that of the second term.

For \( N_S \) fairly greater than \( N_{S_{\text{min}}} \), \( T_0 \) increases with \( N_S \) (Fig. 3(a)). The point is that the less temperature-sensitive component of \( j_{th} \), i.e., \( j_{QD} \), increases with \( N_S \), whereas the more temperature-sensitive component of \( j_{th} \), i.e., \( j_{OCL} \), decreases.

The greater the RMS of relative QD size fluctuations \( \delta \) or the total loss \( \beta \) (i.e., the less perfect the structure), the lower \( T_0 \) at given \( T \) and given other parameters (Fig. 3(b)).

The characteristic temperature depends strongly on \( T \); \( T_0 \) falls off profoundly with increasing \( T \) (Fig. 3(c)). A drastic decrease in \( T_0 \) occurs in passing from the temperature conditions wherein \( j_{th} \) is controlled by radiative recombination in QDs (Fig. 2) to those under which \( j_{th} \) is controlled by radiative recombination in the OCL (Fig. 2). The \( T_0 \) values at \( T = 200 \) and 300 K are 582 and 128 K, respectively (Fig. 3(c)).

We emphasize that the tendency for \( T_0 \) to decrease drastically with \( T \) seems to be in line with the available experimental results [7].

As Fig. 3(c) suggests, at relatively low \( T \) (when \( j_{th} \) is controlled by \( j_{QD} \)), the actual \( T_0 \) differs significantly from that calculated assuming the charge neutrality in QDs.

As our example, we use a GaInAsP/InP heterojunction structure lasing at 1.55 \( \mu \text{m} \) [3]–[6]. A device with OCL thickness of \( b = 0.28 \mu \text{m} \) and an as-cleaved facet at both ends is considered. A Gaussian distribution of the relative QD size fluctuations is assumed. The mean size of cubic QDs is taken to be 150 Å. The surface density of QDs, RMS of relative QD size fluctuations, cavity length, and temperature are taken to be \( N_S = 6.1 \times 10^{10} \text{cm}^{-2} \), \( \delta = 0.025 \text{ (5\%)} \), \( L = 500 \mu \text{m} \), and \( T = 300 \text{ K} \), respectively, unless otherwise specified. The corresponding values of the minimum tolerable surface density of QDs, maximum tolerable RMS of relative QD size fluctuations, and minimum tolerable cavity length required to attain lasing are \( N_{S_{\text{min}}} = 2.1 \times 10^{10} \text{cm}^{-2} \), \( \delta_{\text{max}} = 0.074 \text{ (14.8\%)} \), and \( L_{\text{min}} = 170 \mu \text{m} \), respectively.

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Properties of wide mesastripe InGaAsP heterolasers

Ioffe Physico-Technical Institute, St Petersburg, Russia
e-mail: tarasov@hpld.ioffe.rssi.ru

Abstract. InGaAsP/InP wide mesastripe laser diodes ($\lambda = 1.3–1.55 \mu m$) grown by metal-organic chemical vapour deposition (MOCVD) method have been fabricated. Light-current characteristics and emitting spectra under pulse and continuous-wave (CW) operation have been investigated in 10–60°C temperature range. Laser diode active region overheating of 30–60 K in respect to the copper heatsink at maximum CW drive currents has been determined. Strong influence of the external differential quantum efficiency temperature dependence on CW maximum output power has been established. Optical output powers of 3 W and 2.6 W under CW operation, 9 W and 6.5 W under pulse operation have been reached for a single 100 $\mu m$-wide aperture mesastripe InGaAsP/InP laser diodes emitting at 1.3 $\mu m$ and 1.55 $\mu m$ wavelength, respectively.

Introduction

At present time there is a great interest in laser radiation with high output power. Record-high CW optical output power of 11 W has been reached for 0.98 $\mu m$ laser diodes (LDs) [3–5]. 5 W CW output power in InGaAsP/InP 1.48 $\mu m$ diode lasers with 200 $\mu m$ mesastripe width has been achieved [11]. Such discrepancy in the record values of output power is mainly explained by the difference in the energy band structure of AlGaAs/GaAs and InGaAsP/InP solid solutions [8].

In this paper we investigate the properties of MOCVD-grown InGaAsP/InP separate confinement LDs [9] with the aim to determine the primary factors limiting maximum optical output power.

1. Experimental samples

Separate confinement heterostructure with two strained quantum well active layers was the basic structure for LD fabrication [3–5, 9]. The waveguide thickness of 0.9 $\mu m$ was chosen. Further increase of the waveguide thickness is not reasonable due to high-order optical modes lasing [5]. The waveguide doping level and free carrier concentration in p-type and n-type cladding layers were 10$^{16}$ cm$^{-3}$ and 10$^{17}$ cm$^{-3}$ respectively, as it further reduction would lead to the increase of structure series resistance. Solid solution composition of the waveguide layer was chosen to provide 4$kT$ quantum well depth for electrons. The waveguide bandgaps were defined as 1.25 eV and 1.1 eV for laser heterostructures emitting at 1.3 $\mu m$ and 1.55 $\mu m$ wavelength, respectively.

2. Experimental results and discussion

Light-current characteristics in pulse regime were investigated at 2 $\mu s$ pulse duration. In this case a slight active region overheating is observed only at pump currents higher than 10 A in LDs with 1.5–2 mm cavity length (Fig. 1(a)). However, pulsed optical output power
Fig. 1. (a) CW (curves 1, 3) and pulse (curves 2, 4) output power versus current at 10°C heatsink temperature for LDs emitting at 1.3 \( \mu \)m wavelength \((L = 2 \text{ mm}; \text{curves } 1, 2)\) and 1.55 \( \mu \)m wavelength \((L = 1.2 \text{ mm}; \text{curves } 3, 4)\). (b) CW (curve 1) and pulse (curves 2–8) light-current characteristics for 1.55 \( \mu \)m LD with 500 \( \mu \)m cavity length. Heatsink temperature: 1, 2—11°C, 3—14°C, 4—20°C, 5—34°C, 6—41°C, 7—50°C, 8—56°C.

of 9 W and 6.5 W were reached in laser diodes emitting at 1.3 \( \mu \)m and 1.55 \( \mu \)m wavelength, respectively.

Strong active region overheating resulting in the saturation of light-current characteristics was observed for LDs operating in CW regime (Fig. 1(b)). Nevertheless CW optical output power of 3 W and 2.5 W were obtained in long cavity \((L = 1.5–2 \text{ mm})\) 100 \( \mu \)m-wide mesastripe LDs emitting at 1.3 \( \mu \)m and 1.55 \( \mu \)m wavelength, respectively (Fig. 1(a)). The obtained results are comparable with record output power values of this wavelength range [11].

The temperature of LDs active region was determined in two different ways. The comparison of light-current characteristics of laser diode measured under CW operation at 10°C heatsink temperature and under pulse operation at different temperatures higher than 10°C was carried out (Fig. 1(b)). In the point of intersection of light-current curves the active region overheating was determined as the difference between temperature values of the heatsink. In the second method the active region overheating was calculated from the difference of emitting spectra peak positions under pulse and CW operation.

The value of active region overheating determined by both methods coincided with ±2 K accuracy and was 30–60 K at maximum CW drive currents depending on LD cavity length.

It is necessary to point out, that in the investigated samples we succeeded to reduce the reciprocal series resistance of laser heterostructure down to \(10^{-4} \Omega/\text{cm}^2\).

The dependence of characteristic temperature \(T_0\) on LD cavity length is shown in Fig. 2. This dependence is typical for LDs based on conventional separate confinement heterostructures with thin active region [11]. Even in short cavity LDs in the 30–40 K temperature range the influence of threshold current increase on maximum output optical power is not essential in comparison with the external differential quantum efficiency \((\eta_d)\) temperature dependence (Fig. 1(b)), that coincides with [6].

The dependence of characteristic temperature \(T_1\) on LD cavity length (Fig. 2) has an opposite behaviour compared to the \(T_0\) dependence. \(T_1\) parameter characterizes the tem-
perature dependence of $\eta_d$ [7]. Low value of $T_1$ parameter 140–180 K in LDs on the base of InGaAsP/InP solid solutions is an essential point. The result is drastic decrease of $\eta_d$ with active region overheating observed for both short cavity (Fig. 1(b)) and long cavity (Fig. 1(a)) InGaAsP/InP LDs, substantially limiting maximum output power. In GaAs-based LDs $T_1$ parameter reaches 300–1600 K values [7].

The $1/\eta_d(L)$ dependencies plotted in the $10–50^\circ C$ temperature range allowed to determine temperature dependencies of internal optical losses ($\alpha_i$) and internal quantum efficiency ($\eta_i$) (Fig. 3). To reduce the influence of $\alpha_i$ temperature dependence on $\eta_d$ is possible only by decreasing the $\alpha_i$ value. $\eta_i$ value quantitatively characterizes the presence of carrier leakage and nonradiative recombination channels in heterostructure above threshold. To reduce the influence of $\eta_i$ temperature dependence on $\eta_d$ is possible by decreasing the carrier leakage and nonradiative recombination channels.

**Conclusion**

InGaAsP/InP wide mesastripe lasers have been fabricated and studied. Optical output powers of 3 W and 2.6 W under CW operation, 9 W and 6.5 W under pulse operation have been obtained for 1.3 $\mu$m and 1.55 $\mu$m wavelength, respectively. Under CW operation LD active region overheating of 30–60 K in respect to the copper heatsink has been determined. It has been established, that along with internal optical losses and series resistance the
temperature dependence of the external differential quantum efficiency strongly affects CW maximum output power. Due to low values of $T_1$ parameter this influence is more dramatic for InGaAsP/InP laser diodes. To limit the negative influence of $T_1$ is possible only by reducing the internal optical losses and carrier leakage in laser heterostructure.

References

Anisotropy of light emission from the surface LED based on the type-II ZnSe/BeTe heterojunction

A. V. Platonov†, V. P. Kochereshko†, E. L. Ivchenko†, D. R. Yakovlev‡, G. Reuscher‡, W. Ossau‡, A. Waag‡ and G. Landwehr‡

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physikalisches Institut der Universität Würzburg, 97074 Würzburg, Germany

Abstract. Electroluminescence (EL) spectra are studied in the type-II ZnSe/BeTe light emitting diode (LED) based on a single heterojunction. The light emitted from the surface exhibits a strong in-plane linear polarization along [1\(\bar{1}\)0] crystal axis. The polarization is stable in respect to an increase of the applied voltage and temperature up to 300 K. The experimental data are discussed in the framework of a tight-binding model taking into account a type-II band alignment and lack of common atom in the ZnSe/BeTe heterosystem.

Introduction

It has been shown recently that a single heterojunction on the (001) atomic plane between two zinc-blend semiconductors has to reveal an optical anisotropy in the interface plane due to the lack of the point symmetry of the bulk materials. The effect was demonstrated for number of type-I heterostructures [1]. The experimental results and theoretical calculations show that in the type-I structures the optical anisotropy may reach 20% in absorption and emission spectra. In a recent time similar effects have been observed for symmetrical quantum well structures under external electric field [2] for semiconductors system with type-I band alignment. The effect was called Quantum Confined Pockels Effect.

Semiconductor heterostructures with type-II band alignment, where electrons and holes are separated spatially in the alternating layers, have obvious advantages for the optical study of interface-related phenomena. In our papers [3, 4] we studied spatially indirect transitions in the type-II ZnSe/BeTe heterostructures such as superlattices and/or double-barrier structure and found that in this case the PL spectra are almost totally linearly polarized. In the present paper we perform further investigations of the optical transitions at the heterointerface ZnSe/BeTe. In particularly we measured polarization characteristics of the photoemitting diode based on the single type-II heterojunction ZnSe/BeTe.

Experimental details and results

The type-II heterosystem ZnSe/BeTe allows to fabricate a new type of light emitting diode (LED) which contains only a single ZnSe/BeTe interface. The emission of such a device can be adjusted between 640 nm and 515 nm [5]. The light emission occurs because of a spatially indirect carrier recombination between the ZnSe conduction band and BeTe valence band. Normally such devices based on a type-II heterostructures possess a very low radiative efficiency. However due to the strong carrier confinement at the interface at which the recombination of electrons and holes occurs efficiencies of 0.5% were achieved.

The devices were grown by molecular-beam epitaxy on p-type doped GaAs (100) substrates, which were covered by a thin GaAs buffer layer. In order to reduce the stacking
Fig. 1. Sketch of the band structure of a ZnSe/BeTe type-II LED. Fault density, 4 monolayers of BeTe were deposited before the growth of the diode. The growth was continued with 25 nm of $5 \times 10^{18}$ cm$^{-3}$ $p$-type BeTe. For the doping we used a nitrogen plasma at 300 W at partial pressure of $3 \times 10^{-5}$ Torr. Then the plasma was switched off and 25 nm undoped BeTe was grown followed by 3 nm of ZnSe acting as a quantum well for electrons. After a spacer of 25 nm undoped BeMgZnSe 500 nm lattice matched $n$-BeMgZnSe doped with ZnI$_2$ to $5 \times 10^{17}$ cm$^{-3}$ resulting in bandgaps ranging from 2.85 eV to 3.35 eV respectively were grown. The structure was completed by 10 nm of $n$-ZnSe with an electron concentration of $10^{19}$ cm$^{-3}$ for the ohmic metal contact. The scheme of the structure is presented in Fig. 1.

Figure 2(a) shows electroluminescence emitted from the surface of a typical LED at room temperature. The emitted light is strongly (is about 70%) linearly polarized in the direction $[1 0 \bar{1}]$. The line-width (FWHM) of the spectra was about 100 meV that is 3 times higher than in conventional ZnSe LED’s with ZnCdSe quantum wells. By narrowing the ZnSe quantum well the emission wavelength can be shifted towards shorter wavelengths, for a well thickness of 1 nm luminescence light of 515 nm is emitted while the polarization degree remains the almost constant. The total output was measured in a Ulbricht sphere. For a voltage of 4 V and a current of 15 mA the power was 0.3 mW, which corresponds to an external quantum efficiency of 0.5%. This high efficiency of optical emission in the type-II structures can be explained by a carrier confinement at the interface. Degradation experiments did not show any decrease of efficiency after 1000 hours. Figure 2(b) demonstrates the angle dependence of the LED emission in polar coordinates.

Discussion

To explain the experimental results in [3, 4] authors assumed that PL signal of the spatially indirect transition related to the certain interface in the type-II ZnSe/BeTe heterostructures is almost totally polarized. Using structures with single heterojunction we clearly experimentally shown that this assumption is correct. In the ZnSe/BeTe system the conduction- and valence-band offsets amount to 2 eV and 1 eV, respectively, and the penetration depth of the electron wavefunction into the BeTe layer or for a hole into the ZnSe layer is about 3 Å.

Therefore, in type-II direct-gap ZnSe/BeTe heterostructures the wavefunctions of an electron and a hole participating in the spatially indirect optical transition overlap substantially only over few atomic planes. In this case the calculation of the interband matrix elements requires the knowledge of the microscopic behavior of the wavefunctions at the interfaces which can be obtained by using the pseudo-potential or tight-binding method.

The calculation based on tight-binding method shows that the strong polarization of
Fig. 2. (a) Surface emitted electroluminescence signal detected in two linear polarizations along the [1\bar{1}0] and [110] crystallographic axis. (b) EL angle dependence, polar coordinates.

the spatially indirect transition at the type-II heterojunction may be explained by the fact that contribution of only one atomic plane dominates in the matrix elements [3, 4]. The polarization of the light radiated by the one atomic plane is totally polarized along the chemical bonds lying at the plane. We mentioned here that so strong localization of the optical transition area is an absolutely new result, which is following from the polarization experiment, described above and developed theory.

We also analyze the influence of the interface disorder on the polarization degree. It may be shown that monomolecular fluctuations on the interface don’t lead to remarkable changes in the polarization. On the other hand, the interfacial disorder connected with admixture of chemical bonds or any rearrangement of chemical bonds at the interfaces should be followed by dramatic variation of the polarization.

The reported effect of the optical anisotropy has a potential to become a powerful tool for investigation of the microscopic structure of heterointerfaces with monolayer resolution by means of non-destructive optical methods. We also mentioned here that the effect play an important role for designing any optical devices based on the type-II optical transition.

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References

Unipolar semiconductor lasers on asymmetric quantum wells

Yu. A. Aleshchenko, V. V. Kapaev, Yu. V. Kopaev and N. V. Kornyakov
P. N. Lebedev Physical Institute of RAS, 117924 Moscow, Russia

Abstract. We propose the original design of an active element of quantum unipolar semiconductor laser both for the optical pumping and current injection modes of operation. The peculiarities of the proposed design are strongly asymmetric barriers surrounding a double-well active element. The suppression of intersubband transitions to the lower working subband can be readily achieved if the transformation point of electronic state dimensionality for the lower subband occurs at small momentum. By this means the population inversion conditions in this system can be easily realized. The results of photoluminescence studies of the individual elements of the proposed structure are presented.

Theory

The double QW structure with asymmetric barriers, which permits to suppress sufficiently the intersubband relaxation, was proposed by us earlier [1]. The design of this structure facilitates the achievement of the intersubband population inversion and consequently the reduction of the threshold pump currents and pumping intensity respectively in the current injection [2] and fountain [3] modes of operation. In this case, the situation of both coherent photonic and electronic subsystems is possible as well as lasing even without the population inversion [4].

The asymmetry of the barriers causes an existence of 2D subband of the corresponding QW only in restricted region of 2D wave vector $0 < |k| < k_c$ because of 2D–3D transformation of electronic state dimensionality. The intersubband relaxation is appreciably suppressed if the distance between the 2D–3D transformation point $k_c$ of the lower subband and bottom of the upper subband exceeds the LO phonon energy $E_{LO}$. We propose here the original design of an active element both for the optical pumping and current injection modes of operation. In Figure 1, the band diagram of the laser structure with optical pumping is shown.

The active element comprises two QW of $h_1$ and $h_2$ width separated by narrow barrier. We use the scheme with pumping between $\varepsilon_1$ and $\varepsilon_3$ subbands and lasing between $\varepsilon_3$ and $\varepsilon_2$ subbands. The peculiarity of the proposed design is the insertion of this active element between the asymmetric barriers $U_0$ and $U_1$ ($U_1 \ll U_0$) where $U_1$ lies close to the $\varepsilon_2$ subband. With such an active element, the transformation of electronic state dimensionality for $\varepsilon_2$ subband allows to increase appreciably the phonon relaxation time $\tau_{32}$ and strengthens the inequality $\tau_{32} \gg \tau_{21}$. The latter is necessary for the population inversion achievement.

Taking into account the finite width $L$ of $U_1$ barrier in the real structure, the active element design was optimized on the basis of calculations of phonon intersubband relaxation times $\tau_{ij}$ and dipole matrix elements $z_{ij}$ ($i, j = 1, 2, 3$). The concrete calculations were performed for the structure on the basis of GaAs/AlGaAs system with a molar fraction of Al in the right barrier $x = 0.35$ and that of $U_1$ barrier $x \sim 0.09$. The QW widths in the active region ($h_1 = 82 \text{ Å}, h_2 = 51 \text{ Å}$) were chosen to obtain the energy gap between
\(\varepsilon_3\) and \(\varepsilon_1\) subbands of the order of 120 meV (for pumping with a CO\(_2\) laser), and \(\varepsilon_2-\varepsilon_1\) separation close to \(E_{LO}\) (\(E_{LO} \approx 36\) meV). In this case, \(\tau_{21}\) takes a minimum and reaches 0.5 ps. The width of separating barrier \(b_1\) was chosen to optimize a \(\eta = (\varepsilon_3 - \varepsilon_2)(z_{31}z_{32})^2\) value, which determined the ratio of the gain in the system to the loss [3]. The product \(z_{31}z_{32}\) has a maximum at \(b_1 \approx 14\) Å.

We have found that the insertion of a narrow barrier \(b_2\) of \(U_0\) height between \(h_2\) QW and \(U_1\) barrier allows one to improve the lasing characteristics. This barrier affects \(\tau_{32}\) and \(\tau_{21}\) values and permits to control \(z_{ij}\) values. The product \(z_{31}z_{32}\) increases steadily with \(b_2\) and at \(b_2 = 20\) Å nearly coincides with that of the structure with symmetric barriers (\(U_1 = U_0\)).

One has to increase \(\tau_{32}\) to obtain the population inversion. In the proposed design this is achieved as a result of the escape of electrons of \(\varepsilon_2\) subband with a finite momentum of longitudinal motion to the region of low barrier \(U_1\). This effect occurs at a reasonable large \(L\). In this case, a quasi-continuous (QC) spectrum is formed in this region and the additional mechanism of the electron escape from the upper laser subband \(\varepsilon_3\) appears. Moreover, with appropriate choose of \(L\) the escape of the electrons from \(\varepsilon_3\) subband to the states of the QC spectrum formed over \(U_1\) barrier appears to be suppressed. The described approach allows one to refuse complex injector design in the form of graded Bragg superlattice for quantum cascade laser [2] and simplifies a change-over from the optical pumping to the injection mode of operation.

We carried out numerical calculations of the \(b_2\) dependences of the relaxation times \(\tau_{31}\) and \(\tau_{32}\) for intersubband transitions with a participation of optical phonons, the escape time of electrons from the upper subband to the region of QC spectrum \(\tau_{3con}\), and the total lifetime \(\tau_{3tot}\) for the \(\varepsilon_3\) subband at \(L = 400\) Å and \(U_1 = 81\) meV. There is a flat maximum of \(\tau_{3con}\) at \(b_2 \sim 30\) Å. As this takes place, the total lifetime \(\tau_{3tot}\) is close to its limiting value, and \(\tau_{32} \approx 4\) ps being 2.7 times as much as that for the structure with symmetric barriers.

Figure 2 shows the dependences of the relaxation times on the width \(L\) of the region forming a QC spectrum. The maxima \(\tau_{31}\) and minima \(\tau_{3con}, \tau_{3tot}\) at \(L \approx 340\) and 420 Å are due to the appearance of the level, which is in resonance with \(\varepsilon_3\), in the region of QC spectrum. In this case, the wave function of \(\varepsilon_3\) state penetrates significantly in the region of low barrier \(L\), causing the decrease in the overlap with wave functions of \(\varepsilon_1\) and \(\varepsilon_2\) states and the increase in the overlap with states of QC spectrum. The abrupt change of \(\tau_{3con}\) at \(L = 350\) Å is associated with the approach of the next level of the QC spectrum to the \(\varepsilon_3\) level by a distance equal to \(E_{LO}\) energy. Thus, the optimal value of \(L\) lies in the region of 400 Å.

In order to optimize the laser on the population inversion \(I\) parameter, we wrote equations that describe non-equilibrium kinetic processes in the structure under study. It was shown that the dependence \(I(L)\) keeps a strong non-monotonous character owing to the
Fig. 2. Dependences of the relaxation times on the width $L$ of the region forming a QC spectrum.

sharp modulation of relaxation rates with $L$ for the various subbands.

The presence of QC subbands decreases the value of population inversion. For example, when a single QC subband $\varepsilon_0$ exists, the population inversion is expressed by the equation

$$I = \frac{\tau_{32}}{\tau_{21}} \left( 1 + \frac{\tau_{32}}{\tau_{30}} \right)^{-1},$$

where $\tau_{ij}$ is the electron relaxation time from $\varepsilon_i$ subband to $\varepsilon_j$ one and $\tau_0$ is a full lifetime in $\varepsilon_0$ subband. Obviously, the top limit of equation (1) is reached, when $\tau_{30} \gg \tau_{32}$ and $\tau_{02} \gg \tau_0$. However, not all the QC subbands participate effectively in electron relaxation from the $\varepsilon_3$ subband to $\varepsilon_2$ one. The transitions to QC subbands separated from $\varepsilon_3$ subband with a gap smaller than $E_{LO}$ are forbidden by the law of energy conservation. If the intersubband relaxation time exceeds the intrasubband time, the electrons are accumulated near the subband edges. In this case, the transitions to the $\varepsilon_2$ subband from the QC subbands separated with a gap smaller than $E_{LO}$ are also prohibited. On these grounds, it is possible to suppress significantly the influence of the QC spectrum on the population inversion.

**Experimental results**

To study the concurrence of the mechanisms of interband radiative recombination and tunneling through the narrow barrier, the photoluminescence (PL) was measured of single QW with symmetric barriers one of them having the width that was varied in the range $d = 3-10$ nm. The unipolar regime under consideration is realized when the tunneling time appears to be lower than the recombination time. It should be stressed that the tunneling time in its turn determines the intersubband relaxation time. When narrowing barrier the possibility to alter the lifetime of coupled electronic state in a broad range was demonstrated. The variation of lifetime between two limiting cases when it was determined by tunneling from QW through the barrier ($d = 40$ Å) and by radiative recombination time ($d = 80$ Å) was realized. It was found that 80 Å barrier effectively prevents tunneling. The intensity of QW peak drastically decreases for the structure with 60 Å barrier. This is an evidence of the key role of the tunneling through 60 Å barrier in this structure. There is no QW
contribution in the PL spectrum of the structure with 40 Å barrier and tunneling through the narrow barrier dominates.

The drastic modification of the degree of electronic-wave-function localization in 2D–3D transformation point was demonstrated from comparison of PL spectra of 40 Å GaAs/AlGaAs single QW with strongly asymmetric barriers and of the similar structures but with the additional wider (60 or 80 Å) QW with symmetric barriers coupled with the initial one. The latter structure is a component of the active element of a fountain laser. We have shown that this structure permits one to increase considerably the degree of electronic wave function localization in QW at $k = 0$ that determines the oscillator strength of the working laser transition with only minor variation of the critical value of wave vector of 2D–3D transformation.

The key feature of the proposed scheme is the resonance of the lower subbands in the asymmetric and symmetric QW. This is achieved by the increase of the width of QW with symmetric barriers. For the structure with 60 Å QW the ground subband of this well is sufficiently higher than the ground subband in asymmetric QW and the resonance and thus the transformation point in this subband are absent. Therefore, the transition to this subband from higher states is not suppressed and the ground subband in symmetric QW is highly populated. This is the reason of high intensity of QW peak. For the structure with 80 Å QW the ground state in the symmetric QW appears to be at lower energy and in resonance with the ground subband in asymmetric well. For this reason the ground subband in this structure has the transformation point and the transitions to this subband are suppressed. That is why the intensity of QW peak for this structure is sufficiently weaker. For the structure with single asymmetric QW the discussed effect is weaker due to the lower degree of localization of electronic wave function.

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References


Injection laser threshold from the standpoint of collective resonance

Ioffe Physico-Technical Institute, St Petersburg, Russia
† Present address: Applied Optoelectronics, Inc., 242 Kingfisher Dr., Sugar Land, TX 77478, USA

Introduction

The phenomenon of collective resonance via electromagnetic field during radiative recombination in semiconductor heterostructures provokes rising interest in recent years [1, 2]. Such many-body interactions take place in a group of dipoles having an average distance of less than the wavelength of their radiation [3]. Mutual influence of dipoles leads to their phasing and, as a result, to the formation of a “macro-dipole” with very high radiation efficiency. For the semiconductor material conditions, the superradiance (SR) pulse duration lies in sub-picosecond range. Such pulses can not be directly measured by any modern photodetector, and were discovered only using optical autocorrelation method [4].

To take into account collective resonance interaction of carriers, we must replace usual time dependence

\[ I(t) = \frac{N}{\tau} e^{-t/\tau} \]

by corresponding formula derived by Dicke [5] describing the time profile of the pulse.

It has been recently shown [6] that a spectrum of electroluminescence (EL) of single quantum well (QW) heterostructure at low temperatures can be very precisely described using formula for homogeneous line broadening by the superradiance effect:

\[ R(\hbar \omega) = A \cdot \text{sech} \left( \frac{\hbar \omega - \hbar \omega_0}{\epsilon} \right) \]  (1)

where characteristic energy \( \epsilon = \hbar / \pi \tau_N \) (can be found from experimental spectra) i.e. the characteristic time \( \tau_N = \hbar / \pi \epsilon \). These spectra must have almost exponential rise and fall. To recognize this property of a spectrum, we have to measure it very precisely and to plot spectra in semi-logarithmic scale.

In this work we would like to show how the model of collective resonance can be applied to explain the most important part of laser operation — transition from the spontaneous radiation to lasing. To do this series of spectral and temporal measurements were carried out.

1. Experimental results and discussion

The separate confinement laser double heterostructure (\( \lambda = 970 \) nm) based on InGaAs/GaAs single quantum well (QW) was grown by molecular beam epitaxy (MBE). EL spectra of samples were studied in wide range of temperatures from 77 K up to 300 K under the quasi-CW pump current (3 \( \mu s \), 5 kHz). Some samples were prepared in special geometry: to avoid positive feedback, all edges of the sample were etched and covered by high refractive index black paint. Pump current dependence of the time coherence (\( \tau_C \)) at RT have been investigated in the same manner as in [4].

EL spectra at 77 K measured through the substrate window under the different pump current are shown in semi-logarithmic scale in Fig. 1. Near the lasing threshold (which was

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**Fig. 1.** Current dependence of the EL spectra measured through substrate window at low temperature (InGaAs/GaAs single QW heterostructure) plotted in semi-logarithmic scale.

**Fig. 2.** EL spectrum through substrate window of InGaAs/GaAs single QW heterostructure plotted in semi-logarithmic scale. Two superradiance components of EL spectrum with different characteristic slope are shown.

**Fig. 3.** Collective radiation pulse duration corresponding to components A and B as a function of pump current density.
approximately 1400 A/cm²) EL spectrum has a complex shape that can be presented as a superposition of two superradiance components with different characteristic slope (Fig. 2).

Our estimate of the corresponding duration of components A and B coming from Eq. (1) is plotted as a function of the pump current in Fig. 3. Characteristic time of the component A is practically constant in wide range of pump current. We suppose that component A describes the superradiance in semiconductor without feedback. Component B corresponds to that part of superradiance pulse, which is changed by the laser cavity. Due to rather narrow gain spectrum amplified SR pulse becomes longer up to 70 fs (Fig. 3). Intensity of A component is so small in comparison with B one that it practically could not be detected using time-resolving investigations.

Full width at half-maximum level (FWHM) of the central peak of time coherence, which should corresponds to superradiance characteristic time, is shown in Fig. 4. Below the lasing threshold these results were compared with the characteristic time (τ_N) which was calculated from the low-energy slope of EL spectra at RT (Fig. 5) and also shown in Fig. 4. According to our experiments τ_c approximately in 2 times lower than τ_N. This difference can be caused by the significant inhomogeneous line broadening. As a result the duration of the single superradiance pulse is longer than one estimated from time coherence pattern.
It should be noticed that both $\tau_N$ and $\tau_c$ practically do not depend on the pump current below the threshold, which is in a good agreement with our previous results [6]. At room temperature inhomogeneous line broadening does not allow to distinguish components of luminescence spectra mentioned above. However, narrow and more intensive component B is clearly presented in time coherence pattern above the threshold. It is important that at RT the characteristic time of SR component B is longer than one at 77 K.

2. Conclusions

Mechanism of laser threshold is considered in terms of superradiance. Presence of short coherent pulses has been demonstrated below, at and above laser threshold in wide temperature range. Existence of positive feedback leads to SR pulse becomes longer. Luminescence lineshape above threshold has a complex structure but is well described using the theory of collective recombination. Spectral investigations at low and room temperature, as well as time coherence measurements at RT allow estimating the characteristic time of superradiance pulse in these two modes. The maximum duration of the pulse has been found as low as 300 fs at RT and 70 fs at 77 K.

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References

Electroluminescence study of green Be-contained II–VI lasers


Ioffe Physico-Technical Institute, St Petersburg, Russia
† Physikalisches Institut der Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

Introduction

The interest to study of II–IV laser diodes (LD) emitting in a visible spectral range is mainly caused by a possibility of their application for record high-density compact disks and for creation full-color projection TV (see, e.g., references in [1]). The recent utilization of the novel design conception of the both active region and the structure geometry led to increasing the lifetime of II–IV lasers. It is mainly owing to (a) spatial separation of the radiative recombination and defect-containing regions and (b) the prevention outside penetration and development of extended and point defect [2]. However, the basic emission parameters (threshold current density, quantum efficiency etc.) also defining the commercial application are on the outside of deep investigations.

In this paper we study the main components of the threshold current of room temperature injection lasers based on BeMgZnSe/ZnCdSe low-dimensional heterostructure and explain the characteristic lasing features observed.

1. Experimental

The laser BeMgZnSe/ZnCdSe structure investigated was grown by molecular beam epitaxy (MBE) pseudomorphically to a GaAs(001) substrate at growth temperature of 270–280°C. The MBE growth and composition control of Be-chalcogenides based heterostructures have been published elsewhere [3]. The active region of the laser diode structure contains a (10Å-Be0.05Zn0.95Se/15Å-ZnSe)82 superlattice (SL) waveguide lattice-matched to GaAs as a whole, centered with a 2.6 ML-CdSe/10 nm-ZnSe nanostructure. Details of structural and optical characteristics of the active region have been given elsewhere [2]. The structure also involves 1 μm-thick wider bandgap n- and p-Be0.05Mg0.95Zn0.05Se cladding layers, doped with iodine and nitrogen, respectively, as well as a top ZnSe/BeTe:N modulation doped graded short-period SL capped with a highly p-doped BeTe:N/ZnTe:N contact structure. The 10 nm-BeTelayer was grown to produce low-resistively ohmic contact.

Broad-area lasers with 20 μm-wide stripes were fabricated by standard techniques. Laser characteristics were studied under pulsed excitation (1 μs pulse duration, 1 kHz repetition frequency) at room temperature.

2. Results and discussion

The lifetime of the samples studied was large enough to measure basic electroluminescence (EL) characteristics. We attribute the significant lifetime increasing to using:
(a) fractional monolayer (FM) CdSe/ZnSe active region grown in a special manner which allows to separate spatially defects and radiative recombination area;
(b) the Be chalcogenides, which possess the highest energy of defect-formation among II–VI compounds.
The dependence of the inverse differential quantum efficiency of stimulated emission versus laser cavity length (Fig. 1) demonstrates rather high value (40%) of the internal quantum efficiency of stimulated emission in the Be-contained laser structure. However, still the greater part (60%) of emitted photons is a result of a spontaneous radiation process. In our opinion this value could be significantly reduced (in favor for stimulated emission) by increasing the electronic localization.

Threshold current density dependence of the modal gain ($\Gamma g$) is shown in Fig. 2. It is seen that, in spite of the rather weak rise of $\Gamma g$ with increasing the driving current, the gain is saturated at high values of the current density (3–4 kA/cm$^2$). This, so called gain saturation effect, is typical for the laser structures with an active region based on a single low-dimensional layer. The transmission electron microscopy (TEM) measurements [2] of the structure investigated showed the fractional monolayer origin of the active region which consists of the in situ grown CdSe-based self-organized islands. The surface density of
islands evaluated was about $2 \times 10^{10} - 10^{11} \text{ cm}^{-2}$ for large and small islands, respectively. Probably, the fractional configuration of the active region increases the current density, when gain saturation takes place. As follows from Fig. 2, the maximal value of the modal gain is limited by the value of $\approx 50 \text{ cm}^{-1}$. We suppose that it is mainly caused by not completely optimized waveguide parameters. In addition, the optimization of p-n-junction location will allow to improve the efficiency of carrier injection and to enhance the gain in the structure.

Rather high value of the threshold current and specific behavior of the gain in the structure lead to existence of the characteristic features of lasing. A change of a near field pattern with increasing the pumping current is demonstrated in Fig. 3. It is seen that at low pumping current (under threshold) two peaks with almost equal intensity appear. It should be noted that the light spot clearly distinguished at the laser mirror are located just under the stripe bounds. Above threshold the near field pattern is strongly asymmetrical because the lasing takes place only in one light spot mentioned above. At the same time a far field pattern (Fig. 4) remains uniform with increasing the current and is characterized by only one maximum in both transverse directions, resulting in the single mode operation in
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A wide interval of the pumping current. Such behavior may be caused by anti-guiding effect, which was early observed and investigated for III–V lasers (see, e.g., [4]). It is known that an increase of injection carrier concentration leads to reduce a refractive index. In case of relatively low gain and current spreading in transverse direction gain-guiding could be suppressed. As a result the light is forced out to the bounds of the pumping area.

Analysis of electroluminescence spectra confirms the existence of the anti-guiding effect. Lasing spectra of injection laser with the cavity length $L = 435 \, \mu m$ is shown in the insert of Fig. 1. The indistinct pattern of Fabry–Perot modes may be attributed to the heating of the active region and neighboring layers. Using the known formula connecting the values of cavity length, wavelength, refractive index (for this compound) and dispersion relation of the refractive index allows us to calculate the “expected” mode spacing as 0.074 nm. At the same time, from the experiment Fabry–Perot mode spacing was determined as 0.085 nm. Larger mode spacing means that total refractive index of the waveguide is lower than expected one and this reduction may be caused by injected carriers.

3. Conclusions

The detail EL study of the BeMgZnSe/ZnCdSe injection laser heterostructure has been performed. It has been revealed that the enhanced device lifetime allowed one to investigate the wide spectrum of electroluminescence parameters of the RT green laser. The surface density of CdSe-based islands is shown to be sufficient for creation of the effective green laser emitters. Single mode operation of injection lasers has been demonstrated in the wide pumping current range. Anti-guiding effect has been discussed and should be taken into account in creating the lasers based on II–VI heterostructures.

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References

The power of catastrophic optical mirror degradation in InGaAs/AlGaAs/GaAs QW laser diodes

E. Yu. Kotelnikov†, A. A. Katsnelson†, D. A. Livshits†, W. Richter‡, V. P. Evtikhiev†, I. S. Tarasov† and Zh. I. Alferov†
†Iofe Physico-Technical Institute, St Petersburg, Russia
‡Jena University, Germany

Abstract. We report on the fabrication and characteristics of conventional QW SCH InGaAs/AlGaAs broad area laser diodes. The detailed investigation of 100 µm striped contact laser diodes produced by MBE technique shows the strong dependence of a differential quantum efficiency \( T_1 = 280-420 \text{ K} \) on temperature. This factor gives the strong influence on the output power. The power of catastrophic optical mirror degradation of InGaAs/AlGaAs laser diodes may reach practically the same (20 MW/cm²) data as for Al-free laser diodes. The output power of 6.3 W (CW, 280 K) in InGaAs/AlGaAs QW SCH 100 µm stripe is achieved.

Introduction

The main factor limiting the optical output power of semiconductor lasers is catastrophic mirror degradation (COMD). It caused by overheating of the active region at the mirrors. Last years many papers devoted to the continuous wave very high powers from diode lasers have been published [1–2]. The achievement of record high CW powers stay possible due to the new “broadened waveguide” (BW) construction of laser diodes and application of Al-free semiconductor compounds for laser heterostructures. The increase of waveguide thickness in BW construction decreases the light power density and prevents the mirror overheating. The overheating process is not clear in detail until this time. Some authors believe the overheating due to light absorption in “dead layer” that appears in active region near the facets, some of them connect the overheating with the surface recombination. Botez showed [3] that maximum optical power is limited by surface recombination rate of active region material. In any case the overheating temperature is strongly connected first of all with active region materials [4]. Thus the above mentioned explanations of COMD do not limit the material for the laser heterostructures by Al-free compounds only. We have found that investigation of maximum optical power in SCH QW AlGaAs/GaAs laser diodes with InGaAs quantum well active region is of interest.

1. Experiment

We grew the SCH QW InGaAs/AlGaAs/GaAs laser heterostructure by the TsNa-4 molecular beam epitaxy system on oriented \( n^+ \)-type GaAs (001) substrate. The schematics band diagram of the heterostructure is shown in Fig. 1.

The structure contains 0.8 µm thick Al\(_{0.45}\)Ga\(_{0.55}\)As emitters. The 85 Å In\(_{0.18}\)Ga\(_{0.82}\)As active region is confined by 0.5 µm thick waveguide layers of variable gap AlAs/GaAs superlattice with linearly varying band gap, similarly to the equivalent graded layer Al\(_{0.43}\)Ga\(_{0.57}\)As/Al\(_{0.29}\)Ga\(_{0.71}\)As. For our investigation 100 µm SiO\(_2\) stripe lasers was fabricated.
In Fig. 2 are presented output power vs. current and voltage vs current characteristics recorded at CW condition at temperature 280 K. One can see in power vs current characteristic that maximum power reach 6.3 W at 7.75 A. As far as we know output power 6.3 W is one of the best results for non Al-free lasers. It is difficult to compare different laser because there are too many kind of laser construction. To compare our result with different lasers output power we use an optical power density:

\[
P_{\text{COMD}} = \frac{P_{\text{max}}}{d/(\Gamma W)[(1-R)/(1+R)]},
\]

where \( P_{\text{max}} \) is maximum optical power, \( W \) is stripe width, \( R \) is front facet reflectivity, \( d \) is active region thickness and \( \Gamma \) is optical confinement factor.

Calculated electrical field distribution is shown in Fig. 1 \( d/\Gamma \) ratio obtained from our calculations is 0.52. Under these condition we get \( P_{\text{COMD}} \) above 20 MW/cm². This value is near the best results for Al-free lasers. So we can conclude that Al-free active region in non Al-free waveguide in SCH QW semiconductor lasers demonstrate good \( P_{\text{COMD}} \) value.
At the current higher as 5 A the power–current dependence receive the sublinear character. To find the reason of such behavior we measured the power-current characteristics for laser diodes with different cavity length in a temperature range of \(-10 \ldots +80^\circ C\). We estimated the characteristic temperature \(T_0\) and \(T_1\) that describe accordingly the temperature dependencies of threshold current and differential quantum efficiency. The dependencies of characteristic temperature \(T_0\) and \(T_1\) vs the cavity length of the laser diode are given in Fig. 3.

The value of \(T_0 = 180\,\text{to}\,200\,\text{K}\) are typical for InGaAs/AlGaAs and Al-free laser diodes in all range of cavity length. At the same time it is important to note the value of parameter \(T_1 = 270\,\text{to}\,280^\circ C\) for cavity length more as 3 mm is too low. The reason of such low value of \(T_1\) parameter may be the temperature dependence of internal losses \(\alpha_i\) or the temperature dependence of internal quantum efficiency \(\eta_i\). We extract these dependencies from experimental data (Fig. 4). The main contribution of the temperature dependence of differential quantum efficiency gives the dependence of internal losses on the temperature. So, the reduction of internal optical losses in investigated InGaAs/AlGaAs laser diodes is
mandatory for the achievement of the maximal optical power.

2. Conclusion

By this paper we show that the output power of catastrophically optical mirrors damage of traditional InGaAs/AlGaAs QW SCH laser diodes may have practically the same value as for Al-free laser diodes. The maximum output power of investigated broad area laser diodes of 6.3 W (CW, 280 K) is achieved. This value is controlled mainly by the very high temperature sensitivity of differential quantum efficiency of InGaAs/GaAs laser diodes.

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Intersubband population inversion under resonance tunneling in wide quantum well structures

P. N. Lebedev Physical Institute, Leninsky pr. 53, 117924 Moscow, Russia

Abstract. Theoretical estimates and the results of vertical transport and optical investigations in GaAs/AlGaAs structures show that resonant tunneling can lead efficiently to selective de-population of the levels, resulting in a population inversion and possible stimulated emission due to intersubband transitions between the lowest states in wide-quantum-well structures.

The recent investigations of intersubband infrared quantum cascade and quantum fountain lasers make a considerable step in nanostructure physics development but as to the far infrared range the problem has not been yet solved. In this paper we discuss a version of a far-infrared resonant tunneling laser based on the intersubband transitions in wide quantum well structures (WQWS) with the energy spacing between the two lowest states below the longitudinal-optic (LO) phonon energy. The population inversion is achieved due to the difference in scattering relaxation processes between the lowest states with or without LO phonon emission, and via the selective removal of carriers from the ground state by use of resonant tunneling to the neighboring quantum well [1, 2].

On the base of vertical transport and optical investigation of the structures the relaxation life-time and resonant tunneling rate values as well as the intersubband optical absorption and amplification and the other characteristics are discussed revealing the possibility of intersubband population inversion and far-infrared stimulated emission in the systems. The analysis of the intersubband relaxation in quantum wells is based on the theoretical approach resulting in the analytical expressions for optical phonons, acoustic phonons and electrically charged impurities scattering rates. The vertical transport and photoluminescence investigations are carried out in GaAs/Al_{0.3}Ga_{0.7}As superlattices and asymmetric multiple quantum well structures with central well of width $d_w = 25$ nm ($\epsilon_1 = 7$, $\epsilon_2 = 29$, $\epsilon_3 = 64$, $\epsilon_4 = 113$, $\epsilon_5 = 173$ meV, the lasing transition wave lengths around 60 $\mu$m [1, 2]) revealing the incoherent behavior of the resonant tunneling when it is combined with a relaxation process, resulting in the essential dependence of the tunneling time on the relaxation rate. The space charge effects are demonstrated resulting in disalignment of the resonant states and transformation of the whole resonant tunneling structure under electrical fields, which change dramatically the rate characteristics of resonant tunneling. The experimental evidence of the resonant tunneling as an effective way for selective redistribution of carriers resulting in population inversion throughout the lowest subbands in wide quantum well structure is shown.

Scattering relaxation carrier life-times

The experimentally confirmed theoretical estimates show that in structures considered the optic-phonon carrier relaxation between excited states results in $\tau^{\text{opt}} \approx 0.5$ ps while the
acoustic-phonon intersubband relaxation between the first excited and the ground states results in $\tau_{ac}^{12} \approx 300$ ps at low temperatures [3, 4]. This value strongly decreases in electric field due to ionized-impurity scattering. The estimates of $\tau_{12}$ due to ionized-impurity scattering were made in analytical form taking into account the screening effect by free electrons that, as was shown, plays a noticeable role in structures with rather wide wells. The interaction between an electron and an ionized impurity scatterer centered at $z = z_i$ was described by a potential $\phi(\rho, z, z_i)$ which has been found in quasi-two-dimensional approximation from Poisson’s equations ($q_0$ is reciprocal screening length, $z$ is distance from the ionized impurity in-grown direction). The results of calculation with use of a standard program for computation of the probabilities of intersubband transitions for unscreened ($q_0 = 0$) and screened ionized impurities in GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum wells ($d_w = 25$ nm, $d_b = 4$ nm, $N_s = 10^{10}$ cm$^{-2}$, $T = 4.2$ K) are presented to show that the relaxation time increases noticeably due to screening, decreases distinctly with increasing the well width, depending strongly on the impurity center location in the well. The $\tau_{12}^{imp}$ values in uniformly doped wells averaged over the impurity positions are about 40 ps ($N_d = 10^{16}$ cm$^{-3}$, $N_s = 10^{10}$ cm$^{-2}$) which can be increased using the selective doping (in the well center where the square of the wave function in the first excited state goes to zero). It can be shown that the electron-electron scattering does not play a decisive role in the situation [5].

**Resonant coherent and incoherent tunneling rates and selectivity in structures with broadened energy levels**

In view of the estimates the problem of resonant tunneling becomes to be the very important and calls for particular attention. The experiments show that the resonant tunneling times usually estimated from the energy-level splitting $\tau_{tun}^{coherent} \cong h/\delta\epsilon_{split}$ are as a rule essentially longer. The main reason is the necessity of taking into account the level widths which can be caused, in particular, by carrier relaxation in neighboring well, that makes the tunneling process incoherent. According with the nonstationary quantum perturbation theory the resonant tunneling combined with the fast relaxation process in neighboring well has [6, 7] as a result:

$$\tau_{tun}^{incoherent} \cong \tau_{opt}^{opt} + \frac{h}{\delta\epsilon_{split}} \left( \frac{\gamma}{\delta\epsilon_{split}} \right) \frac{\Delta\epsilon^2 + \gamma^2}{\gamma^2}$$

(1)

where $\Delta\epsilon$ is mismatch of the resonant levels, $\gamma$ — half-width of the final tunneling level. The value $\tau_{tun}$ under exact resonance is proportional to $\delta\epsilon_{split}^2$ and increase with increasing of the level width as $\gamma/\delta\epsilon_{split}$. The experimental vertical transport and photoluminescence investigations are presented [2] to be consistent with (1) resulting in the values $\tau_{tun}$ for tunneling transitions $\epsilon_1 \rightarrow \epsilon_2$ in the modeling structure of about 30 ps ($d_B = 6$ nm), 10 ps ($d_B = 4$ nm), 3 ps ($d_B = 2$ nm) based on the calculated $\delta\epsilon_{split} = 0.5, 1$ and 3 meV.

It is shown with use of the far infrared emission [9], photoluminescence and vertical transport investigations [1, 2] that the lowest subbands are manifested separately in superlattices with $d \leq 35$ nm (the lowest level widths are of order 1–2 meV) revealing the proof that the resonant tunneling can be used for selective depopulation of the lowest levels leading to a population inversion in the structures.

**Space charge mismatching of resonant levels in electric fields**

The vertical transport measurements are presented to reveal a large variety of the effects that extremely change the resonant tunneling behavior and the electrical properties of the
WQWS under electrical fields. These peculiarities have been investigated in detail being caused by the carrier redistribution in the structures leading to a mismatch of the resonant levels and breaking the system down to the areas with different electric field strength (electric-field domains) [1, 2, 9–10]. These effects can be overcome but merit detailed consideration.

Optical intersubband absorption and amplification in wide-quantum-well-structures

The intersubband optical absorption coefficient for transitions between two first excited and the ground subbands in GaAs/$\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ structure evaluated on the base of analytically [2] obtained expression

$$
\alpha_{\text{inter}}^{12} = \frac{512}{9} \frac{e^2 (\epsilon_F - \epsilon_1) \Gamma}{mc\eta \rho \omega \left[ (\epsilon_2 - \epsilon_1 - \hbar \omega)^2 + \Gamma^2 \right]}
$$

is of order $\alpha_{\text{inter}}^{12} = 4 \times 10^3 \text{ cm}^{-1}$ at carrier concentration $10^{16} \text{ cm}^{-3}$ ($d_w = 25 \text{ nm}, d_b = 4 \text{ nm}, \Gamma = 5 \text{ meV}, \epsilon_2 - \epsilon_1 = 22 \text{ meV}, 4.2 \text{ K}$). Here $\epsilon_F$—Fermi energy, $\Gamma$—level width, $\eta$—refractive index, $e$ and $m$—electron charge and effective mass. Taking into account the estimates for free carrier absorption (less than $10 \text{ cm}^{-1}$) [2] and for phonon absorption (less than $10 \text{ cm}^{-1}$) [2] one can conclude that the very small population inversion in the lowest subbands may result in a drastic increasing of the intensity of emission due to transitions between these states. This point plays an essential role appearing to be stimulating for laser investigations not only in WQWS but also in multiple WQWS as well.

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Power conversion efficiency in a quantum dot based diode laser

A. E. Zhukov, A. R. Kovsh, S. S. Mikhrin, N. A. Maleev,
V. A. Odnoblyudov, V. M. Ustinov, Yu. M. Shernyakov,
E. Yu. Kondrat’eva, D. A. Livshits, I. S. Tarasov, N. N. Ledentsov,
P. S. Kop’ev, Zh. I. Alferov and D. Bimberg‡
Ioffe Physico-Technical Institute, St Petersburg, Russia
‡Technische Universität Berlin, D–10623 Berlin, Germany

Abstract. Power conversion efficiency of diode lasers with the active region based on a quantum dot array is under investigation. The model is proposed which allows one to estimate analytically the optimal cavity length corresponding to the maximum power conversion efficiency at a given output power. The model is compared with the experimental results from high–power 0.94–µm diode lasers based on sub-monolayer quantum dots.

Introduction

Power conversion efficiency is one of the most important characteristics of any diode lasers. It depends on both the output power level and the cavity length. The same output power can be achieved from the diodes of various lengths whereas the conversion efficiencies are quite different owing to the differences in the threshold currents and the slope efficiencies. The output power at the point of peak conversion efficiency for the diode of a given length has been found in [1]. However, there can exist some cavity length(s) which corresponds to higher value of the conversion efficiency for the same power level. In the present work, optimization of the diode laser for high–efficient operation is considered as a search for the optimum cavity length, $L_{opt}$, which maximizes the power conversion efficiency for a given (designed) output power. We propose a simple analytical model which enables to determine these parameters for a diode laser with the active region based on array of semiconductor quantum dots (QDs). Also, their relationship with the internal characteristics of the diode lasers, i.e., series resistance, internal loss, transparency current, differential gain, etc. can be quantitatively predicted. Good agreement is observed when applying this model to a high–power 0.94–µm diode lasers based on sub–monolayer InAs/GaAs (QDs).

1. Conversion efficiency

Power conversion efficiency, $\eta_C$, is defined as the ratio of the output optical power from diode facets to the electrical input:

$$\eta_C(P, L) = \frac{P}{IV_0 + I^2R_S},$$

(1)

where $I = P/\eta + I_0$ is the drive current which is necessary to apply for achieving the design output power $\eta$ and $I_0$ are cavity length ($L$) dependent differential efficiency and threshold current, respectively; $V_0$ is the diode turn–on voltage; $R_S = \rho_S/WL$ is the series resistance of the diode with the width of $W$. In opposite to analysis developed in [1], our approach is based on matching the condition $\delta\eta_C(P, L)/\delta L |_{P=\text{const}} = 0$. The solution of this equation
determines the optimum diode length, \( L_{\text{opt}} \), at which the conversion efficiency reaches its maximum, \( \eta_{\text{C max}}(P) \), for the given power, \( P \). It can be shown that this is equivalent to expression:

\[
V_0 C + 2 R_S IC = -I^2 \frac{dR_s}{dL},
\]

(2)

where \( C \equiv dI/dL \mid_{P=\text{const}} \). It is obvious that \( C \) parameter depends on both the power and the cavity length. In general case, this dependence is rather complex because it involves variation of both \( I_{\text{th}} \) and \( \eta \) with \( L \). However, in the case of a diode laser based on an array of QDs, Eq. (2) can be significantly simplified giving a mathematical expressions for \( L_{\text{opt}} \) and \( \eta_{\text{C max}}(P) \) under analysis. The characteristic feature of a QD laser is the linear relationship between threshold current density, \( J_{\text{th}} \), and modal gain, \( g \):

\[
g = \beta(J_{\text{th}} - J_0),
\]

(3)

under condition that \( g \) does not exceed the saturated gain, \( g_{\text{sat}} \). Here \( \beta \) is the differential gain (with respect to current density) and \( J_0 \) is current density at the transparency. Thus,

\[
C = \frac{P \alpha_i}{\eta_0 r} + W \left( \frac{\alpha_i}{r} + J_0 \right),
\]

(4)

where \( \eta_0 \) is the internal differential efficiency, \( \alpha_i \) is the internal loss, \( r = 1/2 \ln(1/R_1 R_2) \), \( R_{1,2} \) is the facet power reflectivities. Substituting Eq. (4) into (2), the optimum cavity length of a QD diode laser, \( L_{\text{opt}} \), and the corresponding maximum conversion efficiency \( \eta_{\text{C max}}(P) \) are finally given by:

\[
L_{\text{opt}} = \frac{P_0}{C \sqrt{1 + B^2}}, \quad \eta_{\text{C max}}(P) = \frac{P}{V_0 P_0 B / (1 + \sqrt{1 + B})^2},
\]

(5)

where \( B = W V_0 / \rho S C \) and \( P_0 = P / \eta_0 + Wr / \beta \) are length independent parameters. Substituting the experimental parameters of the laser structure, such as specific resistance, internal loss, etc one can design the cavity length and facet reflectivities by such manner to achieve the maximum conversion efficiency for the practically desired level of power output. On the other hand, Eq. (5) allowed one to optimize the laser structure for high-efficient operation by estimating the relative importance of different internal parameters. Applicability of the proposed optimization scheme is discusses below, where we consider high-power diode lasers based on sub-monolayer InAs/GaAs QDs.

2. Experiment

Sub-monolayer QDs are formed as a result of molecular beam epitaxial deposition of a short-period InAs/(Al)GaAs superlattice on a GaAs(100) surface with the InAs effective thickness of less than 1 monolayer (ML). High-power operation of 0.94-\( \mu \)m diode lasers with SML QDs has been recently reported [2]. Diode lasers discussed in this work have 100-\( \mu \)m-wide uncoated facets and were tested under CW excitation at 10\( ^\circ \)C. Figure 1(a) shows the experimental power conversion efficiency as a function of the power for two different cavity lengths. It is seen that \( P = 1.9 \) W corresponds to the peak conversion efficiency (50.5%) for \( L = 1.76 \) mm. On the other hand, there exist the other cavity lengths which result in higher conversion efficiency at \( P = 1.9 \) W. In particular, at \( L = 1.04 \) mm the power conversion efficiency of 58% is achieved at 1.9 W. This example shows the importance of the analysis above for optimization of efficiency characteristics. Assuming the transparency
Fig. 1. Efficiency characteristics of 0.94-µm diode lasers based on sub-monolayer QDs. CW regime, 10°C. (a) Power conversion efficiency versus output power for two diode lengths; (b) Optimum cavity length and maximum conversion efficiency at 2 W calculated under assumptions $\beta = 0.06 \text{ cm/A}$, $J_0 = 90 \text{ A/cm}^2$, $\rho_S = 1.244 \times 10^{-4} \Omega \text{ cm}^2$, $V_0 = 1.37 \text{ V}$, $\alpha_i = 2.3 \text{ cm}^{-1}$, $\eta_0 = 1.31 \text{ W/A}$.

Current density of $J_0 = 90 \text{ A/cm}^2$ and the differential gain of $\beta = 0.06 \text{ cm/A}$, Eq. (4) gives a good agreement with the experimental data in the range of $J_{th} < 500 \text{ A/cm}^2$ ($L > 0.5 \text{ mm}$). For the shorter cavities (higher $J_{th}$), the modal gain saturates at $g_{sat} = 21.5 \text{ cm}^{-1}$. This is, to our best knowledge, is the highest saturated gain ever reported for QD lasers of any kind with single QD plane in the active region. Maximum conversion efficiency, $\eta_{Cmax}$, and corresponding optimum cavity length, $L_{opt}$, calculated by is Eq. (5) are shown in Fig. 1(b) as functions of the optical power, $P$. Internal parameters are shown in the caption. It is seen that the optimum cavity length increases with $P$. The maximum conversion efficiency for the power range of 2–4 W, which was achieved in SML diode lasers with uncoated facets, corresponds to the optimum cavity length of about 1 mm. The experimental results (Fig. 1(a)) are in a good agreement with the calculation. The power conversion efficiency in excess of 55% is measured in a wide range of output power (1–2.5 W) in the 1.04-mm-long diode. The maximum conversion efficiency is 59% which is, to the best of our knowledge, is the record high value for any QD lasers. It is worth mentioning that it is only 7% lower the highest result achieved by well established QW laser technology [3, 4].

Fig. 2. Calculated dependence of the maximum power conversion efficiency at 2 W on the differential gain and the transparency current density. The other parameters are shown in the caption of Fig. 1.
As an example of relationship between internal laser parameters and efficiency characteristics, Fig. 2 shows the dependence of $\eta_{\text{max}}^C$ at 2 W on the differential gain, $\beta$, and the transparency current density, $J_0$. Both these parameters are closely related to the QD active region design. Figure 2(a) demonstrates that $\eta_{\text{max}}^C$ steeply decreases if the differential gain becomes lower than $\beta \sim 0.05 – 0.06$ cm/A. Differential gain of a QD laser is mostly governed by inhomogeneous broadening of QD states. For the structure under investigation the inhomogeneous broadening of the ground state transition was estimated to be 25 meV. This corresponds to $\beta$ of 0.06 cm/A, which is our experimental result. According to the data of Fig. 2(a) this differential gain still results in high efficiency. On the one hand, less uniform QD array (lower $\beta$) could not provide high conversion efficiency. On the other hand, maximum conversion efficiency is practically unchanged with $\beta$ higher than $\sim 0.09$ cm/A. Thus, formation of more uniform QD array can hardly improve significantly the efficiency characteristic. According to Fig. 2(b), $\eta_{\text{max}}^C$ weakly depends on the transparency current density, which, in the turn, is linearly proportional to the surface density of QDs. This means that the QD density can be noticeably increased without a significant effect on the efficiency. On the other hand, formation of denser QD array is strongly desired to improve the saturation gain and decrease the internal loss.

3. Conclusions

In summary, the power conversion efficiency of QD diode lasers is studied. Analytical model proposed allows us to evaluate the optimum cavity length which maximizes the conversion efficiency at the given output power. The maximum conversion efficiency and the optimum cavity length were calculated as function of the power as well as internal parameters of the active region. The calculation results are in a good agreement with experimental efficiency data on high–power 0.94–$\mu$m diode lasers based on sub-monolayer quantum dots.

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References

Quantum well DFB laser having a curved grating structure

G. S. Sokolovskii†, E. U. Rafailov‡, A. G. Deryagin†, V. I. Kuchinskii†, D. J. L. Birkin‡ and W. Sibbett‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ School of Physics and Astronomy, University of St Andrews, North Haugh, St. Andrews, Fife KY16 9SS UK

Abstract. Theoretical investigation of novel DFB laser with “curved-grooves” diffraction grating (c-DFB) is presented. Calculations show the proposed design provides spectral selectivity of conventional DFB resonator and output beam focusing in the plane of p-n-junction. The curved-grating DFB-laser design is predicted to be most attractive for the quantum well structures.

At present there is a great interest in high power single-mode single-frequency QW laser sources. Applications of these lasers ranges from medicine (e.g. NMR tomography) to telecommunication (pumping of Er-doped fibre amplifiers for fibre optical communication). Up-to-date single-element diode lasers based on a narrow index-guiding stripe are limited in power to less than 500 mW by either catastrophic optical damage or thermal overheating of the facet. Although higher powers may be achieved by increasing the stripe width, multiple lateral modes generally appear. Other approaches aimed at improving the diffraction-limited output power include arrays of antiguided narrow-stripe lasers, unstable resonators and master oscillator/power amplifier (MOPA) structures. Of these, only MOPA’s have demonstrated diffraction-limited operation to output powers of >1 W, but the technology required to fabricate MOPA’s is quite complex.

We present a novel type of distributed feedback laser with a “curved-grooves” diffraction grating (c-DFB), which is expected to give spectral and spatial output beams that are superior to those currently obtained from an angled diffraction grating (α-DFB). Figure 1 is a simplified schematic of the c-DFB laser [1]. The use of a c-DFB will combine not
Fig. 2. The modelled performance of the c-DFB laser.

Fig. 3. Plot of the longitudinal intensity distribution for various coupling levels for c-DFB laser with $\rho_0 = L$ (solid lines) and conventional DFB laser (dashed lines).

only the high-power of a broad stripe device and spectral control provided by a diffraction grating but also output beam focusing in the plane of p-n-junction. A diffraction grating with curved-grooves can be designed to concentrate the output emission to a focus determined by the curvature of the grating. Figure 2 shows the calculated c-DFB laser output field distribution in the plane of p-n-junction for a single transverse mode. Laser beam focusing in the direction orthogonal to the plane of p-n-junction can be simply obtained by a cylindrical lens mounted on the laser heat-sink.

The other advantage of the proposed c-DFB laser structure is the flat lateral intracavity intensity distribution, as it is shown in Fig. 3. The strongly non-uniform intensity distribution in the cavity of the conventional DFB laser causes the effect of spatial hole burning preventing high-power single-frequency laser operation. Spectral selectivity of c-DFB laser resonator is the same as that of a conventional DFB. These allow us to predict the improved spectral selectivity of the proposed c-DFB laser at high power compared to the conventional DFB laser design.
Figure 4 represents the calculated dependence of the focal distance and beam waist size on the grating curvature $\rho_0$ for a wavelength 1 $\mu$m. The c-DFB laser design allows adjustment of the structural parameters to obtain the beam properties necessary for different applications: minimal beam waist size and short focal distance for coupling with single-mode optical fiber or long beam waist for pumping of nonlinear crystal.

In summary we have proposed the construction of a novel DFB laser with “curved-grooves” diffraction grating. The theoretical analysis shows that this laser can combine not only the high-power of a broad stripe device and the spectral control provided by a diffraction grating but also output beam focusing in the plane of p-n-junction. The maximum efficiency of the curved grating device is predicted to be achieved at the high-power operation (which imply high photon density) i.e. in QW lasers.

References

Interaction between Landau levels of different two-dimensional subbands in GaAs

D. Yu. Ivanov†, E. Takhtamirov‡, Yu. V. Dubrovskii†, V. A. Volkov‡, E. E. Vdovin†, L. Eaves§, P. C. Main§, M. Henini§, D. K. Maude¶, J.-C. Portal¶, *, #, J. C. Maan∥ and G. Hill†
† Institute of Microelectronics Technology RAS, 142432 Chernogolovka, Russia
‡ Institute of Electronics and Radioengineering RAS, Moscow, Russia
§ The School of Physics and Astronomy, University of Nottingham, Nottingham NG7 2RD, UK
¶ Grenoble High Magnetic Field Laboratory, MPI-CNRS, BP166 38042 Grenoble Cedex 9, France
∥ High Field Magnet Laboratory, Research Institute for Materials, University of Nijmegen, 6525 ED Nijmegen, The Netherlands
* Department of Physics, University of Sheffield, Sheffield S3 3JD, UK
# Institut Universitaire de France
§ INSA, F31077 Toulouse Cedex 4, France

Abstract. Tunneling studies measurements between strongly disordered two-dimensional electron systems in a magnetic field parallel to the current are presented. Two-dimensional electron systems are formed by Si delta doping of GaAs on both sides of the AlGaAs barrier. Strong interaction between Landau levels of different two-dimensional subbands in GaAs have been observed as anticrossing of related peak positions with magnetic field. The splitting of the interacted Landau levels is about 10 meV, which could not be explained by non-parabolicity of the conduction band in GaAs. The possible reasons of the observed interaction are discussed.

In the case of a spherical energy-band model, there is no interaction between Landau levels of different two-dimensional subbands and intersection of two Landau levels is allowed [1]. Interaction between Landau levels was observed previously only in tunneling studies of surface quantization in narrow gap PbTe [2].

In this work we present tunneling studies measurements between strongly disordered two-dimensional electron systems (2DES) in a magnetic field parallel to the current. Due to the strong elastic scattering assisted tunneling in the structures the amplitude of the peaks related to the tunneling between Landau levels was of the same order of magnitude for processes both with Landau level index conservation and without. It gave us chance to study interaction between Landau levels. The main observation was that interaction between Landau levels of different two-dimensional subbands in GaAs is very strong and the observed splitting was about 10 mV, about the same as was observed in PbTe [2]. The possible reasons of the observed interaction are also discussed.

The MBE-grown sample was a single barrier GaAs/Al_{0.4}Ga_{0.6}As/GaAs heterostructure with a 12 nm thick barrier. The barrier was separated from the highly-doped, bulk contact regions by 50 nm thick, undoped GaAs spacer layers. To form the 2DES we used Si donors sheets with concentration of $3 \times 10^{11}$ cm$^{-2}$ located 5 nm from each side of the barrier. The tunneling transparency of the main barrier was much lower than that of the spacer regions, so that most of the applied voltage is dropped across the barrier. Measurements of the Shubnikov–de-Haas (SdH) like oscillations in the tunnel current gave electron sheet
concentrations approximately equal to the donor doping levels. Samples were fabricated into mesas of diameters 100–400 μm. The typical electron mobility in the 2DES is about μ = 1000 cm²V⁻¹s⁻¹ at 4.2 K.

Figure 1 shows the differential tunnel conductance G, at 4.2 K, measured using standard lock-in techniques, versus external voltage V_b at various magnetic fields up to 15 T. In zero magnetic field (lowest curve in Fig. 1) the differential conductance has a peak at zero bias and two pronounced shoulders at higher voltages of both polarities. We argue that the zero voltage peak reflects resonant tunneling between the ground states of the 2DESs, and that the shoulders are due to resonant tunneling between the emitter ground state (ground 2D subband, n = 0) and first exited state (excited 2D subband, n = 1) in the collector 2DES. The observation of a pronounced maximum at zero bias in zero magnetic field indicates that, despite the relatively large number of scattering centres in the 2D layers, the conservation of in-plane momentum is important for the tunneling process. The evolving structure in the curves with increasing magnetic field reflects resonant tunneling between different Landau levels, which will be discussed below.

Around B = 6 T, which is close to ν = 2 for our sample, experimental G(V_b) curves show (see Fig. 1) a pronounced minimum at zero voltage. With a further increase of the magnetic field, the minimum of the differential conductance at zero bias gradually becomes a maximum. For B higher than 8 T, a dip at zero bias appears within this maximum, reflecting the gap in the tunneling density of states around the Fermi level of the 2D electron systems. The details of the equilibrium tunneling processes around zero bias with magnetic field have been discussed before [3].

In this work, we concentrate on the evolution of the structure related with tunneling between different Landau levels and the appropriate peaks to be considered below are marked by circles in Fig. 1. The fan diagram of the peak positions on the voltage scale versus magnetic field is shown in Fig. 2. For simplicity we only consider peaks at negative bias voltage. In fact, observed structure is perfectly symmetrical around zero bias except
Fig. 2. Peak positions on the voltage scale as a function of magnetic field. Circles, squares and triangles represent experimental data and are discussed in the text. Curves “1” and “2” represent expected peak position calculated in the absence of the interactions between Landau levels. Lines “A” and “B” represent tunneling between Landau levels with $\Delta N = 1$ and $\Delta N = 2$ correspondingly in the case of the ideal Landau level quantization.

In a field higher than 12 T circles correspond to the tunneling between 1st Landau level ($N = 0$) of the ground subband state in the emitter ($n = 0$), and 2nd Landau level ($N = 1$) of the ground subband state in the collector ($n = 0$), without Landau level index conservation, i.e. $(n = 0, N = 0) \rightarrow (n = 0, N = 1)$ tunneling. The dash line labeled “A” has a slope equal to $L\hbar\omega_C$, where $\hbar\omega_C$ is the cyclotron energy, $L$ is the leverage factor equal to 1.28 for our structure, and presents position of the peaks for $\Delta N = 1$ tunneling in the case of ideal Landau level quantization, the dash line labeled “B” presents position of the peaks for $\Delta N = 2$ tunneling in ideal case correspondingly. For broadened Landau levels the tunneling differential conductance which was just measured in our studies reflects the joint density of states at the Fermi levels of the emitter 2D electron system. In this case the calculated position of the peaks for $(n = 0, N = 0) \rightarrow (n = 0, N = 1)$ tunneling is shown by curve labeled “1”. The position of the peaks related with $\Delta N = 0$ processes do not depend on magnetic field. Evidently the peak position for $(n = 0, N = 0) \rightarrow (n = 1, N = 0)$ should be presented by vertical line labeled “2”, which coincide with position of the peak reflected resonant tunneling between ground 2D state in the emitter and first excited state 2D state in the collector without magnetic field. Without interaction between Landau levels lines described peak positions of the different processes versus magnetic field dependencies should intersect as the curves “1” and “2” do. In contrast, experimental dependencies presented as circles and squares in Fig. 2 exhibit obvious anticrossing, which is a manifestation of the interaction between Landau levels ($n = 0, N = 1$) and ($n = 1, N = 0$) of the different subbands in the collector. The Landau level splitting is about 10 meV. Some peculiarities is evident also around point where line labeled “B” intersect vertical line “2”. These peculiarities indicate that interaction between Landau levels ($n = 0, N = 2$) and ($n = 1, N = 0$) in the collector 2DES also takes place, but details of this interaction is out of the experimental accuracy. For generality we also indicated position of the peaks around zero voltage (triangles in Fig. 2) which appeared...
due to the development of the tunneling gap at Fermi level in magnetic field by triangles.

Let us discuss possible mechanisms of strong (~10 meV) anticrossing of the Landau levels \( N = 1 \) from the ground subband \( n = 0 \) and \( N = 0 \) from the excited subband \( n = 1 \). Naturally, such mechanism should mix longitudinal and transverse motion of electrons in the 2D layer. The first reason is connected with the weak non-parabolicity of the electron spectrum in GaAs, the more so as the qualitatively similar anticrossing was observed in 2D layer of highly non-parabolic material PbTe [2]. But in the latter case the strong anticrossing may be caused by the fact that the main axes of the constant energy ellipsoid of the conduction band minimum (L-point of the Brillouin zone) are not along the direction of the structure growth. It does not take place when concerning GaAs, and the theoretical estimation of contribution of the non-parabolicity yields only ~1 meV—too small a value.

The second reason of the strong anticrossing may be connected with the effect of the chaotic potential \( V(r) \) which is not weak in our case. In the two-subband \((n = 0, 1)\) approximation after averaging \( V(r) \) over the longitudinal motion the contribution of the potential takes the matrix form

\[
\begin{pmatrix}
V_{00}(r_\parallel) & V_{01}(r_\parallel) \\
V_{10}(r_\parallel) & V_{11}(r_\parallel)
\end{pmatrix}
\]  

(1)

Here the diagonal elements \( V_{00}(r_\parallel) \) and \( V_{11}(r_\parallel) \) describe chaotic motion in the plane of 2D layer in the subbands \( n = 0 \) and \( n = 1 \). It is the functions that correspond the broadening of the Landau levels which is large enough in our “dirty” system (~10 meV). The non-diagonal elements \( V_{01}(r_\parallel) \) and \( V_{10}(r_\parallel) \) push the states from the subbands \( n = 0 \) and \( n = 1 \) apart. If the characteristic length of variation of \( V(r) \) relatively to the normal to 2D layer is of the order of the “width” of 2D layer (which sounds reasonable), the value \( V_{01}(r_\parallel) \) is not small, and it may be responsible for the observed effect. One more mechanism of the anticrossing connected with the many-particle interaction is under consideration.

Thus we have investigated tunneling processes between strongly disordered 2D electron systems in a quantized magnetic field parallel to the current. The manifestation of the strong interaction between Landau levels of different two-dimensional subbands in GaAs have been observed experimentally.

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References


2D symmetry and pinning of quantum-Hall “strip phase”

E. E. Takhtamirov and V. A. Volkov
Institute of Radioengineering and Electronics of RAS, Mokhovaya 11, 103907 Moscow, Russia

Abstract. Recent experiments on (001) GaAs/AlGaAs structures revealed a new phase in the excited Landau levels. The results can be related to spontaneous formation of unidirectional charge density waves (“strip phase”). We address to the unsolved problem: why stripes are pinned coherently across the sample along [110]. Developing the Kroemer’s idea about the role of $C_{2v}$ symmetry in this system we show that for (001) $A_3B_5$ single heterojunction the conduction band in-plane effective mass is anisotropic. This anisotropy and the optical anisotropy in heterostructures with different cations and anions have the same origin. Although very weak, the difference in the conduction band effective masses along the crystallographic [110] and [1 ̅10] axes may define the direction of the stripes. The results agree with the experiments in tilted magnetic fields.
constructed many-band system of envelope-function equations preserves information about the lowered symmetry of the heterostructure ($C_{2v}$) as compared to the symmetry of the host materials ($T_d$). The light hole-heavy hole mixing at the center of 2D Brillouin zone is one of the consequences of the symmetry lowering [9]. This mixing leads to the giant optical anisotropy (with the same principal axes [110] and [110]) in heterostructures composed of materials with different cations and anions observed experimentally [10]. It is obvious that the symmetry $C_{2v}$ should manifest itself in EM equation for conduction band too.

However, the derived in [8] one-band effective Hamiltonian for electrons lost $C_{2v}$ symmetry information. To construct the effective Hamiltonian having $C_{2v}$ symmetry means that now we should go beyond the approximation used in [8] and consider smaller but important here anisotropic terms. Because of the limiting accuracy of the method [8], we might not allow for them if we were interested merely in more precise calculation of the conduction band states. Such contributions may be taken into account just because they provide the symmetry lowering—the effect which is absent without them. So, as we are not interested here in terms of symmetry higher than $C_{2v}$, the effective Hamiltonian may include only the leading potential and kinetic energy terms (used in standard EM approximation) and the leading anisotropic term which will be found below.

We may obtain the correct Hamiltonian having $C_{2v}$ symmetry via the method of invariants. Setting the spin-orbit interaction aside (we will not discuss it here) one may conclude, that the information about $C_{2v}$ symmetry should be found in the kinetic energy operator. If we assume that the in-plane components of momentum $\hat{p}_x[[100]$ and $\hat{p}_y[[010]$, then the most general form of the quadratic in the in-plane momentum kinetic energy operator has the following form

$$\hat{T} = \frac{\hat{p}_x^2 + \hat{p}_y^2}{2m^*} + A \frac{1}{2}(\hat{p}_x \hat{p}_y + \hat{p}_y \hat{p}_x).$$  \hspace{1cm} (1)

Here $m^*$ is the conduction band EM, $A$ defines the in-plane EM anisotropy. From the derived in [8] many-band system of envelope-function equations we have indeed

$$\mathcal{A}(z) = \delta(z) \sum_{n,n'} 2 \{ s \mid \hat{p}_x \mid n \} D_{nn'} \{ n' \mid \hat{p}_y \mid s \} + 4D_{ss} \{ n \mid \hat{p}_x \mid n \} \{ n' \mid \hat{p}_y \mid s \} \frac{m_0^2 (\epsilon_s - \epsilon_n) (\epsilon_s - \epsilon_{n'})}{m_0^2 (\epsilon_s - \epsilon_n) (\epsilon_s - \epsilon_{n'})}.$$  \hspace{1cm} (2)

Here $\delta(z)$ is the Dirac $\delta$-function, $z = 0$ defines the position of the heterointerface, $\{ n \mid \hat{p}_a \mid n' \}$ is $\alpha$-component of the interband (for bands $n$ and $n'$ at $\Gamma$-point) matrix element of momentum, $s$ is the conduction band index, $m_0$ is the free electron mass, and $\epsilon_n$ means the $n$th band edge energy of a host material. The parameter

$$D_{nn'} = \sum_{j=\pm1, \pm2, \ldots} \frac{\{ n \mid \delta U \sin(4\pi j z/a) \mid n' \}}{4\pi j/a} \int_{-\infty}^{\infty} \frac{dG(z)}{dz} \cos \left( \frac{4\pi j z}{a} \right) dz.$$  \hspace{1cm} (3)

Here $a$ is the lattice constant. The appearing in the above expression functions are defined the way the crystalline potential of the heterostructure is

$$U(r) = U_1(r) + G(z)\delta U(r),$$  \hspace{1cm} (4)

$U_1$ is the crystalline potential of one of the materials of the heterojunction, and $U_2 = U_1 + \delta U$ is that of another one. Note, that parameter $D_{XY}$ defines the light hole-heavy hole
mixing at the center of 2D Brillouin zone, here $X$ and $Y$ are indexes of the Bloch functions of the valence $\Gamma_{15}$ band edge (transforming like $x$ and $y$ under symmetry operations of the group $T_d$) [8, 9].

If we perform the 45°-rotation of the coordinate system, so that in new coordinates $x[[110]$ and $y[[110], and then add magnetic field $B$ with the gauge of the vector potential $A = (B_y z, -B_z z + B_x x, 0)$, the conduction band Hamiltonian will take the form (without spin-orbit interaction)

$$\hat{H} = \frac{\hat{p}_z^2}{2m^*} + V(z) + \frac{1}{2} \left( \frac{1}{m^*} - A(z) \right) \left( \hat{p}_x + \frac{e}{c} B_z z \right)^2 + \frac{1}{2} \left( \frac{1}{m^*} + A(z) \right) \left( \hat{p}_y - \frac{e}{c} B_x z + \frac{e}{c} B_z x \right)^2.$$  \hspace{1cm} (5)

Here $V(z)$ is the conduction band potential, $e$ is the absolute value of the electron charge and $c$ is the speed of light in vacuum.

For 2D electron gas the in-plane magnetic field may be treated as a small perturbation [11]. To the second order in $B \parallel$ this leads to the diamagnetic energy shift and an increase (for ground electric subband) of EM in the direction perpendicular to the direction of the in-plane magnetic field. Then the native anisotropy of EM may be treated as a small perturbation too. For simplicity we assume that $B \parallel$ is parallel either $[110]$ or $[110]$ so that $B_x B_y = 0$.

Including all terms to the second order in $B \parallel$ and first in $A$ we obtain for the zeroth subband

$$\hat{H}_0 = E_0 + \frac{e^2}{2m^*c^2} \left( B_x^2 + B_y^2 \right) \left( \langle z \rangle_{00}^2 - \langle z \rangle_{00}^2 \right) + \frac{1}{2m^*} \left( 1 - \frac{\Delta_N}{2} - \frac{B_x^2}{B^2} B_0 \right) \left( \hat{p}_x + \frac{e}{c} B_y \langle z \rangle_{00} \right)^2 + \frac{1}{2m^*} \left( 1 + \frac{\Delta_N}{2} - \frac{B_y^2}{B^2} B_0 \right) \left( \hat{p}_y + \frac{e}{c} B_x \langle z \rangle_{00} \right)^2.$$  \hspace{1cm} (6)

The values of the native and magnetic field induced anisotropies of EM are

$$\Delta_N = 2m^* \langle A(z) \rangle_{00}, \quad \Delta_B = \frac{2e^2 B^2}{m^*c^2} \sum_m \langle | \langle z \rangle_{0m} \rangle_0 \rangle^2.$$  \hspace{1cm} (7)

Here $E_n$ is the $n$th electric subband edge energy at $B = 0$. Using the rough estimations below let us evaluate $\Delta_N$ and $\Delta_B$ at $B || = 0.5$ T (in experiment [2] such in-plane magnetic field, when $B ||$ along [110], makes initially anisotropic magnetoresistance essentially isotropic; higher $B ||$ reorient the direction of low resistance).

$$\langle A(z) \rangle_{00} \sim \frac{2\langle s | \hat{p}_x | X \rangle D_{XY} \langle Y | \hat{p}_y | s \rangle}{m^* E_g^2} \langle \delta(z) \rangle_{00} \sim \frac{D_{XY}}{m^* E_g} 10^{-2} \AA^{-1}.$$  \hspace{1cm} (8)

Here $E_g = 1.5$ eV is the band gap, $m^* = 0.067 m_0$, and $D_{XY} \sim 0.2$ eVÅ (in [9] it was found $D_{XY} \sim 0.5$ eVÅ for GaAs/AlAs). For $\Delta_B$ we need also

$$\sum_m \langle | z_{0m} \rangle_0 \rangle^2 \sim \frac{50 \AA^2}{0.01 \text{eV}}.$$  \hspace{1cm} (9)
Finally we have

\[ \Delta_N = 0.27 \cdot 10^{-2} = 0.27\%, \quad \Delta_B = 0.33 \cdot 10^{-2} = 0.33\% . \]  \hspace{0.5cm} (10)

Matching of these values coming from quite different mechanisms is very good. Now we
may conclude that as the in-plane magnetic field \( B_1 \approx 0.5 \) T has a strong influence on
the direction of the stripes and the value of the ratio \( R_{xx}/R_{yy} \), the native anisotropy of the
in-plane EM can be that mechanism making the stripes coherent over the macroscopic size
of the studied samples. If so, from the experiment [2] we may conclude that \( \langle \hat{A}(z) \rangle_{00} < 0 \).
Moreover, knowing the parameters of the structure more definite (or getting the magnitude
of the magnetic field induced EM anisotropy from experiments) one may deduce \( \langle \hat{A}(z) \rangle_{00} \)
from the dependence of the magnetoresistance anisotropy versus in-plane magnetic field
[2]. The magnitude of the parameter that governs the native in-plane EM anisotropy may
become one of the characteristics of the heterointerface [8].

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References
Cyclotron resonance quantum Hall effect detector

B. A. Andreev†, I. V. Erofeeva†, V. I. Gavrilenko†, A. L. Korotkov†,
A. N. Yablonskiy†, Y. Kawano‡ and S. Komiyama‡

† Institute for Physics of Microstructures of RAS, GSP-105,
603600 Nizhny Novgorod, Russia
‡ Department of Basic Science, The University of Tokyo,
Komaba, Meguro-ku, Tokyo 153, Japan

Abstract. Far IR photoresponse of QHE device operating at cyclotron resonance has been investigated. The possibility of the detector band tuning at the simultaneous increase of the magnetic field and the 2D electron concentration (the latter due to the persistent photoconductivity after band-gap illumination) is demonstrated. Time characteristics of the response has been studied.

Introduction

Far infrared (FIR) photoresponse of high mobility two-dimensional (2D) electrons in GaAs/AlGaAs heterostructures under cyclotron resonance (CR) conditions has been the subject of several studies (see, for example [1, 2]). In high magnetic fields when the Fermi level \(E_f\) lies in localized states between two adjacent Landau levels the Hall resistance \(R_H\) is quantized to a multiple of \(\frac{h}{e^2}\) and the longitudinal resistance \(R_{xx}\) vanishes. The finite \(R_{xx}\) emerges when electrons and holes are photoexcited in delocalized states near the level centers above and below \(E_f\). Therefore quantum Hall effect (QHE) devices may serve as an excellent CR detector in FIR range. In the present work the possibility of the detector tuning as well as its time characteristics were investigated.

1. Experimental

The sample under study was a long Hall bar fabricated from high mobility (\(\mu_{4.2\, \text{K}} = 8 \times 10^5 \, \text{cm}^2/\text{Vs}\)) with a width of 50 \(\mu\text{m}\) and a length of 170 mm patterned in zig-zag shape and fitted into an area \(4 \times 4\, \text{mm}^2\) [1]. All measurements were carried out at \(T = 4.2\, \text{K}\). The sample placed in the center of superconducting solenoid was biased by d.c. current of 3 \(\mu\text{A}\). FIR radiation was guided to the sample by stainless steel light pipe. Black body source (\(T = 600\, ^\circ\text{C}\)) was used to reveal the detector sensitivity bands in the whole range of the magnetic fields (up to 6 T). The spectral study was carried out using BOMEM DA3.36 FT spectrometer. The tuning was provided the simultaneous increase of the magnetic field (and correspondingly CR frequency) and the concentration of 2D electrons by band-gap illumination of the sample by near IR (\(\lambda \approx 0.9\, \mu\text{m}\)) radiation of GaAs light emitting diode (LED). The increase of the concentration resulted from the illumination persists after the diode switching off up to the thermal recycling of the sample (persistent photoconductivity effect [3]). Time characteristics of the detector response were studied using broad band FIR emission of hot holes in InGaAs/GaAs multiple-quantum-well (MQW) heterostructure (see, for example [4]) excited by pulsed lateral electric field (about 10 \(\mu\text{s}\) in duration).

2. Results and discussion

Magnetic field dependencies of longitudinal resistance \(R_{xx}\) and photoresponse on the broad band black body source radiation of QHE device (measured by two-terminal scheme) are shown in Fig. 1. 2D electron concentration obtained from the period (in \(1/B\) scale)
Longitudinal resistance $R_{xx}$ and photoresponse on the broadband black body source radiation of QHE device versus the magnetic field.

Evolution of SdH oscillations under the LED radiation, $n$ is a number of pulses. Arrows show the minimum positions corresponding to $\nu = 8$.

Photoresponse spectra measured at magnetic fields near the $R_{xx}$ minimum at $\nu = 6$. Spectra 1–6 correspond to increasing numbers of LED radiation pulses.

of Shubnikov–de Haas (SdH) oscillations is $n_s \approx 2.8 \times 10^{11}$ cm$^{-2}$. It is clearly seen that the response occurs near $R_{xx}$ minima, i.e. at the even values of the filling factor $\nu = 2, 4, 6, 8, 10$ etc. Spectral investigation of the response shows that it consists of sharp CR line of 2 to 3 cm$^{-1}$ in width ($m_e \approx 0.068m_0$, cf. [5]). The response is greater for the lower values of the filling factor mainly due to the increase of the spectral density of the blackbody radiation with the frequency. The absolute measurements of the response at $\nu = 4$ and $\nu = 6$ gave the same value $S_V \approx 10^4$ V/W at NEP $\approx 10^{-11}$ W/Hz$^{1/2}$ that is comparable with the existing semiconductor photoelectric detectors.

Figure 2 illustrates the evolution of SdH oscillations with the increase of the LED radiation amount (number of pulses of 500 $\mu$s in duration). It is clearly seen that $R_{xx}$ minima corresponding to definite values of the filling factor shift to higher magnetic fields. This is a result of the increase of 2D electron concentration due to the persistent photoconductivity effect. The maximum shift reaches 80% in the samples under investigation that opens the possibility of continuous tuning of the detector sensitivity band. Arrows in Fig. 2 indicate the shift of the minimum corresponding to $\nu = 8$ that moves to the higher magnetic fields (and correspondingly frequencies) covering the broad range spreading over initial (before band-gap illumination) position of the minimum corresponding to $\nu = 6$. The tuning is illustrated by Fig. 3 where the photoresponse spectra measured at the magnetic fields near
Fig. 4. Time dependences of photoresponse at \( \nu = 6 \) and \( \nu = 4 \) (a) and \( \nu = 2 \) (b). The dash line is the oscillogram of FIR emission puls.

The \( R_{\text{xx}} \) minimum (\( \nu = 6 \)) are presented. It is clearly seen that the photoresponse consists of the narrow CR line (FWHM about 2 to 3 cm\(^{-1}\)). By simultaneous increasing of band-gap illumination amount and the magnetic field the line is tuned to higher frequencies, its FWHM being approximately the same. Such tuning demonstrates the possibility to utilize QHE detector as spectral analyzer for the FIR range.

Another important characteristic of the detector is its response time. The results of the preliminary investigation of the detector time characteristics are shown in Fig. 4. Operating at the magnetic fields corresponding to the filling factor values \( \nu = 6 \) and \( \nu = 4 \) the detector exhibits rather fast response (\( \tau \leq 5 \mu\text{s} \)) and the response oscillograms repeat that of the voltage pulse applied to the emitter. At the same time at higher magnetic fields at \( \nu = 2 \) the characteristic time determined from the response decay after emitter voltage switching off is much longer, about 200 \( \mu\text{s} \). Moreover it is clearly seen from Fig. 4 that at \( \nu = 2 \) the response continues to increase after the emitter voltage switching off and reaches its maximum with some delay. Such behavior can be naturally explained by the arising in high magnetic field (at \( \nu = 2 \) in contrast to \( \nu = 6 \) and \( \nu = 4 \)) the localized states in between the centers of Landau levels which are responsible for the quantum Hall effect. The significant part of electrons and holes generated by FIR radiation above and under the Fermi level respectively seems to be excited in long living localized states which do not participate in d.c. conductivity. The excited carriers have to relax into delocalized states thus resulting in the above delay of the photoresponse maximum.

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References

Localization length determination for the two-dimensional electrons in the $\delta$-doped GaAs/AlGaAs heterostructures from acoustical studies of the quantum Hall regime

Yu. M. Galperin†, I. L. Drichko†, A. M. Diakonov†, I. Yu. Smirnov†
and A. I. Toropov‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
* Centre for Advanced Studies, 0271 Oslo, Norway and Department of Physics, University of Oslo, 0316 Oslo, Norway
‡ Semiconductors Physics Institute of SD of RAS, 630090 Novosibirsk, Russia

Introduction

In the disordered two-dimensional systems at low temperatures (quantum Hall effect regime) the electrons are localized and the conductivity is of a hopping type. The electronic wavefunction in this case is characterized by a single dimension: the localization length. The localization length is usually obtained from $dc$ conductivity measurements when the variable-length hopping conductivity takes place. In the case of the integer Hall effect (for the magnetic fields corresponding to the middle of the Hall’s plateau) the static ($dc$) conductivity is zero. The high-frequency hopping conductivity manifests itself in this case and it is due to the electronic transitions between the localized states with close energies situated in the vicinity of the Fermi level. The states effective for such transitions form compact pairs, rather distant from each other. There are no transitions between these pairs, that is why there is no current in a static field. However, the high-frequency electric field producing transitions within the pairs, polarizes them, thus the high-frequency conductivity appears to be non-zero. As it has been pointed out by Efros [1], the high-frequency hopping conductivity of the two-dimensional electrons is a complex quantity $\sigma_{xx} = \sigma_1 - i\sigma_2$ and $\sigma_2 > \sigma_1$, $\sigma_1 \propto \xi$, where $\xi$ is the localization length.

Hence, the study of $hf$-conductivity provides one more method to determine the localization length and which is important, in a magnetic field when the $dc$-conductivity is zero. Acoustical methods have proved to be very effective for the study of the $hf$-hopping conductivity. One measures the attenuation $\Gamma$ and the relative velocity change $\Delta V/V$ of a surface acoustic wave (SAW) both attributable to the SAW interaction with the two-dimensional electrons of a heterostructure. The detailed account of the acoustical method is given elsewhere [2]. The simultaneous measurement of $\Gamma$ and $\Delta V/V$ makes it possible to determine $\sigma_1$ and $\sigma_2$ without any need of electric contacts.

Experimental results and discussion

In [3, 4] $\Gamma$ and $\Delta V/V$ have been measured for a SAW at 30 MHz in GaAs/AlGaAs heterostructures with $n = (1.3–2.8) \times 10^{11} \text{ cm}^{-2}$ in a temperature range of 1.5–4.2 K and in a magnetic field up to 7 T. Figure 1(a) shows the $\sigma_1$ and $\sigma_2$ magnetic field dependencies at $T = 1.5$ K. $\sigma_1$ and $\sigma_2$ have been calculated from $\Gamma$ and $\Delta V/V$ using the relations of [3, 4]. The $\sigma_1$ and $\sigma_2$ dependencies on $\nu$, the filling factor, for different temperatures and at $\nu$ near to $\nu = 2$ are presented in Fig. 1(b) ($H = 5.5$ T, $\nu = nch/eH$, $c = 3 \times 10^{10}$ cm/s, $h = 6.62 \times 10^{-27}$ erg $\cdot$ s, $e = 4.8 \times 10^{-10}$ CGS). One can see from this figure that for
Fig. 1. (a) The experimental dependences of $\sigma_1$ and $\sigma_2$ on the magnetic field $H$, $T = 1.5$ K, $f = 30$ MHz; (b) Dependences of $\sigma_1$ and $\sigma_2$ on the filling factor near $\nu = 2$ at $T = 1.5 - 4.2$ K, $f = 30$ MHz; (c) The dependences of $\lg(F_1) = \lg(\sigma_1 - \sigma_1^{eq})$ and $\lg(F_2) = \lg(\sigma_2 - \sigma_2^{eq})$ vs $\nu$ near $\nu = 2$, $T = 1.5$ K.

$v = 2$ the relation $\sigma_2 > \sigma_1$ earlier predicted by Efros for the case of relaxation $hf$-hopping conductivity is really demonstrated experimentally.

In our case we also deal with the relaxation, or phonon-assisted, $hf$ absorption due to phonon-assisted transitions which lead to a lag of the levels populations with respect to microwave-induced variation in the inter-level spacing. One can obtain

$$\sigma_1 = \pi^2 g^2 \xi^3 \omega_e^4 \left( \mathcal{L}_T + \mathcal{L}_\omega / 2 \right)^2 / (2 \varepsilon_s).$$

Here $g$ is (constant) single-electron density of states at the Fermi level, $\xi$ is the localization length of the electron state, $\mathcal{L}_T = \ln J / kT$, $J$ is a typical value of the energy overlap integral which is of the order of the Bohr energy, while $\mathcal{L}_\omega = \ln(\gamma_0/\omega_c)$; $\varepsilon_s$ is the dielectric constant of GaAs. Eq. (1) is valid provided the logarithmic factors are large. Note that (1) is similar to the one obtained in Ref. [1], however it differs by some logarithmic factors and numerical factor 1/4.

Analysis of $\sigma_2(\omega)$ is a bit more complicated because virtual zero-phonon transitions give a comparable contribution. The analysis gives the following ratio,

$$\frac{\sigma_2}{\sigma_1} = \frac{2L_{cw} (L_T^2 + L_T L_\omega + L_\omega^2 / 12) + 4c L_T^2 L_c}{\pi (L_T^2 + L_T L_\omega + L_\omega^2 / 4)}.$$  

Here $L_c = \ln(\hbar \omega_c / kT)$, $\omega_c$ is the cyclotron frequency, while $c \geq 1$ is a numerical factor depending on the density of states in the region between the Landau levels. Using the estimate for $\gamma_0$ from Ref. [5],

$$\gamma_0 = 4 \pi e^2 K^2 kT / \varepsilon_s h^2 V,$$

valid for the piezoelectric relaxation mechanism, as well as other parameters relevant to the present experiment, one concludes that in the hopping regime $\sigma_2 \geq \sigma_1$. This conclusion
agrees with the experimental results obtained for the middles of the Hall plateaus at 5.5 T and 2.7 T and make us sure that the conductance mechanism in these regions is hopping.

Given an experimental value of $\sigma_1$, one can obtain from Eq. (1) the localization length $\xi$ provided the single-electron density of states, $g$, is known for for given values of magnetic field. This quantity has been obtained from the temperature dependence measurements of thermally-activated dc conductivity [6, 7]. It has been shown that for small even filling factors the density of states in the plateau regions is finite and almost field-independent.

Using the density of states versus mobility curve from Ref. [6] obtained for a sample similar to ours, we estimate the DOS as $g = 2.5 \times 10^{24} \, \text{cm}^{-2} \cdot \text{erg}^{-1}$. On the other hand, according to Ref. [7], the DOS as a function of magnetic field $H$ can be expressed by the interpolation formula

$$g(H) = g_0 \left[ 1 + \sqrt{\mu H} \right],$$

where $\mu$ is the mobility of the 2D-electrons while $g_0 = m/(\pi \hbar^2)$ is the 2D DOS at $H = 0$. From Eq. (3) we obtain for $H = 5.5$ T the DOS $g = 1.7 \times 10^{24} \, \text{cm}^{-2} \cdot \text{erg}^{-1}$.

Using the first estimate for the DOS one obtains $\xi = 6.5 \times 10^{-6}$ cm, that is about 1.6 times greater than the spacer thickness, $l_{sp} = 4 \times 10^{-6}$ cm. On the other hand, it is the spacer width, which characterizes the random potential correlation length in the 2DEG layer. Hence, this fact contradicts to our interpretation of the experimental results in terms of pure nearest-neighbor pair hopping.

To solve the controversy, we assume that the $hf$ hopping conductivity of the 2DEG channel is shunted by the hopping along the doping Si $\delta$-layer.

This assumption can be substantiated as follows. Let us suppose that in the middle of the Hall plateau $\sigma_1$ is entirely determined by the hopping conductivity along the Si $\delta$-layer. Such a contribution is weakly dependent on magnetic field because the latter is too weak to deform substantially the wave functions of Si-dopants. Then the contribution to the Si- $\delta$-layer’s contribution to $\sigma_1$ associated with 2D-layer is just a difference between the experimentally measured $\sigma_1$ in a given $H$ and its value at $v = 2$.

Let us analyze dependences of the differences $F_1 \equiv \sigma_1 - \sigma_1^{\nu=2}$ and $F_2 \equiv \sigma_2 - \sigma_2^{\nu=2}$ on the filling factor $\nu$. The plots of $\lg F_1$ versus $\nu$ are shown in Fig. 1(c). Both curves tend to straight lines, and in this way they can be extrapolated to $\nu = 2$. On the other hand, as it has been mentioned, at small even $\nu$ the shunting of 2DEG by Si $\delta$-layers is important. Consequently, $F_1$ are determined by the contributions of the 2D electron channel. Indeed, the Si- $\delta$-layer’s contributions to $\sigma_1$ and $\sigma_2$ at $\nu = 2$ are $4 \times 10^{-7} \, \Omega^{-1}$ and $2.4 \times 10^{-6} \, \Omega^{-1}$, respectively. These quantities are more than order of magnitude larger than the quantities $F_1 = 10^{-8} \, \Omega^{-1}$ and $F_2 = 5 \times 10^{-8} \, \Omega^{-1}$ obtained by extrapolation of the experimental curves. Using the extrapolated values of $F_1$ and $F_2$ to extract the 2DEG contributions to $\sigma_1$ and $\sigma_2$, one can calculate the electron localization length at $\nu = 2$ from Eq. (1).

It should be noticed here that the experimental ratio $F_2/F_1 = 5$ is close to the theoretical value 4.2 coming from Eq. (2). The localization length at $\nu = 2$ obtained in this way is $\xi = 2 \times 10^{-6}$ cm, which is a half of the spacer width. This estimate makes realistic the “two-site model” which we have extensively used. Construction similar to that of Fig. 1(c) ($T = 1.5$ K) has been performed for all temperatures of the 1.5–4.2 K range. The temperature dependence analysis of $\sigma_2$ and $\sigma_1$ obtained as an extrapolation for $\nu = 2$ has shown that both components do not depend on $T$. This fact confirms the existence in this region of a hopping $hf$ conductivity (see Eq. (2)). Both $\sigma_1$ and $\sigma_2$ begin to grow at $T > 3.2$ K. The idea that the 2D $hf$-conductivity of heterostructures in magnetic fields corresponding to small $\nu$ is shorted up by the hopping conductivity along the Si-$\delta$-layers
could be confirmed by simple calculations of the localization length in the Si-δ-layers. Calculations with the use of Eq. (1) provide the value $\xi = 7 \times 10^{-7}$ cm, which is of the same order as the interatomic distance in a δ-layer. So one could use the same term “two-site model” when considering the hopping conductivity along the δ-layer. It should be emphasized, however, that from the above value of $\xi$ in 2D channel the hopping length $r_{\omega}$ is estimated 1.4 $\times$ 10$^{-5}$ cm. Consequently, there is an interplay between hops to the nearest and more remote neighbors. A more rigorous theory for this situation should be worked out. This theory should also explain why the dependences $\sigma_1(H)$ and $\sigma_2(H)$ in the vicinity of $\nu=2$ appear different – the $\sigma_1(H)$-dependence is more pronounced than the $\sigma_2(H)$-one. According to the “two-site model”, both dependences are determined by the $\xi(H)$ dependence and should be similar. Indeed, the ratio (2) is almost field-independent. It follows from the experiments that there exists an additional mechanism leading to the pronounced decrease of $\sigma_2$ as the Fermi level falls into the extended states. A probable mechanism is thermal activation of electrons from the Fermi level to the upper Landau band, leading to a decrease of the number of pairs responsible for the hopping conductivity, and to a screening of the electric field of SAW.

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References

Temperature emerging of combination frequencies in quasi-2D
Shubnikov–de Haas effect

N. S. Averkiev, L. E. Golub and S. A. Tarasenko
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The theory of the Shubnikov–de Haas effect is presented for quasi-2D systems when several subbands of size quantization are occupied. The temperature behavior of oscillation spectrum is considered. It is shown that the main harmonics with frequencies defined by subband fillings damp exponentially and the non-damping oscillations with differential frequencies proportional to energy gaps between subbands manifest themselves. They are harmonics, which became dominating at the following increasing of temperature.

It is known that the Shubnikov–de Haas effect displaying in the conductivity oscillations under quantized magnetic fields is caused by consequently crossing of the Fermi level by Landau subbands. The period of oscillations is found proportional to the carrier concentration what allows to use this phenomenon for its determination. Temperature increasing leads to broadening of carrier distribution and to suppression of the conductivity oscillations.

The qualitatively new situation takes place in two-dimensional (2D) structures where two or more subbands of size quantization are occupied. In this case, the oscillations appear with several frequencies determined by filling of subbands. It is important that the harmonics corresponding to the excited subbands exist even at relatively small degree of their filling due to intersubband scattering. Besides, the oscillations with frequencies proportional to energy gaps between size-quantized levels may appear in quasi-2D systems. The peculiarity of these harmonics is that they do not damp with temperature increasing [1].

The aim of this communication is the theoretical consideration of the magnetoconductivity temperature behavior in the presence of intensive intersubband scattering. We investigate an electron gas in 2D structure with two occupied subbands. Scattering is assumed to be isotropic and a spin splitting in a magnetic field is neglected.

In 2D systems, the Shubnikov de Haas oscillations are observed in fields $\omega_c \tau \leq 1$, where $\omega_c$ is the cyclotron frequency and $\tau$ is the momentum relaxation time determined by both intra- and intersubband scattering. To calculate the conductivity tensor, it is convenient to use the Green function formalism. The main and the combine frequencies appear in the first and second order in the parameter $\exp(-\pi/\omega_c \tau)$ respectively.

The Green function of quasitwodimensional electron gas at finite temperature in the Matsubara technique has in energy representation the form

$$ G_j(\epsilon_m, \xi_{j,n}) = [i\epsilon_m - \xi_{j,n} - \lambda_j'(\epsilon_m)]^{-1} $$

where $\epsilon_m = \pi T(2m + 1)$, $T$ is the temperature in energy units, $m$ is a whole number, $\xi_{j,n} = \hbar \omega_c (n + 1/2) - \mu_j$ is the energy distance between the $n$-th Landau level in the $j$-th subband and the chemical potential level. The self-energy parts, $\lambda_j'(\epsilon_m)$, are found from the system of integral Dyson equations and has the following form in the second order in
\[ x_j(\epsilon_m) = -\sum_{j'=1}^{M} \frac{i\hbar}{\tau_{jj'}} \left\{ 1 + 2 \exp \left( -\frac{\pi}{\omega_c \tau_j} \right) \exp \left[ 2\pi i \left( \frac{\mu_{j'}}{\hbar \omega_c} - \frac{1}{2} \right) \sign m \right] \right. \\
\times \exp \left( -\frac{2\pi |\epsilon_m|}{\hbar \omega_c} \right) + \left( 1 - \frac{2\pi}{\omega_c \tau_j} \right) \exp \left( -\frac{2\pi}{\omega_c \tau_j} \right) \\
\left. \times \exp \left[ 4\pi i \left( \frac{\mu_{j'}}{\hbar \omega_c} - \frac{1}{2} \right) \sign m \right] \exp \left( -\frac{4\pi |\epsilon_m|}{\hbar \omega_c} \right) \right) \sign m . \] (2)

Here \( \tau_{jj'} \) and \( \tau_{jj''} \) are the times of intra- and intersubband scattering in the zero magnetic field, and \( \tau_j \) is the total scattering time in the \( j \)th subband.

Calculating the polarization operator with help of (1), one can obtain the expression for the conductivity tensor. At zero temperature, the main contribution appears from the first order in \( \exp(-\pi/\omega_c \tau) \) terms [2]. The terms of higher orders are small and therefore are not observable. However, at raising of temperature, the amplitudes of the main harmonics decrease exponentially and the non-damping with temperature oscillations of higher orders may manifest themselves. In the second order in \( \exp(-\pi/\omega_c \tau) \), the former have the form

\[ \sigma_{xx}^{(nd)} = \sum_{j=1}^{M} \frac{N_j e^2 \tau_j/m}{[1 + (\omega_c \tau_j)^2]^{1/2}} \left\{ \frac{2\tau_j}{\tau_{jj'}} - 1 + (\omega_c \tau_j)^2 \left\{ 1 - \frac{6\tau_j}{\tau_{jj'}} \right\} \right\} \\
\times \sum_{j' \neq j} \frac{\tau_j}{\tau_{jj'}} 2 \cos \left( \frac{2\pi \mu_{jj'} - \mu_{jj'}}{\omega_c} \right) \exp \left( -\frac{\pi}{\omega_c} \left( \frac{1}{\tau_j} + \frac{1}{\tau_{jj'}} \right) \right) \\
\times \sum_{j' \neq j} \frac{\tau_j}{\tau_{jj'}} 2 \cos \left( \frac{2\pi \mu_{jj'} - \mu_{jj'}}{\omega_c} \right) \exp \left( -\frac{\pi}{\omega_c} \left( \frac{1}{\tau_j} + \frac{1}{\tau_{jj'}} \right) \right) \\
(3)

\[ \sigma_{xy}^{(nd)} = -\sum_{j=1}^{M} \omega_c \tau_j \sum_{j' \neq j} \frac{N_j e^2 \tau_j/m}{[1 + (\omega_c \tau_j)^2]^{1/2}} \left\{ \frac{2\tau_j}{\tau_{jj'}} - 1 + (\omega_c \tau_j)^2 \left\{ 3 - (\omega_c \tau_j)^2 \right\} \right\} \\
\times \sum_{j' \neq j} \frac{\tau_j}{\tau_{jj'}} 2 \cos \left( \frac{2\pi \mu_{jj'} - \mu_{jj'}}{\omega_c} \right) \exp \left( -\frac{\pi}{\omega_c} \left( \frac{1}{\tau_j} + \frac{1}{\tau_{jj'}} \right) \right) \\
(4)

where \( m \) is the carrier effective mass and \( N_j \) are the concentrations in subbands. At further increasing of temperature, these oscillations dominate.

In Figure 1 the dependences of the resistance, \( \rho_{xx} \),

\[ \rho_{xx} = \frac{\sigma_{xx}}{\sigma_{xx}^2 + \sigma_{xy}^2} \\
(5)\]
on magnetic field are presented for the structure with two occupied levels of size quantization. The components \( \sigma_{xx} \) and \( \sigma_{xy} \) include the terms of zeroth, first and temperature-non-damping second order in the parameter \( \exp(-\pi/\omega_c \tau) \). At calculation we assumed that the carrier concentration in the excited subband is relatively small, \( N_2/N_1 = 0.1 \), intersubband scattering is intensive, \( \tau_1 = \tau_2 \), \( \tau_1/\tau_{12} = 0.5 \), and the condition of good conductor is executed in both subbands: \( \mu_1 \tau_1/\hbar = 50, \mu_2 \tau_2/\hbar = 5 \).

Figure 1(a) corresponds to the case of zero temperature. The oscillations from both subbands are clear. The low-frequency harmonic exists due to intensive intersubband scattering.
Fig. 1. Magnetoresistance oscillations in the presence of intensive intersubband scattering at different temperatures. (a) $T = 0$, (b) $T = 2T_D$.

In Figure 1(b) the case of $T = 2T_D$ is presented, where $T_D = h/2\pi\tau_1$ is the Dingle temperature. The amplitudes of the main and differential harmonics are comparable, and therefore the beats are observable because the differential frequency is close to the main one due to the small filling of the excited subband.

In conclusion, the effect of temperature emerging of combine frequencies in the quasi-2D Shubnikov–de Haas effect is described consequently in the communication.

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References


Electron spin relaxation in zinc-blende heterostructures

N. S. Averkiev†, L. E. Golub†‡ and M. Willander‡

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physical Electronics and Photonics, Department of Microelectronics and Nanoscience, Chalmers University of Technology and Göteborg University, SE-412 96, Göteborg, Sweden

Abstract. Spin relaxation in-plane anisotropy is predicted for heterostructures based on zinc-blende semiconductors. It is shown that it manifests itself especially brightly if the two spin relaxation mechanisms (Dyakonov–Perel and Rashba) are comparable in efficiency. It is demonstrated that for the quantum well grown along the [001] direction, the main axes of spin relaxation rate tensor are [110] and [1 10].

Introduction

Spin relaxation processes have significant effect in optical and kinetic properties of semiconductors. They play important role in optical orientation of electrons and nuclei [1] and in anomalous magnetoresistance caused by weak localization [2]. Now, big interest to spin dynamics and relaxation exists due to a spin transistor creation attempts. Both theoretical calculations and experimental data analysis have been carried out assuming that one spin relaxation mechanism dominates only. Therewith in spite of the strong anisotropy of spin-orbit scattering, the relaxation times of spin lying in the plane of a heterostructure with zinc-blende lattice turn out to be independent on orientation with respect to crystallographic axes.

This communication is devoted to an investigation of spin relaxation processes in real heterostructures when several mechanisms of spin-orbit scattering exist. We show that contributions of these mechanisms interfere and their simultaneous action leads to the strong anisotropy of spin relaxation even in the plane of a quantum well (QW).

1. Theory

In zinc-blende semiconductors, spin relaxation of electrons is well known to be due to spin-orbit splitting of conduction band. In a bulk crystal, the splitting is cubic in wave vector. In a QW structure, the corresponding Hamiltonian has to be averaged over the motion along the growth axis. We consider the QW grown along z-direction parallel to [001] and choose x and y directions coinciding with crystallographic axes. At relatively small carrier concentrations, one can neglect cubic in 2D wave vector terms and the Hamiltonian has the form:

\[ H_1 = a_1 (\sigma_x k_x - \sigma_y k_y) . \]

Here \( \sigma_i \) \( (i = x, y) \) is the Pauli matrix, \( k_i \) is the wave vector component in the plane of the QW. \( a_1 \) is a constant which is determined by both bulk properties of the semiconductor and the value of \( k_z^2 \) averaged over the z-motion.
In asymmetrical heterostructures, there is a contribution to the Hamiltonian which is absent in the bulk [3]:

\[ H_2 = a_2 (\sigma_x k_y - \sigma_y k_x), \]  

(2)

where \( a_2 \) is the constant determined by heterointerface properties. It is equal to zero in a symmetrical structure and is proportional to the barrier height difference in an asymmetrical QW or to the electric field in a triangular QW. It is possible to change the value of \( a_2 \) by varying a gate voltage applied to the system or by another changing of the structure symmetry.

The relationship between the constants \( a_1 \) and \( a_2 \) may be different in different systems. \( a_1 \) may be much larger than \( a_2 \) [4], much less than \( a_2 \) [5] or they may be comparable [6].

The most interesting case realizes when the constants \( a_1 \) and \( a_2 \) are equal to each other by absolute value:

\[ a_1 = \pm a_2. \]

In this case, the total spin-orbit Hamiltonian is:

\[ H' = H_1 + H_2 = a_1 (\sigma_x \mp \sigma_y)(k_x \pm k_y). \]  

(3)

Therefore for electrons with the spin along [110] or [1\( \bar{1} \)0], \( H' = 0 \) for any value of the wave vector, \( \mathbf{k} \). It means that the spin relaxation time caused by both mechanisms, (1) and (2), is infinite. In other words, the spin relaxation mechanisms due to splittings (1) and (2) suppress each other totally.

In the general case, when \( a_1 \) and \( a_2 \) are arbitrary, we have obtained the following equations for spin dynamics [7]:

\[ \dot{S}_z = -\frac{S_z}{\tau_z}, \quad \dot{S}_x \pm \dot{S}_y = -\frac{S_x \pm S_y}{\tau_{\pm}}. \]  

(4)

Here

\[ \frac{1}{\tau_z} = C (a_1^2 + a_2^2), \quad \frac{1}{\tau_{\pm}} = \frac{C}{2} (a_1 \pm a_2)^2, \]  

(5)

and \( C \) is determined by properties of the scattering potential and electron distributions in spin sublevels. Note that the equation (4) is valid at times longer than the momentum relaxation time but shorter than the spin relaxation times [7].

2. Discussion

It is seen from Eq. (5), that if \( a_1 = \pm a_2, \tau_\pm = \infty \), and the other time, \( \tau_z \), is equal to \( \tau_z \). Besides, one can see the spin relaxation anisotropy even in the plane of the heterostructure. All three times, \( \tau_+, \tau_- \) and \( \tau_z \) are different in the general case.

One can also see from (5) that at only one spin relaxation mechanism, when \( a_1 = 0 \) or \( a_2 = 0 \), spin relaxation is isotropic in the plane of the heterostructure:

\[ \tau_+ = \tau_- = 2 \tau_z. \]  

(6)

It means that, despite the cubic anisotropy included into the Hamiltonian \( H_1 \) or \( H_2 \), it averages and does not exhibit itself in spin relaxation. In the presence of both spin relaxation mechanisms, on the contrary, the cubic anisotropy does lead to the difference between the spin relaxation times for spin lying in the plane of the heterostructure (see (5)).
The spin relaxation anisotropy results from the initial $T_d$ symmetry of the zinc-blende semiconductor. For this reason, the similar effect can take place in a strained bulk crystal. The corresponding Hamiltonian linear in 3D wave vector, $\mathbf{k}$, and components of an elastic strain tensor, $u_{ij}$, has the form:

$$H'(u) = A_1 u_{ii} \left( \sigma_{i+1} k_{i+1} - \sigma_{i+2} k_{i+2} \right) + A_2 u_{ij} \left( \sigma_{i} k_{j} - \sigma_{j} k_{i} \right).$$

(7)

Here $i, j = x, y, z$, $i + 3 \rightarrow i$, $A_1$ and $A_2$ are constants. Deriving the spin dynamics equations for this spin-orbit Hamiltonian, one can obtain three different spin relaxation times. It can be shown that the maximum anisotropy may be achieved if

$$A_1 u_{xx} = A_1 u_{yy} = -\frac{A_1 u_{zz}}{2} = A_2 u_{xy}/3$$

(8)

with the rest of $u_{ij} = 0$. Therewith two spin relaxation times are equal to each other and the third is infinite. Note that the tensor $u_{ij}$ determined by (8) may be obtained by applying two uni-axial strains along the axes [001] and [110] and they are not restricted to uni-axial strain along any axes.

3. Conclusion

The possibility for spin relaxation suppression was noted in Ref. [8] for a QW grown along [110] direction when the spin is oriented along the same axis. The present work shows that the spin relaxation rate also decreases for [110] direction, but in a QW grown in the symmetrical direction [001]. Therefore this decrease takes place for the spin lying in the plane of the QW.

Analyzing weak localization effect, the authors of Ref. [9] showed that the mechanisms (1) and (2) suppress each other in anomalous magnetoresistance, but they are additive in spin relaxation. The present analysis shows that the suppression occurs in the spin relaxation also. Besides, we have found that spin relaxation is anisotropic even in the plane of the QW.

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Low field negative magnetoresistance in double layer structures

G. M. Minkov†, A. V. Germanenko†, O. E. Rut†, O. I. Khrykin‡, V. I. Shashkin‡ and V. M. Danil’tsev‡
† Institute of Physics and Applied Mathematics, Ural State University, 620083 Ekaterinburg, Russia
‡ Institute of Physics of Microstructures of RSA, 603600 N. Novgorod, Russia

Abstract. The weak localization correction to the conductivity in coupled double layer structures was studied both experimentally and theoretically. The statistics of the closed paths was determined from analysis of the magnetic field and temperature dependencies of the negative magnetoresistance for magnetic field perpendicular and parallel to the structure plane. The comparison of the results with the results of computer simulation of carrier motion at scattering shows that inter-layer tunneling plays decisive role in the weak localization.

The tunneling between 2D layers is one of fundamental features of double layer structures. It changes the quantum corrections to the conductivity, especially in a magnetic field parallel to the 2D plane.

It is well known [1] that the interference of electron waves scattered along closed trajectories in opposite directions (time-reversed paths) produces a negative quantum correction to the conductivity. An external magnetic field, \( \mathbf{B} \parallel \mathbf{n} \), where \( \mathbf{n} \) is the normal to 2D layer, gives the phase difference between pairs of time-reversed paths which is proportional to the area enclosed \( \varphi = 2\pi \mathbf{BS}/\Phi_0 \) and thus destroys the interference. The magnetic field dependence of the negative magnetoresistance is determined by the statistics of the closed paths: the area distribution function, \( W(S) \), and area dependence of the average length of closed paths, \( \bar{L}(S) \) [2]. Just these functions can be obtained both from the experimental data [3] and from computer simulation of carrier motion at scattering [2].

In a magnetic field parallel to the 2D layer (\( \mathbf{B} \perp \mathbf{n} \)) the product \( \mathbf{BS} \) is equal zero therefore the magnetic field does not destroy the interference and the negative magnetoresistance is absent in single layer structures for this magnetic field orientation [4].

In coupled double layer structures, the tunneling between layers leads to arising of the closed paths where an electron moves over one layer then over another one and returns to the first one. For this paths the product \( \mathbf{BS} \) in the parallel magnetic field is non-zero and negative magnetoresistance in parallel magnetic field has to appear.

The double well heterostructure GaAs/In\(_x\)Ga\(_{1-x}\)As was grown by Metal Organic Vapor Phase Epitaxy on semi-insulator GaAs substrate. The heterostructure consists of a 0.5 \( \mu \)m-thick undoped GaAs epilayer, a Si \( \delta \)-layer, a 75 \( \AA \) spacer, a 100 \( \AA \) In\(_{0.08}\)Ga\(_{0.92}\)As well, a 100 \( \AA \) barrier of undoped GaAs, a 100 \( \AA \) In\(_{0.08}\)Ga\(_{0.92}\)As well, a 75 \( \AA \) spacer, a Si \( \delta \)-layer and 1000 \( \AA \) cap layer of undoped GaAs. The measurements were performed in the temperature range 1.5–4.2 K at low magnetic field up to 0.4 T with discrete \( 10^{-4} \) T for two orientations: (i) the magnetic field was perpendicular to the structure plane (\( \mathbf{B} \parallel \mathbf{z} \)), (ii) the magnetic field was parallel to the structure plane and current (\( \mathbf{B} \parallel \mathbf{x} \)). Additional high field measurements were also made to characterize the structures. The electron densities in the wells have been
determined from the Fourier analysis of the Shubnikov–de Haas oscillations and consist of 4.5 × 10¹¹ cm⁻² and 5.5 × 10¹¹ cm⁻². The Hall mobility was about 5000 cm²/(V·s).

The magnetic field dependencies of the in-plane magnetoconductivity for magnetic field perpendicular (\( \Delta \sigma(B_z) \)) and parallel (\( \Delta \sigma(B_x) \)) to the structure plane are presented in Fig. 1. One can see the negative magnetoresistance is observed for both magnetic field orientations and the effects are comparable in magnitude in contrast to the case of single layer structure. For \( B < 0.3-0.5 \) T the main contribution to the negative magnetoresistance comes from the interference correction. In this case the magnetic field dependence of the negative magnetoresistance is determined by the statistics of closed paths \([5,2]\) and for double layer structure it can be written as follows

\[
\sigma(B_i) = \sigma_0 + \delta \sigma(B_i) = \sigma_0 - 4\pi l^2 G_0 \int_{-\infty}^{\infty} dS_i W(S_i) \exp \left( -\frac{\tilde{L}}{l_{\psi}} \right) \cos \left( \frac{2\pi B_i S_i}{\Phi_0} \right).
\]

Here, \( i = x, z \), \( G_0 = e^2/(2\pi^2\hbar) \), \( \sigma_0 \) is the classical Drude conductivity, \( l = v_F\tau \), \( l_{\psi} = v_F\tau_{\psi} \), \( v_F \) is the Fermi velocity, and \( \tau, \tau_{\psi} \) stand for the momentum relaxation and phase breaking time, respectively. The value of \( \tilde{L} \) is the function of \( S \) and \( l_{\psi} \) and it was defined by Eq. (6) in Ref. [2]. It should be noted that this expression is valid when the probability to escape the plane at a collision (\( t \)) is less than 0.5.

Thus, for the magnetic field perpendicular to the structure plane, \( \mathbf{B} = B_z \), the contribution to the magnetoresistance is determined by \( z \)-component of \( S \) only and for parallel magnetic field, \( \mathbf{B} = B_x \), it is determined by \( x \)-component of \( S \).

One can see from the above expression that the Fourier transform of \( \delta \sigma(B)/G_0 \)

\[
\Phi(S_i, l_{\psi}) = \frac{1}{\Phi_0} \int_{-\infty}^{\infty} dB_i \frac{\delta \sigma(B_i)}{G_0} \cos \left( \frac{2\pi B_i S_i}{\Phi_0} \right) = 4\pi l^2 W(S_i) \exp \left( -\frac{\tilde{L}}{l_{\psi}} \right)
\]

carries an information on \( W(S_i) \) and \( \tilde{L}(S_i, l_{\psi}) \). Because the value of \( l_{\psi} \) tends to infinity when the temperature tends to zero, the extrapolation of \( \Phi(S_i, l_{\psi}) \)-vs-\( T \) curve to \( T = 0 \) gives the value of \( 4\pi l^2 W(S_i) \). The \( \tilde{L}(S_i, l_{\psi})/l_{\psi} \) ratio for given \( S_i \) can be then obtained as \( \ln(4\pi l^2 W(S_i)) - \ln(\Phi(S_i, l_{\psi})) \). In more detail this method of analysis of experimental data was described in Ref. [3].
The area distribution functions and area dependence of the ratio \( \bar{L}(S_x)/\bar{L}(S_z) \) are plotted in Fig. 2 and Fig. 3. The significant difference in \( W(S_z) \) and \( W(S_x) \) dependencies stands out. The \( W(S_z) \) curve shows a much steeper decline at \( S_z > (0.8 - 1) \times 10^{-10} \text{ cm}^2 \). Other feature of the statistics of the closed paths in double layer structure is the fact that for given \( S \) the values of \( \bar{L}(S_x) \) are significantly larger than \( \bar{L}(S_z) \). It is clear that the behaviour of enclosed areas \( S_z, S_x \) is analogous and that at \( S_x = S_z \), \( W(S_x) > W(S_z) \) and the average length of the trajectories \( L(S_x) \) is greater than \( L(S_z) \).

As was shown in Ref. [2] the area distribution function, the area dependence of average lengths of closed paths, and the weak localization magnetoresistance can be obtained by the computer simulation of a carrier motion over 2D plane. In our case the model double layer system has been conceived as two identical plains with randomly distributed scattering centers with a given total cross-section. We take into account that after every collision the particle has two possibilities: it transits from one plane to another with a probability \( t \) and moves over the second plane or it remains in the plane with probability \( (1 - t) \), changing the motion direction only.

The simulation results for the inter-layer transition probability \( t = 0.1 \) are presented in Figs. 1–3. It is clearly seen that the used theoretical model reproduces all the peculiarities of the experimental data: \( W(S_z) \) is close to that obtained experimentally; \( W(S_x) \) shows steep decline at large \( S \); and the \( \bar{L}(S_x)/\bar{L}(S_z) \) ratio is close to experimental results. In Fig. 1 the calculated magnetoconductance is presented for both magnetic field orientations.
for temperatures 1.5 K and 4.2 K. Perfect agreement is evident. Thus, the results presented show that the inter-layer tunneling plays decisive role in the weak localization in coupled double layer structures.

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[4] In reality, in a magnetic field parallel to the 2D layer the negative magnetoresistance is observed, but the magnitude of the effect is significantly less. In the structures with one subband filled the effect arises due to asymmetry of the wave function or scattering potential with respect to the center of 2D layer [Vladimir I Fal’ko, J. Phys.: Condens. Matter 2 3797 (1990)].
Physics of quantum well and quantum dot infrared photodetectors

V. Ryzhii
Computer Solid State Physics Laboratory, University of Aizu,
Aizu-Wakamatsu, 965-8580, Japan

Abstract. We review the recent studies of physical effects in quantum well and quantum dot infrared photodetectors utilizing intersubband transitions.

Introduction

Electron and hole processes in semiconductor superlattices have been the topic of extensive experimental and theoretical studies for almost three decades. This is due to a variety of interesting physical effects in such systems and their device applications. A great deal of attention has also been paid to the electron (hole) phenomena in multiple quantum well (QW) structures with a weak coupling between QWs. These QW structures are used in infrared photodetectors utilizing intersubband transitions [1]. QW infrared photodetectors (QWIPs) on the base of AlGaAs/GaAs and other heterostructures utilizing intersubband absorption have been successfully implemented for wavelengths in the range 4−28 µm [1, 2]. The application of large QWIP arrays in infrared cameras is in the developing stage (see, for example [3]). The basic physics of QWIPs has been well documented [1, 4, 5]. However, as shown recently, the spectrum of physical effects arisen in QWIPs under different conditions is wider than expected and their in-depth understanding and utilization need further thorough investigation. Despite QWIPs have been successful in different applications, the replacement of QWs by arrays of quantum dots (QDs) in infrared photodetectors (QDIPs) is very promising. QDIPs have been predicted to have some important advantages over QWIPs [6] such as the sensitivity to normally incident radiation, lower dark current, and higher photoelectric gain. Different InAs/GaAs, InGaAs/GaAs, InGaAs/InGaP, and Si/Ge QDIPs have been recently fabricated and experimentally studied by several groups [7–16]. In this paper we review the recent results concerning physical aspects of QWIP and QDIP operation focusing primarily on the electron transport, capture, and injection phenomena in such devices.

1. Principles of QWIP and QDIP operation

The QWIP structure consists of a number of doped QWs separated by relatively thick undoped layers forming the inter-well barriers. Such a QW structure is clad by doped layers playing roles of the emitter and collector contacts. In QDIPs, arrays of QDs are used instead of QWs. Generally, the principles of operation of QWIPs and QDIPs are similar. Actually, their operation principles are also similar to those of the photodetectors utilizing impurity excitation. The excitation (thermal- and photoexcitation) of electrons from the bound states in QWs or QDs ensures the occurrence of mobile electrons propagating over the continuum states. The energies of intersubband bound-to-bound and bound-to-continuum transitions are in the mid- or far infrared ranges of spectrum. Under applied voltage, the generation of mobile electrons results in the electron current across the QW or QD structure.
additional ionization of QWs or QDs due to the increase of the excitation rate (say, because of the increase in the flux of infrared photons) leads to the redistribution of the electric potential in the structure. This, in turn, gives rise to the increase in the electric field near the emitter contact. The latter leads to strengthening of the electron injection from the emitter contact. The total electron current is determined by both the excited and injected electrons. In the steady-state conditions, the concentration of electrons in QWs (QDs) and, hence, the space charge in the structure, are maintained by the balance between the electron excitation and the capture of previously excited and injected electrons. The type of the injection from the emitter contact to the QWIP (QDIP) active region depends on their band alignment. The emitter layer in QWIPs is primarily made of a material with narrower energy gap than that of the inter-well barriers, so that the injection of electrons into the QWIP active region is associated with the tunneling through the barrier separating the emitter contact layer and the extreme (first) QW [1]. Contrary, in QDIPs the emitter layer is usually made of the same material as the material of the barriers between QDs [7–16]. Because of this, the electron injection in such QDIPs is due to thermionic injection over the barrier created by the charged QD layer (enriched by electrons) adjacent to the emitter contact [6].

2. Heating of electrons and their capture

As the capture processes in QWIPs and QDIPs play an important role in the performance of these devices, the dependence of the capture rate on the local value of the electric field can be a significant factor. Apart from the effect of electric field on the photoelectric gain, it essentially influences the spatial distribution of the self-consistent electric field in the device active region. There are two mechanisms of the effect of electric field. First, the local electric field influences the probability of such elementary processes as the unbound-bound phonon-assisted electron transitions. Secondly, the electric field in the QWIP (or QDIP) active region usually leads to a significant heating of electrons. As a result, the fraction of low energy electrons which can be captured, say, with the emission of optical phonons drastically decreases with increasing electric field. Taking into account that the direct field effect is not so strong in the range of normally used electric fields, the electron heating can be the most significant one. The results of ensemble Monte Carlo particle study of the heating mechanism yield nearly exponential dependence of the electron capture rate on the electric field [17] which is consistent with the experimental data [18, 19]. A strong electric-field dependence of the capture rate can be one of the most important factors determining the QWIP current-voltage characteristics [20] both in dark conditions and under illumination. Residual donors in the barriers and nonuniform distribution of donors in QWs (associated with features of the growth processes and leading to an asymmetric form of the QWs [21]) can markedly complicate the electron heating and, hence, the electric-field dependence of the capture rate [22].

3. Contact and space charge effects

Early models of QWIPs with multiple QWs are based on the assumption that the electric-field distribution in the active region is uniform [23, 24]. Such simplified models assume that the emitter contact is perfectly injecting [23], i.e., it injects as many electrons as needed by the QW structure. Hence, the injected current density should be that which ensures the required concentration of mobile electrons and, consequently, the required rate of electron capture into QWs to compensate the excitation of electrons from the QWs. A real emitter contact yields such an injected current density if the contact electric field has a proper value which can significantly differ from the average electric field in the active region. An
appropriate electric field at the emitter contact is created by a space charge in the structure arisen due to the difference in the concentrations of bound electrons and donors. The space charge in donor doped QW (and QD) structures can be either positive or negative. The existence of a space charge in the active region results, in general, in the nonuniformity of the electric field. It has been demonstrated using numerical simulations [25–27], that in QWIPs with a large number of QWs, the region of nonuniform electric field can be relatively narrow and located near the injecting contact. However, in QWIPs in which the local electric field weakly affects the excitation of electrons from the QWs and their capture into the QWs (in particular, due to a strong nonlocality of the electron heating), the electric-field distributions can be essentially nonuniform but fairly smooth with the scale of nonuniformity comparable to the active region thickness [28]. Thus, strongly nonuniform distributions, like distributions with high electric field domain near the emitter, are associated with relatively strong dependences of the electron excitation and capture rates upon the local value of the electric field. As a result, the electric-field distribution in a QWIP and, consequently, some its characteristics are determined both by the emitter contact parameters and the field dependences of the rates. However, the overall characteristics of QWIPs with a large number of QWs can be rather insensitive to the emitter contact parameters in a wide range of applied voltage [28, 29]. In contrast, the contact and space charge effects in QWIPs with a moderate number of QWs can be essential [28] (see also [20]) giving some flexibility for the optimization of such devices. In addition, properties of the emitter contact can manifest themselves in nonlinear effects in QWIPs, for example, at a high power of incident infrared radiation. Some of the features associated with the contact and space charge effects can reveal in QDIPs as well [30].

4. Recharging instability and periodic domains

As usually reasoned (see above), the electric-field distributions in QWIPs are monotonic. They correspond to rather smooth distributions of the potential. A novel effect in multiple QW structures with uncoupled QWs under infrared excitation — the formation of periodic and near periodic electric-field and charge domains — has been predicted recently using an ensemble Monte Carlo particle method [31, 32]. This effect is associated with the excitation of the QW recharging waves. The origin of the recharging waves, predicted for the first time in compensated semiconductors with deep traps [33, 34], is associated with the existence of two groups of electrons (mobile and bound) and the electron exchange between the groups. It is natural that the periodicity of the QW locations leaves its imprint. As shown previously [35, 36], the transient photocurrent in QWIPs includes a slow component, which is attributed to the recharging processes. More detailed study using ensemble Monte Carlo particle method which takes into account nonequilibrium and nonlocal effects reveals additional features of the QWIP response to step-like pulses of infrared radiation including oscillatory transients [32, 37]. The development of the recharging instability leads either to the establishment of periodic or near periodic electric-field and charge structures with the period equal to twice the QW structure period or to pronged chaotic pulsations. The formation of periodic or near periodic electric-field and charge structures can significantly influence the operation of QWIPs, particularly, at high power excitation level. A study of such a self-organization in QWIPs is important in view of their application for photomixing and heterodyne detection [38]. As QWIP high frequency operation is primarily limited by electron transit-time [39, 40] and velocity overshoot effects [41, 42], a strong periodic nonuniformity of the electric field pronouncedly affecting the electron dynamics can be very significant factor. The transition from smooth monotonic electric-field distributions
to periodic one’s occurs with increasing intensity of infrared radiation when the photoexcitation rate overcomes the rate of thermoexcitation [43]. This effect can be responsible for low power nonlinearities in QWIPs observed experimentally [27].

5. Application of QWIPs and QDIPs in pixelless infrared imaging devices

Recently, the concept of integrated QWIP and light emitting diode (LED) has been proposed [44] and realized [45] to convert long wavelength infrared radiation into short wavelength one. Further development of this idea has led to the concept of QWIP-LED pixelless imaging device [46, 47]. In this device, long wavelength infrared radiation absorbed in the QWIP due to intersubband transitions generates mobile electrons. A fraction of them is injected into the device LED part causing short wavelength infrared radiation from the active layer where the electrons injected from the QWIP recombine with holes. If the incident radiation is nonuniform (input long wavelength infrared image), the electron current injected into the LED and, hence, the intensity of output short wavelength infrared radiation are nonuniform as well. Thus, the spatial distribution of the output radiation repeats the form of the distribution of the photocurrent in the QWIP which, in turn, repeats the spatial distribution of incoming radiation. Physical effects both in the QWIP and LED parts of such imagers limiting their characteristics were analyzed in Refs. [47, 48]. It has been shown that when the number of QWs in the QWIP part of the QWIP-LED imager is sufficiently large, the electron spreading in this part is insignificant. As a consequence, the main factors limiting the efficiency of up-conversion and the quality of the image conversion are associated with the processes in the LED part of the imager, namely with low external LED efficiency (due to the total internal reflection of a significant portion of the generated short wavelength infrared photons) and with the photon recycling effect in the latter [48] (increasing the external efficiency but leading to an additional smearing of the image [49]). The replacement of a QWIP by a QDIP in such a QDIP-LED pixelless imaging device [30] can be beneficial for the enhancement of its performance.

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References

Resonant states and terahertz generation in strained semiconductors and semiconductors nanostructures

I. N. Yassievich†, M. S. Kagan‡ and K. A. Chao§

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institute for Radioengineering and Electronics RAS, 103907 Moscow, Russia
§ Department of Theoretical Physics, Lund University, S-223 62, Lund, Sweden

1. Introduction

Semiconductors lasers are very attractive for applications as sources of coherent radiation in the terahertz (THz) spectral region which has become an area of intensive research. The first pulsed semiconductor lasers with wavelengths in the range 100–300 µm were developed in the mid-1980s using emission from p-Ge under the simultaneous action of strong electric and magnetic fields at liquid-helium temperatures (see, for example, [1] and works cited there). A cascade laser based on intraband transitions in narrow quantum wells has been recently developed and can, in principal, operate at wavelengths from the mid-IR range to 100 µm [2].

The stimulated emission of p-Ge subjected to uniaxial compression was observed in electric fields [3]. By analyzing the emission spectrum, it was concluded that the stimulated emission is associated with the appearance of a resonant acceptor state as a result of strain-induced splitting of the fourfold degenerate acceptor ground state [4]. This system is of unquestionable interest for developing a new type of unipolar resonant states lasers for THz range. Recently, continuous-wave operation of a such laser with possibility of broad band tuning in the range 2.5–10 THz has been demonstrated [5]. The splitting of acceptor states can be realized also in heterostructures due to internal strain and/or size quantization. In this case the splitting and the position of resonant states can be controlled by the QW width, alloy composition and/or the external transfer electric field applied. The first observation of stimulated emission from strained SiGe QW structures doped by boron has been recently observed.

Here we present a short review of theoretical investigation of resonant states induced by shallow acceptors in uniaxially strained semiconductors, mechanism of population inversion induced by external electric field, experimental results, and consideration of possible nanostructure for THz resonant states lasers.

2. Resonant states induced by shallow acceptors in uniaxially strained semiconductors

For cubic-lattice semiconductors like Ge and Si, the top of the valence band occurs in the Γ point and it is fourfold degenerate. The corresponding wavefunctions \( u_m \) are transformed according to the \( \Gamma^+ \) representation of double point group \( O_{mh} \). Here \( m = \pm 3/2 \) and \( \pm 1/2 \) are the \( z \) component of the total angular momentum of the hole at the Γ point. An uniaxial stress lowers the symmetry of the crystal. A stress parallel to the [001] direction changes the point group symmetry from cubic \( O_h \) to tetragonal \( D_{4h} \). The \( \Gamma_6^+ \) -representation is split into two irreducible representations: \( \Gamma_{6h}^+ \) for the Bloch functions \( \hat{u}_{\pm 3/2} \) and \( \Gamma_{6l}^+ \) for the Bloch functions \( \hat{u}_{\pm 1/2} \). Consequently, the valence band splits into a heavy-hole (hh) and a light-hole (lh) subband with tops located below and above the initial position respectively. The
energy difference between the valence-band tops $E_{\text{def}} = \alpha S$ where for Ge $\alpha = 6$ meV/kbar. If a stress is parallel to the $[001]$ crystallographic direction, $\alpha = 4$ meV/kbar.

Since a shallow acceptor is attached to the valence band edge, in uniaxially strained semiconductor, both its fourfold degenerate ground state and excited states are separated into two doubly degenerate levels. Under a sufficiently strong stress $S \geq S_0$ that the valence band splitting is larger than the acceptor binding energy, two separated series of localized and resonant acceptor levels are formed. Each acceptor level attached to the heavy-hole subband overlaps with the light-hole subband $E_{\text{lh}}(\vec{k})$ and forms a resonant state via its hybridization with the extended Bloch states. The theoretical consideration of these resonant states has been done in papers [4, 6]. The calculated energy positions of the split valence band tops ($E_{\text{lh}}$ and $E_{\text{hh}}$) of strained Ge and the split ground acceptor levels ($E_g$ and $E_s$) of Ga are presented in Fig. 1 as a function of stress applied along the $[001]$ axis. In this case, the threshold stress $S_0 = 2.2$ kbar. A resonant state can be characterized by a complex energy $E_0 - i\Gamma/2$ with imaginary part $\Gamma$ that determines life-time $h/\Gamma$ of a hole in resonant state. In Fig. 2, the calculated resonant width $\Gamma/2$ is presented as function of $E_0$ for Ga in Ge (here $E_0$ is measured from the top of the $E_{\text{lh}}(\vec{k})$ subband).
3. Lasing in strained p-Ge

3.1. Proposed mechanism of population inversion

The intracenter population inversion is formed in enough strong electric fields, when practically all holes occur in the extended states of the light-hole subband and localized acceptor states are empty due to the impact ionization. The free holes are accelerated by the external electric field applied. The scattering of the hot holes at the resonant state \( M = \pm 3/2 \) results in trapping of holes with energy close to the energy \( E_0 \) for a time interval of \( h/\nu \). A theory of transport phenomena in the presence of resonant states and the calculation of their population as function of the external electric field and stress have been produced by Odnoblyudov et al. [6, 7].

3.2. Experimental results

The THz emission from Ga-doped Ge crystals at liquid-He temperature was measured; Ga concentration varied from \( 3 \times 10^{13} \) to \( 10^{14} \) cm\(^{-3} \). Crystals of size \( 1 \times 1 \times 10 \) mm\(^3\) were cut in [001] or [111] crystallographic direction; uniaxial stress and electric field were applied parallel to the long axis of the samples. The optical resonator was formed by the parallel lateral faces of the crystal. THz emission was registered with a cooled Ga-doped Ge photodetector, with a grating monochromator. As was shown in [4], the spectrum of emitted radiation consists of four peaks which were identified as the hole transitions between the resonant level \( E_s \) split-off from the ground acceptor state and the local states in the energy gap. The analysis based on calculations of energy levels positions and strain dependence of the frequency related the main emission peak to the transition from the resonant \( E_s \) state to the first excited local state of p-type symmetry. The frequency increased with stress applied (see Fig. 3).

It was found [5] that for a crystal with optical resonator planes parallel within 0.5–4 arc/min one can only observe THz emission in pulsed regime. The minimum electric field necessary to obtain emission is 2.5 kV/cm in this case. To reach cw lasing a high-quality optical resonator was necessary. For stress applied along the [001] axis and for the resonator planes parallel within 20 arcsec or better, cw THz lasing was observed at which the main peak was corresponds to the frequency of about 2.5 THz. Fig. 4 shows the bias voltage dependence of the current and THz emission intensity at frequency of 2.5 THz. The cw regime was realized at a bias voltage below 10 V. The minimum values of the bias voltage

![Graph](image)

**Fig. 3.** Stress dependence of the frequency of the main emission peak for stress applied along the [111] (■) and [001] directions [5].
and current necessary for lasing were 2 V and 5 mA, respectively. The maximum emitted power estimated was about 1 mW in pulse regime and at least of 1–2 µW in the cw regime.

![Graph](image)

**Fig. 4.** The bias voltage dependence of the current and THz emission intensity for stress along the [001] axis. The cw regime was realized at bias voltage below 10 V [5].

The concept of emission with participation of resonant states is supported by several additional facts. The minimum pressure at which stimulated emission could be observed corresponds exactly to the threshold pressure $S_0$ at which the split-off acceptor level $E_s$ arrives into the continuous spectrum for both [001] and [111] directions. On the other hand, the intensity of the stimulated emission decreases sharply at pressure along the [001] axis of 8 kbar. Depopulation of the resonant state begins at this pressure due to possible emission of optical phonon.

4. Nanostructures suitable for THz lasing

Doped SiGe/Si and InGaAs/AlGaAs QW structures are considered as promising systems to realize inversion phenomena due to shallow acceptor states which are split-off by size quantization and strain without any external stress. The intense Thz emission of stimulated character has been observed in Si/SiGe/Si QW structures doped with boron. It has been demonstrated that stimulated intra-center emission is possible in a strain single-QW inclined by a transverse potential. The special paper of Altukhov et al will be presented on this problem. The 2D structures provide the unique possibility to employ $A^+$-centers which can exist in thermal equilibrium here in contrast to the bulk where they can appear only due to external excitation. We present also some ideas about the possibility to use QW-surface states and states induced by acceptors in barriers.

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References


Terahertz emission of SiGe/Si quantum wells doped with shallow acceptors

I. V. Altukhov†, M. S. Kagan†, V. P. Sinis†, S. G. Thomas‡, K. L. Wang‡, K.-A. Chao§, A. Blom§, M. O. Odnoblyudov¶ and I. N. Yassievich¶

† Institute of Radioengineering and Electronics, Russian Academy of Sciences, Moscow, Russia
‡ University of California, 66-147KK Engineering IV, Los Angeles, CA 90095, USA
§ Department of Theoretical Physics, Lund University, S-223 62 Lund, Sweden
¶ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. THz emission of stimulated character is observed in Si/SiGe/Si quantum well (QW) structures doped with boron. The resonance cavity formed by well parallel QW structure planes owing total internal reflection is necessary for the emission. The model of possible population inversion of strain-split acceptor levels is proposed.

Introduction

The stimulated terahertz emission of uniaxially stressed p-Ge has been shown to be due to a population inversion of strain-split acceptor levels, one of them being in the continuum creating a so-called resonant state. The inversion mechanism consist in depopulation of the ground state of an acceptor (being in the gap) by strong electric field while the resonant state (being in the continuum) is filled to some degree because of exchanging carriers with valence band states. The intra-center population inversion seems to be rather general since the resonant states can arise by quite different reasons. In particular, they should exist in strained QWs where acceptor states are split with no external stress due to internal strain and/or size quantization. In this report we present the data of intense terahertz emission of boron doped Si/Ge, Si1−x/Si QWs and propose an inversion mechanism similar to that in bulk p-Ge.

1. Experimental

The p-type Si/Ge, Si1−x/Si QW structures MBE-grown pseudomorphically on the n-type Si substrate and selectively doped with boron were studied at the temperatures of 4 to 100 K. The Ge content x in SiGe alloy was 0.15. The SiGe layer of 20 nm thickness was δ-doped with boron in the QW middle with the concentration of 6 × 10^{11} cm^{−2}. It was sandwiched between undoped Si buffer (130 nm wide) and cap (60 nm) layers. Two boron δ-layers with B concentration of 4 × 10^{11} to 10^{12} cm^{−2} positioned within the buffer and cap layers on the distance of 30 nm from each QW interface were aimed to supply holes into the QW. The buffer δ-layer should also supply holes to form a p–n junction between the p-layers and the n-substrate. High voltage pulses of 0.3 μs duration (at luminescence measurements) or small dc voltage (at transport measurements) were applied along the QW to the ohmic contacts. The contacts were deposited on the p-side of the structure so that the p–n junctions prevented from a current by-pass through the substrate. Terahertz luminescence was registered by a cooled gallium-doped Ge photodetector sensitive above 10 meV. We studied terahertz emission of the structures arisen at high electric fields applied.
Fig. 1. Photodetector signal vs current.

Fig. 2. Temperature dependence of lateral conductivity of SiGe QW at different voltages from 0 to 1.5 V.

An intense emission caused by Joule heating has usually been observed at the end of a 1.5 to 2 kV voltage pulse. However, in QW structures with well parallel polished opposite lateral (narrow) planes to form an optical resonator due to total internal reflection, the intense emission arose at essentially smaller fields, at the leading edge of the voltage pulse, and could not be, therefore, of thermal origin. Figure 1 shows the radiation intensity in dependence on the current along the quantum well. The radiation and current was registered at voltages above 300 V, it is the threshold of an impact ionization of acceptors in the QW. It is seen that there is a jump in radiation intensity which can be by orders of magnitude greater than spontaneous emission intensity. The intense emission exists in some range of currents and out of that the radiation-current curve is of the same character; this confirms non-thermal origin of the emission.

The intense radiation was coming out mainly from the narrow QW planes, that is natural for the resonator used. It was demonstrated by means of cutting filters that the wavelength of the emission is in the range from 50 to 100 µm. Of course, to make sure finally that the intense emission is stimulated, a spectrum of the emission should be measured.

To understand the cause of possible population inversion, we studied hole transport along the QW. Figure 2 shows the temperature dependence of a conductivity, $\sigma$, along the SiGe layer. One can see two activation-law regions in the curves. It has been shown by
Fig. 3. Potential distribution across the QW for zero and 0.2 V transverse voltage applied between the QW and the surface.

Fig. 4. Scheme of inversion.

means of Hall-effect and magnetoconductivity measurements [2] that the low-temperature part of the curves is due to the thermally activated hopping over neutral B centers. At higher temperatures, the conductivity is shown to be due to thermal activation of holes from the ground to strain-split B states following by hole tunneling into the QW valence band. The tunneling is possible due to inclining valence band profile by a transverse potential caused by hole capture at surface states which makes the surface charged.

This model is confirmed by transverse field-effect measurements. Besides of voltage applied along the QW, the voltage across the QW was applied. Different curves in Fig. 2 correspond to various transverse voltages. The lowest one is for zero transverse voltage. The external potential applied across the QW essentially increases the lateral low-T conductivity and decreases the activation energy, i.e. holes are transferred by transverse electric field from Si cup-layer surface into QW populating B levels.

The calculations of a valence band profile across the QW structure, free carrier concentration in the QW and acceptor population at various transverse voltages were performed by solving Poisson and Schrödinger equations with taking into account pinning the Fermi energy on the Si cup-layer surface. The energy gap, $\Delta$, between the valence band top and the Fermi level fixed at the surface was regarded as a fitting parameter. The results are shown in Fig. 3. The best fit to the experiment was at $\Delta = 0.5$ eV.

The data presented permit us to propose the mechanism of a population inversion of strain-split acceptor levels. The scheme of the inversion is shown in Fig. 4. The transverse
potential caused by a surface charge inclines the valence band profile so that the split-off state of an acceptor becomes resonant. The electric field applied along the QW depopulates the ground state of an acceptor due to impact ionization while the split-off state can be filled to some degree because of tunnel exchange with the valence band continuum. Thus, this model is similar to that in uniaxially stressed bulk p-Ge [1].

2. Conclusion

The intense terahertz emission of boron doped strained SiGe QWs was observed. The stimulated character of the emission is confirmed by an existence of the threshold electric field and by the necessity of a resonator. The data obtained show that the strained quantum-sized semiconductor structures doped with acceptors are promised for lasing at THz frequencies.

References


Mid infrared range laser based on intersubband transitions and resonant Auger processes in quantum wells

L. E. Vorobjev†, G. G. Zegrya‡ and D. A. Firsov†
† St Petersburg State Technical University, St Petersburg 195251, Russia
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. A new type of mid infrared semiconductor laser based on intersubband optical electron transitions in type II quantum wells with electrical or optical pumping is suggested. Inversion of population is created due to special shape of quantum well and resonant Auger recombination providing additional pumping of excited level.

Semiconductor lasers of mid infrared range (MIR, \(\lambda > 4 \mu m\)) have a large number of applications. Development of MIR injection lasers based on interband electron-hole optical transitions comes up against the fundamental problems. The main difficulty is the increase of Auger recombination with decrease of forbidden gap. Because of this in last years the possibility of development of MIR lasers based on intersubband (intraband) optical electron transitions in quantum wells (QWs) is intensively explored. Up to now, among the numerous ideas only two was realized. It is the unipolar quantum cascade laser (QCL) [1] based on idea of Kasarinov and Suris [2] and the unipolar fountain laser (FL) with optical pumping [3]. However, the fabrication of QCL needs a complicated technology. The necessity of powerful optical pumping at wavelength closed to the one of generation restricts the practical use of FL.

The interband cascade lasers (ICL) based on interband electron transitions in type II quantum wells [4] represent a special class of lasers. QWs in them include Sb-containing semiconductor solid solutions. Up to now lasers of this type have emission wavelength not exceeding 4 \(\mu m\). Nevertheless, the threshold current of ICL \(J_{th}\) is significantly less in comparison with QCL, so they seem more perspective.

In the present work a new type of laser based on intersubband carrier transitions and resonant Auger processes in Sb-based type II QWs is offered. Usually, nonradiative Auger recombination plays a negative role in injection lasers but for the first time in this laser the resonant Auger recombination is the important positive factor.

The mechanism of creation of population inversion (PI) is illustrated in Fig. 1. QW for electrons has a shape of asymmetrical funnel. The energy differences \(E_{e3} - E_{e2}\) and \(E_{e2} - E_{e1}\) exceed the energy of optical phonon \(h\omega_{LO}\). So, the electron lifetime \(\tau_3\) on level \(e3\) genetically coupled with wide part of QW is great due to weak overlapping wave functions of levels \(e3 - e2\) and \(e3 - e1\). At the same time the wave functions of levels \(e2\) and \(e1\) are strongly overlapped. The electron lifetime \(\tau_2\) on level \(e2\) is approximately equal to the time of intersubband electron transition with emission of optical phonon. As it follows from calculations, \(\tau_3\) is approximately an order more. This difference between lifetimes assists the creation of population inversion between levels \(e3\) and \(e2\) (\(n_3 > n_2\), where \(n_i\) is the electron concentration on level \(i\)). PI can be destroyed due to \(e - e\) interaction if the concentration \(n_1\) becomes great. It may be prevented by fast depopulation of level \(e1\)

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connected with Auger recombination. Under the resonance the Auger recombination related lifetime can be sufficiently small. Moreover, the resonant Auger processes additionally pump the level $e_3$ (as it is shown by vertical arrows in Fig. 1). So, there are two positive factors arising due to resonant Auger processes: concentration pinning at lowest energy level and additional pumping of excited level.

A set of rate equations describing the variation of electron concentration at levels $e_1$, $e_2$ and $e_3$ under pumping current density $J$ has a form:

$$
\frac{dn_3}{dt} = \eta J \frac{e}{A_3} - \frac{n_3}{\tau_{32}} + \frac{n_1}{\tau_{31}} - \frac{n_1}{\tau_A}
$$

(1)

$$
\frac{dn_2}{dt} = \eta J \frac{e}{A_2} + \frac{n_3}{\tau_{32}} - \frac{n_2}{\tau_{21}} + n_1 \beta
$$

(2)

$$
\frac{dn_1}{dt} = \eta J \frac{e}{A_1} + \frac{n_2}{\tau_{21}} + \frac{n_3}{\tau_{31}} - 2 \frac{n_1}{\tau_A} - n_1 \beta.
$$

(3)

Here $\eta$ is the electron (hole) loss factor due to $e^{-}h$ recombination beyond the QWs ($\eta < 1$); $A_i$ determine the capture of electrons on levels ($A_1 + A_2 + A_3 = 1$, accordingly to calculations $A_3 \gg A_1, A_2$); $\tau_{ij}$ are the electron lifetimes related with intersubband $i \rightarrow j$ transitions with emission of LO phonons (it should be noted that intrasubband lifetimes $\tau_{ii} \ll \tau_{ij}$); $\tau_A$ is the lifetime related to the resonant Auger process ($\tau_A^{-1} \propto n_1 p_1 \approx n_1^2$).

Last terms in (2), (3) describe thermal excitation ($\beta \propto \tau_A^{-1} \exp[-(E_{e_2} - E_{e_1})/k_B T]$).

Stationary solution of (1-3) gives for PI value and for concentration $n_1$:

$$
n_3 - n_2 = \eta \frac{J}{e} \left[ (A_1 + A_2 + 2A_3) \frac{\tau_{31}}{\tau_{32} + \tau_{31}} - A_2 \frac{\tau_{21}}{\tau_{31}} - n_1 \exp \left( \frac{E_{e_2} - E_{e_1}}{k_B T} \right) \right]
$$

(4)

$$
n_1 = \tau_A \eta \frac{J}{e}.
$$

(5)
Amplification coefficient due to intersubband optical electron transitions $e_3 \rightarrow e_2$ is given by:

$$\alpha_{32} = \frac{4\pi^2 e^2 (n_3 - n_2) \omega_{32}}{c \sqrt{\varepsilon} L_W} |Z_{32}|^2 \frac{1}{\pi} \frac{\gamma}{\gamma^2 + (\hbar \omega - \hbar \omega_{32})^2},$$

(6)

where $\varepsilon$ is high frequency dielectric permittivity, $L_W$ is QW width, $Z_{32}$ is the coordinate matrix element, $\gamma$ is line broadening.

To obtain MIR stimulated emission the following condition has to be satisfied:

$$\Gamma \alpha_{32} = \frac{1}{l_R} \ln \left( \frac{1}{R} \right) + \alpha_{\text{loss}},$$

(7)

where $\Gamma$ is optical confinement factor, $l_R$ is resonator length, $R$ is mirror reflection coefficient, $\alpha_{\text{loss}}$ describes losses in waveguide and active layers.

Calculations give the value of $\alpha_{32}$ approximately 68 cm$^{-1}$ for 10 layers of QWs embedded in $i$-layer of the structure (waveguide for $\lambda \approx 10 \mu$m with $\Gamma \approx 0.1$) under losses $\alpha_{\text{loss}} = 0$, $R \equiv 0.36$, $l_R = 1.5$ mm. Let now estimate the threshold current $J_{th}$ or threshold intensity of interband ($h\nu > E_{e3} - E_{hh1}$) optical pumping $P_{\nu}^{th}$ for stimulated MIR emission. Accordingly to calculations the electron lifetimes in QW are: $\tau_{32} \equiv 6$ ps, $\tau_{21} \equiv 0.4$ ps, $\tau_{31} \equiv 20$ ps. Assuming the temperature is low (thermal excitation is negligible: $E_{e2} - E_{e1} \gg k_B T$) we obtain: $\eta J_{th} \equiv 2 \cdot 10^4$ A/cm$^2$ or $P_{\nu}^{th} \equiv 4 \cdot 10^3$ W/cm$^2$ at $\lambda_{\text{pump}} = 0.5 \mu$m. The last value is rather small. For structure area $S = 1.5$ mm $\times$ 200 $\mu$m the threshold intensity $P_{\nu}^{th} \cdot S = 12$ W. This intensity can be provided with semiconductor laser.

The level of concentration $n_1$ is very important for threshold current (or threshold intensity of optical pumping). This concentration can be calculated from (5).

Let now calculate $\tau_A$. In order to determine the probability of Auger recombination or Coulomb electron excitation from ground state ($e_1$) to excited state ($e_3$) we used 4-band Kane model. The electron and hole wave functions are found in terms of superposition of band states of $s$- and $p$-types. Rate of electron Auger excitation is calculated in frames of second order of perturbation theory on electron-electron interaction:

$$G = \frac{2\pi}{\hbar} \sum |M|^2 \delta(E_{e1} + E'_{e1} - E_{e3} - E_{hh1}) f_{e1}(k) f_{e1}(k') f_{hh1}(k'),$$

(8)

where $f_{e1}$, and $f_{hh1}$ are the electron and hole distribution functions, $E_{e1}$, $E'_{e1}$ are the initial electron states and $E_{e3}$, $E_{hh1}$ are the final ones. The summation is over all initial and final states. The matrix element of Coulomb electron interaction can be expressed as

$$M = \frac{4\pi e^2}{\kappa_0} \int \frac{d^3q}{(2\pi)^3} \frac{1}{q^2} I_{12}(q) I_{34}(-q),$$

(9)

where

$$I_{12}(q) = \int \psi^*_{e1}(r) \psi_{e3}(r) e^{iqr} d^3r,$$

$$I_{34}(-q) = \int \psi^*_{e1}(r') \psi_{hh1}(r') e^{-iqr'} d^3r'.$$

(10)
and \( \mathbf{q} \) is the momentum transferred at Coulomb interaction. It should be noted that \( I_{12}(\mathbf{q}) \) is proportional to overlap integral of localized electron states and \( I_{34}(\mathbf{-q}) \) is proportional to overlap integral of localized electrons and holes.

Taking in (9) integration over \( \mathbf{q} \) and using an explicit form of electron and hole wave functions we can obtain the final expression for matrix element. Let us next substitute the expression obtained for matrix element in (8). Taking in (8) integration over initial and final particle states we obtain the expression for inverse time of electron Coulomb excitation under resonant conditions:

\[
\frac{1}{\tau_A} = \frac{G}{n_1} = \frac{8\pi}{\hbar} \frac{E_B}{k_B T} \left( \frac{E_{e1} T}{k_B T} \right)^2 \left( \frac{m_c}{m_h} \right)^2 |d_{13}|^2 n_1 p_1. \tag{11}
\]

Here \( n_1 \) and \( p_1 \) are the concentrations of 2D electrons and holes; \( m_c \) and \( m_h \) are effective masses of electrons and holes; \( d_{13} \) is dipole matrix elements of electron transition from ground state \( (e_1) \) to excited state \( (e_3) \); \( E_B = m_c e^4 / 2\hbar^2 k_0 \) is Bohr energy of electron; \( \lambda_{E_g} = h / (2m_c E_g)^{1/2} \); \( \tilde{E_g} = E_{e1} - E_{hh1} \).

Using (5), (11) and substituting the parameters for InAs/GaSb QW for \( J_0 \) or \( P_0^h \) we obtain \( \tau_A \approx 10^{-11} \) s and \( n_1 \approx 2.3 \cdot 10^{11} \) cm\(^{-2} \). It is rather small value allowing do not take into account influence of electron collisions from different subbands on PI formation.

It should be noted that the dependence of \( \tau_A \) upon resonance detuning is weak if detuning \( (E_{e3} - E_{e1}) - (E_{e1} - E_{hh1}) < k_B T \). So, it is not necessary to satisfy precisely the resonance conditions what simplifies structure growth.

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References

Optical absorption and birefringence in GaAs/AlAs MQW structures due to intersubband electron transitions

L. E. Vorobjev†, S. N. Danilov†, I. E. Titkov†, D. A. Firsov†, V. A. Shalygin†, A. E. Zhukov‡, A. R. Kovsh‡, V. M. Ustinov‡, V. Ya. Aleshkin§, B. A. Andreev§, A. A. Andronov§ and E. V. Demidov§
† St Petersburg State Technical University, St.Petersburg 195251, Russia
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia
§ Institute for Physics of Microstructures, Nizhny Novgorod 603600, Russia

Abstract. For the first time the electrooptical phenomena due to intersubband electron transitions were investigated in GaAs/AlAs MQW structures. These structures are purposed for creation a mid infrared laser of new type. Experimental results on electron redistribution between size-quantization levels under electron heating were obtained up to electric field of 3500 V/cm.

Mid infrared (IR) semiconductor lasers (λ = 4 – 15 µm) can find extensive applications in different fields. The development of physics and technology of low-dimensional structures opens up new possibilities of the development of mid IR lasers. Many attempts have been made to find structures with quantum wells in which an intraband population inversion between the size-quantization levels can be produced. Studies in this field have already resulted in the development of quantum cascade lasers [1] and fountain lasers with optical pumping [2]. However, a search for the new semiconductor lasers is still under way. Recently a new simple intraband laser scheme was proposed [3]. It is based on hot electron phenomena in GaAs/AlAs MQW structures under lateral transport, namely intervalley transfer and real space transfer of electrons. It should provide lasing in mid and far IR ranges. In the present paper an attempt of experimental study of electron redistribution between size-quantization levels under electron heating was taken. For the first time the electrooptical phenomena due to intersubband electron transitions were investigated in GaAs/AlAs MQW structures.

The structure under study was grown up by MBE method and consisted of 100 periods of 10 nm GaAs quantum wells and 2.5 nm AlAs barriers. The middle layer (5 nm width) of each quantum well was doped by Si (ND = 6 × 10^{17} cm^{-3}). Low field electron mobility was about 1000 cm^{2}/Vs at T = 77 K. Such small mobility (compared to MQW structures selectively doped in the barriers) points to significant electron scattering on impurities.

![Fig. 1. Scheme of Γ–X transfer and electron accumulation in X subband under heating lateral electric field. Thin arrows denote radiative transitions.](image-url)
Fig. 2. Equilibrium absorption and birefringence spectra of GaAs/AlAs MQW structure at different temperatures. The experimental absorption spectra are restored taking into account the spectrometer resolution. The birefringence spectra are calculated with the help of Kramers–Kronig relations.

The considered MQW structure is such that the lowest electronic level of the system is the GaAs $\Gamma$-valley level, while the lowest level in the AlAs layer is the so-called $XZ$-valley level (Fig. 1).

Energy distance between $\Gamma_1$ and $\Gamma_2$ levels is 117.4 meV at $T = 77$ K. Experimental absorption spectra demonstrate strong absorption peak due to $\Gamma_1 - \Gamma_2$ transitions (Fig. 2). A weak peak near $h\nu \simeq 150$ meV is probably connected with optical transitions $\Gamma_2 - \Gamma_3$. The intensity of this peak increases with the temperature due to the increase of the electron concentration in $\Gamma_2$ subband.

In accordance with selection rules, $\Gamma_1 - \Gamma_2$ optical transitions take place for light of $p$-polarization only. That is why there is a linear birefringence in the spectral range corresponding to these transitions, and refractive indexes for light waves of $p$- and $s$-polarization are not equal each other: $n_p \neq n_s$. We calculated the equilibrium spectra of birefringence magnitude ($n_p - n_s$) with the help of Kramers–Kronig relationship and experimental spectra of optical absorption ($\alpha_p$):

$$n_p - n_s = \frac{c}{\pi} \int \frac{\alpha_p(\omega')d\omega'}{\omega'^2 - \omega^2}. \quad (1)$$

The calculated equilibrium birefringence spectra are shown in Fig. 2. As it was predicted in [3], sufficiently strong lateral electric field can produce the electron population inversion between $\Gamma_2$ and $\Gamma_1$ levels, as well as between $XZ$ and $\Gamma_1$ levels (Fig. 1). Electrooptical phenomena are convenient tools to study experimentally the electron redistribution between these levels. We investigated the modulation of optical absorption and birefringence in strong lateral electric field with the help of CO2-laser. Its operating wavelength $\lambda = 10.6 \mu m$ is corresponding to the spectral range of $\Gamma_1 - \Gamma_2$ intersubband transitions. To increase the sensitivity we used multipass waveguide geometry for the measurements, so that the total length of the optical way through the MQW layers was 17.7 $\mu$m. The
incident laser beam excited the waves of both $p$- and $s$-polarization in the MQW layers. We measured the amplitude modulation as well as the modulation of phase retardation for these waves. Experimental technique was the same as it was described in [4]. Electron heating was achieved with pulse lateral electric field ($\Delta t = 200$ ns).

The results of electrooptical experiments are presented in Fig. 3. At a temperature of 100 K there is a great decrease of optical absorption in electric field of 3500 V/cm. The magnitude of decrease is $\Delta \alpha_p = -1100$ cm$^{-1}$, i.e. 25% of the equilibrium value of the absorption coefficient $\alpha_p$. At the same field the magnitude of birefringence $\Delta |n_p - n_s|$ is about 0.1.

There are two reasons of the modulation observed. In addition to the redistribution of electrons between the size quantization levels, which we are interested in, we must also take into account the shift of the levels under electron heating due to exchange interaction effect. The role of exchange interaction in intersubband absorption was investigated in [5]. As one can see from Fig. 4, under electron heating the line of intersubband absorption undergoes a “red” shift due to exchange interaction effect.

As it is clear from Fig. 2, in the case of our experiment at $T = 100$ K and $\lambda = 10.6$ $\mu$m the line shift gives a great contribution to birefringence modulation, whereas redistribution
of electrons between the size quantization levels gives a main contribution to absorption modulation. So we have extracted from our electrooptical measurements the value of spectral line shift and the rate of electron redistribution independently. At maximal electric field $E = 3500$ cm the magnitude of “red” shift is 0.7 meV, and relative decrease of difference $n_{\Gamma_1} - n_{\Gamma_2}$ is about 25\%, where $n_{\Gamma_1}$ and $n_{\Gamma_2}$ are the electron concentrations in $\Gamma_1$ and $\Gamma_2$ subbands respectively. Similar value of “red” shift of absorption line can be estimated for investigated structure with the help of [5] and [6] taking into account relatively small electron mobility in our structure. The observed magnitude of $n_{\Gamma_1} - n_{\Gamma_2}$ is close to calculated one [3], so we can predict that population inversion between $\Gamma_1$ and $\Gamma_2$ subbands will appear at much higher fields: $E > 8$ kV/cm.

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References

Precise micro- and nanotubes formed by scrolling
Langmuir–Blodgett/GaAs/InGaAs films

V. Ya. Prinz, V. A. Seleznev, L. L. Sveshnikova and J. A. Badmaeva
Institute of Semiconductor Physics RAS SB, 13 pr. Lavrenteva,
Novosibirsk, 630090, Russia

Abstract. The paper is devoted to further development of previously advanced approach in nanostructuring. This approach is based on using few monolayer-thick bifilms for scrolling 3D heterostructures from them. In this work, initially flat strained GaAs/InGaAs bilayers with Langmuir-Blodgett films deposited onto them were rolled up in tube-shaped scrolls. It is shown that, giving local parts of the surface of the resulting tubes hydrophilic or hydrophobic properties, one can fabricate a perfectly ordered array of free-standing nanotubes.

The present-day nanotechnologies develop along two entirely different lines. The first line is extension of well-established microstructuring methods to nanoscale-size region. The technological methods used here include conventional lithography, ion implantation, and, in some cases, even mechanical grinding. The processes employed here are usually described in terms of film deposition, patterning and etching steps, and often include a planarization procedure for preparing circuit wiring. However, this approach encounters many difficulties. For example, in lithographic processes, there is a limitation of fundamental character on the sizes of obtainable electronic components. In addition, fabrication of 3D nanostructures here is highly problematic.

The second approach in nanostructuring invokes procedures to be performed over individual atoms and molecules or over their highly ordered ensembles. In this case, transformation of initial objects into a more complex 3D structure is implied. This approach often employs various self-forming procedures. However, the possibility of easy assembling electronic components in a circuit here appears to be doubtful.

As initial object for preparation of 3D nanostructures, we propose here to use semiconductor nanotubes with precisely controllable sizes. In addition to already proposed technological procedures, we describe here some new methods intended for fabrication of assembled arrays of 3D nanostructures.

Recently, self-formed semiconductor nanotubes have been fabricated, and the possibility of precise control over their parameters has been shown [1–3]. The fabrication process is based on using thin highly strained InGaAs/GaAs heterolayers that roll up in scrolls after being debonded from substrate (after selective etching of an underlying AlAs sacrificial layer). It has been shown that the layers in the resulting tube stick together forming a monocrystalline tube wall. Here, the tube diameter $D \approx d \cdot a / \Delta a$ is determined by the bilayer thickness $d$ and by the mismatch of lattice parameters $\Delta a / a$. In this manner, InGaAs/GaAs tubes with tube diameters $D$ ranging from 3 nm to 10 $\mu$m and lengths as large as 1 mm were fabricated.

In the present work, hybrid micro- and nanotubes scrolled from initially planar structures were fabricated for the first time which contain a strained InGaAs/GaAs bilayer with a thickness of several monolayers and a Langmuir–Blodgett (LB) film.
Figure 1 shows a schematic flow-chart representation of the procedure intended for production of hybrid micro- and nanotubes. The initial planar structure contains a strained InGaAs/GaAs bilayer with a thickness of several monolayers and an LB film also several monolayers in thickness (for instance, two). As the bifilm gets free of bonding with substrate, the interatomic forces in it start acting to increase the interatomic distance in the compressed InAs layer and to decrease it in the tensile-stressed GaAs layer (Fig. 1(a)). The elastic forces $F_1$ and $F_2$ are oppositely directed, and they give rise to a non-zero moment of forces $M$, which tends to bend the bilayer. Under the action of this moment, the initially planar bilayer scrolls in a tube with the LB film clamped between the tube turns.

In this way, we obtained multiturn tubes which can be considered as radial superlattices (see Fig. 1(b)). It should be noted that the tubes thus formed could be prepared fixed to a desired place of the substrate.

The LB films used in this study was cadmium bencenate ($[\text{CH}_3(\text{CH}_2)_{20}]_2\text{Cd}$) films prepared by covering the free surface of a $4 \cdot 10^{-4}$ mole/liter cadmium chloride aqueous solution with a behenic acid solution in hexane. The prepared monolayer covering was transported from the liquid surface onto an InGaAs/GaAs heterostructure under a surface tension of $30 \text{ mN/m}$ at $T = 23 \pm 1^\circ \text{C}$. The transport velocity was $1.5 \text{ cm/s}$ [4]. The surface of the GaAs layer onto which the LB film was transported was hydrophilic, as well as the surface of the detached InAs layer. An important point is that the LB technique permits preparation of ordered multilayers of organic molecules with a definite number of layers. To bond the LB film to the surfaces of the scrolled layers, we used films composed of even numbers of monolayers (namely, 2, 4, 6, and 20 monolayers). The thickness of each monolayer in the LB film was $30.4 \pm 0.4 \text{ Å}$. The inside diameter of the produced tubes is dependent on the characteristics of the InGaAs/GaAs layer used [1, 2]. In this study, tubes with inside diameters ranging from 80 nm to 8 $\mu\text{m}$ were fabricated. A simplest example of application of the described technology is shown in Fig. 2.

On complete removal of the sacrificial layer, we obtain a free-standing tube. The following important property of such tubes is noteworthy: being placed onto a water surface, they float freely on it and, if necessary, can be arranged in ordered arrays (of the LB film type).

The surface of a free-standing tube can be made either hydrophobic or hydrophilic, or be given spatially alternating hydrophobic/hydrophilic properties, which can, in turn, be used for exerting precise control over the geometry of nanoobject ensembles prepared as
described above.

For example, if a tube is prepared in such a manner that one its part has a hydrophilic and other a hydrophobic surface, then, on a water surface, it will float half-submerged into water. An ensemble of such tubes freely floating on a water surface can be used to prepare LB-like films of more intricate geometries.

In conclusion, it is shown that the LB technique can be used for fabrication of intricate 3D structures which cannot be prepared by any other technology.

Acknowledgements

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References

MBE grown Ge nanostructures on CaF₂/Si(111)

O. P. Pchelyakov†, L. V. Sokolov†, M. M. Moisseeva‡ and N. S. Sokolov‡
† Institute of Semiconductor Physics, Novosibirsk, Russia
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Germanium nanocrystals were grown on CaF₂/Si(111) by molecular beam epitaxy. Specific features of Ge and CaF₂ growth have been analysed in this work, using electron diffraction and atomic force microscopy. Well-pronounced Ge quantum dots were observed in case of growth on thick CaF₂ buffer.

Introduction

Heteroepitaxial films with Ge islands of small sizes (quantum dots) are very attractive because Ge is a prospective semiconductor for new devices based on quantum effects. Ge quantum dots or structures with 3-D electron confinement have potential applications in advanced electronic and optoelectronic devices, e.g. single electron transistors and multispectral detectors. In optoelectronic applications, germanium nanostructures can successfully compete with traditional AIIIBV materials [1, 2]. Such structures are also interesting because their growth techniques are compatible with usual silicon microelectronic technology. However, there are some drawbacks of Ge/Si nanostructures. Because silicon has relatively narrow band gap, only shallow quantum wells can be created using this heterocouple. Therefore quantum effects in these heterostructures are important only at low temperatures. Moreover, some of the Ge islands may luminesce at the Si absorption edge. To avoid the above-mentioned problems, the Ge dots studied in the present work were grown on CaF₂/Si(111) heteroepitaxial substrates. The previous reports on Ge growth on CaF₂ or Ge/CaF₂ heteroepitaxy starting from the initial stages when average coverage by Ge did not exceed a few monolayers (0.5–1 nm).

1. Experimental technique and pre-growth Si treatment

Ge/CaF₂/Si(111) and CaF₂/Ge/CaF₂/Si(111) heterostructures were grown in a research MBE system at the Institute of Semiconductor Physics, Novosibirsk, Russia. After standard chemical cleaning, the silicon substrates were loaded into the growth chamber and cleaned thermally at 830°C in ultra high vacuum. This procedure allows obtaining atomically clean Si(111) surfaces with a 7 × 7 superstructure. The fluoride and Ge were evaporated from graphite crucibles. Due to molecular mode of CaF₂ sublimation, the stoichiometry of the grown layers was kept naturally. The growth rate of CaF₂ or Ge layers was about 1 nm/min for a few nanometers films and about 15 nm/min for thick (over 100 nm) films. The CaF₂ buffer growth temperature was kept within the 700 to 780°C range. Germanium films were grown at 350°C.
2. In situ RHEED measurements

At the first stage, CaF$_2$ buffer layer was grown on clean 7 × 7 Si(111) substrates. The buffer layer thickness was 100–200 nm. The growth modes and crystalline quality of the fluoride layers were monitored in situ using reflection high energy electron diffraction (RHEED). To avoid undesirable influence of electron beam on growth processes its intensity and exposure time were kept as low as possible. One can see a RHEED pattern taken after 200 nm CaF$_2$ growth in Fig. 1(a). The picture demonstrates two-dimensional diffraction typical for a single crystal film with the smooth surface.

![RHEED pattern](image)

Fig. 1. RHEED patterns from: (a) CaF$_2$ surface at electron beam azimuth [11\bar{2}]; (b) the same structure after Ge deposition, electron beam azimuth [\bar{1}10].

Ge was grown at the second stage of heteroepitaxy. The substrate temperature was chosen about 350°C because a Ge film has no defects at this temperature [5], and diffusion current is too low to produce large islands [6]. A RHEED pattern does not change at first, but as an average film thickness increases to 11.5 nm, bright spots appear on the streaks in Fig. 1(b). This implies, that the growth proceeds in the Stransky–Krastanov mode. Germanium growth on CaF$_2$ is quite likely to start in the layer-by-layer mode, and then the strain relaxes resulting in coherent islands on the surface. At the final stage, we covered the most of the structures by a thin CaF$_2$ cap layer to protect Ge from oxidation after the growth.

3. Surface morphology studies by AFM

The main method of surface morphology studies was atomic force microscopy (AFM), because of CaF$_2$ radiolysis under electron beam in the electron microscope. The surface morphology measurements have been carried out using a P4-SPM-MDT microscope produced by NT-MDT (Zelenograd, Russia). The samples were studied at the ambient conditions in the tapping mode. The typical radius of the cantilever’s tip produced by Silicon MDT was about 20 nm. The resonance frequency was in the 20–60 or 250–500 KHZ ranges.

Figure 2(a) shows Ge small islands situated on atomically flat CaF$_2$ triangular shape terraces resulting from thermal stress relaxation in thick fluoride layers on Si(111) [7]. The average radius of Ge islands does not exceed 20 nm. This value is close to the expected lateral resolution limited by the sharpness of the AFM tip. The average height of the islands is less than 10 nanometers.

Increasing Ge deposition time, at the same substrate temperature (~350°C), one could obtain more pronounced Ge nanocrystals (Fig. 2(b)). The typical lateral size of Ge islands was about 70 nm and the height was about 30 nm. The average thickness of Ge, that assembles in the islands, is 10 nm.

Summarizing the above, we have demonstrated MBE-growth of Ge nanocrystals on
Fig. 2. AFM images of Ge/CaF$_2$/Si(111) structures having various average thickness of deposited Ge: (a) 1 nm, (b) 10 nm.

CaF$_2$ surface. Their size and density can be controlled by choosing proper growth conditions. Optical and electrophysical characterization of the Ge dots is underway.

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References

Novel Cd(Se,Te)/BeTe nanostructures: fabrication by molecular beam epitaxy and properties

S. V. Ivanov†, G. Reuscher‡, T. Gruber‡, T. Muck‡, V. Wagner‡, J. Geurts‡, A. Waag‡, G. Landwehr†, T. V. Shubina†, N. A. Sadchikov†, A. A. Toropov† and P. S. Kop’ev†

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physikalisches Institut der Universität Würzburg, D-97074 Würzburg, Germany

Recently different II–VI wide-gap systems such as CdS/ZnS [1], CdTe/ZnTe [2], CdSe/ZnSe [3–5] have been tested for a possibility to fabricate quantum dot (QD) structures with high binding energy of zero-dimensional excitons, with the latter two systems being studied more intensively. Green RT lasers using as an active region a sheet of CdSe-based QD-like islands in a ZnSe matrix have been fabricated [6], demonstrating extremely low thresholds and enhanced degradation stability as compared to quantum well (QW) lasers. Typical features of all above mentioned QD systems are a common anion sublattice and, as a consequence, a type-I band alignment.

In this paper we report for the first time on fabrication by molecular beam epitaxy (MBE) and investigation of structural and optical properties of a new class of II–VI wide gap nanostructures based on ultra-thin CdSe insertions in a BeTe matrix, which have no common atoms at the CdSe/BeTe interface and are characterized by a type-II band structure and by the same lattice mismatch (∼7%) as for the CdSe/ZnSe heteropair. It is expected that higher bond strength of BeTe can suppress significantly interdiffusion processes characteristic for CdSe/ZnSe [7] (like e.g. in ZnSe/BeTe multiple QWs [8]), as well as affect on the CdSe island formation mechanism by the intentional growing of highly strained BeSe or CdTe interfaces. Schematic band alignment diagram of the system is presented in Fig. 1.

Four CdSe/BeTe MQW structures have been grown on (100)GaAs substrates at 300°C by a conventional MBE mode in a Riber 2300 setup. A 5 monolayer (ML) thick BeTe buffer layer was initially grown at 350°C on a c(4×4) surface of a GaAs buffer layer. BeTe and CdSe growth rate gradients across the wafer and specular spot intensity (SSI) during the nanostructure growth were examined in situ by a specially designed RHEED system. Structural properties of the MQW structures were analysed using high resolution x-ray diffraction (XRD) measurements using a Philips X’pert diffractometer equipped with a four-crystal Ge 220 monochromator. Photoluminescence (PL) measurements were performed at 77 K and RT using 1 mW He-Cd (325 nm) and 2.5 mW Ar+ (457 nm) lasers.

All the structure parameters estimated from XRD and RHEED are summarized in Table.

<table>
<thead>
<tr>
<th>Sample</th>
<th>CdSe nominal thickness, ML</th>
<th>1st interface</th>
<th>2nd interface</th>
<th>MQW period, nm</th>
<th>Number of periods</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.0</td>
<td>BeSe-0.5ML</td>
<td>BeSe-0.5ML</td>
<td>4.5</td>
<td>10</td>
</tr>
<tr>
<td>B</td>
<td>1.0</td>
<td>CdTe-SSI max.</td>
<td>CdTe-SSI 1st min.</td>
<td>5.0</td>
<td>5</td>
</tr>
<tr>
<td>C</td>
<td>1.0</td>
<td>CdTe-SSI max.</td>
<td>CdTe-SSI max.</td>
<td>6.0</td>
<td>5</td>
</tr>
<tr>
<td>D</td>
<td>1.5</td>
<td>CdTe-SSI max.</td>
<td>CdTe-SSI max.</td>
<td>6.0</td>
<td>10</td>
</tr>
</tbody>
</table>
Due to the huge lattice mismatch of the interface compounds with GaAs: \(-10\) and \(+15\%\) for BeSe and CdTe, respectively, their critical thickness should be below 1 ML. Therefore, fractional MLs of BeSe and especially CdTe, intentionally grown at a BeTe/CdSe interface, may serve as local compressive or tensile stressors for enforced CdSe nucleation on the BeTe surface. Sample A containing BeSe interfaces demonstrates a dramatic decrease in SSI during growth of both interfaces, which cannot be completely recovered by the following 1 ML CdSe and 4.5 nm BeTe layers. This results in disturbed structural quality and strongly suppressed PL intensity at 77 K.

The RHEED SSI variation during growth of one MQW period of samples C and D with CdTe interfaces are shown in Fig. 2. As has been observed in additional experiments which will be reported elsewhere, CdTe growth on BeTe involves three stages characterized by the specific SSI behavior: (i) an abrupt drop to a 1st SSI minimum likely corresponding to a maximum morphological and stress disorder introduced by pseudomorphically deposited CdTe, (ii) partial recovering and smoothing the interface, resulting in the intermediate SSI maximum, and (iii) final SSI reduction accompanied by a streaky to spotty RHEED pattern transformation indicating an onset of 3D growth. The whole SSI variation is limited by the CdTe thickness of \(\sim0.5\) ML (for strained CdTe), as estimated from XRD analysis of CdTe/BeTe MQWs. As is seen from Fig. 2, sample C has both CdTe interfaces corresponding to the SSI maximum (stage II), with CdTe nominal thickness being 0.25 and 0.15 ML, respectively, whereas the first CdTe interface in sample D is intentionally interrupted at SSI minimum (stage I), i.e. at \(\sim0.15\) ML. One should mention that sample B having the second CdTe interface slightly beyond the SSI maximum (\(\sim0.20\) ML) shows gradual decrease in the integral SSI during the growth, resulting finally in poor structural quality and PL intensity. Contrary to that, the samples C and D demonstrate non-degraded SSI behavior during the whole structure growth, which is reflected in bright PL both at 77 and 300 K.

Figure 3 presents PL spectra of these samples in a logarithmic scale, taken at 77 K at
different points along the Cd gradient on the surface (solid lines) and at RT at the point with a maximum Cd(Se,Te) thickness (dotted line). The parameters presented in Table 1 for both structures also correspond to the point where Cd flux is maximal. Low temperature PL spectra of both samples involve two separated lines, with an integral PL intensity being much higher for sample D having larger CdSe thickness. The high-energy line dominating the spectra of sample C is narrower and shifts to blue between 2.2 and 2.45 eV with the CdSe thickness decrease, while the approximately twice broader low-energy band, dominant for sample D, is practically fixed at $\sim$1.8 eV at any point on the sample surface. Additionally, strong redistribution of the PL intensity from high- to low-energy lines accompanies the Cd(Se,Te) thickness reduction for the sample C demonstrating also much larger blue shift of the high-energy line. Both of these effects are less pronounced in sample D, although they develop in a reverse direction versus Cd(Se,Te) thickness. A RT PL band in both structures follows the dominant lines, which may serve as an evidence of their intrinsic excitonic nature. However, further experiments, e.g. time resolved PL, are necessary to confirm this claim.

Summarizing these findings, the high-energy line can be assigned to a uniform Cd(Se,Te)/BeTe type-II QW, correlating well with the smooth 1st CdTe interface corresponding to the point with the maximum Cd flux (Fig. 2, sample C). The low-energy line is likely attributed to CdSe-based islands with a wide lateral size distribution. The dominance of this line in sample D agrees well with the highest non-uniformity of the 1st CdTe interface, stimulating the Cd(Se,Te) islands nucleation, at the point of maximum Cd flux related to the RHEED pictures in Fig. 2. A decrease in Cd amount along the sample surface obviously leads to a decay of integral PL intensity (from black to grey). Simultaneously, the 1st CdTe interface thickness also decreases, changing CdSe nucleation conditions. For sample C the reverse situation is observed. These speculations are additionally confirmed by XRD data presented for sample C in Fig. 4. The clearly observed MQW satellites and oscillation fringes strictly corresponding to the number of MQW periods indicate a high crystalline quality and uniformity of the Cd(Se,Te)/BeTe MQW structure, whereas the high
satellite intensity evidences a rather high compositional contrast in the MQW, i.e. less pronounced interdiffusion processes. Contrary to that, XRD data on sample D does not reveal the MQW satellites at all, which evidences the significantly disordered Cd(Se,Te) sheets.

In conclusion, CdSe/BeTe type-II nanostructures have been grown by MBE on GaAs substrates and studied by RHEED, XRD and PL for the first time. These structures demonstrate extremely high sensitivity to the interface bond type (BeSe or CdTe), with the CdTe interfaces providing much higher structural quality and PL intensity. Formation of the first CdTe interface, controlled by RHEED at the 0.1 ML level, causes radical variation of the structure morphology at a similar CdSe nominal thickness in the 1.0–1.5 ML range. It may be changed from the well defined homogeneous Cd(Se,Te) QW in the case of the smooth interface to the array of CdSe-based nanoislands (or QDs) coupled by much thinner QW in the case of non-uniform locally strained interface. However, final conclusion on the intrinsic morphology of these structures needs high resolution transmission electron microscopy studies, which are currently under the way.

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II–VI wide-gap heterostructures still remain the most natural candidates for blue-green laser diode (LD) applications, although demonstrating so far a non-sufficient device lifetime (less than 500 hours [1]), because this spectral range (<450 nm) is hardly reachable by III-nitrides based lasers. It is generally believed now that the large stress accumulated in a lattice-mismatched quantum well (QW) active region (like ZnCdSe one) causes enhanced multiplication and diffusion of extended and point defects. This effect in combination with a low threshold of defect formation, characteristic of ZnSe-based compounds, seems to be responsible for a so called “slow” LD degradation. Thus, the need in low-stress high-stiffness QW structures for the active region of blue-green II–VI lasers and light-emitting diodes stimulates a search of new suitable materials.

Recently a new material for the laser active region — BeCdSe — has been proposed [2]. Despite expectations of a very wide instability region for this alloy, it has been predicted that stable Be$_x$Cd$_{1-x}$Se alloys with composition lattice-matched to GaAs ($x \sim 0.46$) can be grown by molecular beam epitaxy (MBE) until a relaxation of elastic stress in the layer occurs. The high Be content in this alloy is believed to make the crystalline lattice much harder than in commonly used ZnCdSSe alloys. Since previous study was mainly focused on $x = 0.0-0.33$ range, only rough extrapolation of an energy gap ($E_g$) versus $x$ dependence to the lattice-matched composition region has been made, taking optical bowing parameter equal to 3 eV. It gave nearly equal $E_g$ values for ZnSe and Be$_{0.44}$Cd$_{0.56}$Se alloy, leaving questionable the possibility to obtain the lattice-matched BeCdSe/ZnSe QW suitable for laser applications.

In this paper we study optical and structural properties of the Be$_x$Cd$_{1-x}$Se/ZnSe QWs grown by MBE in the composition range close to the compound lattice-matched to GaAs. Bright low-temperature (80 K) photoluminescence (PL) in the 2.45–2.66 eV range, preserving up to 300 K, has been obtained for $x = 0.36-0.49$ estimated from reflection high energy electron diffraction (RHEED) and x-ray diffraction (XRD) measurements. Much stronger optical bowing in this alloy system has been found, as compared to previously estimated one.

Structures with both single QW (SQW) and multiple QWs (MQW) were grown either in a conventional MBE mode or by a sub-monolayer digital alloying (SDA) technique at a substrate temperature $T_s = 300^\circ$C on (001) GaAs substrates, using a solid source MBE setup (Riber 2300). Except for BeCdSe/ZnSe QWs of either 2 or ~10 nm thick, separated by ~10 nm ZnSe barriers, the structures involve 50–100 nm-Zn$_{0.97}$Be$_{0.03}$Se claddings and a thin BeTe buffer layer grown at 350°C. Structural properties of the multi-layer samples
were analysed by a dynamic simulation of (004) XRD $\theta - 2\theta$ rocking curves obtained using a Philips X'pert diffractometer equipped with a four-crystal Ge 220 monochromator. PL spectroscopy was performed using a 325 nm excitation line of a He-Cd laser. Growth rate and alloy composition were additionally controlled by RHEED oscillation technique, as shown in Fig. 1 where a ratio of the BeTe buffer to the BeCdSe QW growth rates (in monolayer/s) gives a rather accurate estimation of the alloy composition.

To study a modification in BeCdSe optical properties with Be content, two 2 nm-BeCdSe/9 nm-ZnSe MQW structures with significant grade of the Be content along a wafer surface were grown without rotation in MBE and SDA modes. As the XRD data analysis demonstrates the Be content variation across the MBE structure surface from $x = 0.40$ to $x = 0.48$ (Fig. 2(a)), with the maximal composition corresponding to the point of RHEED registration, PL peak gradually shifts to higher energies (Fig. 2(b)), still remaining at 160 meV below $E_g$ of ZnSe. Another important feature of the PL spectra is an existence of a pronounced maximum in the peak intensity versus Be composition, relating to a pseudomorphic composition of $x = 0.46-0.47$, as estimated from XRD data. PL spectra of the SDA structure with the Be content changing from 0.36 to 0.49 demonstrate the same behavior, with the most intensive PL peak being placed at the same energy of 2.65 eV. Since strain-induced defects reduce a quantum efficiency of QW structures, one could expect an existence of the PL peak corresponding to a non-strained BeCdSe layer, which is appreciably more intensive than others related to the QWs with increasing lattice mismatch. One should mention that both MQW structures demonstrate very bright PL of nearly the same intensity, independently of the growth mode, although XRD analysis shows higher structural quality of SDA MQW structure even at minimum $x = 0.36$.

Despite the rather large lattice mismatch variation in the MQW structures, with the lowest $x = 0.36$ being somewhat at the boundary of the BeCdSe instability region thermodynamically calculated in [2], no pronounced signature of phase separation has been observed probably due to the small QW thickness as compared to the critical one. It is confirmed additionally by two 2 nm SQW structures with $x \sim 0.35$ and $x \sim 0.4$, whereas a decrease in $x$ below 0.33 causes already a pronounced phase separation even in 2 nm QWs [2]. Contrary to that, a 10 nm-BeCdSe/10 nm-ZnSe MQW structure, also grown with the intentional Be composition gradient, has revealed a dramatic disturbance of structural properties due to the phase separation at relatively small deviation of $x$ from the lattice-
matched composition, as observed in situ by RHEED and ex situ by XRD and PL and will be reported in details elsewhere. This effect is probably caused by exceeding the QW critical thickness, followed by elastic stress relaxation resulting in dramatic CdSe-BeSe phase diagram transformation [2].

Figure 3 presents an estimation of the BeCdSe $E_g$ versus $x$ behavior, which has been done using the PL data on all ZnSe/BeCdSe SQW and MQW structures discussed in this work (shown by squares and triangle). Additionally, PL data on 0.8 nm BeCdSe/ZnSe SQWs from [2] are given by circles. One should note that the point with $x \sim 0.33$ can be regarded only as a very rough value due to an onset of the phase separation process. Solid curve is a result of theoretical estimation of the BeCdSe energy gap bowing in accordance with

$$E_g(\text{BeCdSe}) = (1-x)E_g(\text{CdSe}) + xE_g(\text{BeSe}) - x(1-x)C,$$

where $C$ is the bowing parameter estimated as 4.5 eV in this case. This value is even higher than 3.0 eV found in [2], using an extrapolation of experimental slope obtained for the narrow 0.8 nm QWs with $x$ at the CdSe-rich composition side. The new data allow us to consider the lattice-matched BeCdSe/ZnSe QWs as very promising structures for the active region of II–VI blue-green laser, which can provide sufficient electronic confinement. The question of band-offsets in the BeCdSe/ZnSe system still remains unclear, but

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Fig. 2. (a) XRD $\theta-2\theta$-rocking curves of BeCdSe/ZnSe MQW structure taken from two opposite points along the grade of Be content: experimental (solid curves) and theoretical simulations (dashed curves). (b) PL spectra (80 K) of the same structure taken along the grade of Be content. A and B denote points where XRD measurements were performed (see Fig. 2(a)).
ZnSe/BeCdSe QW is expected to be of type-I due to the absence of blue shift of the PL peak with a ~150 time increase in an excitation power.

In summary, SQW and MQW structures of different thicknesses (2–10 nm), based on a new II–VI compositional material Be$_x$Cd$_{1-x}$Se with a Be content ranging from 33% to 50% have been grown by MBE using different growth modes. The structures have demonstrated bright PL up to room temperature and no phase separation phenomena in the vicinity of $x = 0.46$ corresponding to a composition lattice-matched to GaAs. Large energy gap bowing ($C = 4.5$ eV), refined using new experimental PL data, permits one to consider BeCdSe alloys with large Be content as suitable materials for a QW active region of ZnSe-based blue-green laser diodes.

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The influence of Bi doping of the InAs/GaAs quantum dots on morphology and photoelectronic properties of the heterostructures obtained by MOVPE

B. N. Zvonkov§, I. A. Karpovich†, N. V. Baidus§, D. O. Filatov‡, S. V. Morozov† and Yu. Yu. Gushina‡
† University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia
‡ Research and Educational Center for Scanning Probe Microscopy, University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia
§ Physical-Technical Research Institute, University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia

Abstract. Bi doping of InAs quantum dots layer during growth by MOVPE depresses coalescence of nanoclusters and advances obtaining more uniform distribution of quantum dots size. Bi itself is practically not being incorporated into the QD material. Its role consists in limitation of In and As atoms diffusion mobility on the growing layer surface.

Introduction

One of the serious problems in growing the self-assembled quantum dot structures (QDSs) by MOVPE is to avoid coalescence of nanoclusters usually occurring at the growth temperatures which are optimal for their optical properties. Coalescence results in increasing of dispersion of the QDs in size, in decreasing their surface density and in formation of considerable number of relatively big clusters with continuous spectrum in QD layer [1]. We studied possibility of depressing the coalescence by Bi doping of QDs layer during growth. It was suggested that presence of bigger Bi atoms with less mobility on the growth surface will result in limitation of diffusion mobility of In and As atoms and to depress the coalescence.

Experimental

The GaAs/InAs QDSs were grown on (100) GaAs semi-insulating substrate with misorientation 3° towards [110] by atmospheric pressure MOVPE. The n-GaAs buffer layer was grown at 600°C. Then temperature has been decreased down to 530°C, then InAs QD layer was deposited. Trimethylindium and arsine were introduced into the reactor alternatively for 6 and 4 sec. respectively with the 4 sec. intervals between the cycles. Total number of the cycles was 10, the estimated InAs layer thickness was 1.5 nm (about 5 monolayers (ML)). Bi doping was made during the QD layer deposition by sputtering of the Bi target placed in the cold reactor zone (∼12 cm away from substrate) by a Q-switch YAG pulsed laser. The estimated surface density of Bi is ∼10^{14} cm^{-2}. A number of structures both with and without 15 nm GaAs cap layer for optical and morphological investigations respectively were grown. Morphology of the QD layers was investigated by Atomic Force Microscopy (AFM) using TopoMetrix “Accurex” TMX-2100 AFM in contact mode. Also photoluminescence (PL) at 77 K and capacitive photovoltage (CPV) spectra at 300 K were measured.
Results and discussions

The Bi doping of the QD layer during growth suppresses coalescence. It is especially clear if In concentration in the InAs layer is not too much.

Figure 1 shows the surface topography of the QD layer without GaAs cap layer with (a) and without (b) Bi doping. The other growth parameters were the same. In the first case the dots are highly uniform in lateral size (41 ± 2 nm) as well as in height (5.8 ± 0.2 nm). Although there is almost no big clusters the surface density of QDs was rather low (4 × 10⁹ cm⁻²). Increasing the In concentration in the QD layer increases QD density, but efficiency of coalescence suppression becomes less at the same concentration of Bi. Possibly optimization of In and Bi flows will allow to increase QD surface density without surface morphology degradation.

The PL spectra of the undoped QDS have FWHM about 60 meV at 77 K. The Bi doped QDS have more narrow peaks with FWHM 40 meV (Fig. 2, curve 1) as well as the abnormal broad ones with width up to 200 and even 300 meV (curve 2). Appearance of the ones can be ascribed to fluctuations of Bi surface concentration and therefore to fluctuations in QD size in certain growth regimes. The lowest peak energy \( h\nu_m \approx 1.0 \text{ eV} \) at 77 K has been observed. This value fits into the 1.3 μm band important for optronics applications. Such a position of a PL peak cannot be ascribed to presence of Bi in the QD material, because the QDs grown without Bi doping also show PL in this band [2]. Because of big covalent radius Bi atoms are likely to be hustled away to the growth surface or to InAs/GaAs interface. Therefore, another mechanism of Bi influence on self-assembled growth of the QD array, namely due to changing of the surface energy [3] should be proposed.
On the doped QDS without cap layer as low values of $h\nu_m \approx 0.75$ eV (Fig. 2, curve 3) at 77 K have been observed for all samples. The correspondent ground transition energy at 300 K is $\approx 0.67$ eV. Shift of the PL maximum for structures with surface QDs can be explained by partial elastic strain relaxation in the QD [4].

Usually photoelectric sensitivity of the QDs at 300 K can be observed only in the QDS with thin GaAs covering layer ($\sim$15 nm) so that the QD layer is built in the depleted region of the surface barrier. The strong electric field favor emission of the electron-hole pairs from the QDs. But if the QD layer is situated on the surface, photoluminescence and photosensitivity can be observed only if the QD density is high ($\sim 10^{10}$ cm$^{-2}$) due to a high surface recombination rate.

CPV spectra of the QDS are shown in Fig. 3. Photosensitivity threshold corresponds to PL peak position (Fig. 2) taking into account the thermal band gap shift $\approx$80 meV. The ground transition and thin structure at high energies due to transitions to higher excitation levels are well resolved.

Conclusions

In conclusion, the results of this work show that Bi doping of InAs QDs is an effective tool for improving the morphology and photoelectric properties of the QDS obtained by MOVPE.

Acknowledgements

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References

Electroluminescence properties of a new asymmetric AlSbAs/InAs/II–VI double heterostructure grown by MBE

Ioffe Physico-Technical Institute, St Petersburg, Russia

Recently new physical approach and new energy band design has been proposed by us to create high-power mid-infrared lasers with improved performance [1]. It can be achieved in a laser structure with high asymmetric barriers for electrons and holes at the interfaces between a narrow-gap active layer and wide-gap cladding layers.

In this case the narrow-gap active layer sandwiched between the wide-gap layers forms type I heterojunctions with them, whereas the n- and p-confining layers form a type II heterojunction pair and have energy gap much higher than the photon emission energy (Fig. 1). In such heterostructure a strong overlap of electron and hole wave functions in the type I quantum well (QW) active layer placed at the type II heterointerface takes place. It leads to the higher optical gain and quantum efficiency as in conventional type I double heterostructure lasers, but with substantial suppression of both injection and non-radiative losses. In the proposed device a low threshold current and its weaker temperature dependence are expected. Total current is expected to be determined mostly by a radiative recombination in the active region of the laser structure.

![Fig. 1. Schematic band diagram of asymmetric laser heterostructure design.](image)

There exist only three III–V compounds to design the proposed laser structures, which could provide high enough barriers for electrons and holes simultaneously: AlAsSb-InAs-InP. However, the latter is strongly lattice-mismatched to the former two, making practically impossible to grow a pseudomorphic laser structure with a strong hole confinement in the InAs active layer by any of state-of-art technological methods (LPE, MBE, MOCVD).

A novel approach, which we propose in this paper, consists in combining III–V and II–VI compounds in one laser heterostructure, which allows one to achieve the necessary large valence band offset \( \Delta E_V \), keeping the whole structure pseudomorphic. A new asymmetric double heterostructure including AlAsSb/InAs (as a III–V part) and CdMgSe/CdSe (as a II–VI part), which is proposed as an active region for the modified mid-infrared laser, was grown for the first time by MBE on p\(^+\)-InAs (100) substrate.

The III–V part grown at a substrate temperature \( T_S = 480^\circ C \) consists of a 0.1 \( \mu \text{m} \) thick p\(^+\)-InAs:Be buffer layer followed by a 20 nm thick p-AlAsSb:Si barrier and, finally, of an
undoped 0.6 µm-InAs layer ($n<10^{17}$ cm$^{-3}$). A thin As cover was used as a passivating layer to protect the surface from oxidation in atmosphere, when transferring the structure to a separate II–VI MBE chamber through the air. After removal of the As cap by annealing the wafer at $T_S<480^\circ$C, a Cd(Mg)Se growth was initiated with a formation of In-Cd interface by a 5 s exposure of the (4 × 2)In-rich InAs surface controlled by RHEED to a Cd flux. Further, a migration enhanced epitaxy (MEE) mode was employed at $T_S = 200^\circ$C to grow $\sim$10 nm of CdMgSe nucleation layer. No 3D growth stage was observed. The rest of the II–VI structure, involving a nominally undoped 50 nm-CdMgSe layer followed by 0.3 µm of n-type CdMgSe:Cl and then by 10 nm-CdSe:Cl, was grown in MBE mode at $T_S = 280^\circ$C under the (2 × 1)Se-rich conditions. ZnCl$_2$ was used as an n-dopant source. The electron concentration of $n \sim 4 \times 10^{17}$ cm$^{-3}$ was obtained for the CdMgSe layer from C–V measurements. Mg mole fraction was estimated as 12% from x-ray diffraction (XRD) measurements which also confirm a pseudomorphic nature of the II–VI layers. The cross-sectional scanning electron microscopy image of the sample is presented in Fig. 2(a).

To measure photoluminescence (PL), we used single-grating monochromators and different excitation sources for different spectral regions. InGaAs cw laser diode emitting at 950 nm was used to excite PL in the III–V part of the structures, responsible for infra-red (IR) spectral region, while a 325 nm line of a cw He-Cd laser was used to excite PL from Cd(Mg)Se. For electroluminescence (EL) studies, the mesa-stripe samples were processed by a standard photolithography with diameters of a mesa and a contact of 300 and 50 µm, respectively. EL spectra were measured using a MDR-4 grating monochromator and a lock-in amplifier. A liquid N$_2$-cooled InSb photodetector was used for detection. Spontaneous EL spectra were measured both under quasi-cw conditions with pulse duration of $\tau = 2.5$ ms and filling factor of 1/2 and in a pulsed mode with a pulse duration $\tau = 1-10$ µs and a repetition rate $f = 10^3-10^4$ Hz.

Figure 2(b) summarizes the PL spectra measured at 77 K in the structure. Two relatively narrow peaks are visible at 0.410 eV and 2.086 eV, which are attributed to the near-band-edge radiative recombination in InAs and CdMgSe layers, respectively. To estimate the band-offset at the InAs/CdSe interface we used the “model-solid theory” of Van de Walle [2], which suggests the type II band line-ups. InAs represents a $\sim$60 meV potential barrier for electrons at the bottom of the CdSe conduction band, whereas the heavy hole band offset at the interface is as large as $\sim$1.42 eV. An incorporation of the large enough content of Mg is expected to change the situation at the InAs/CdMgSe interface from type II to type I. Indeed, the peak energy of the CdMgSe emission band taken from the spectrum in Fig. 2(b) allows an estimation of the CdSe/CdMgSe band gap difference as about 350 meV ($E_G$ of
MBE cubic CdSe has been found to be 1.74 eV at 80 K [3]). One can expect that at least a half of this value falls on the conduction band offset $\Delta E_C$, resulting in the strong type I band alignment at the InAs/CdMgSe interface with $\Delta E_C$ at least larger than 110 meV and $\Delta E_V \sim 1.6$ eV. Thus, the AlAsSb/InAs heterointerface with well known $\Delta E_C = 1.28$ eV value and the InAs/CdMgSe heterointerface probably can prevent the electron and hole leakage from the InAs active layer.

The intense electroluminescence has been found at both 77 and 300 K as shown in Fig. 3. The EL spectrum at 77 K contains the single emission band with a photon energy maximum at $h\nu = 430$ meV and FWHM = 40 meV. The emission band had a weak asymmetric shape with abrupt high-energy edge. The room temperature EL spectrum contains also the single emission band with a photon energy maximum at $h\nu = 396$ meV and FWHM = 68 meV, although the peak has a reverse asymmetry with a noticeable short-wavelength tail. The photon energy of the spontaneous EL is close to the PL energy maximum observed in Fig. 2(b).

With the temperature increase from 77 to 300 K, the maximum EL intensity decreases just by 7–10 times, which evidences a weaker temperature dependence of the spontaneous emission in this structure compared to conventional InAsSbP/InAs-based laser structures. The dependences of EL intensity on a drive current at cw and pulsed modes were studied both at low and room temperatures (Fig. 4). The dependence of spontaneous emission at 77 K exhibits the behavior close to a superluminescence regime.

In conclusion, a new double heterostructure with high asymmetric band offsets, based on a combination of III–V (AlAsSb/InAs) and II–VI (CdMgSe/CdSe) heterostructures, has
been proposed for the first time as the active region of mid-infrared laser and successfully grown by MBE. Intense longwavelength electroluminescence has been observed both at low (77 K) and room temperature. Weak temperature dependence of spontaneous emission from the structure serves an evidence of the effective carrier confinement in the InAs layer due to high potential barriers in conduction ($\Delta E_C = 1.28$ eV) and valence ($\Delta E_V \sim 1.6$ eV) bands.

Acknowledgements

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References

Epitaxial growth and characterization of MnAs/Si(111) nanoscale magnetoelectronic heterostructures

A. G. Banshchikov, R. V. Pisarev, A. A. Rzhevsky, N. S. Sokolov, Ahsan M. Nazmul† and M. Tanaka†
Ioffe Physico-Technical Institute, St Petersburg, Russia
† The University of Tokyo, Bunkyo-ku, Tokyo 113-8654, Japan

Abstract. Molecular beam epitaxy was used to grow thin (6–12 nm) ferromagnetic MnAs films on a Si(111) substrate. The characterization of the film structural and magnetic properties was carried out using X-ray, RHEED, AFM and magneto-optical methods.

Introduction
In recent years, epitaxial ferromagnetic thin films on semiconductors have become a subject of intensive studies [1, 2]. It is expected that such systems can lead to a new class of devices which integrate magnetic, semiconducting and optical properties. Ferromagnetic manganese arsenide (MnAs) films on silicon (Si) and gallium arsenide (GaAs) substrates are attractive for such applications due to excellent magnetic properties and a high Curie temperature (40°C). Much progress has lately been made in the study of relatively thick MnAs layers [3, 4]. At present, many research groups focus their efforts on the growth of nanoscale ferromagnetic films on semiconductors because these can display new spin-dependent kinetic and magneto-optical phenomena [5].

In this paper we report on MBE growth, epitaxial structure and magnetic characterisation of MnAs nanoscale films.

1. Growth
Manganese arsenide thin films having the thickness (6–12 nm) were grown in a conventional III–V MBE system (ULVAC MBC-508) with a Mn effusion cell. We used Si(111) substrates with 1.5° misorientation. After chemical treatment [6] and drying in N2 gas flow, the Si substrates were introduced into the MBE growth chamber and annealed at ∼900°C at As4 flow for a few minutes. With the As4 flow kept on, the MnAs epitaxial growth was started by supplying Mn flow. The typical growth rate was about 0.8 nm/min. The crystalline quality of the substrates and ferromagnetic film layers was monitored in situ by RHEED.

The structural characterization of the (6–12 nm) MnAs/Si(111) samples was made by X-ray diffraction measurements (θ–2θ method). The X-ray results for the B1118 film (d = 10 nm) are shown in Fig. 1. One can see MnAs[0001] (at 15.5°), MnAs[0002] (at 31.3°) and MnAs[0004] (at 65.3°) peaks in addition to intensive Si(111) and Si(222) peaks. These peaks indicate that the film grows epitaxially in the direction of c-axis. The average thickness (d) was defined as the amount of material deposited on the Si substrate.

The surface film morphology was measured with a Digital Instruments atomic force microscope operating in the contact mode. The surface morphology features for 3 films is depicted in Fig. 2. In Fig. 2(a), the thin film B1105 has a uniform distribution of MnAs across the substrate surface. Hexagon-like islands appear with increasing average
Fig. 1. X-ray diffraction spectrum of a MnAs film (B1118, \( d = 10 \) nm) grown on Si(111). The assignment of dominant peaks is shown.

Fig. 2. AFM images of MnAs films: (a) B1105 (350°C; 1.2; 6 nm), (b) B1118 (400°C; 1.2; 10 nm), (c) B1083 (350°C; 2.0; 12 nm). In brackets: the substrate temperature, As4-to-Mn flow ratio and the average thickness, respectively.

thickness (for B1118 and B1083), as can be seen in Fig. 2(b),(c). In Fig. 2(b), the islands have heights up to 50 nm and lateral dimensions in the range from 50 nm to 100 nm. Figure 2(c) demonstrates a tendency for larger island sizes up to the heights of 50–100 nm.

2. Magneto-optical studies

The magneto-optical Kerr rotation effect was measured in the longitudinal and polar geometry [7] when linearly polarized light was reflected by the films. The measurements were carried out at the wavelength of 0.63 \( \mu \)m (He–Ne laser) in a magnetic field up to 0.7 T at room temperature. The measurement sensitivity of rotation was \( \sim 5'' \). Figure 3(a) shows the Kerr hysteresis loops measured in the longitudinal geometry in the B1083 and B1105 films. The loops are saturated. Figure 3(b) demonstrates the azimuth dependence of the Kerr rotation magnitude in the B1083 film. The dependence is mostly isotropic. The measurements in the polar geometry using a solenoid to create a field of 0.05 T have shown that the value of the polar effect in different films is by about a factor of ten smaller than in the longitudinal geometry and no saturation is reached. Figure 4(a) illustrates the dependence of the longitudinal Kerr effect on the average film thickness.

3. Discussion and conclusions

The measurements of Kerr hysteresis loops clearly indicate the ferromagnetic character of the MnAs films. The azimuthal dependencies of the Kerr effect show that the films can be
Fig. 3. (a) The Kerr hysteresis loops in the longitudinal geometry for p-polarized light at the incidence angle of $\varphi \approx 15^\circ$ in the B1083 and B1105 films. (b) The azimuth dependence of the longitudinal Kerr rotation in the B1083 film.

Fig. 4. The longitudinal Kerr rotation as a function (a) of the average film thickness; (b) of the total intensity of MnAs X-ray peaks.

characterized as having “easy plane” anisotropy with magnetization in the film plane. Since MnAs has the hexagonal NiAs structure, this type of anisotropy apparently follows from the (111) orientation of the substrate, which provides a high epitaxial growth symmetry. The dependence presented in Fig. 4(a) can be understood taking into account the fact that the films studied are much thinner than the light penetration depth into the sample. In this case, the Kerr effect is proportional to the thickness of the magnetic layer. An analysis of the AFM images shows that the Kerr rotation magnitude depends on the film morphology. The effect is larger in films where MnAs islands display hexagon-like patterns and where the dimensions of the islands are larger. We suggest that these islands form the magnetic fraction of the film. To prove this suggestion, we have replotted the data of Fig. 4(a) as a function of the magnetic fraction evaluated from the total intensity of MnAs X-ray diffraction peaks normalized to the integral Si substrate peak intensity. The results are presented in Fig. 4(b). The data in Fig. 4(b) are well described by the linear dependence crossing the thickness axis close to zero.

Thus, we have grown thin (6–12 nm) MnAs films on Si (111) by MBE. X-ray diffraction analysis has shown that the MnAs growth plane is (0001). The film surface morphology was studied by AFM. The AFM images indicate that MnAs hexagon-like islands are formed on the Si substrate. The magneto-optical studies have shown that the films are ferromagnetic with the “easy-plane” type of anisotropy. The combined analysis of RHEED, X-ray diffraction, AFM and magneto-optical studies allows us to conclude that hexagon-like islands form
the magnetic fraction and can be attributed to MnAs.

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References

Influence of annealing on the formation of InGaAs quantum dots in GaAs matrix during metal organic chemical vapor deposition

N. A. Cherkashin, N. A. Bert, N. N. Ledentsov, I. V. Kochnev, V. M. Lantratov and Yu. G. Musikhin
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Structure investigations of InGaAs quantum dots (QD) grown by metal organic chemical vapor deposition (MOCVD) on GaAs substrate at various growth conditions were carried out by transmission electron microscopy. Incomplete covering of InGaAs QD layer by a thin GaAs spacer grown at 480°C followed by annealing at 600°C is shown to result in removal of developed defect structure observed in unannealed samples. The growth of three InGaAs QD layers separated by GaAs spacers using the same treatment algorithm for each InGaAs QD layer led to perfect QD system formation.

Introduction

During the last several years self-organized nano-sized domains (so-called quantum dots) have been attracting much attention [1, 2]. In particular, QD present the ultimate spatial confinement of charge carriers and to date have been applied as an active media in modern semiconductor devices, i.e. lasers [1]. Currently molecular beam epitaxy is widely used to grow structures with InAs or InGaAs QDs on GaAs substrate [1]. The MOCVD growth process for QDs in InAs/GaAs system has not been developed yet [2]. The aim of this work is to specify the influence of the growth conditions on the formation of InGaAs QDs during the growth on GaAs substrate by MOCVD.

1. Specimens and methods

The structures under study were grown by MOCVD using equipment with a horizontal resistively heated reactor at low pressure (76 Torr). Three kinds of structures with a single InGaAs QD layer and two kinds of structures with three InGaAs QD layers separated by GaAs spacers were grown. The AlGaAs and GaAs buffer layers were subsequently grown at 480°C on GaAs substrate (001) for all structure kinds. To grow the sample with the single QD sheet InGaAs layer was deposited for 3 sec to be transformed into QDs and cover AlGaAs and GaAs layers of 15 nm thickness each were then grown at 480°C. The main difference between the first and second or third types of structures is that the InGaAs QD layer was partially overgrown by 5 nm GaAs layer followed by annealing at 600°C and that the deposition of the AlGaAs and GaAs cover layers was performed at 600°C. The difference between the second and the third types of structures is in an annealing time of 10 or 20 min, respectively. To grow the sample with three QD sheets three InGaAs layers separated by 120 nm GaAs spacers were deposited at 480°C for 3 sec each, annealed and covered at 600°C by AlGaAs and GaAs layers of 15 nm thickness each. The main difference in the growth procedure for this structure and the second one is that the annealing at 600°C for the second structure was repeated three times after incomplete 5 nm GaAs covering of each InGaAs QD layer. Investigations were carried out using transmission electron
microscope (TEM) Philips EM-420 operating at 100 kV accelerating voltage. Specimens were prepared both in (001) plan view and (110) cross section. The conventional technique of TEM specimen preparation including mechanical treatment (grinding and polishing) and final sputtering by Ar$^+$ ions with an energy of 4 keV at 14° ion beam tilt from the sample surface was used.

2. Results and discussion

To obtain a distinct contrast of epitaxial layers of semiconductor heterostructures with zinc-blend lattice the dark-field (DF) technique of image formation with operating reflection (002) of which the amplitude is proportional to the difference between atom scattering factors of A and B sublattices is widely used. Analyzing (001) plan-view bright-field (BF) and (110) cross-section dark-field images obtained in two-beam conditions with operating reflections (220) and (002), respectively, allowed us to evidence QDs presence by the characteristic contrast. Along with QDs, three-dimensional (3D) dislocated islands showing characteristic moiré pattern were also detected in the sample with the single InGaAs QD layer grown without annealing. Using the value of GaAs (220) interplanar distance and the average island moiré period the average lattice constant of 3D islands was estimated to be 5.98 Å. Taking into account a residual strain in the 3D islands the obtained value is believed to correspond to the InAs lattice constant. The 3D island density is $\sim 10^8 - 10^9$ cm$^{-2}$. The island lateral size varies from 50 to 100 nm. At the same time no developed defects were observed in the structure with the annealed InGaAs QD layer. Table 1 shows the results of image analysis including average QD density, lateral size and height for the samples with the single InGaAs QD layer. Thus annealing of InGaAs QD layer partly covered by 5 nm GaAs spacer leads to improving crystal quality and changing QD shape, i.e. decreasing QD height and increasing QD lateral size. To explain the obtained data one needs to analyze the growth process of material lattice mismatched to the substrate. If the sum of layer surface energy and interface energy does not exceed substrate surface energy the deposition of small InGaAs quantity on the GaAs (001) surface leads to the planar strained InGaAs layer formation. The subsequent material deposition results in an increase of a layer elastic strain energy. To decrease this energy the layer can spontaneously transform to an array of three-dimensional islands on the residual wetting layer (Stranski–Krastanow (SK) growth mode). These islands have the pyramidal shape [1] and is known to be QD. But actually three-dimensional dislocated islands significantly exceeding QDs in size also appear. The reasons for these islands to occur are not clear yet but the substrate surface undulations and the presence of other surface peculiarities which are difficult to avoid in MOCVD may be the nucleus of dislocated islands. The presence of dislocations in islands may cause an enhanced material accumulation on them. The average thickness of the GaAs spacer used for overgrowth of QDs is about 5 nm which is insufficient for complete covering of

Table 1. Data obtained from TEM images of InGaAs QD layer.

<table>
<thead>
<tr>
<th>The structure type</th>
<th>Average QD density, cm$^{-2}$</th>
<th>Average QD lateral size, nm</th>
<th>Average QD height, nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unannealed</td>
<td>$1.3 \times 10^{10}$</td>
<td>17</td>
<td>11</td>
</tr>
<tr>
<td>Annealed (600°C)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$t_{\text{ann}} = 10$ min</td>
<td>$2.3 \times 10^{10}$</td>
<td>20</td>
<td>9</td>
</tr>
<tr>
<td>Annealed (600°C)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$t_{\text{ann}} = 20$ min</td>
<td>$2.4 \times 10^{10}$</td>
<td>20</td>
<td>7</td>
</tr>
</tbody>
</table>
QDs having ~11 nm height according to our observations. Under annealing with growth interruption In migrates from the top of pyramidal QD and distributes on the GaAs surface because of a high surface migration ratio of In. The changing QD shape is due to In migration and diffusion processes. Indium redistribution within an annealed layer is also the reason for dislocation disappearing on the interface of wetting layer and an island.

TEM study of the structure with three InGaAs QD layers separated by GaAs spacers and post-growth annealing revealed the developed defects presented all over the InGaAs QD layer-GaAs spacer system (Fig. 1). Three-dimensional dislocated islands were the origin of these defects starting with the first InGaAs QD layer. The presence of QDs was confirmed by the characteristic contrast which presented at the TEM images. The QD height is about 13 nm. Considerably decreasing QD density from the bottom layer up to the top one as well as a failure of planar growth are observed in (110) cross-section images. It is believed to be do to the influence of defects in the bottom InGaAs QD layer which accumulate the subsequent InGaAs deposit. The observed defects propagate all three layers and suppress In redistribution within each layer to eliminate defects under post-growth annealing. The InGaAs QD sheets showing dark contrast separated by 125 nm GaAs layers were observed in the (110) cross section image of the structure grown with intermediate annealings of each QD layer (Fig. 2). The characteristic contrast from QD presents in the images of each layer. The average QD density, lateral size and height calculated for the upper layer from the (001) plan-view image (Fig. 3) are $1.5 \times 10^{10}$ cm$^{-2}$, 19 nm, 10 nm respectively. One
can observe thin layers with dark contrast at the 10−11 nm distance from the QD sheets. These thin layers are believed to be InAs layers (Fig. 2). Comparing the distance from QD sheet to the observed thin InAs layer, the QD height and the GaAs spacer thickness specified by the growth regime we consider the changing QD shape and the thin InAs layer appearance to be the effect the In redistribution from QD tops onto the GaAs spacer surface during annealing after incomplete QD covering by GaAs spacer.

3. Conclusion
To summarize the main results we can formulate the following conclusions:

— In the case of InGaAs deposition at 480°C without an additional treatment at elevated temperature both perfect QDs and large three-dimensional dislocated islands are formed;
— Intermediate annealing after incomplete covering of QDs by GaAs spacer leads to defect elimination;
— The developed defect structure exists in the system consisting of three InGaAs QD sheets separated by GaAs spacers where annealing was applied after the deposition of all three periods;
— Applying growth-annealing procedure for each QD sheet results in perfect QD system formation with maintaining crystal quality of the whole structure.

Thus the intermediate annealings during InGaAs QD growth in GaAs matrix by MOCVD method improve the QDs and whole structure quality and can be introduced in QD-InAs/GaAs laser structure fabricating process.

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References
Deformation and viscouse flow in nano-imprinting

M. V. Chukalina, V. N. Matveev, V. V. Sirotkin, A. A. Svintsov and S. I. Zaitsev
Institute of Microelectronics Technology RAS, 142432 Chernogolovka, Russia

Abstract. The paper is concentrated on the theoretical consideration of imprinting process and of polymer flow during embossing of the master. Another activity was related to development of experimental techniques for performing imprinting.

Introduction

New technologies for mechanical pattern transfer in sub-100 nm regime with aim to replace conventional lithography in semiconductor mass production (on area 10 cm × 10 cm) are now under extensive investigations. Mechanical imprint of a stamp (mold) in plastic polymer or printing with “inks” forming self-assembled monolayers (SAMs) are in the center of interest.

During imprinting resist changes its shape under a stamp, this leads to extraordinary situation when a part of material should be transferred on distances about 10 cm through narrow (less than 100 nm) canal. Due to polymer nature of the resist one could expect additional difficulties if molecular size becomes comparable with canal width.

1. Estimation of printing time and force

An order of magnitude of imprinting force was obtained considering simple situation (a round flat stamp of radius $R$ moves with velocity $u$ into resist layer of thickness $h$ and viscosity $\eta$) solved firstly by Reynolds on the base of Navier–Stokes equations.

![Fig. 1. A round flat stamp (of radius $R$) moves with velocity $u$ into resist layer of thickness $h$ and viscosity $\nu$.](image)

On the base of Reynolds solution estimations of force ($F = \frac{3\pi \eta u R^4}{2h^3}$), time of imprinting $T$, pressure distribution, velocity distribution were obtained. Main conclusion from the analysis is that it is not possible to perform imprinting of 10 cm stamp for reasonable time ($F$ is about $10^7$ kg).

2. Optimization methods for design imprinting structures

One of the way to avoid enormous high force or time of imprinting is decreasing of polymer material transport. Practically it means design of structure with special cavities to gather extra material during imprinting. This means that some additional elements should be added to functional features of the structures during design to prevent flow on large distance. Some practical methods for the design and structure optimization are suggested and discussed.
3. Development of the numerical/analytical tool for approximate description (information capacity is $10^{12}$–$10^{14}$ pixels)

An information capacity of the structures is very high and could be estimate as ratio of a structure size ($\sim 10$ cm) to minimal feature (10–100 nm). This gives $10^{12}$–$10^{14}$ pixels. It is clear that it is not possible to describe and simulate objects of such sizes. Some approximate methods should be developed for description/simulation.

One of ideas is to use natural hierarchy of microelectronics structures. On this way an equation of viscose flow for small units of the lowest level should be solved then considering the smallest units as structureless elements consider flow at higher spatial and temporal scale for the units of higher level and so on. This way could be combined with another idea to take into account relief of the stamp effectively considering viscosity dependent on coordinates.

Imprinting could be roughly divided into two stages, first (with short matter transfer distance) is filling of stamp space (cavities) by resist. After exhausting free stamp space
further imprinting results to large distance of matter distance through narrow channels. Analyzing of the first stage is important to estimate duration of the stage. One more important reason is determination of resist shape near internal sharp edges when influence of surface tension and polymer coils size become essential.

To meet the goals formulated a numerical methods were developed for the two stages analysis on base of non-stationary and stationary Navier–Stokes equations and first simulations were performed.

Figure 2 shows the velocity field near the “ridge” in non-stationary stage where as Fig. 3 shows velocity distribution in stationary stage. Other characteristics of the flow were calculated: the pressure $P$ and the non-zero velocity components $V_x$ and $V_z$. The calculation demonstrate the lengths of hydrodynamic stabilization for both velocity components are $O(h)$, where $h$ is the depth of the canal. Note that the stabilized flow is characterized by $V_x = 4(h/L)[1 - (z/h)](z/h)$ and $V_z = 0$ in total correspondence to Reynold's solution.

The calculation were performed for different sizes of ridge, similar characteristics were calculated for trench of different sizes. Special calculation were made for $Re = 10^{-2}–10^2$. It was shown that flow practically independent on $Re$ for values below $10^{-1}$. So one important consequence of calculations is conclusion that nonlinear term Navier–Stokes equations could be removed from equations.

4. Polymer chain size effects

Polymer matter has some specific features which become essential at scales considered in nano-imprinting. Roughly polymer could be considered as matter consisting of 'particles' with radius $L$ related to polymer length $N$

$$L = L_0 N^{\frac{1}{2}}. \quad (1)$$

For molecular weight 950 K and 50 K this gives (at molecular weight of monomer equal 100 and effective length $L_0 = 1$ nm)

$$L = 20–100 \text{ nm.}$$

The particle size $L$ is comparable with resist thickness and imprinting structure details. But at this scale Navier–Stokes equations should be used with some accuracy. Viscosity is now dependent on distance from the walls and is not further local characteristics.

![Fig. 4. Polymer resist comprises a melt of coils.](image1)

![Fig. 5. Consideration of polymer coil size $L$ results to remarkable change in velocity profile at resist thickness $h$ comparable with $L$.](image2)
5. Viscosity near wall and Reynolds problem for polymer flow

To calculate nonlocal viscosity we supposed that any contact with the wall makes polymer chain immobile so only mobile part of polymer chains can contribute in flow. Effectively this means that viscosity becomes lower near the wall. Let $W(z)$ is a probability for a chain to avoid the wall for the chain starting at point $z$ from the wall so only part $W(z)$ can take part in movement and viscosity $\eta(z)$ is reduced in comparison with bulk value $\eta_b$, $\eta(z) = \eta_b / W(z)$.

Function $W(z)$ was calculated considering polymer chain conformation (polymer coil) as a trace of random walk consisting of $N$ steps. Mathematical model for $W(z)$ calculation is non-stationary diffusion equation with absorption on the boundary. It gives for small $h/L$

$$W(z) = \sin \left( \frac{\pi z}{h} \right) \exp \left[ - \left( \frac{\pi L}{h} \right)^2 \right].$$

Velocity, pressure distributions and force $F_p$ were obtained for Reynolds problem, for example force

$$F_p = \frac{1}{2} \pi \eta u R^4 \exp \left[ \left( \frac{\pi L}{h} \right)^2 \right]$$

increases exponentially instead of power $(1/h^3)$ law in classical Reynolds solution.

As result one could expect impossibility to deform resist layer after reaching some thickness close to size $L$. It is seen immediately (due to (1)) that usage of resist with lower molecular weight could reduce the thickness of incompressible layer. Another way is to use monomer liquid with photo- (thermo-) polymerization after imprinting.

6. Development of technology components for imprinting

Technological steps developed for imprinting realization consist of several steps.

**Electron lithography.** Test structures were designed and fabricated with minimal features of 300 nm by electron beam lithography.

**Etching/development.** After thin Al deposition and lift off the samples were dry etched to depth about 1 $\mu$m.

**Imprinting.** The most important step was made in development of imprinting, a stage with controllable heating (up to 200$^\circ$C) was fabricated.

First experiments with imprinting showed that transfer could be performed but it showed as well some resist pollution in trenches. Further optimization of regimes are desirable.

**Diagnostics.** For control of shape transfer optical and scanning electron microscopes were used. It is planned to use a specially developed device with diagnostic possibilities close to AFM mode is used for control as well.

Acknowledgments

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GaN/Al₂O₃ epilayers grown by MBE with a controllable nitrogen plasma composition

V. N. Jmerik, V. V. Mamutin, T. V. Shubina, M. G. Tkachman, V. A. Vekshin, V. V. Ratnikov, A. V. Lebedev, S. V. Ivanov and P. S. Kop'ev
† Ioffe Physico-Technical Institute, St Petersburg, Russia

Molecular beam epitaxy with N₂ plasma activation (PA MBE) is a promising technique to fabricate III-nitride nanostructures. The chemical activity of N₂ molecules in the PA MBE is increased due to excitation of molecular electron-vibration states as well as dissociation and ionization [1]. However, difficulties in the effective realization and control of these processes under ultra-low pressure conditions typical for MBE result in a low growth rate (much less than 1 µm/h) and imperfections in the epitaxial layers, induced by high-energy particles. To characterize material quality of GaN, photoluminescence (PL) measurements are widely used along with x-ray diffraction (XRD) studies. Particularly, the relative PL intensities of a useful band-edge line (BL) near 3.46–3.47 eV and a parasitic PL bands near 3.26–3.27 eV (DAP recombination line), as well as near 2.2 eV (YL line) are rather informative [2].

Nitrogen activators using both a radio frequency (RF) inductive discharge and that with electron cyclotron resonance excitation have been commonly employed in PA MBE [3]. In addition, we have proposed a new type of N₂ activator with an RF capacitively-coupled magnetron (RF-CCM) discharge [4, 5]. Using the RF-CCM activator with the discharge localized in the coaxial inter-electrode gap permitted us to obtain the GaN growth rate (\(\nu_g\)) up to 0.5 µm/h with reasonable quality. However, some growth rate lowering was observed during the exploitation of the RF-CCM operated in the “localized” mode at high RF-power (∼200 W) and a short distance between the activator and a substrate (∼4 cm).

In this paper, we demonstrate a new mode of the RF-CCM operation with the relatively intense plasma glowing extended into the growth chamber forwards the sample substrate. The “extended” mode shows good reproducibility and provides the GaN growth rate as high as 1.8 µm/h. Moreover, we demonstrate a possibility to control the GaN optical quality by a variation of the activated N₂ composition and a growth temperature.

GaN epitaxial films were grown on Al₂O₃(0001) substrates in a home made EP 1203 MBE setup equipped with the 13.56 MHz RF-CCM activator operating at a maximum power of 150 W and an axial magnetic field of 500 G in a coaxial design, as described in details elsewhere [5]. Unlike the “localized” mode employing a multi-hole output activator aperture, the single-hole one with a diameter of 3.5 mm was used for the “extended” mode. Typical distance between the activator aperture and the substrate was 7 cm. Nitrogen mass flow rate (\(Q_N\)) up to 30 sccm was controlled by measurements of N₂ background pressure in a growth chamber, which was lower than 1 mTorr. The MBE setup was equipped with one (in the most of the experiments) or two turbomolecular pumps with total effective pumping rates of 350 and 600 l/s, respectively. Thermal cleaning at 900°C, nitridation of the sapphire substrates at 850°C and growth of a GaN buffer layer (∼20 nm) at 350°C were followed by the growth of a main layer (∼2 µm) in a 500–700°C temperature range. All the processes were monitored in situ by reflection high energy electron diffraction.
Optical emission spectra (OES) of the RF-CCM discharge were also recorded in situ. In the “localized” mode the emission from a discharge chamber reflected by a Si mirror plate mounted on a substrate holder was registered, while in the “extended” mode the direct measurements of the glowing plasma volume in the growth chamber were used. PL spectra of the GaN epilayers were obtained at 77 K with excitation by a 325 nm line of a 1 mW He-Cd laser. XRD measurements were performed using CuKα radiation.

Preliminary testing of the RF-CCM has shown that the single-hole aperture enables one to excite the magnetron discharge in the “extended” mode with the OES intensity one order of magnitude higher than that in the “localized” mode (Fig. 1). Besides, higher intensities of the first and second positive series, related to excited N₂ molecules, as compared to the first negative one, related to N₂ ions, allow us to assume a lower electron temperature ($E_e$) in the “extended” mode [5]. Therefore, one may expect a higher content of metastable N₂ molecules and N-atoms impinging onto the substrate. In addition, a strong dependence of $E_e$ on N₂ mass flow rate has been found. The inset in Fig. 1 shows that the OES intensity ratio ($\xi$) of the ion to molecular lines at 391.4 and 380.4 nm, respectively, directly proportional to $E_e$, is raised towards low $Q_N$ values. It should be noted that electrical probe measurements revealed charged particles in the activated beam in this mode in contrast to the “localized” one.

The main technological result of employing the “extended” mode of the RF-CCM is the greatly enhanced GaN growth rate, which can be regulated from 0.2 to 1.8 $\mu$m/h, depending on the Ga flux, the distance to a substrate and $Q_N$. In addition, this mode provides the possibility to control the material quality of GaN films by variation of activated nitrogen beam composition. To illustrate this, the set of growth runs was carried out at different $Q_N$ and, hence, different $E_e$ (shown by (a)–(d) points in the inset in Fig. 1). The experiments were done at moderate values of $Q_N$ and Ga fluxes, when $v_g$ (about 0.7 $\mu$m/h) was determined by Ga flux, i.e. the films were grown under the N-rich conditions.
A variation of the optical properties of the GaN epilayers is illustrated in Fig. 2. One may see that to obtain the intense band edge emission with relatively suppressed parasitic lines, the discharge must be kept at the optimal $Q_N$ corresponding to $\xi = 1.2$ (Fig. 2(d)). Indeed, in the case of high $E_e$ ($\xi > 1.2$, Fig. 2(a)) both the content and energy of the ions in the activated beam are increased, resulting in a generation of non-radiative defect complexes [1]. Furthermore, high energy ions can be considered as a possible reason of the YL in the PL spectra, as was demonstrated in [6] and in our previous studies on the “localized” mode [7], where an absence of the YL was explained by a good separation of the high energy ions from the N$_2$ activated beam. Currently, the YL is considered as being related to Ga vacancies (V$_{Ga}$) [8], oxygen contamination and/or to point defects decorating dislocations [9, 10]. We believe that the ions serve as a trigger in the processes of the complex defect generation.

In the regime with low $E_e$ corresponding to the inessential ion component ($\xi < 1.2$, Fig. 2(b)), the YL may be completely suppressed. However, this regime is not optimal due to the domination of the DAP band in the PL spectra. This band is attributed to a 225 meV acceptor in GaN, which has commonly been assumed to have an intrinsic origin related to the V$_{Ga}$ [2]. Indeed, the high $Q_N$ results in a non-stoichiometric growth with the enhanced Ga vacancies generation. Additionally, at this conditions a molecular regime of the gas flow is disturbed, resulting in three-dimensional growth.

The appearance of the BL corresponds to the medium $E_e$, $\xi = 1.2$, Fig. 2(c). The increase in the substrate temperature from 500 to commonly used 700$^\circ$C permits us to enhance the BL with the better ratio of the BL/DAP(YL) intensities (Fig. 2(d)). The BL line with relatively narrow FWHM of 36 meV dominates in this PL spectrum. The reasonable quality of the former film is confirmed by (0002)GaN XRD rocking curves with FWHM values of 994 and 67 arcsec in $\omega$- and $\theta - 2\theta$ scans, respectively. In conclusion, our experiments have shown that the discovered “extended” mode in the RF CCM can provide a high MBE GaN growth rate. The comparative analysis of the OES and PL spectra reveals the optimal composition of a N$_2$ activated beam, which permits us to compromise on the
ratio between BL and parasitic lines.

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References

Monte Carlo simulation of quantum dots formation during heteroepitaxy

I. G. Neizvestny, L. N. Safronov, N. L. Shwartz, Z. Sh. Yanovitskaja
and A. V. Zverev
Institute of Semiconductor Physics SB RAS, 13 pr. Lavrenteva,
Novosibirsk, 630090, Russia

Abstract. Formation of quantum dots (QD’s) during heteroepitaxial growth was investigated by
Monte Carlo simulation. Shwoebel barriers for explanation of QD’s growth kinetic were suggested. Two different barriers for hops to the upper and lower layers were introduced in our model. Ranges of these barriers for QD’s formation were estimated.

Heteroepitaxial growth of highly strained structures offers the possibility to fabricate islands
with very narrow size distribution — quantum dots (QD’s). The quality, size and uniformity
of dots are known to be dependent on deposition conditions [1]. Experimental data indicates
Ge island array to be a kinetic-controlled one in contrast to InAs/GaAs system of arrays
of 3D coherently strained islands whose formation is governed by thermodynamics [1, 2].
Kinetic theories of ordering emphasize the crucial role of inhomogeneous strain fields in
the vicinity of 3D islands. Strain field dependence on island dimensions and deposited
dose were demonstrated elsewhere [1, 3]. Strains arising at the interface between substrate
and growing island during heteroepitaxial growth as in Ge/Si system are schematically
illustrated in Fig. 1.

![Fig. 1. Schematic drawing illustrating strained heteroepytaxial island (b) of material (a) on the
substrate (c) with lattice parameter of lower size. Darker color corresponds to higher stress.]

Figure 2 illustrates compression influence on energy for edge atoms in Si_{5} cluster. Ter-
ssoff potential [4] with parameter [5] was used in our calculation. As could be seen from
Fig. 2 energy minimum of atom situated at the origin of the coordinates increases and shifts
under pressure. It might be assumed that stress changes binding energy of edge atoms lead-
ing to variation of diffusion hops for atoms at tops and walls of 3D islands. We investigate
the kinetic of QD’s formation during heteroepitaxial growth using standard solid-on-solid
(SOS) model [6] as well as 3D model epitaxy on diamond-like crystal (111) surfaces [7].
Monte Carlo simulations of self-assembling process were recently reported [8–11]. How-
ever in these works only initial stages of 3D islands growth were considered. We simulated
3D islands growth during multilayer deposition and took into account peculiarities of edge atoms by introducing additional step edge barrier $E_{st}$ (Schwoebel barrier) in our model. This barrier has to be surmounted by an atom in addition to the surface diffusion barrier if adatom crosses an island edge. So the jump rate to cross an island edge is changed by a factor $P = \exp(-E_{st}/kT)$. Contrary to [10] we introduce two barriers for interlayer hops to the upper layer ($2 \rightarrow k$, $4 \rightarrow m$) and to the lower layer ($3 \rightarrow k$, $4 \rightarrow n$) in Fig. 1. Such asymmetry in Schwoebel barrier was found in Si$_{1-x}$Ge$_x$/Si(001) system [12]. These two barriers correspond to the model parameters $P_{up}$ and $P_{down}$. $P_{up}$ and $P_{down}$ varied in the following ranges: $0.2 < P_{up} < 5$, $0.1 < P_{down} < 1$ ensuring growth conditions associated with three mechanism of growth: layer-by-layer, 3D island and islands with wet layer growth (Stranski–Krastanov). Values of $P_{up} > 1$ mean that atom hops to the sites in the upper layer is more probable than to the neighbor sites in the same layer.

Crosshatched region in Fig. 3 corresponds to Stranski–Krasstanov growth mechanism while area lower shaded region is consistent with layer-by-layer growth and higher — with QD’s formation. Region (1) was obtained using SOS model for cubic crystal and region (2) using 3D model for diamond-like crystal. 3D islands appear only for $P_{down} < 1$, and no 3D are grown for $P_{down} > 1$, even for $P_{up}/P_{down} > 1$. For the shaded region QD’s formation takes place after some dose deposition. At the initial stage of growth islands of the second layer appear after coalescence of the first layer islands. Ratio of whole number of up hopping atoms to down hopping ones increases with time. This increase results from the fact that not only $P_{up}/P_{down}$ value determine 3D islands formation but numbers of up, down or lateral target sites for hopping atoms. Rather sharp size distribution of 3D islands observed in Fig. 4(c) (in comparison with Fig. 4(b)) is due to nucleation in the second layer.
Fig. 4. The simulated surfaces (160 × 160 atomic sites) using SOS model after 1.9 ML deposition for adatom diffusion length $\lambda = 20$ a.s., $b = 0.01$ (parameter $b = \exp(-E_b/kT)$ determines bond energy $E_b$ for given temperature) (a) $P_{up} = 1, P_{down} = 1$; (b) $P_{up} = 0.8, P_{down} = 0.2$; (c) $P_{up} = 1, P_{down} = 0.1$.

Fig. 5. The simulated surfaces (100 × 100 atomic sites) using 3D model after 2 ML deposition at $T = 725$ K, $P_{up} = 3, P_{down} = 0.3$: (a) critical coverage $\Theta_{cr} = 0.01$; (b) $\Theta_{cr} = 0.1$; (c) (1) — average 3D islands height dependence on $\Theta_{cr}$, (2) — thickness of flat film.

before coalescence of islands in the first layer. Islands nucleated earlier are distributed more uniform.

Figure 5 demonstrates simulation results of QD’s formation using 3D model for diamond-like crystal. Dependence of Schwöbel barrier on layer number was not taken into account in these calculations. However barrier dependence on average size of island was introduced in 3D model. We suggested barrier to be dependent on deposited dose in the following way: $P(\Theta) = [P_0 - 1]/\Theta_{cr} \cdot \Theta + 1$ for $\Theta < \Theta_{cr}$ and $P(\Theta) = P_0$ for $\Theta > \Theta_{cr}$, where $P_0 = \exp(-E_{sc}/kT)$, $\Theta$ is layer coverage, $\Theta_{cr}$ is critical layer coverage. After 2 ML deposition with Schwöbel barriers corresponding to $P_{up} = 3$ and $P_{down} = 0.3$ quiet different 3D islands appear for two different $\Theta_{cr}$. If $P$ achieves it’s maximum value $P_0$ after 1% monolayer coverage we get rather high sharp islands (Fig. 5(a)) and if $P = P_0$ after 10% monolayer coverage — islands become flatter (Fig. 5(b)). Dependence of average islands height on critical coverage $\Theta_{cr}$ is shown in Fig. 5(c), curve 1. Curve 2 corresponds to film thickness in the case of layer-by-layer growth. Later on we are going to introduce barrier dependence on atomic layer number of growing island.

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Simulation of pores sealing during homoepitaxy on Si(111) surface

I. G. Neizvestny, N. L. Shwartz, Z. Sh. Yanovitskaja and A. V. Zverev
Institute of Semiconductor Physics RAS SB,
13 pr. Lavrenteva, Novosibirsk, 630090, Russia

Abstract. Simulation of homoepitaxy process on porous Si(111) using 3D Monte Carlo model was carried out to give estimation of necessary dose for complete pores overgrowth of different sizes, porosity and various deposition flux intensities for given temperature.

Porous silicon (PS) is one of the most promising materials for Si-based emitting devices that can be useful in integrated optoelectronics [1, 2]. Recently an approach to fabricate a compliant substrate for growing high-quality heteroepitaxial layers was proposed [3]. A double layer of PS obtained from heavily doped p-type Si was used as the substrate. A low-porosity top layer and a bottom layer with high porosity act as a mechanical damper between heterostructure and a massive support and allow to grow GeSi film without misfit and threading dislocation. The same idea was used when growing GaAs layer on thin Si layer over porous Si substrate [4]. Usually there are only circumstantial evidences that porous substrate is overgrown [5, 6]. Number of deposited layers necessary for complete pores healing is of great importance for high quality heteroepitaxial growth on porous substrates.

3D model of epitaxy on the surfaces of diamond like crystals [7] was applied for analysis of epitaxy on (111) porous silicon surfaces. Our lattice model can take into account atoms from surface region up to hundred atomic layers in the depth. One monolayer could contain up to 10^5 atoms. The main event in the model is diffusion hop, with atom hopping to the empty lattice site in the first, second or third coordination spheres. Adsorption process is considered to be independent on diffusion. Voids and vacancies in the bulk, multilevel steps and deep pores on the surface are possible. The abandonment of solid-on-solid (SOS) principle permits atoms movement along the walls of steps and pores.

Simulation of homoepitaxy process on porous Si(111) was carried out giving estimation of necessary dose for complete pores overgrowth of different sizes, porosity and deposition rate. Simulations were carried out in the following ranges of substrate parameters: porosity 13 ≤ P_s ≤ 64%, pores diameter: 3.5–14 nm and rather wide ranges of deposition rate, according to experimental works [3, 5, 6]. In the model deposition rate is introduced through the universal parameter n_{dif} ∼ D/V, where D is diffusion coefficient and V - deposition rate. Simulation results of epitaxial growth on porous surface with 100 × 100 atomic sites (a.s.) at T = 1073 K [5, 6] for different surface porosity, pores sizes and deposition rate V = 0.004–40 nm/min are presented in Fig. 1–4. For given temperature one can estimate deposition rate according to the formulae V = v · \exp(-E_d / kT) / n_{dif}, where E_d is activation energy for surface diffusion (eV), V is deposition rate (bilayer per second, BL/s), v is vibration frequency (1/s), k is Boltzmann’s constant, T is substrate temperature (K). Taking v = 10^{13} s^{-1}, E_d = 1.75 eV [7], T = 1073 K for n_{dif} = 10^5 1/BL one obtains V = 0.6 BL/s.
Fig. 1. Homoeptaxy simulation on porous Si(111) surface at $T = 1073$ K for $n_{\text{dif}} = 10$ (high deposition rate). Pores diameter is 10 atomic sites, height is 20 BL, surface porosity $P_s = 26\%$. (a) initial surface top view; (b) initial cross-section AA along [011] direction; (c) cross-section AA after 1.5 BL deposition; (d) cross-section AA after 3.1 BL deposition.

Fig. 2. Dependence of minimum number of deposited bilayers necessary for complete pores overgrowth $N_c$ on porosity $P_s$ for small pores for high and low deposition rates: pores diameter 10 a.s. $T = 1073$ K, (1) — $n_{\text{dif}} = 10$ ($V = 6000$ BL/s), (2) — $n_{\text{dif}} = 10^5$ ($V = 0.6$ BL/s).

Plan view and cross-section along [011] direction for the surface with small pores and surface porosity $P_s = 26\%$ before and after deposition with high deposition rate could be seen in Fig. 1. For these conditions it was necessary 3.1 BL of silicon to be pores completely overgrown. Figure 2 demonstrates dependence of minimum number of deposited bilayers necessary for complete pores healing ($N_c$) on surface porosity $P_s$ for low and very high deposition rates and small pores. For high deposition rate $N_c$ is independent of porosity and for low $V$ $N_c$ increases with $P_s$. This dependence is clear: for high deposition rate atoms reaching pores are gathered only from small region around each pore limited by migration length. For porosity less than 65\% and deposition rates under examination migration length is less than average distance between pores. For low deposition rate all adatoms on the surface are involved in healing process, so the higher porosity the less numbers of atoms are left on the surface (portion of atoms falls to the bottom of the pores). Figure 3 demonstrates dependence of $N_c$ on $n_{\text{dif}}$ for three different pores sizes. For large pores strong dependence of flux intensity is observed. Large number of deposited layers $N_c$ is necessary for complete pores sealing for high deposition rate ($N_c > 40$ BL) while for low flux $N_c$ is rather small and has only weak dependence on pores diameter. In the latter case $N_c$ is determined by deposited dose rather than diffusion, so dose limited regime takes place. Figure 4 illustrates $N_c$ dependence on porosity for different pores sizes (Fig. 4(a)) and on pores size for different deposition rates (Fig. 4(b)). $N_c$ independence of porosity for high deposition rate is evident from curves Fig. 4(a) and was discussed above. Linear dependence of $N_c$ on pore diameter for low deposition rates is clear from curves 2, 3 while curve 1 ($n_{\text{dif}} = 10$, corresponds to high deposition rate) demonstrates superlinear dependence. Cross-sections along [011] direction before (a) and after deposition (b-c) on porous Si(111) surface with
Fig. 3. Dependence of $N_c$ on $n_{\text{dif}}$ for different sizes of pores. $T = 1073$ K, porosity $P_s = 26\%$, pores diameter: (1) — 40 a.s., (2) — 20 a.s., (3) — 10 a.s.

Fig. 4. $T = 1073$ K (a) Dependence of $N_c$ on porosity $P_s$ for high deposition rate: ($n_{\text{dif}} = 10$) for different sizes of pores: pores diameter: (1) — 40 a.s., (2) — 20 a.s., (3) — 10 a.s.; (b) Dependence of $N_c$ on pores size for porosity $P_s = 26\%$ and different growth rate: (1) — $n_{\text{dif}} = 10$, (2) — $n_{\text{dif}} = 10^3$, (3) — $n_{\text{dif}} = 10^5$.

Fig. 5. Homoepitaxy simulation on porous Si(111) surface at $T = 1073$ K for high and low deposition rates, surface porosity $P_s = 50\%$, pores diameter 40 a.s., height 20 BL. (a) initial cross-section along [011] direction; (b) cross-section after 37.7 BL deposition for high deposition rate ($n_{\text{dif}} = 10$); (c) cross-section after 9.6 BL deposition for low deposition rate $n_{\text{dif}} = 10^5$.

The differences in sealing process are evident from these figures. For high deposition rate (Fig. 5(b)) overgrown pores have cone shape with convex up bottom and $N_c$ is found to be rather large. In this case migration length is small and adatoms create islands as between pores on the surface as on pores bottom. Atom flux in our simulation is perpendicular to the surface. Overhanging walls arising during growth process are beneficial for whiskers formation along pores walls near the bottom. Quite different situation one has for low deposition rate (Fig. 5(c)). At the initial stage of growth pores shape keeps nearly invariable, only diameter of each pore is decreasing due to larger migration length of adatoms moving along pores walls. Final sizes of buried pores in the latter case are noticeable smaller.
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References

Non-linear theory of alloy phase separation in open systems: 
Kinetic phase transitions between 1D and 2D structures

V. A. Shchukin and A. N. Starodubtsev
Ioffe Physico-Technical Institute, St Petersburg, Russia

Spontaneous formation of nanometer-scale composition-modulated structures is a common phenomenon for III–V and II–VI semiconductor alloys [1]. These structures are mostly being formed in open systems during the epitaxial growth. Theoretical understanding has so far been achieved of earlier stages of growth where the linear stability analysis of the homogeneous alloy growth applies [2–4]. In semiconductors, bulk diffusion coefficients are, typically, negligibly small compared to surface ones. Then, fluctuations of composition are created at every surface atomic layer and are frozen once this layer is overgrown by subsequent layers. Frozen fluctuations of composition from the entire film affect, via long-range strain fields, migration of surface adatoms. This interaction can result in a kinetic instability of a homogeneous alloy growth against fluctuations of composition. However, to describe a final structure formed during the growth, a non-linear theory is needed.

In our earlier paper [5] we have solved a non-linear problem for the final structure of a growing alloy in a weak segregation regime close to the onset of the instability, i.e. in the vicinity of the solid line at the linear stability phase diagram of Fig. 1(a). In the diagram of Fig. 1(a), at high temperatures \( T \), the instability is hindered by the mixing entropy of the alloy. At low \( T \), the surface diffusion coefficient decreases, \( D(T) = D_0 \exp(-E_a/k_B T) \), where \( E_a \) is the activation energy. Then adatoms are buried by the incoming flux faster than migrate along the surface, that hinders the instability. Due to elastic anisotropy of zinc-blend semiconductors, the instability occurs for the first time, for the growth on a (001) substrate, for composition fluctuations with the wave vector in an elastically soft direction [100] or [010]. The solution of a non-linear problem in a weak segregation regime [5] shows a principal possibility of the formation of either a 1D structure modulated along [100] or along [010] or a 2D structure modulated in both [100] and [010] directions. However, a
2D structure occurs only in a very narrow temperature interval which is hard to realize experimentally. Intending to explain the formation of commonly observed 2D modulated structures, in the present paper we seek the final state of the alloy growth in a strong segregation regime, i.e., in the entire region of the $T-v$ phase diagram where the homogeneous growth is unstable. We consider the growth of an alloy $A_{1-c}B_cC$ by molecular beam epitaxy (MBE) on an atomically rough surface. The alloy is lattice-matched on average to the (001) substrate. The growth proceeds via deposition of atoms on the surface, surface migration of atoms in a stress- and composition-dependent chemical potential, and incorporation of atoms into the growing crystal, desorption being neglected. Let the composition equal $c(r) = \bar{c} + \phi(r)$, the average composition being $\bar{c} = 1/2$, and the surface profile equal $h(x, y) = vt + \zeta(x, y)$, where $v$ is the average growth velocity controlled by the deposition flux. Coupled kinetic equations describe a joint evolution of composition fluctuations $\phi(r)$ at the advancing surface and of the surface profile fluctuations $\zeta(x, y)$,

$$
\begin{align*}
\frac{\partial \zeta}{\partial t} &= \frac{D_S}{k_B T} \nabla_i \nabla_j \left[ a \frac{\delta F}{\delta \zeta} - \phi \frac{\delta F}{\delta \phi} \right] \\
\frac{\partial \phi}{\partial t} &= \frac{D_c}{k_B T} \nabla_i \nabla_j \frac{\delta F}{\delta \phi} - \left[ v + \frac{\partial \zeta}{\partial t} \right] \frac{\delta F}{\delta \phi}.
\end{align*}
$$

Here $F$ is the total Helmholtz free energy, $D_S^i_j$ is the diffusion coefficient tensor related to the evolution of the surface, $D_c^i_j$ is the one related to the substitutional diffusion of alloy components on the surface, and $a$ is the lattice parameter.

Fig. 2. Coupling between composition modulation and surface profile. (a) Two separate bulk alloys; (b) Composition-modulated structure with the planar surface; (c) Composition-modulated structure with a non-planar surface; (d) Lattice parameters of bulk alloys with compositions $c_1$ and $c_2$; (e) Lattice parameters of coherently conjugated composition domains in a stressed system with the planar surface; (f) Lattice parameters of coherently conjugated domains in a partially relaxed system with a non-planar surface; (g) Elastic driving force to phase separation: atoms A(B) are attracted by A(B)-rich domains; modulation of composition $\phi$ and of the lattice parameter $a$; (h) modulation of the lattice parameter at the planar surface (dashed line) and at a non-planar surface (solid line).

For the lattice-matched alloy growth, the composition instability and the morphological one are not coupled in a linear regime [3, 5]. A key property of the non-linear regime is that both instabilities are coupled (Fig. 2). Let the structure consist of alternating domains with compositions $c_1 < \bar{c}$ (A-rich domains) and $c_2 > \bar{c}$ (B-rich domains) and the binary AC have smaller lattice parameter than BC. Then, for bulk alloys $A_{1-c_1}B_{c_1}C$ and $A_{1-c_2}B_{c_2}C$, one has $a_0(c_1) < \bar{a} = a(\bar{c}) < a_0(c_2)$ (Fig. 2(d)). In the structure of Fig. 2(b), domains
are coherently conjugated. Then, A-rich domains are stretched with respect to intrinsic lattice parameter of bulk $A_{1-c}B_{c}C$, i.e. $a(c_1) > a_0(c_1)$. At the same time, they are compressed with respect to the average lattice parameter of the structure $\bar{a}$. B-rich domains are compressed with respect to the lattice parameter of bulk $A_{1-c}B_{c}C$, and stretched with respect to $\bar{a}$. The surface consists of alternating domains under tensile and compressive stress and is unstable against undulations, if the modulation wavelength exceeds a certain critical value. Then the energy gain due to elastic relaxation exceeds the energy cost of a non-planar surface profile, and the surface will consist of troughs over domain boundaries and crests in the center of each composition domain (Fig. 2(c)). Due to elastic relaxation, lattice parameters of A-rich and B-rich domains are shifted towards their intrinsic values, $a_0(c_1)$ and $a_0(c_2)$ (Fig. 2(f)).

Figures 2(g) and 2(h) illustrate the reaction of surface undulations on phase separation. Atoms B having larger atomic radius prefer to incorporate to surface regions which are stretched with respect to the average lattice parameter $\bar{a}$, i.e., to B-rich domains (see, e.g., [4]). Similarly, A atoms prefer to incorporate to A-rich domains. Elastic driving force to phase separation is proportional to the actual variation of the lattice parameter, $a(c_2) - a(c_1)$. Since this variation increases due to surface undulations (Fig. 2(f)), the elastic driving force to phase separation increases, too.

To obtain stable steady-state solutions of kinetic equations (1), we have solved these equations numerically and have checked the stability of solutions by integration of kinetic equation over time in the vicinity of the steady state solutions.

Figure 1(b) displays the calculated steady-state phase diagram of the alloy growth in case of isotropic surface diffusion. The modulation period is known to increase with the decrease of the growth velocity $v$ [6]. The interplay between surface and elastic energies favors surface undulations at large period, i.e., at low $v$. A non-planar surface favors a 2D structure modulated in both [100] and [010] directions versus a 1D structure modulated in either [100] or [010] direction. The effect is similar to that for strained islands in lattice-mismatched systems where 2D structures (pyramids) provide a more efficient elastic relaxation than 1D structures (prisms) [7]. The diagram contains the regions of the homogeneous growth, of the growth of 2D structures, and of the growth of 2D structures, and the region where both 1D and 2D structures can grow. In the latter region, the actual final structure of the growing alloy depends on initial conditions. To obtain the structure for any particular initial conditions, one needs to integrate kinetic equations (1) over the entire time of evolution. We do not address this problem here.

In Fig. 1(b) we use typical material parameters for III–V semiconductors. Elastic moduli are $c_{11} = 1.0 \times 10^{12}$ erg/cm$^3$, $c_{12} = c_{44} = 0.5 c_{11}$, the surface energy is 50 meV/Å$^2$; the lattice mismatch between pure AC and BC is 7%; the critical temperature is $T_c = 1000$ K, the activation energy for surface diffusion is $E_a = 1.5$ eV. Growth velocities marked in Fig. 1(b) are: $v_1 = 1.2$ Å/s, $v_2 = 4.0$ Å/s and $v_3 = 96$ Å/s. The growth velocity $v_3$, at which the instability is suppressed, is higher than typical ones in MBE. The growth velocities $v_1$ and $v_2$ corresponding to the kinetic phase transition between the growth of a 1D structure and the growth of a 2D structure, are just of the order of typical ones in MBE. Thus, the described kinetic phase transition corresponds to realistic growth velocities and can indeed be observed.

A kinetic phase transition between the growth of a 1D structure and the growth of a 2D one has been observed by Ueda et al. [8] in the MBE growth of InAlAs alloy lattice-matched to InP(001) substrate. At the growth velocity $v = 3$ Å/s, the growth temperature has been varied. At 440 and 470 °C, a 1D modulation in [110] direction is formed. The growth at
higher temperatures, 500, 530 and 560 °C reveals a 2D modulated structure in [100] and
[010] directions.

To address this experiment we take into account the anisotropy of the surface diffusion
\(D_{[110]} \neq D_{[110]}\) and construct the linear stability phase diagram of Fig. 1(c). At high
temperatures, close to the onset of instability, the interval of wave vectors \(k_0\) of unstable
fluctuations is rather narrow. Then, the wave vectors of the most unstable fluctuations \(k_0^{(0)}\)
are parallel to elastically soft directions [100] and [010]. As the temperature decrease,
the interval of \(k_0\) corresponding to unstable fluctuations becomes wider, the role of elastic
anisotropy decreases, and the role of diffusion anisotropy increases. The orientation of \(k_0^{(0)}\)
deviates from [100] and [010] towards the direction of fast diffusion [110].

Finally, the direction of \(k_0^{(0)}\) coincides with [110]. In this case, the growth will
presumably result in a 1D modulated structure. Calculated phase diagram of Fig. 1(c) is in
qualitative agreement with experimental results. A detailed consideration given in Ref. [9],
allows to reach a quantitative agreement and to fit the kinetic phase transition tempera-
ture \(T_1\).

To conclude, the steady state phase diagram of Fig. 1(b) applies to any alloy. We explain
a large variety of structures, e.g., the 1D structure in InAlAs modulated along [110] [10],
the 2D structure in InGaAsP modulated in [100] and [010] directions [11], and a kinetic
phase transition in InAlAs between the growth of a 2D structure modulated along [100] and
[010] and the growth of a 1D structure modulated along [110] [8]. Our results demonstrate
a possibility to tune the structure between 1D and 2D ones by varying growth velocity and
temperature.

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References

MBE-grown CaF$_2$ nanostructures on Si(001)

N. S. Sokolov and S. M. Suturin
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Atomic force microscopy and reflection high energy electron diffraction have been used to study initial stages of CaF$_2$ epitaxial growth and formation of nanostructures on Si(001). A variety of nanostructures has been observed including ultra-thin two-dimensional layers at 750°C, quasi one-dimensional wires at 650°C and well-ordered dots of almost equal size at lower growth temperatures. An attempt has been made to understand the influence of the growth parameters on the orientation and shape of the resulting nanostructures.

Introduction

Fabrication of new solid state nanostructures is known to offer unique opportunities in creation of new micro-, opto- and magnetoelectronic devices. However, traditional lithographic procedure is very difficult to use in forming nanostructures with characteristic size of less than 10 nm, that are necessary for quantum well devices operating at room temperature. Therefore self-assembled nanostructures are very attractive. One of suitable candidates to be used as a lithographic mask is CaF$_2$. This compound is chemically inert and well matched to Si lattice. When deposited on Si it causes no intermixing at CaF$_2$/Si interface. Recently it has been demonstrated that fabrication of CaF$_2$ nanostructures is possible both on Si(001) [1] and Si(111) [2] substrates. In this work, we applied reflection high energy electron diffraction (RHEED) and atomic force microscopy (AFM) to study initial stages of CaF$_2$ epitaxial growth and formation of epitaxial nanostructures on Si(001) in a wide range of growth conditions.

1. Experimental

Fluoride nanostructures were grown in a custom MBE system. After standard chemical cleaning, Si substrate was transferred into growth chamber and thermally cleaned at 1250°C for ~2 min. Then it was kept at 1000°C for 15 min to flatten the surface. The RHEED pattern of the prepared substrate showed a well pronounced 2 $\times$ 1 superstructure which is characteristic of the clean Si(001) surface. The fluoride was deposited at the rate of 2–3 nm/min from the molecular beam formed by sublimation of single-crystal CaF$_2$ pieces from an amorphous carbon crucible. The nanostructures were grown at relatively high temperatures (450–750°C) and in most cases after cooling down to room temperature were capped with 10 monolayers (ML) of CaF$_2$ for protection from humidity and oxidation. The surface morphology was studied out of the MBE system with atomic force microscope produced by NT-MDT (Zelenograd, Russia). The microscope was operated in the tapping mode at cantilever resonance frequency of 300–400 kHz.

2. Experimental results

It is known that clean Si(001) surface is dimerized to show 2 $\times$ 1 reconstruction. On a vicinal surface the dimerization changes orientation by 90° on every single atomic step. This results
Fig. 1. The surface morphology of Si(001) substrate after the pregrowth treatment.

Fig. 2. Surface morphology of Si(001) surface with 1 ML of CaF₂ deposited at (a) 450°C, (b) 550°C.

Fig. 3. Surface morphology of Si(001) surface with 1 (a) and 6 (b) ML of CaF₂ deposited at 650°C.

in alternating of $2 \times 1$ and $1 \times 2$ reconstructions across the surface. Figure 1 shows typical surface morphology of Si(001) substrates which have passed high-temperature cleaning and 1000°C treatment. One can see 100–200 nm wide terraces separated by single atomic steps and step bunches consisting of up to 8 single atomic steps. The surface morphology drastically changes after 1 ML of CaF₂ is deposited on the substrate at 450°C. Figure 2(a) shows domination of 20–40 nm wide rectangular islands 4–8 nm high with the sides parallel to [110] and [110] directions. One can also see several [110]-aligned narrow stripes attached
to some of the islands. At higher growth temperature (550°C) instead of the rectangular islands we observe series of long wires (Fig. 2(b)). Most of the wires run along [110] direction, however some of them are orthogonal to it. It is noteworthy that the wires never cross each other, but rather form a T-shape joint. Increasing the growth temperature to 650°C, one obtains the structure with even longer [110]-aligned wires (Fig. 3(a)). The wires exceed several microns in length being only 3–8 nm in height. The measured width \( \sim 20 \) nm, however, may be larger than the actual one because of the finite sharpness of the cantilever tip. A much higher density of the wires is observed if CaF\(_2\) coverage is increased up to 6 ML at the same growth temperature (Fig. 3(b)). Further increase of the growth temperature to 750°C at the same coverage (6 ML) leads to a considerable decrease in the wire density. Surface steps of tooth-saw shape with edges parallel to [110] and [1\,1\,0] are clearly observed between the wires (Fig. 4).

3. Discussion and conclusion

Taking into account that the (111) surface has minimal surface energy in the fluorite structure [3], it is natural to suppose that the side facets of the nanostructures shown in Figs. 2–4 are formed by \{111\} planes. In principle, there are two possible ways to build such a nanostructure. The first one is to form a pyramid or a hut with (100) base and \{111\} facets. One can expect that such an island will be elastically strained in the interface plane, because the mismatch between CaF\(_2\) and Si lattices is less than 2%. The other possible way of an island growth is with the CaF\(_2\) [110] direction normal to the Si substrate surface. In this case, the lattice mismatch along one of the Si \langle 1\,1\,0 \rangle directions is the same as for the pyramid \(< 2\%\), however in the perpendicular direction, the mismatch can exceed 45%. Therefore, one can expect that this type of the islands will be partially or completely relaxed in the direction of their width.

The data presented above as well as in [1, 4] show that at the initial stages of CaF\(_2\) growth on Si(001) the latter type of islands dominate on the surface. Their epitaxial relations (CaF\(_2\) [110] // Si[001], CaF\(_2\)[1\,1\,0] // Si[1\,1\,0] for the islands aligned along [1\,1\,0] and CaF\(_2\)[1\,1\,0] // Si[001], CaF\(_2\)[1\,1\,0] // Si[1\,1\,0] for [110]-aligned islands) are confirmed by both HRTEM measurements [1] and our RHEED observations which showed the patterns similar to the observed in [5] during CaF\(_2\) growth on Si(110). The island nucleation is likely being influenced by surface reconstruction on Si. It is known [5] that diffusion on the 2 × 1 reconstructed Si surface is highly anisotropic. Diffusion is much easier along the rows of dimers than across them. Provided the diffusion length exceeds the terrace width, the most
likely terraces for island nucleation are those where the diffusion prevails along the terrace rather than across it. The reason for this is that the admolecule concentration is higher at the terraces with a longitudinal diffusion. However in the case of short diffusion length (low growth temperature) the nucleation on both types of terraces will be equally probable. It is believed that the island is nucleated with the well-matched CaF$_2$ [110] direction parallel to the dimer rows on the Si terrace. Once the nucleation occurs the island extends along the well-matched direction keeping constant width along poorly matched CaF$_2$ [100] direction. An additional reason for the one-dimensional growth is that the diffusion is much easier along the dimer rows. That means that the island orientation is determined at the nucleation stage. At high growth temperature the nucleation occurs at only one sort of reconstructed terraces, leading to the same orientation of all the islands on the surface (Fig. 3, 4). At lower growth temperature both 2 × 1 and 1 × 2 terraces are allowed for nucleation which results in growth of both [1T0]- and [110]-aligned wires (Fig. 2(b)). Due to the lower admolecule diffusion length, these wires are much shorter than the ones grown at higher temperature. Decrease in the wire density with growth temperature (Fig. 3, 4) can be explained by the increase in the rate of fluorine depletion taking place during formation of CaF$_2$/Si interface. As it was shown in [4] it is accompanied by the transition from the formation of elongated islands, similar to the wires observed in our work, to the two-dimensional lateral growth related to the formation of CaF$_2$ layer at the interface. This process leads to the rearrangement of the steps which become better ordered (compare Fig. 1 and Fig. 4).

In conclusion, we have shown that strong anisotropy of 2 × 1 and 1 × 2 domains on Si(100) surface, as well as the temperature dependence of the interface fluorine depletion rate results in the formation of a variety of CaF$_2$ nanostructures including quasi-zero-dimensional dots and quasi-one-dimensional wires.

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Photoluminescence studies of InAs/InSb nanostructures grown by MBE

Ya. V. Terent’ev†, A. A. Toropov†, B. Ya. Mel’tser†, V. A. Solov’ev†, S. V. Ivanov†, P. S. Kop’ev†, B. Magnusson‡ and B. Monemar‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Department of Physics and Measurement Technology, University of Linköping, S-581 83 Linköping, Sweden

Semiconductor heterostructures based on narrow gap III–V compounds and their alloys are under intensive study as highly promising materials for mid-IR optoelectronic devices. InSb nanostructures are of peculiar interest having the narrowest band gap among these materials. During the past few years considerable success was achieved in the technology of InSb nanostructures embedded in different matrices. In particular, a thin layer of InSb was formed on GaAs substrates by MBE and optimal growth conditions were found for the formation of the nanometer-scale dots [1]. Photoluminescence (PL) of these objects has been studied in Ref. [2]. MBE growth of InSb embedded in GaSb was studied in [3], demonstrating a maximum red shift of an InSb quantum dot (QD) related PL peak to 1.7 µm for the InSb critical thickness of 1.7 monolayer (ML). Finally, self-assembled InSb QDs in GaSb, grown by metalorganic vapor phase deposition, were reported in Ref. [4]. However, until recently there have been no publications devoted to InSb/InAs nanostructures, although this system is of great interest, owing to its potential ability to emit or detect light in the spectral range 3–5 µm.

In this paper we report on the first MBE growth of InSb submonolayer insertions in an InAs matrix and PL studies of this heterostructure. The active region of the sample grown for PL studies consists of a 0.2 µm InAs layer centered with a 0.5 ML InSb insertion. The buffer structure involving a 0.5 µm InAs layer followed by a 20 nm thick AlSb barrier was grown at a substrate temperature $T = 480^\circ$C on an n-InAs substrate having an electron concentration $n \sim 2 \times 10^{16}$ cm$^{-3}$. The active region was grown at $T = 420^\circ$C. The existence of an optimum temperature ($T \sim 400^\circ$C) for MBE growth of GaInSb/GaSb strained quantum wells (QWs) has recently been proved by PL studies [5]. Degradation of optical properties was observed, either due to the formation of Sb clusters at lower temperatures, or due to structural defects arising from the enhanced group V molecule reevaporation from the surface at higher temperatures [6]. One should stress especially that the main problem here is intermixing of group V elements at the interfaces. Since we used conventional solid sources for As and Sb, the growth rate of the InSb/InAs active region was chosen five times lower as compared to that of the InAs buffer layer, with respective reduction of the As flux intensity [7], which allowed us to suppress the As incorporation into the InSb insertion.

The band alignment of the InAs/InSb heterostructure is predicted to be type-II broken gap [8]. In particular, the estimated valence-band offset for the strained InSb layer on InAs $\Delta E_v$ equals to 910 meV. The respective band diagram of the InAs/InSb/InAs active region is shown in Fig. 1. Note that due to 7% lattice mismatch between InSb and InAs either the formation of QW or self-organized growth of QDs can take place, depending on the amount of evaporated InSb. It will be shown further that the InSb submonolayer in our sample can be most probably regarded as a QW.
PL measurements of the structure were performed in the temperature range from 2 to 102 K using for excitation an Ar$^+$ laser operating at 514.5 nm. The excitation power varied from 50 to 300 mW and the laser spot size was about 2 mm. A Bomem DA8 Fourier transform spectrometer equipped with an InSb detector was used for registration of PL spectra.

A relatively narrow PL peak (FWHM ~ 20 meV) is observed at an energy less than the InAs band gap (see Figs. 2, 3). The PL maximum shifts towards higher energies with increasing excitation density (Fig. 2), whereas the temperature rise up to 102 K causes mainly a decrease of PL intensity (Fig. 3). The observed blue shift of the PL maximum with increasing excitation density is inherent for type-II structures and results from the dipole layer formation caused by spatial separation of non-equilibrium holes confined in the InSb layer and electrons confined in a triangular QW in the nearby InAs region [9]. The fact that the energy of the PL maximum does not follow the known temperature dependence of the InAs band gap also confirms that PL originates from radiative recombination between holes in the InSb insertion and electrons weakly confined by electrostatic interaction in the InAs barrier. The relatively small values of the PL peak width indicates, in our opinion, the formation of a QW in the structure since PL from QDs exhibits generally a significantly wider peak due to the inhomogeneous distribution of the dot sizes (see e.g. [2]).

In summary, we have grown a nanostructure containing an InSb QW in an InAs matrix,
which objectively demonstrates the type-II band alignment. The PL emission ability of this structure near 4 µm is demonstrated. This work is the initial step in the fabrication and studies of InSb/InAs nanostructures and further structural and optical investigations are currently in progress.

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Optical properties of InGaAsN/GaAs quantum well and quantum dot structures for longwavelength emission

B. V. Volovik†, A. R. Kovsh†‡, W. Passenberg‡, H. Kuenzel‡, Yu. G. Musikhin†, V. A. Odnoblyudov†, N. N. Ledentsov†, D. Bimberg‡ and V. M. Ustinov†
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Heinrich–Hertz–Institut für Nachrichtentechnik Berlin GmbH, Einsteinufer 37, 10587 Berlin, Germany
§ Technische Universität Berlin, Hardenbergstr. 36, 10623, Berlin, Germany

Introduction

Lasers emitting in the wavelength range near 1.3 μm are the key components of the fiber optic communication systems. Currently these are fabricated using InGaAsP or InGaAlAs heterostructures on InP substrates. However, these devices have some essential disadvantages: poor temperature stability due to the small conduction band offset [1], low thermal conductivity and difficulties in fabricating highly reflecting distributed Bragg reflectors (DBR) due to the low difference of the refractive indices. The last disadvantages are particularly important for vertical cavity surface emitting lasers (VCSELs). This stimulates attempts to fabricate high quality 1.3 μm lasers on GaAs substrates. The best results are obtained using InGaAs/GaAs self assembled quantum dots [2, 3] and using InGaAsN alloy insertions in a GaAs matrix [4–7].

An addition of several percents of Nitrogen into (In)GaAs alloy was shown to result to the strong bandgap decrease due to the large bandgap bowing parameter in the GaAs–GaN system [4]. The insertion of N into InGaAs also leads to a strain compensation, and the growth of InxGa1−xNyAs1−y layers lattice matched to GaAs substrate is possible for x ∼ 3y. A high temperature stability of the threshold current density in the lasers based on this system was predicted theoretically due to the large conduction band offset in the InGaAsN/GaAs heterostructures [4]. Fabrication of the surface–emitting lasers is possible using well developed technology of AlGaAs/GaAs DBR.

1.3 μm lasers based on InGaAsN insertions in GaAs have been fabricated by several research groups [5, 6]. The CW emission at room temperature with a low threshold current density of 400 A/cm² and high output power of 2.7 W were reported [6]. The vertical cavity surface emitting laser (VCSEL) emitting at 1.18 μm was also reported [7].

The main problem in the InGaAsN epitaxy is a large difference in the atom sizes of As and N, which makes difficult the uniform alloy formation. The incorporation of large amount of N into GaAs leads to a phase separation [8]. Even in the case of low N contents, the main problem in the growth of InGaAsN/GaAs heterostructures is a degradation of the material quality when Nitrogen concentration is increased [9]. In molecular beam epitaxy (MBE) one of the reasons of this is the surface quality degradation due to the high energy ions originating from plasma nitrogen source which is necessary to obtain the active nitrogen pieces [10]. So, the optimization of growth conditions is required to achieve an efficient 1.3 μm emission and to improve the laser performance. In this work we investigate the optical properties of (In)GaAsN/GaAs heterostructures grown by MBE, and show the possibility to realize the 1.3 μm emission using the different structure parameters.
1. Experiment

The investigated structures were grown by MBE on GaAs semi-insulating substrates using standard solid state cells of the group III elements. High volume solid state cell with a cracking area was used to create the As molecules. The active Nitrogen radicals were created by gas Nitrogen flux through the standard radio frequency plasma source. The plasma source power was changed in the range of 75–150 W. Two types of samples were grown: the thick (0.2 $\mu$m) GaAsN layers and InGaAsN quantum wells, placed in the center of 0.2 $\mu$m GaAs layer confined from the both sides by AlAs/GaAs superlattices. The growth temperature was 500°C for N–containing layers. Photoluminescence (PL) was excited by an Ar$^+$ laser ($\lambda \sim 514.5$ nm, 100 W/cm$^2$) and detected by a cooled Ge photodiode. Transmission electron microscopy (TEM) studies were carried out in a Philips EM 420 microscope operated at 100 kV.

2. Results and discussion

First, the Nitrogen incorporation efficiency in the GaAsN layers was studied. In Fig. 1 the results of x–ray diffraction measurements are shown for the samples grown at the different growth rates. The plasma source power was 75 W for both samples. To estimate Nitrogen content in the layers we used the method described in [11] and the elastic constants values from [12]. One can see that the lower growth rate leads to an increase in the N mole fraction in the layer. Moreover, Nitrogen incorporation rate $S_N = V_{gr} \times [N]$, where $V_{gr}$ is layer growth rate, $[N]$ is Nitrogen content in the layer, remains almost constant with growth rate variation. These data are in good agreement with the dependencies reported in [13], where it was also shown that N incorporation efficiency is independent on the growth rate.

Next, we investigated the samples with InGaAsN/GaAs insertions. The data obtained by x-ray diffraction were used to estimate the N concentration in these samples. In Fig. 2 the PL spectra of the samples with In$_{0.25}$Ga$_{0.75}$AsN insertions are shown. One can see that the increase of the plasma source power from 0 to 75 W leads to the PL line redshift of $\sim$80 meV. Assuming that the N content in GaAsN and InGaAsN at the same source power are also similar, we estimate that this shift corresponds to $[N] \sim 0.5\%$. Thus, using a linear approximation, we obtain the dependence of PL maximum on the N content with the line slope of 160 meV/($\%$). This result is in good agreement with the data reported in [8]. Thus, we estimate the maximum N content in our samples as $\sim 1.3\%$.

We investigated the optical properties of InGaAsN–GaAs structures with a different In

![Fig. 1.](image-url) (a), (b) Rocking curves for GaAsN layers grown at the different growth rates with plasma source power of 75 W. (c) Dependence of N incorporation rate on the growth rate.
Fig. 2. (a) PL spectra of InGaAsN QWs grown at the different plasma source power (marked near the spectra). (b) PL spectra of the (115 W) sample before and after postgrowth annealing (700°C).

Fig. 3. PL peak positions at room temperature for samples with different In and N concentration in QW.

Fig. 4. PL spectra for the sample with InGaAsN QDs. In the inset — the plane-view TEM image of this sample.

and N composition. The PL peak positions are shown in Fig. 3. For small Indium and Nitrogen composition PL emission looks similar to emission in InGaAs–GaAs quantum well (QW) structures. For higher Indium and Nitrogen content PL broadens significantly. The increase of plasma source power leads to a continuous PL redshift at any given In composition in the InGaAsN. At the maximum power of 150 W and In content of 35%, the PL line at 1.32 μm was observed. The increase in N concentration also leads to a significant PL intensity decrease, which is typical for InGaAsN layers. The improvement of PL intensity of InGaAsN QWs is possible by using high temperature postgrowth annealing [10]. We investigated the annealing effect on the optical properties of our structures. The annealing was carried out in the growth chamber under As flux. The annealing at 650°C (1 hour) did not change neither PL intensity, nor peak position. Using the 700°C anneal leads to the 3 times increase in integral PL intensity, while the corresponding PL blueshift was rather small (20–25 meV for different structures) (see Fig. 2(b)). Thus, the postgrowth annealing gives a possibility to improve the quality of the structures without significant
changes in their electronic spectrum.

We note that to get 1.3 µm emission one needs to use both higher Indium and Nitrogen compositions. From the point of view of PL lineshape and linewidth the properties of the samples resemble PL properties of QD-like structures. We investigated PL in InGaAsN samples with higher In composition of 53%. TEM images show that dense array of rather large (lateral size (23 nm) QDs is formed in the structure (see inset in Fig. 4.) The PL spectra of this structure is shown in Fig. 4. The 3D localization is confirmed by the fact that the PL linewidth does not change with the temperature variation from 10 to 300 K (FWHM ≈ 45 meV). We note that PL wavelength in this sample is 1.32 µm, in spite of rather small N concentration in this sample. Thus, the significant redshift thus can be attributed to both QD formation and Nitrogen incorporation. Longwavelength emission from InGaAsN structures using relatively small N concentrations may lead to a significant increase in the PL intensity and, hence, to an improvement of the laser performance.

To conclude, we investigated the optical properties of heterostructures with InGaAsN/ GaAs QW–like and QD–like insertions. GaAsN and InGaAsN layers with relatively high nitrogen content (more than 1%) were grown. The long wavelength emission up to 1.32 µm at room temperature was realized. TEM and optical studies confirm formation of quantum dots for the case of higher indium concentrations.

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Luminescence enhancement in indirect band-gap semiconductors with quantum confinement structures

Y. Shiraki
Research Center for Advanced Science and Technology (RCAST)
The University of Tokyo 4-6-1 Komaba, Meguro-ku, Tokyo 153-8904, Japan

In order to obtain efficient light emission from indirect band-gap semiconductors, we have systematically investigated the correlation between electronic states and optical properties in three kinds of novel quantum confinement structures.

The first one is a neighboring confinement structure (NCS) in which a pair of thin layers of spatially indirect band-gap semiconductors is sandwiched by a wider band-gap barrier layers. The usefulness of this structures, that is, increase of luminescence, especially no-phonon emission, and thermal stability of luminescence was demonstrated for GaP/AlP NCS structures where the GaP/AlP pair was sandwiched by AlGaP barrier layers. The luminescence was proven to be further enhanced by growing the structure on InGaP pseudo-substrates. This enhancement can be attributed to Γ–X band mixing and the increase of wavefunction overlap caused by the strain. These NCS structures were demonstrated to be applicable to Si/Ge heterostructures and systematic energy shift and significant luminescence enhancement were clearly observed.

The second structure is a quantum well which contains an ultrathin AlP layer at the center of type-I indirect GaAsP/GaP quantum wells (QWs) so that electrons are effectively localized into the well region. Insertion of 1-ML of AlP into the center of GaAsP QWs was found to drastically increase luminescence intensity. The study on As composition and AlP thickness dependencies as well as numerical calculation revealed that exciton localization is preferable for enhancement of luminescence when the Xz band rather than Xxy bands constructs the quantum well.

An ultimate structure for exciton localization in indirect band-gap materials is the quantum dot structure. Formation of Ge dots on Si layers was investigated and it was found that well-controlled dot array can be realized by employing electron-beam lithography and selective epitaxial growth of gas source MBE of Ge on Si layers with SiO2 masks. The numbers and size of Ge dots are dependent on growth conditions, especially the size of SiO2 windows, reflecting migration length of Ge atoms. Significant luminescence is observed from the dot array and the attempt to reduce the dot size is now under progress to enhance the quantum confinement effects in the dots.
Observation of electromagnetically induced transparency in a three-subband quantum well system

K. L. Vodopyanov†, G. B. Serapiglia, E. Paspalakis, C. Sirtori‡ and C. C. Phillips
Blackett Laboratory, Imperial College, London, SW7 2BZ, UK
† At present: INRAD, Inc., Northvale, NJ 07647, USA
‡ LCR Thomson-CSF, Domaine de Corbeville, 91404 Orsay Cedex, France

Abstract. The phenomenon of Electromagnetically Induced Transparency is demonstrated for the first time in a subband quantum well system. Applying strong coupling field which is two-photon-resonant with the 1–3 intersubband transition, produces dramatic change in the 1–2 intersubband absorption profile. This effect can be accounted for in terms of 1–2 and 2–3 dipoles being driven into coherence by a strong coupling field and is similar to Fano-type destructive interference of probability amplitudes.

Strong resonant optical field, applied to a 3-level atomic system, may create an ensemble of phase-coherent atoms, with interesting properties, caused primarily by the quantum interference between probability amplitudes of the atomic states. The study of such a system has led, in the last decade, to prediction and experimental demonstration of exciting (and sometimes counter-intuitive) effects, such as electromagnetically induced transparency (EIT) [1] and lasers without inversion (LWI) [2]. The most recent examples include observation of ultraslow (∼10⁻⁷ s) propagation of light in a Bose-condensed atom system [3].

To our knowledge, observations of EIT have so far been restricted to comparatively sharp transitions in atomic vapours [1]. In these experiments, the condition that the Rabi frequency, \( \Omega = \mu E / \hbar \) (where \( \mu \) is a transition dipole matrix element and \( E \) is an amplitude of the coupling EM field) is greater than the linewidths is readily achieved. Although there is much theoretical interest in EIT and LWI in semiconductor quantum wells (QW’s), the observation of coherent phenomena in these systems is complicated by their large (∼100 fs⁻¹) dephasing rates.

Devices based on intersubband (ISB) transitions in QW’s offer a great potential for applications in lasers and nonlinear optics (NLO) because of their inherent advantages, such as large electric dipole moment, high NLO coefficients, and the possibility to engineer key microscopic properties, such as energy levels and wavefunctions, matrix elements and even decay rates [4]. Gigantic NLO coefficients in asymmetric double QW’s (ADQW’s) can also be created, via bandgap engineering [5], and efficient optical frequency conversion can be achieved [6]. However, the full potential of this system could not be realized because of the in-built resonant ISB absorption. Coherent population control in ADQW will make it possible to engineer efficient NLO devices which, by analogy with atomic systems [7], utilize an absorption transparency while maintaining gigantic, resonantly enhanced NLO coefficients.

EIT typically involves a three-level system, having two dipole-allowed transitions (\(|1\rangle \rightarrow |2\rangle \) and \(|2\rangle \rightarrow |3\rangle \)) and a third (\(|1\rangle \rightarrow |3\rangle \)) dipole forbidden transition (Fig. 1). Driving this with a 'coupling field', of angular frequency \( \omega_c \), resonant with the \(|2\rangle \rightarrow |3\rangle \) dipole
Fig. 1. Electromagnetically Induced Transparency in atoms. Driving a three-level system with a coupling field (Ωc, resonant with the |2⟩→|3⟩ dipole transition) establishes a coherent superposition of the probability amplitudes and makes quantum interference possible, thus eliminating absorption in the |1⟩→|2⟩ line center (b). Here (γ₁₂ is |1⟩→|2⟩ dephasing rate (inverse linewidth).

Fig. 2. (a) Transition energies in the 3-level QW structure (T = 30 K) and (b) schematic of the pump-probe mid-IR experiment.

transition establishes a coherent superposition of the probability amplitudes and makes quantum interference possible. In this case (|1⟩→|2⟩ transitions occur through two coherent paths (|1⟩→|a⟩ and (|1⟩→|b⟩, where the ‘dressed states’ are given by |a⟩ = (|3⟩−|2⟩)/√2 and |b⟩ = (|3⟩+|2⟩)/√2, whose transition probability amplitudes interfere destructively and cancel the original absorption at energy difference E₁₂.

In the present work we study a three-level QW system where, according to selection rules, ISB transitions 1–2 and 2–3 are allowed and 1–3 is dipole-forbidden. The QW sample consists of forty symmetric 10 nm n-doped (nₓ = 6 × 10¹¹ cm⁻²) In₀.₄₇Ga₀.₵₃As wells with 10 nm Al₀.₄₈In₀.₵₂As barriers, lattice matched to an undoped InP substrate [8]. Transition energies (Fig. 2(a)) were E₁₂ ≈ 129 meV and E₂₃ ≈ 160 meV (at T = 30 K) and matrix elements (µ₁₂ = 2.34 nm and (µ₂₃ = 2.64 nm.

A double pass 45⁰ waveguide geometry was used to couple p-polarized light to ISB transitions (Fig. 2(b)). Independently tunable (λ ~ 6–12 μm) synchronised coupling (ωc) and probe (ωp) laser pulses, with similar temporal profiles (FWHM ~ 70–80 ps), were generated in separate erbium-laser-pumped Optical Parametric Generators [9] based on ZnGeP₂ and CdSe nonlinear crystals.

Peak intensity of the incoming mid-IR coupling field reached 20 MW/cm². Taking into account that only Eₓ — the component of the optical field (i.e. normal to the QW layers) couples the ISB transitions, this corresponds to the Rabi frequency Ωₓ = µ₁₂Eₓ/h ≈ 5 meV. The coupling and the probe beams were separated by 10⁵; the probe beam intensity
was $10^3$ times weaker than the coupling intensity, and a 300 $\mu$m pinhole was attached to the sample facet — to assure the beam overlap. Linear 1–2 absorption spectrum at $T = 30$ K (partly shown in Fig. 3, dotted line) was best fitted with the Lorentzian shape with FWHM $\sim5$ meV. Thus the coupling Rabi frequency was of the same order as the ISB linewidth. At $T = 30$ K, only the lowest state was populated so that the $E_{23}$ transition was not seen and $E_{23}$ energy were measured using the induced absorption method [8].

While tuning the coupling energy to $\hbar \omega_c = E_{12}$ produced almost complete saturation of the $E_{12}$ absorption line, the most interesting effect was observed when the coupling beam frequency was tuned half-way between $E_{12}$ and $E_{23}$ (‘two-photon’ 1–3 resonance, $\hbar \omega_c \sim E_{13}/2$). A pronounced (67% reduction in absorption) narrow (FWHM $\sim3.6$ meV) dip appears in the absorption spectrum (thick line in Fig. 3). Its spectral narrowness, the non-obvious relationship between its position (126 meV) and the coupling photon energy (144 meV), both suggest an origin in quantum interference. Moreover the transparency feature disappeared when the coupling laser was tuned to its ‘mirror image’ with respect to $E_{12}$ position ($\hbar \omega_c = 116$ meV, thin line in Fig. 3).

We theoretically treat the three-level system within a density-matrix formalism where we assumed an intersubband relaxation time $\tau = 1$ ps [10] and non-diagonal relaxation rates $\gamma_{12} = \gamma_{23} = \gamma_{13} = 5$ meV. In agreement with experiment, the strongest quantum interference effect is predicted when the coupling frequency is tuned to $\omega_c = E_{13}/2$ (Fig. 2). When this occurs, the model finds ‘dressed state’ energies of 132 and 116 meV for the superposition states $\ket{a}$ and $\ket{b}$. These two states are coherently coupled by the two-photon coupling field [11]. Absorption at $\sim125$ meV, half way between these two ‘dressed states’, is the result of two interfering pathways which contribute to the absorption dipole moment with opposite sign, giving strong absorption cancellation.

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References


Non-monotonous temperature dependence of the spectral maximum of photoluminescence in CdS/ZnSe superlattices

S. A. Tarasenko†, A. A. Kiselev†, E. L. Ivchenko†, A. Dinger‡, M. Baldauf‡, K. Klingshirn‡ and H. Kalt‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institute for Applied Physics, Karlsruhe University, D-76128 Karlsruhe, Germany

Abstract. Macro- and micro-photoluminescence (PL) spectroscopy is applied to investigate exciton localization in cubic CdS/ZnSe type-II superlattices (SL) in the temperature range between 5 K and 35 K. The non-monotonous shift of the macro-PL maximum with increasing temperature reveals the kinetic contribution of acoustic-phonon-assisted exciton hopping processes. The kinetic theory of localized excitons has been generalized from zero to finite temperatures allowing for exciton hopping from low-energy to high-energy localized sites as well as for the upwards-downwards correlation. The results of kinetic theory are compared with those of computer simulation and the experimental data.

In this work the properties of localized excitons of several cubic CdS/ZnSe superlattices have been investigated using spatially integrated (macro-PL) and spatially resolved (µ-PL) photoluminescence spectroscopy. The band alignment of this heterostructure is of type-II with the conduction and valence band offset-s of about 800 meV and 500 meV, respectively: the CdS (ZnSe) layers form the potential wells for the electrons (holes) [1]. We concentrate here on the macro- and µ-PL results of a 200 period (19 Å/19 Å) CdS/ZnSe superlattice grown by molecular-beam epitaxy on GaAs(001) [2]. The PL spectra discussed in the following were excited by the 351 nm line of an Ar⁺-ion laser and measured with a spatial resolution of about 1 µm (µ-PL) or about 50 µm (macro-PL).

The macro-PL spectrum is displayed together with the PLE spectrum in the inset in Fig. 1. In Fig. 2(a) µ-PL spectra are shown, which have been recorded at two different temperatures. Besides a broad PL background, narrow line emission is observed which is typical for strongly localized excitons. Full circles in Fig. 1 show the measured temperature dependence of the relative shift for the macro-PL maximum

$$
\Delta E_m = \hbar \omega_m(T) - \hbar \omega_m(0) - \delta E_g(T),
$$

where $$\omega_m(T)$$ is the spectral position of the PL maximum at the temperature $$T$$, $$\delta E_g(T)$$ is the temperature shift of individual narrow lines in the µ-PL spectra which follows the temperature dependence of the SL band gap. The value of $$\Delta E_m$$ obviously represents a purely kinetic contribution to the shift.

For numerical simulation we generated two-dimensional, random, uniform distributions of localization sites on a square cell of sufficiently large size with the chosen energy density of states, $$g(\varepsilon) g_0 \exp (-\varepsilon / \varepsilon_0)$$. The PL spectrum was calculated as a sum of contributions of individual localized sites, each one broadened by a Lorentzian. For the simulation of macro-PL spectra we generated ≥ 2000 configurations to obtain a sufficiently smooth spectrum. In order to take into account qualitatively the inhomogeneous broadening, the macro-PL
Fig. 1. Temperature dependence of the shift of macro-PL maximum defined by Eq. (1) and related to the low-temperature PL Stokes shift equal to 18 meV for a CdS/ZnSe (19 Å/19 Å) SL. Full circles, experiment; solid line, computer simulation. In inset: The PL and PLE spectra are compared revealing a Stokes shift of 18 meV.

Fig. 2. (a) μ-PL spectra (spatial resolution ≈ 1 μm) measured from a CdS/ZnSe (19 Å/19 Å) SL at T = 5 K and 20 K. (b) Computer-simulated μ-PL spectra for T = 0 K and T = 22 K. The shift δE(T) is ignored.

spectra, simulated for a fixed value of the exciton mobility edge E₀, were convoluted with a Gaussian describing the distribution of E₀. For the simulation of μ-PL spectra (see Fig. 2(b)) we took 200 subsystems each containing 100 localized-exciton sites randomly distributed with equal probability within a square area and with the weight g(ε) in energy. The exciton mobility edge in each subsystem was chosen randomly in accordance with the Gaussian distribution.

The parameters chosen to calculate the PL spectra are as follows: \( \omega_0 \tau_0 = 3 \times 10^4 \), \( \pi (a/2)^2 g_0 \epsilon_0 = 0.2 \) (the notations are the same as in [4]). The inhomogeneous broadening is described by the Gaussian with a width 3.660. At zero temperature the PL maximum occurs at \( E_0 - h \omega_m = 3.76 \epsilon_0 \). Since the PL Stokes shift equals 18 meV (see the inset in Fig. 1) we obtain for \( \epsilon_0 \) the value ≈ 4.8 meV.

We have generalized the kinetic theory [3, 4] of localized excitons from zero to low but finite temperatures. At \( T = 0 \) each site is characterized by two parameters: the localization energy \( \epsilon = E_0 - E_L \) (\( E_L \) is the localized-exciton excitation energy \( E \)) and the distance \( r \) to the nearest lower-lying site. At finite temperature each site is characterized by three parameters, \( \epsilon, \epsilon' \) and \( r \), where \( \epsilon' \) is the localization energy of the optimum site for the local...
hopping process. If the probability to occupy such a site is denoted by \( f(\varepsilon, \varepsilon', r) \), then the energy distribution \( N(\varepsilon) \) which determines the PL spectral intensity is given by

\[
N(\varepsilon) = g(\varepsilon) \int_0^\infty d\varepsilon' \int_0^\infty dr P_\varepsilon(\varepsilon', r) f(\varepsilon, \varepsilon', r) .
\] (2)

Here \( P_\varepsilon(\varepsilon', r) \) is the distribution of optimum neighbors in energy and space. We propose a rather simple and effective form of the approximate kinetic equation for \( f(\varepsilon, \varepsilon', r) \) and write it as \([3]\)

\[
\left[ \frac{1}{\tau_0} + \bar{w}(\varepsilon, \varepsilon', r) \right] f(\varepsilon, \varepsilon', r) - \int_0^\infty d\varepsilon_1 \int_0^\infty dr_1 \frac{g(\varepsilon_1)}{g(\varepsilon)} P_{\varepsilon_1}(\varepsilon_1, r_1) \bar{w}(\varepsilon_1, \varepsilon, r_1) f(\varepsilon_1, \varepsilon, r_1) = \Gamma_0 .
\] (3)

The notations used are as follows: \( \tau_0 \) is the recombination time, \( \Gamma_0 \) is the generation rate from delocalized states, \( \bar{w}(\varepsilon, \varepsilon', r) = w(\varepsilon, \varepsilon', r) p(\varepsilon, \varepsilon', r) \), \( w(\varepsilon, \varepsilon', r) \) is the exciton transfer rate for the \( \varepsilon \rightarrow \varepsilon' \) transition between the sites \( O \) and \( O' \) separated by a distance \( r \); for \( \varepsilon < \varepsilon' \), the factor \( p(\varepsilon, \varepsilon', r) \) is set to 1 and, for \( \varepsilon > \varepsilon' \) it is equal to the probability that near the sites \( O, O' \) there exists a localization site \( O'' \) satisfying the following two conditions: (i) the upward hopping process \( O \rightarrow O' \) is more probable than the process \( O \rightarrow O'' \) and (ii) the hopping \( O' \rightarrow O'' \) is more probable than the downward process \( O' \rightarrow O \). The factor \( p \) takes into account that, at nonzero but low temperatures, the major part of excitons which participate in the upwards transition \( O \rightarrow O' \) return back from the site \( O' \) to the site \( O \) and either recombine on the site \( O \) or relax from \( O \) to other localization sites different from \( O' \). Note that in \([3]\) the difference of \( p \) from 1 is ignored.

In Fig. 3(a) we present the relative shifts of the PL maximum as a function of temperature calculated neglecting inhomogeneous broadening. The dashed curve is calculated by using numerical solution of Eq. (3) for \( p \equiv 1 \). One can observe a remarkable discrepancy of the dotted curve from the solid curve which is the exact result of computer simulation. The dotted curve in Fig. 3(a) is calculated by using the proposed approach to account for the upwards-downwards correlation. It shows a good agreement with the results of computer simulation. The calculated non-monotonous behaviour of the Stokes shift with temperature was reported by Zimmermann et al. \([5]\) and Baranovskii et al. \([6]\). Physically, it can be explained taking into account that, at \( T = 0 \), the photoluminescence is dominated by

![Fig. 3](image_url)

**Fig. 3.** Relative shift of the macro-PL maximum as a function of temperature calculated neglecting (a) and taking into account (b) inhomogeneous broadening. Solid lines, computer simulation; dotted lines, the proposed kinetic theory; dashed curve, the kinetic theory \([3]\) which ignores upwards-downwards correlation.
excitons finding themselves on accidentally isolated localization sites acting as pores. For such sites the lifetime with respect to hopping to the nearest lower-energy neighbor exceeds the recombination time, $\tau_0$. At finite temperatures, an exciton trapped by an effective pore has a possibility to be shaken down as it hops first to the nearest higher-energy neighbor and then to another lower-energy site. It is the factor $p(\epsilon, \epsilon', r)$ that estimates the probability for this possibility to be realized.

Figure 3(b) presents the temperature-induced shift of the PL maximum after the PL spectrum is convoluted with the Gaussian. The deviation between the solid and dashed curves can be interpreted in the following way. If the PL spectrum calculated for the homogeneous mobility edge is symmetrical with respect to the maximum then the inhomogeneous broadening makes no effect on the spectral maximum position. However, if the PL spectral maximum calculated for a fixed $E_0$ is asymmetrical, the inhomogeneous broadening results in a shift of the maximum position. At zero temperature the both calculated PL spectra almost coincide in shape. Hence the difference between the solid and dashed curves in Fig. 3(b) appears because the homogeneous macro-PL spectral shape is temperature dependent and, at final temperatures, asymmetries of the simulated PL spectrum and of that calculated by using the kinetic theory are different.

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Experimental determination of the energy distribution function of hot holes in InGaAs/GaAs quantum well heterostructure

V. Ya. Aleshkin†, A. A. Andronov†, A. V. Antonov†, V. I. Gavrilenko†, D. M. Gaponova†, Z. F. Krasil’nik†, D. G. Revin†, B. N. Zvonkov‡ and E. A. Uskova‡

† Institute for Physics of Microstructures RAS, Nizhny Novgorod, Russia
‡ Physical-Technical Institute of Nizhny Novgorod State University, Russia

Abstract. The symmetrical part of the hot hole distribution function in MQW In$_{0.21}$Ga$_{0.79}$As/GaAs heterostructure is obtained from the spectra of the modulation of the fundamental absorption by application of high electric field pulses.

Introduction

To now there are two experimental methods to determine the distribution function of hot carriers in bulk semiconductors. The first method is based on measurements of the modulation of free-hole intervalence-band absorption of infrared radiation by high electric field. This method was successfully used to obtain both symmetrical part of the distribution function and the degree of its anisotropy for hot holes in $p$-Ge [1, 2]. The second method is based on measurements of the modulation of fundamental absorption edge by high electric field in degenerate semiconductors. The last method was used to study high electric field effect on the electron distribution function under Fermi level in degenerate $n$-GaAs [3, 4].

The possibility to determine the distribution function of hot carriers in quantum wells by the second method was experimentally demonstrated in our previous work [5]. In contrast to the case of bulk semiconductors in a quantum well the distribution function of hot carriers can be obtained for energy which exceeds Fermi level as well. However, only estimations for hole effective temperatures were made in [5] and the distribution function was not obtained. In the present work the changes of the symmetrical part of the distribution function of hot holes in high electric field have been determined in In$_{0.21}$Ga$_{0.79}$As/GaAs heterostructure. To make it we measured destruction of Burstein-Moss effect (modulation of the interband optical absorption) by application of high electric field pulses and used the corresponding mathematical data processing.

1. Experiment

The investigated structure was grown by MOCVD technique on semi-insulating GaAs and contains 20 QWs In$_{0.21}$Ga$_{0.79}$As ($d_{QW} = 10.5$ nm) separated by 60 nm GaAs barrier layers. Two delta-layers of Zn were introduced 5 nm apart both sides of each QW. The measured by Hall technique ($T = 300$ K) hole concentration and mobility in QW were $p_s \approx 1.7 \cdot 10^{11}$ cm$^{-2}$ and 315 cm$^2$/V s, respectively.

Photoluminescence (PL) and modulation of the light transmittance were measured at $T = 4.2$ K. The lateral pulsed electric field ($E$) up to 1.9 kV/cm 3 to $10 \mu$s in duration was applied to the structure via strip electric contacts. PL was excited by cw Ar$^+$ laser. In transmittance experiments the light from halogen lamp was dispersed by monochromator, guided to the sample by optical fibre and detected by Ge-diode placed behind the sample.
The measured transmittance signal was proportional to the difference between the intensities of light passed through the sample without and under applied electric field.

In PL spectrum there is only one peak at $E = 0$ due to the optical transitions between the ground electron and heavy hole subbands. We suppose that the width of PL peak measured at 4.2 K ($\approx 18$ meV) is determined by fluctuations of quantum well parameters. The measured and calculated PL spectra are shown in Fig. 1(a). To calculate PL spectrum we took into account the content fluctuations in QWs. We described the fluctuations of the In fraction by the Gaussian distribution function with average value $\bar{x}$ and dispersion $\sigma_x$. Values for $\bar{x}$ and $\sigma_x$ were obtained by fitting the measured and the calculated PL spectra. It is clearly seen from Fig. 1(a) that these spectra practically coincide for $\bar{x} = 0.21$ and $\sigma_x = 0.006$.

At zero electric field the hole concentration in quantum wells is high enough to shift the edge of the fundamental absorption due to the Burstein–Moss effect. Hole heating results in the changes of the hole distribution and the light absorption around edge. The measured modulation of light transmittance by electric field is presented in Fig. 1(b). PL spectrum at $E = 0$ is also given in Fig. 1(b) for comparison. It is clearly seen from Fig. 1(b) that the electric field increases transmittance in the short wavelength region and decreases it in the long wavelength one. The transmittance modulation is observed starting from low enough electric field of 40 V/cm. Both positive and negative transmittance modulations rise with the electric field, being approximately the same up to 200 V/cm. In high enough electric fields the negative modulation of the transmittance dominates. Variation of the electric field practically does not change the maximum and the minimum positions in the transmittance modulation spectra.

2. Discussion

Taking into account content fluctuation in QWs we can write the transmitted light intensity modulation $\Delta I(h\omega)$ in the following form

$$\Delta I(h\omega) = \frac{(1 - R) I}{\sqrt{2\pi \sigma_x}} \int_{-\infty}^{\infty} \exp \left( -\frac{(x - \bar{x})^2}{2\sigma_x^2} \right) \Delta f_h \left( m \frac{m_h}{m_e} (h\omega - \varepsilon(\bar{x}) - B(x - \bar{x})) \right) dx$$

where $R$ is the reflection coefficient, $I$ the intensity of incident light, $\Delta f_h$ the modulation of the symmetrical part of the hole distribution function, $m = m_e m_h / (m_e + m_h)$, $m_{e,h}$...
the electron and hole masses, $\varepsilon(x)$ the energy of optical transition in QW $\text{In}_x\text{Ga}_{1-x}\text{As}$, $B = d\varepsilon(x)/dx$ for $x = \bar{x}$. Eq. (1) is valid if $\sigma_x$ is small and sizes of regions where $x$ is nearly constant are greater then sizes of the space charge regions in QW. The last condition means that the effect of $x$ fluctuation on the hole concentration in QW is negligible. We suppose that both these conditions are satisfied and neglect influence of the Zn concentration fluctuation on light transmission.

To find the dependence $\Delta f(\varepsilon_h)$ on the hole energy $\varepsilon_h$ from measured dependence $\Delta I(h\nu)$ it is necessary to solve integral Eq. (1). But it is impossible to find the correct general solution of this equation if $\sigma_x \neq 0$. However, one can find approximate solution of (1). Indeed, Green’s function of integral operator (1) can be written in the form

$$G(y - y') = \lim_{a \to 0} \left[ G_a(y - y') \right].$$

$$G_a(y) = \frac{1}{2\pi(1 - R)} \int_{-\infty}^{\infty} \frac{\exp(iky)}{\exp\left(-\frac{(k\sigma_x B m)^2}{2m^*} + ak^2\right)} dk.$$  

(2)

Now we can find approximate solution of (1) using $G_a$ for nonzero $a$ as approximate Green’s function:

$$\Delta f_h(\varepsilon_h) = \int_{-\infty}^{\infty} G_a(y - \varepsilon_h) \Delta I \left( \frac{y m_h}{m} + \varepsilon(\bar{x}) \right) dy.$$  

(3)

Dependencies $\Delta f_h(\varepsilon_h)$ determined from experimental data by the use of expression (3) ($a = 0.03$) are shown in Fig. 2. Since the absolute values of $\Delta I$ were not measured the obtained dependencies $\Delta f_h(\varepsilon_h)$ are not normalized. We can find the corresponding coefficient from the requirement that the minimal value of $\Delta f_h$ is of the order of $-1$ at the highest electric field ($E = 1900$ V/cm).

From Fig. 2 one can see that integral of $\Delta f_h(\varepsilon_h)$ over hole energy approximately equals zero at low fields. This reflects the conservation of the hole number in the first subband. This integral is negative at high fields (>380 V/cm) due to the decrease of the hole number in the first subband. Note that holes in excited subbands make a weak contribution.
to the light modulation due to selection rules for optical transitions. The minimal value
of $\Delta f_h$ (negative modulation) tends to the saturation at high electric fields. It is clear
that limit for this minimum is $-1$ that corresponds to a full hole escape from low-energy
states. Dependencies $\Delta f_h(\epsilon_h)$ in logarithmic scale for energies exceeding maximum are
presented in the insert to Fig. 2. It is clearly seen that for relatively low energies (up to
20–30 meV) these dependencies are approximately linear, and further change irregularly.
Linear region corresponds to Maxwell’s type of the hole distribution function. The temper-
atures determined from these linear dependencies are: $T(E = 380\, \text{V/cm}) = 110 \pm 30\, \text{K}$,
$T(630\, \text{V/cm}) = 90 \pm 25\, \text{K}$, $T(1260\, \text{V/cm}) = 110 \pm 15\, \text{V/cm}$, $T(1580\, \text{V/cm}) =
125 \pm 15\, \text{K}$, $T(1900\, \text{V/cm}) = 127 \pm 15\, \text{K}$. In high energy region ($\epsilon_h > 30\, \text{meV}$) there
are significant errors in determination of $\Delta f_h(\epsilon_h)$ due to low experimental accuracy in
measurements of weak signals.

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Raman and hot electron-neutral acceptor luminescence studies of electron-optical phonon interactions in GaAs/Al$_x$Ga$_{1-x}$As quantum wells

K. W. Sun†, H. Y. Chang‡, C. M. Wang†, T. S. Song†, S. Y. Wang‡ and C. P. Lee‡
† Department of Electronic Engineering, Feng Chia University, Taichung, Taiwan, R.O.C.
‡ Department of Electronics Engineering and Institute of Electronics, National Chiao Tung University, Hsin Chu, Taiwan, R.O.C.

Abstract. Using two optical techniques, we have studied the hot electron-optical phonon interactions in GaAs/Al$_x$Ga$_{1-x}$As multiple quantum wells. Raman scattering measurements at 15 K are presented for the Al composition of $x = 0.3, 0.5, 0.7$ and $1.0$. The GaAs-like and AlAs-like phonon frequencies of the first-order modes are also measured as a function of Al compositions. The optical phonon energies emitted by the photoexcited electrons in quantum wells are determined by using hot electron-neutral acceptor luminescence techniques. It is shown that the relaxation of hot electrons in the quantum wells is dominated by the GaAs LO phonon emission for small $x$, but by AlAs-like LO phonons for larger Al composition.

Introduction

There has been a great number of experimental and theoretical studies focused on optical phonons in quantum wells and their interactions with electrons. Raman scattering has been proven as a versatile and efficient tool for probing long-wavelength and short-wavelength lattice dynamics of ternary alloys. The electron–phonon interactions in semiconductor alloys have also been studied by using time-resolved Raman spectroscopy. In addition to Raman scattering techniques, it is well known that the radiative recombination of photoexcited carriers with the neutral acceptors can be used to study the hot carrier relaxation processes. The relaxation of hot electrons through optical phonon emission in bulk GaAs [1, 2] and heterostructures [3, 4] has been extensively studied using above techniques. Sapega [5] has demonstrated that, for quantum wells with large barrier widths, the energy relaxation mechanism for hot electrons is dominated by the AlAs phonons, for smaller barriers, emission via GaAs phonon is more important. By using conventional hot electron luminescence techniques, E. Ozturk et al. [6] have demonstrated that in GaAs/AlAs the dominant electron relaxation mechanism is via the interaction with the AlAs interface mode for a device having a well width of 80 Å. But for a similar GaAs/Al$_{0.23}$Ga$_{0.76}$As structure, the GaAs phonons provide the energy relaxation.

1. Experimental techniques

In this work, we first use Raman spectroscopy to determine the optical phonon energies in GaAs/Al$_x$Ga$_{1-x}$As quantum wells samples with Al composition of $x = 0.3, 0.5, 0.7$ and $1.0$. With the measurements of the energy separation of peaks in the hot electron-neutral acceptor luminescence spectra and the LO phonon energies retrieved via Raman experiments, we then analyze the type of optical phonon emitted by hot electrons during relaxation processes in the quantum wells. The samples investigated were grown by molecular-beam
Fig. 1. Raman spectra of four GaAs/Al$_x$Ga$_{1-x}$As multiple quantum wells and bulk GaAs samples at 15 K in the back scattering geometry for incident wavelength of 514.5 nm. The peak labeled GaAs mode is the LO phonon arising from the GaAs wells. The other two peak labeled GaAs like and AlAs-like mode are related to the Al$_x$Ga$_{1-x}$As barrier layers.

Fig. 2. The AlAs-like LO phonon frequency (square) and GaAs-like LO phonon frequency (circle) as a function of the Al composition for $0 < x < 1$ at incident wavelengths of 514.5 and 655 nm.

2. Results and discussion

Figure 1 shows the Raman spectra for the (50/120) Å quantum wells of four different Al composition excited with Ar$^+$ laser. On the bottom of the spectra we have placed the Raman spectrum of the bulk GaAs sample for comparison. The GaAs LO phonon mode is at 36.7 meV and, for the Al$_x$Ga$_{1-x}$As layers, the optical phonons display a two-mode behavior: the GaAs-like (whose energy is below the GaAs LO phonon energy) and AlAs-like modes (whose energy is below the AlAs LO phonon energy). Our detection system is not capable of resolving the splitting of the GaAs LO phonon into confined modes and there is also no evidence of scattering from interface phonons.

In Fig. 2 we have plotted the AlAs-like and GaAs-like phonon frequencies as a func-
The Al composition at two excitation wavelengths. The AlAs-like phonon frequencies approach those of the phonons in AlAs as $x$ approach 1. On the other hand, the GaAs-like phonons have frequencies approach those of the phonons in GaAs as $x$ approach zero. We found no dependence of the phonon frequencies with the excitation wavelength. We have also measured the anti-Stokes Raman spectrum, but find no evidence related to the phonon absorption by photons. We attribute this to the vanishingly small thermal occupation of the LO phonon modes at very low temperature.

In Fig. 3 we have shown the hot electron-neutral acceptor luminescence spectra of four samples. The principles of this technique [6] are shown in the inset of Fig. 3. The peak labeled “unrelaxed” peak in each spectrum corresponds to recombination of electrons, from the state at which they were created, with a neutral acceptor. The peak labeled “1” represents electrons recombining with neutral acceptors after emitting one LO phonon. The power density of the laser used for the excitation was low enough so the main mechanism of energy relaxation in the sample studied is the emission of optical phonons and the phonon-plasmon coupling can be ignored. In order to demonstrate the change of the luminescence spectra with different Al compositions, we have centered the first unrelaxed peaks in the spectra for all four samples. The separation of the “unrelaxed” peak and “1” peaks in the spectra should allow one to determine the energy of the phonons emitted by hot electrons during the relaxation processes. In order to determine the energy separation more accurately, we first subtract the background (which was originated from the band-to-band recombination) from the spectra and the energy spectra of the two remaining peaks were then fitted by Gaussian distributions. The energy difference between the two peaks is plotted for all the four samples as a function of Al mole fraction as shown in Fig. 4. For the samples with larger $x$, the energy separation in the spectrum approaches 400 cm$^{-1}$, a value in the AlAs phonon regime.

The monotonic increase of the energy separation between the peaks (the phonon energies emitted by hot electrons) in the hot electron luminescence spectra (as shown in Fig. 4) suggests that the coupling strength between hot electrons and AlAs-like phonons is becoming stronger as the Al composition is increased. Therefore, we can estimate the emission strength of AlAs-like LO phonons relative to the GaAs LO phonons by taking into
account the optical phonon energies measured in the Raman experiments and the energy separations in the hot electron luminescence spectra. In the case for $x=0.3$, the energy separation of the peaks is about $29 \text{ cm}^{-1}$ larger than the energy of the GaAs LO phonons. This indicates that although interaction with the GaAs LO phonon is strong, there is still a significant contribution from the AlAs-like LO phonon. However, for $x = 1.0$, the spectra are dominated by AlAs-like LO phonons and the energy separation are very close to the AlAs LO phonon mode.

3. Conclusion

In conclusion, we have observed phonons in the present Raman scattering and hot electron-neutral acceptor luminescence investigation of the GaAs/Al$_x$Ga$_{1-x}$As multiple quantum wells. In the Raman scattering experiments, the dependence of the mode frequency on the Al composition is the important factor in distinguishing the phonon modes from the bulk optical phonons. We have also demonstrated that, for smaller $x$ in the barrier, the emission of the GaAs optical phonon mode is stronger. But for the largest $x$ investigated, the energy relaxation of hot electrons is dominated by the AlAs-like phonon.

Acknowledgments

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References

External electric field effect on energy level positions in a quantum well

A. P. Boltnev, N. N. Loiko, M. M. Rzaev and N. N. Sibeldin
Lebedev Physical Institute RAS, Leninsky pr. 53, 117924 Moscow, Russia
e-mail: rzaev@sci.lebedev.ru

Abstract. The effects of an electric field on quantum well (QW) potential profile and position of bound states there are investigated. It is shown, that the electric field applied on a symmetrical quantum well transforms it into asymmetrical one, and at strong electrical field — into triangular one, and the energy of a bound state decreases with an increase of field as long as the level of energy remains in quadrangular well region. In strong electrical fields the energy level are shifted into triangular well region, that determines the increasing of bound state energy concerning the well middle with field increasing.

The external electric field applied on quantum well structure changes their potential profile and, therefore, level positions there. The symmetric QW in presence of an electric field becomes asymmetrical, and the wide rectangular QW can be transformed in triangular one with corresponding energy spectrum. The considerable attention [1–6] was given to a problem of study of an electric field influence on an energy distribution in QW structures. It is known that the energy spectrum of QW structure could be determine by means of photo or electroluminescence measurements [1, 3]. Disadvantages of these methods are concerned, firstly, with involving to the luminescence process two types of charge carriers (electrons and holes), that considerably complicates a problem of an evaluation of electron or hole energy spectrum, secondly, for many QW structures it is rather difficult to observe the luminescence. The technique recently offered by us [7], allows to determine energy level positions by applying electric field on the quantum well structure for hole and electron wells independently.

In the present paper we study the influence of an electric field on energy level positions in QW structures.

The measurement of a level position in QW structure under external electric field was carried out with the help of a procedure [7]. The technique is based on a screening effect of an external electric field applied on structure by charge carriers localized in the well. Low-frequency capacitance–voltage (CV) measurements of QW structures are made. If a width and depth of well are known, the procedure allows to reconstruct the QW potential profile and to determine an energy level position by a given magnitude of an external electric field.

The experimental study were performed on Si/Ge structures with a Schottky barrier. Samples were grown in conventional molecular beam epitaxy (MBE) system “Katun”. The Si and Ge layers were deposited on n-doped Si (100) substrate with resistivity of 7.5 Ωcm. First a 90 nm-buffer layer was deposited at temperature of 850°C. Then temperature of the substrate was reduced up to 500°C and Ge layer with thickness of 2a = 5 nm was grown. Finally the 20 nm-thick undope Si cap was deposited. The CV measurements performed on this structure have shown that Ge layer in the given structure forms an electron quantum well with depth of $W = 65$ meV. If we assume an electron effective mass in Ge layer equal
to an electron effective mass of bulk material ($m^* = 0.22m_0$), we obtain in QW only one level with an energy $W_x = 23$ meV counted from the well bottom.

Figure 1 shows the external electric field effects on the energy level position in QW. The level position is counted from middle of the well bottom (point 2 in Fig. 3). In Fig. 1 we can see that in weak electric field region the energy of a bound state in QW decreases. Then at an electric field of $E = 10^5$ V/cm the level energy counted from middle of the well bottom begins to increase. The same result have been obtained in [2] although in [1, 3, 4, 6] have been found, that with increasing of an electric field the level energy decreases. In Ref. [2] the influence of the electric field on level positions in QW with various depths was studied theoretically. The Schrödinger equation in an effective mass approach for a particle with a charge $e$ and mass $m^*$ in the quantum well of width $2a$ and depth $W$ was solved and it was revealed that for QW of depth $W = 70$ meV in present of strong electric fields the energy of level counted from the middle of well bottom increase. Physically such behavior of level position changes in shallow quantum well remained not clear.

The reason of such unusual behavior of level position in QW at the applied electric field increasing is due to QW structure potential profile changes. The interpretation of changes in energy level position and potential profile of OW with applied external electric field becomes easier if we plot energy level positions for following well regions (counted from
Potential profile of quantum well and bound state position there as a function of applied electric field.

Schottky barrier, see Fig. 3): the point 1 is the most distant from barrier well wall; the point 2 is in the middle of well; the point 3 is the nearest to the barrier well wall. This is illustrated in Fig. 2 for cases mentioned above (curves 1, 2, 3 respectively). The curve 1 differs from the curve 2 on magnitude of a potential energy $(eaE)$, and the curve 3 differs from a the curve 2 on magnitude $(-eaE)$, where $e$ is an electron charge, $a$ is QW half-width, $E$ is electric field. Thus if we know a level position in a quantum well, and we take into account, that a potential of the well bottom in points 1 and 3 differs from each other on magnitude $(2eaE)$, it is possible to find the potential profile of QW and positions of a level there for a given value of an electric field.

Let us consider level positions as a function of energy for electric fields of $E_a = 0 \text{ V/cm}$; $E_b = 0.75 \times 10^5 \text{ V/cm}$, $E_c = 0.9 \times 10^5 \text{ V/cm}$, $E_d = 1.5 \times 10^5 \text{ V/cm}$. At $E_b$ the level energy in QW is equal to zero, if the energy is counted from a point 3. The energy level for $E_c$ is a minimum if the energy is counted from point 2. The field of $E_d$ is a random sampling, that allow define the QW potential profile and energy level positions in the present of strong electric fields. In Fig. 3 are shown QW profiles for all four cases mentioned above.

It follows from Fig. 1 that for electric field of $E_b$ the rate of energy decreasing with increase of field falls, thus the energy level from rectangular well region transform in triangular one, and for electric field of $E_c$ the level is deeply buried in triangular region of QW. With an increase of electric field the triangular region of well becomes narrower the level energy arises (see Fig. 1).

Thus in the presence of external electrical field a considered symmetrical well becomes asymmetrical, and moreover triangular if the electric field is strong and the energy of a bound state decreases with the field increase (Fig. 1) as long as the energy level remains in quadrangular region of the well. In strong electrical fields the energy level is shifted into triangular region of the well, that defines the increasing of bound state energy concerning the well middle with an electrical field increasing.

Acknowledgements

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References
Multiband coupling and electronic structure of short-period (GaAs)$_n$/((AlAs)$_n$ (001) superlattices

G. F. Glinskii, V. A. Lakisov, A. G. Dolmatov and K. O. Kravchenko
Saint-Petersburg Electrotechnical University, St Petersburg, 197376, Russia

Abstract. It is shown that effective mass approximation together with short range interface corrections can be used to calculate short-period abrupt (GaAs)$_n$/((AlAs)$_n$ (001) superlattices. The results obtained are in excellent agreement with results of pseudopotential calculations (A. Zunger et al.) in a wide range of superlattices periods (from $n = 1$ to $n = 20$).

Introduction

Short-period (GaAs)$_n$/((AlAs)$_n$ (001) semiconductor superlattices (SL) is a convenient model for theoretical and experimental analysis of transformation of heterostructure type from type I to type II. Depending on the number of monolayers, the conduction band minimum of SL is mainly formed either by $\Gamma_{\text{IIa}}$ state of the conduction band of GaAs or by $X_1$ state of the conduction band of AlAs.

In a series of recent publications [1–4] pseudopotential method was used for direct numerical calculations of electronic structure of (GaAs)$_n$/((AlAs)$_n$ (001) SL. However possibility of using of the effective mass approximation (EMA) to analyze short-period abrupt SL was questioned due to absence of interband coupling in that theory. Nevertheless, it was shown by Burt in [5] that this coupling is taken into account in the standard Luttinger–Kohn envelope-function theory and mainly arises due to rapid change of potential at the interface. In the framework of the EMA this coupling gives rise to short range interface corrections (SRIC) in effective Hamiltonian, which modify interface boundary conditions for envelope functions and lead to interband and intraband coupling of the states. Coefficients of this coupling can be determined from a microscopic theory or introduced phenomenologically.

For GaAs/AlAs (001) interface SRIC were analyzed in [6–10]. However, up to the present time there is a lack of unified opinion concerning the coupling coefficients. According to [6–7] these corrections lead only to $\Gamma_{155} - \Gamma_{155}$, coupling in the valence band and to $\Gamma_{1} - X_{32}$, $X_{1x} - X_{1y}$ and $X_{3x} - X_{3y}$, couplings in the conduction band. In contrast to these results, analysis of pseudopotential Hamiltonian [8] shows that $X_{1x} - X_{1y}$ coupling is zero and $X_{1x} - X_{32}$ coupling is non-zero. In [9], where tight binding model was used, it was shown that $\Gamma_{1} - X_{12}$ coupling can exist only when there are some interface imperfections. However magnetotunneling measurements [10] indicate that both $X_1$ and $X_3$ bands contribute to the $\Gamma - X_2$ and $X_4 - X_5$ couplings.

In the present work the EMA together with SRIC (EMA + SRIC model) was for the first time used for systematic analysis of short range corrections that arise at GaAs/AlAs (001) interface. Using the method of invariants in $T_2^2$ space-group, we obtained 7-band $\Gamma_1 \oplus^* X_1 \oplus^* X_3$ and 6-band $\Gamma_8 \oplus \Gamma_7$ multiband effective Hamiltonians, which include SRIC in all orders of perturbation theory. These Hamiltonians were used to calculate electronic structure of short-period (GaAs)$_n$/((AlAs)$_n$ (001) SL. Comparing our results with the results of pseudopotential calculations [2, 4] we determined the coupling coefficients.
It was shown that when one takes into account the short range corrections in the framework of the EMA+SRIC model it allows one to adequately describe energy structure of electron and oscillating character of $\Gamma - X$ anticrossing in the whole range of studied SL periods (from $n = 1$ to $n = 25$).

1. Multiband coupling in effective Hamiltonians

Hamiltonian of $A_3B_5$ heterostructure with single heterointerface $AB/A'B$ can be presented in the following form

$$H = \frac{p^2}{2m_0} + \frac{1}{2} \left[U_2(x) + U_1(x)\right] + f(n(x - a)) \Delta U(x). \quad (1)$$

Here $U_{1,2}(x)$ are the crystal potentials on the left and right sides of the interface respectively, which have the same period and symmetry; $f[n(x - a)]$ is an odd step-function, modulating periodic potential; $n$ is a unit vector normal to the interface; $a$ is Bravias lattice vectors determining positions of atom B at the interface plane.

In the framework of the EMA+SRIC model multiband Hamiltonian contains the following SRIC terms

$$H_{m,n,j}^{(a,b)}(x) = \left\{ \alpha, n, k^0_\alpha \right\} f[n(x - a)] \Delta U(x) \left\{ \beta, m, k^0_\beta \right\} \delta[n(x - a)]. \quad (2)$$

Here $\left\{ \alpha, n, k^0_\alpha \right\}$ and $\left\{ \beta, m, k^0_\beta \right\}$ are Bloch states, which correspond to $k^0_\alpha$ and $k^0_\beta$ points of the Brillouin zone of a virtual Ga$_0.5$Al$_0.5$As crystal; $\alpha$ and $\beta$ denotes bands (including irreducible representations of the space-group $T^2_d$). Matrix element in the right hand side of Eq. (2) determines coefficients of interband and intraband couplings. Since $\Delta U(x)$ is an invariant in the translation group, non-zero contribution into the coupling of these states is produced only by the following rapid Fourier oscillations of $f[n(x - a)]$

$$f^s[n(x - a)] = \sum_{q_\perp, G} f(q_\perp) e^{i q_\perp \cdot [n(x - a)]} \delta_{q_\perp, n, k^0_\alpha - k^0_\beta + G}, \quad (3)$$

where $f(q_\perp)$ is a Fourier transform of $f(nx) = f(x_\perp), f(x_\perp = 0) = f(q_\perp = 0) = 0$. In the (GaAs)$_n$/(AlAs)$_n$ (001) SL interfaces are located at the distance $na = n(a_0/2)$ from the zero coordinate ($a_0$ is a lattice constant). According to Eq. (3) there are only two perturbation operators that are odd under inversion of $n$ (these operators have different signs for $AB/A'B$ and $A'B/AB$ interfaces), which give a non-zero contribution into $\Gamma - \Gamma$, $X - X$ and $\Gamma - X$ couplings in the first order of the perturbation theory. These operators transform according to respectively $\Gamma_{15}(\Gamma_{15})$ and $X_3(X_{3\pi})$ irreducible representations of the space-group $T^2_d$ (in which $\Delta U(x)$ is invariant)

$$\hat{V}_0^{(0)\Gamma_{15}} = \Delta U(x) \sum_{m=1}^{\infty} 2i f \left(\frac{4\pi}{a_0} m\right) \sin \frac{4\pi}{a_0} m z \quad (4)$$

$$\hat{V}_0^{(0)X_3} = \Delta U(x) \sum_{m=0}^{\infty} 2i f \left[\frac{2\pi}{a_0} (2m + 1)\right] \sin \frac{2\pi}{a_0} (2m + 1) z \cos \pi (2m + 1) n. \quad (5)$$

Therefore matrix of multiband EMA+SRIC effective Hamiltonian will include only odd under inversion of $n$ corrections of the first order, which have symmetry of $\Gamma_{15}(\Gamma_{15})$ and $X_3(X_{3\pi})$. These lead to $\Gamma_{15} - \Gamma_{15y}$, $\Gamma_1 - X_3$, $X_1 - X_3$ and $X_3 - X_3$
couplings, which fully agrees with [8]. In the framework of a model with limited number of bands, effective Hamiltonian will include corrections of higher orders, which arise due to interactions with other bands. Corrections of even orders are even under inversion of $n$ and have symmetry of $\Gamma_1, \Gamma_{12}(\Gamma_{12,1}), X_1(X_{1z})$. Corrections of odd orders are analogous to the corrections of the first order. Even $\Gamma_1$ and $\Gamma_{12}(\Gamma_{12,1})$ corrections are diagonal, but $X_1(X_{1z})$ corrections are off-diagonal and lead to $\Gamma_1 - X_{1z}$ and $X_{1x} - X_{1y}$ couplings.

2. Electronic structure of (GaAs)$_n$/[(AlAs)$_n$]

EMA + SRIC method was used to calculate energy structure of electrons ($\Gamma_1 \oplus X_1 \oplus X_3$ model) and of holes ($\Gamma_8 \oplus \Gamma_7$ model) in (GaAs)$_n$/[(AlAs)$_n$] (001) SL with $n = 1 \ldots 25$. Reducing of bulk crystalsymmetry $T^2_{2d}$ to SL symmetry $D^5_{2d}$ leads to a split of energy states

\[
\begin{align*}
T^2_d & \rightarrow D^5_{2d} \\
\Gamma_1 & \rightarrow \Gamma_1 \\
\Gamma_7 & \rightarrow \Gamma_7 \\
\Gamma_8 & \rightarrow \Gamma_6 \oplus \Gamma_7 \\
X_1 & \rightarrow \Gamma_1 \oplus X_1 \oplus X_3 \oplus X_5 (even \ n) \\
X_3 & \rightarrow \Gamma_1 \oplus X_1 \oplus X_3 \oplus X_5 (odd \ n) \\
\end{align*}
\]

Figure 1 shows how energy of the two lowest conduction states of (GaAs)$_n$/(AlAs)$_n$ (001) SL.

![Graph showing energy of the two lowest conduction states of (GaAs)$_n$/(AlAs)$_n$ (001) SL.]

Table 1. Coupling coefficients (eVÅ) for $\Gamma_1 \oplus X_1 \oplus X_3$ model.

<table>
<thead>
<tr>
<th>Coupling coefficients</th>
<th>(\alpha - \beta)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Gamma_1 \rightarrow \Gamma_1$</td>
<td>-0.452</td>
</tr>
<tr>
<td>$X_{1z} - X_{1z}$</td>
<td>0.051</td>
</tr>
<tr>
<td>$X_{3z} - X_{3z}$</td>
<td>0</td>
</tr>
<tr>
<td>$\Gamma_1 \rightarrow X_{1z}$</td>
<td>-0.101</td>
</tr>
<tr>
<td>$\Gamma_1 \rightarrow X_{3z}$</td>
<td>-0.562</td>
</tr>
<tr>
<td>$X_{1z} - X_{3z}$</td>
<td>0.539</td>
</tr>
</tbody>
</table>
the best matching of our results with $n = 1$ and $n = 2$ with the results of the pseudopotential calculations [4] (dots in Fig. 1). It can be seen that there is an excellent agreement of our results for other periods ($n = 3 \ldots 20$) with the results obtained in [4]. This proves equivalency of the two following approaches: EMA+SRIC and empirical pseudopotential models. However the EMA+SRIC method, which we used, is much easier and does not require huge computational efforts.

References

Dynamical Kerr effect in a quantum-well AlGaAs/GaAs structure under circular optical excitation

A. V. Kimel†, V. V. Pavlov†, R. V. Pisarev†, V. N. Gridnev†, V. P. Evtikhiev†, I. V. Kudryashov† and Th. Rasing‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Research Institut for Materials, University of Nijmegen, The Netherlands

Abstract. Time-resolved magneto-optical Kerr effect is used for studying the ultrafast spin dynamics in a semiconductor heterostructure AlGaAs/GaAs in the subpicosecond time range. Results are analyzed on the basis of the semiconductor Bloch equations.

Introduction

In recent years, there has been a growing interest in the investigation of the spin dynamics down to subpicoseconds in semiconductors and semiconductor heterostructures. In particular, the processes governing the coherence between excited and ground states of photo-carriers are of utmost interest for ultrafast electronics. Various techniques have been used in the past for the study of spin dynamics, such as photoluminescence or Faraday effect [1, 14, 3]. However, the magnetooptical Kerr effect has been rarely applied for the studies of semiconductors, though it is a standard practice in linear and nonlinear optics of ferromagnets [4]. The Kerr effect (like the Faraday effect) is an alternative for materials that do not produce any luminescence at the wavelength of interest. Moreover, both Kerr and Faraday effects allow one to study spin dynamics on a broad spectral and thermal range. Compared to Faraday the Kerr effect allows one to study samples outside their transmission region as well as thick samples.

In this paper we illustrate our technique based on the time-resolved dynamical Kerr effect with femtosecond resolution by investigating a GaAsAl/GaAs heterostructure.

1. Theory

The change of the polarization state of the probe pulse upon reflection is described by the following expression [2]

$$\epsilon + i \theta \propto \int dt \, E^*(t) \{ P^{++}(t) - P^{--}(t) \},$$

where $\epsilon$ is the Kerr ellipticity and $\theta$ is the Kerr rotation. The nonlinear polarization $P^{++} = P_x + i P_y$ ($P^{--} = P_x - i P_y$) is induced by the right (left) circularly polarized light. Eq. (1) is valid if the thickness of the reflecting layer is much smaller than the wavelength of light. This condition is fulfilled in the case of reflection from a quantum well structure.

We assume that low-excitation approximation is applicable [6]. Then, the induced macroscopic polarization $\mathbf{P}(t)$ is related to the wave-vector-dependent interband optical polarization $P_{sj}(\mathbf{k})$ for the transition between the conduction band with spin $s = \pm(1/2)$ and the valence band with angular momentum $j = \pm(3/2), \pm(1/2)$ by the equation

$$\mathbf{P}(t) = \sum_{\mathbf{k}, s, j} |\psi_k(0)|^2 \mathbf{d}_{sj} P_{sj}(t, \mathbf{k}),$$

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where $d_{sj}$ is the dipole matrix element, $|\psi_k(0)|^2$ is the Sommerfeld enhancement factor. The interband polarization $P_{sj}(k)$ is calculated by solving the semiconductor Bloch equations ($\hbar = 1$)

$$\frac{dP_{sj}(k)}{dt} = \left[-i\varepsilon_{sj}(k) - \frac{1}{T_2}\right]P_{sj}(k) + i\Omega_{sj}(k)[1 - n_s(k) - n_j(k)],$$

$$\frac{dn_j(k)}{dt} = i\sum_s [\Omega_{sj}(k)P^*_j(k) - \Omega^*_{sj}(k)P_{sj}(k)] - \frac{n_j(k)}{T_1},$$

and a similar equation for $n_s(k)$. Here $\varepsilon_{sj}(k) = k^2/2\mu_{sj} + E_g - \omega$, $\mu$ is the reduced mass, $E_g$ is the band gap, $\omega$ is the central frequency of the laser field, $T_1$ and $T_2$ are the phenomenological relaxation times for population and polarization, respectively, $\Omega_{sj}(k)$ is the renormalized Rabi energy.

The laser field has the form $E(t) = E(t)e^{-i\omega t}$ with the amplitude $E(t) = e_+E_1(t) + e^0_+(e_+ + e_-)E_2(t)$, which describes the relative phase of the pump and probe fields. We take into account only transitions from the heavy-hole (hh) valence subband to the conduction band. The reasons for that are the large frequency detuning with respect to the light-hole (lh) transitions in the quantum well and the $1/\sqrt{3}$ smaller matrix element of the lh transition. For transitions from hh subband $d_{3/2,1/2} = -de_+$ and $d_{3/2,-1/2} = -de_-$, where $d$ is the magnitude of the dipole moment. In this model the $e_+$ component of the probe field only interacts with the $e_+$ component of the pump field.

We solve Eqs. (3), (4) numerically and calculate the macroscopic polarization $P(t)$. We seek for a solution which is linear in $E_2$ and of second order in $E_1$. In accordance with experimental conditions we average the solution over the relative phase $\phi$. This greatly reduces the complexity of the problem. Finally, by using relation (1) we calculate the Kerr ellipticity $\epsilon$ and the rotation $\theta$.

2. Samples and experiment

The $Ga_{0.7}Al_{0.3}As/GaAs$ double quantum well structure was grown by MBE on GaAs (001) substrate and growth was monitored by RHEED [5].

The measurement technique uses a differential detection scheme, which is extremely sensitive to small changes in polarization of probe beam and insensitive to laser intensity fluctuations or fast reflectivity changes. By modulating the helicity of the circularly polarized pump beam without changing its intensity one can strongly diminish the effect of the pump beam on the complex refraction index. For the experiment we used Ti-Sapphire laser (100 fs pulses, repetition rate 82 MHz) in the spectral range of 1.44–1.61 eV. The pump and probe pulses were focused on the sample to a spot with a diameter of about 100 $\mu$m. The pump fluence was approximately 1 $\mu$J/cm$^2$. For the study of dynamical Kerr effect the polarization of the pump pulses between two circular polarizations with opposite helicities was modulated using a photoelastic modulator. For the measurements of reflectivity dynamics the intensity of the linearly polarized pump beam was modulated instead of the polarization.
Fig. 1. The dynamics of reflectivity and Kerr rotation, induced by linearly and circularly polarized excitations, respectively.

3. Experimental results and discussion

The dynamics of the reflectivity and the Kerr rotation, which are induced by linearly and circularly polarized excitations, respectively, is presented in Fig. 1 for the central photon energy of the laser pulse at 1.44 eV. One can see that the Kerr rotation dynamics is characterized by an exponential dependence with a decay time of 73 ps. On the other hand, the reflectivity does not relax on this time scale. This is in agreement with the fact that for excited carriers the spin-relaxation time is much shorter than the energy relaxation time. This comparison shows that the dynamical Kerr effect is sensitive to the spin relaxation and insensitive to the population of the excited states, at least in this time regime.

Dynamical Kerr rotation with femtosecond resolution is presented in Fig. 2 for a set of energies in the range 1.45–1.544 eV. The observed effect is characterized by an additional decaying component with a delay time less than 200 fs. Moreover, the dynamical Kerr rotation shows an oscillatory behavior in the region 1.46–1.48 eV.

In Fig. 3 the spectral dependence of the induced Kerr rotation is shown for two delay times $t = 0$ and $t = 500$ fs. In the first case both relaxation components are present. In the second case only one component with the long relaxation time remains. In the inset the similar spectral dependencies for the dynamical reflectivity are presented. One can see that the spectra of the dynamical Kerr effect vary in time.

The fast and long relaxing parts of the induced Kerr rotation have been compared with similar data for undoped and heavily n-doped bulk GaAs. The decay times for these three samples are of the same order of magnitude. The amplitude of the dynamical Kerr effect in the quantum well structure is one order of magnitude larger than in undoped and heavily n-doped bulk GaAs. The fast and long relaxing parts we explain by scattering the spin of holes and electrons, respectively.

The spectral dependencies of the dynamical Kerr effect are completely different from similar data in heavily doped bulk GaAs. The simulation of the experimental data was done by solving the optical Bloch equations (3), (4).

We have found that the nonmonotonous behavior of the Kerr effect as a function of delay time in the vicinity of the excitonic resonance can be explained by the exciton-exciton interaction. Analyzing the behavior of the Kerr rotation and Kerr ellipticity separately we conclude that exciton-exciton interaction reveals not only as the local field effect but also as the interaction induced dephasing time.
Fig. 2. The dynamical Kerr rotation spectra as a function of time delay in the femtosecond time domain.

Fig. 3. The spectral dependence of the induced Kerr rotation for two delay times $t = 0$ and $t = 500$ fs.

Acknowledgements
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References
An analytic kinetic approach to Zener interminiband transitions in superlattices

P. Kleinert† and V. V. Bryksin‡
† Paul-Drude-Institut für Festkörperlektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Starting from an equation-of-motion analysis of the density matrix, phenomenological kinetic equations are derived that allow a study of semiclassical intraband and quantum-mechanical tunneling contributions to the current densities. An analytical expression is obtained, which describes the field dependence of the electron densities in the upper and lower minibands. It is shown that a population inversion does not occur as long as the scattering time for thermal generation is larger than the interminiband relaxation time. Numerical results for the current density are discussed.

1. Introduction

Recently, there have been intensive investigations of the influence of external electric fields on semiconductor superlattices (SLs). The physics of biased SLs is extremely rich due to the large number of parameters that can be controlled quite independently. Most of the interest has been caused by the need of understanding the interplay of Bloch oscillations, Zener tunneling between Bloch bands, and interaction effects. From a theoretical point of view the simplest models involve only a single miniband. This approach leaves out all interesting interminiband phenomena, which are captured most simply in a two-band tight-binding model. The purpose of the present work is to study the nonequilibrium carrier kinetics by an equation-of-motion analysis of the density matrix and a derivation of an expression for the current, from which semiclassical intraband and quantum-mechanical tunneling contributions can be identified. Starting from a microscopic theory, we derive phenomenological kinetic equations by using the constant relaxation-time approximation. The dynamics of electrons is treated in a simple two-band SL model. Former theoretical studies [1, 2] of unipolar devices will be extended by taking into account all relevant horizontal and vertical electron transitions. In addition, the widely used Esaki–Tsu model [3] is extended to two-band systems.

2. Theoretical model

The quantum-kinetic equation for the Wigner transformed elements of the density matrix $f_{\nu}^{\nu'}(k)$ (with $\nu, \nu'$ being miniband indices), whose explicit spatial dependence describing field domains is not taken into account, is given by [4]

$$\left\{ \frac{e}{\hbar} E \nabla_k + \frac{i}{\hbar} [\varepsilon_{\nu'}(k) - \varepsilon_{\nu}(k)] \right\} f_{\nu}^{\nu'}(k)$$

$$+ \frac{i e E}{\hbar} \sum_{\mu} \left[ Q_{\mu\nu}(k) f_{\mu}^{\nu'}(k) - Q_{\nu'\mu}(k) f_{\nu}^{\mu}(k) \right] = \sum_{\mu\mu'} \sum_{k_1} f_{\mu}^{\mu'}(k_1) W_{\nu\nu'}^{\mu\mu'}(k_1, k),$$

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where the matrix elements
\[ Q_{\mu\nu}^{\prime}(k) = \sum_{K} \chi_{\mu}^{\prime}(k + K) \nabla_{k_{z}} \chi_{\nu}^{*}(k + K) \] (2)
determine the wavefunction overlap calculated from the SL envelope functions \( \chi_{\mu} \). The \( Q \)-term in Eq. (1) describes electric-field-induced vertical transitions via Zener tunneling.

We consider a two-band model characterized by the tight-binding minibands
\[ \varepsilon_{1}(k) = \frac{\hbar^{2}k_{\perp}^{2}}{2m} + \frac{\Delta_{1}}{2} (1 - \cos k_{z}d), \] (3)
\[ \varepsilon_{2}(k) = \frac{\hbar^{2}k_{\perp}^{2}}{2m} + \varepsilon_{g} + \frac{\Delta_{2}}{2} (1 + \cos k_{z}d). \] (4)
\( \Delta_{1} (\Delta_{2}) \) is the bandwidth of the lower (upper) miniband, and \( \varepsilon_{g} \) the energy gap between the minibands. \( k_{\perp} \) is the quasi-momentum of the lateral electron motion. To lowest order in the coupling constant, only the transition probabilities \( W_{11}, W_{22}, W_{12}, W_{21} \) have to be accounted for. We will study the kinetic equation (1) in the constant relaxation time approximation by making use of the replacement
\[ \text{Re} \ W_{11}^{22}(k', k) = \text{Re} \ W_{22}^{11}(k', k) \rightarrow -\delta_{k'k}/\tau \] (5)
for the interminiband transition probabilities. The horizontal interminiband transitions via thermic generation \( W_{12}^{12} \) and recombination \( W_{21}^{21} \) are described by relaxation times \( \tau_{1} \) and \( \tau_{2} \), respectively. Finally, for the description of intraminiband transitions, we use the extended Esaki–Tsu model \[3\].

Focusing on high electric fields, where \( \Omega > 1/\tau_{\text{eff}} \) (\( \Omega = eEd/\hbar \) is the Bloch frequency and \( \tau_{\text{eff}} \) an effective scattering time), and taking into account the periodicity of \( f^{\prime}_{\nu}(k) \) along the field direction \( k_{z} \), we switch to the Wannier–Stark representation by Fourier transforming the distribution function \[4\]
\[ f^{\prime}_{\nu}(k) = \sum_{l=-\infty}^{\infty} e^{ilk_{z}d} f_{\nu}^{\prime}(k_{\perp}, l). \] (6)

We neglect the smooth \( k_{z} \) dependence in \( Q_{\mu\nu}^{\prime}(k) \) and obtain the following analytical result for the electron density \( n_{2} \) of the upper miniband
\[ n_{2} = \frac{2(\Omega \tau)^{2} A_{0} + \tau/\tau_{1}^{\prime} + \tau/\tau_{2}^{\prime}}{4(\Omega \tau)^{2} A_{0} + \tau/\tau_{1}^{\prime} + \tau/\tau_{2}^{\prime}}, \quad A_{0} = \sum_{l \neq 0} \frac{q(l)^{2}}{(l\Omega \tau - \Omega_{g} \tau)^{2} + 1}. \] (7)

In the expression for \( A_{0} \), we introduced the abbreviations
\[ q(l) = (-1)^{l} Q_{12} J_{l} \left( \frac{\Delta_{1} + \Delta_{2}}{2\hbar \Omega} \right), \quad \Omega_{g} = \frac{\varepsilon_{g} + (\Delta_{1} + \Delta_{2})/2}{\hbar} - \Omega(Q_{22} - Q_{11}). \] (8)

with \( J_{l} \) denoting the Bessel function. In the absence of thermal generation \( \tau_{1}^{\prime} \rightarrow \infty \) and under the condition \( n_{2} \ll 1 \), we recover the result \( n_{2} \approx 2\Omega^{2} \tau_{1}^{\prime} A_{0} \) derived already in \[4\]. From Eq. (7), it is seen that population inversion occurs only under the unrealistic condition \( \tau_{2}^{\prime} > \tau_{1}^{\prime} \) independently of the value of the electric field strength.
Fig. 1. (a) Electric-field dependence of the carrier density for the upper (solid lines) and lower (dashed lines) minibands. The bandwidth of the lower (upper) miniband is $\Delta_1 = 5$ meV ($\Delta_2 = 40$ meV). The two minibands are separated by an energy gap of $\epsilon_g = 100$ meV and the temperature is $T = 300$ K. We use identical intraminiband relaxation times $\tau_1 = \tau_2 = 0.1$ ps and the following values for the interminiband scattering times $\tau = 0.3$ ps and $\tau' = 20$ ps. For the solid curves from bottom to top, the interminiband recombination time $\tau'_2$ is given by 1, 3, and 5 ps. The positions of Zener resonances at $l\Omega = \Omega_g$ ($l = 1, 2$) are marked by thin vertical lines.

(b) The same as in (a) for the relative current density $j_z/j_0$ with $j_0 = e n_s (\Delta_1 + \Delta_2)/2\hbar$. $n_s$ is the carrier sheet density.

An analytical expression for the current density is obtained from [4]

$$j_z = -\frac{n}{E} \sum_{kk'} \left[ \epsilon_1(k_z) f_1^{k_z}(k'_z) W_{11}^{11}(k'_z, k) + \epsilon_2(k_z) f_2^{k_z}(k'_z) W_{22}^{22}(k'_z, k) 
+ \epsilon_1(k_z) f_2^{k_z}(k'_z) W_{21}^{21}(k'_z, k) + \epsilon_2(k_z) f_1^{k_z}(k'_z) W_{12}^{12}(k'_z, k) \right], \quad (9)$$

where $n$ denotes the total electron density.

3. Numerical results and discussion

Numerical results for the carrier densities and the relative current $j_z/j_0$ are shown in Fig. 1(a) and (b). The set of parameters used in the calculation is given in the caption. The matrix elements $Q_{\mu\mu'}$ have been estimated with wavefunctions of a quantum well with infinite potential barriers. A characteristic feature in these graphs is the appearance of Zener resonances at $l\Omega = \Omega_g$, which are shifted to larger field strengths with increasing energy gap $\epsilon_g$ so that at low electric fields the well-known Esaki–Tsu current-voltage characteristics [3] is recovered. The lineshape of the Zener resonances depends sensitively on the miniband widths. Whereas the carrier density $n_2$ is a function of the sum $\Delta_1 + \Delta_2$, pronounced current resonances occur only for narrow lower and broad upper minibands. With increasing interminiband relaxation time $\tau$ the peaks become sharper. When the generation and recombination lifetimes are identical ($\tau'_1 = \tau'_2$), we obtain from Eq. (7) $n_2 = 0.5$ independently of the electric field strength. As long as thermal generation can be neglected ($\tau'_2 < \tau'_1$), a population inversion cannot occur in the considered SL model.

References


Band alignment in ZnCdTe/ZnTe and ZnCdSe/ZnSe SQW structures grown on GaAs(100) by MBE

V. I. Kozlovsky†, Yu. G. Sadofyev† and V. G. Litvinov‡

† P. N. Lebedev Physical Institute RAS, 53, Leninsky Pr., 117924, Moscow, Russia
‡ Ryazan State Radioengineering Academy, Gagarina 59/1, 390000, Ryazan, Russia

Abstract. MBE-grown ZnTe/CdZnTe/ZnTe and ZnSe/ZnCdSe/ZnSe strained single quantum well (SQW) structures with non-doping layers were investigated by cathodoluminescence (CL) and deep level transient spectroscopy (DLTS). Obtained DLTS and CL results were used for the estimation of the conduction band offset parameter $Q_C$.

1. Introduction

One of the major parameters of SQW structures is the energy band offset $\Delta E_C$ (conduction band), $\Delta E_V$ (valence band), which determines the heights of potential barriers for electrons and holes. However, these parameters for structures that we investigated are determined insufficiently reliably. Thus, the data of conduction band offset value, $Q_C = \Delta E_C / \Delta E_G$, ($\Delta E_G = \Delta E_C + \Delta E_V$ is gap discontinuity), for the Zn$_{1-x}$Cd$_x$Te/CdTe heterojunction are known from the literature being changed from 0.775 up to 1.2 [1, 2] and for Zn$_{1-x}$Cd$_x$Se/ZnSe one from 0.64 up to 0.83 [3], respectively. One of the reasons of such data scattering can be essential dependence of edges of allowed bands on built-in elastic strains. These strains appear due to mismatching of crystal lattice parameters and their distribution on layers depends on concrete structure composition.

2. Experiment

In this paper the band offsets in strained ZnTe/Zn$_{1-x}$Cd$_x$Te/ZnTe and ZnSe/Zn$_{1-x}$Cd$_x$Se/ZnSe SQW structures are experimentally determined by the low-temperature cathodoluminescence (CL) and electrical current deep level transient spectroscopy (DLTS) methods. Studied samples were grown by molecular beam epitaxy (MBE) on $n^+$-GaAs(100) substrates misoriented on $3^\circ$ to a $\langle 110 \rangle$ direction. Main parameters of grown structures are given in the left part of Table 1. CL spectra were recorded at temperature of 14 K, electron energy of 10 keV, e-beam current density of 0.1 mA/cm$^2$ and an electron beam spot diameter of 1 mm. In the case of DLTS the thermal evaporation of indium on the GaAs substrate back and nickel on the ZnTe (ZnSe) cap layer was used to prepare diode-like structures. The area of the nickel electrodes was 1 mm$^2$. All samples had high electrical resistance because of self-compensation effects in the epitaxial layers. Capacity of structures did not change with a bias voltage. For this reason, traditional DLTS variant based on a relaxation of the structure capacity [4] could not be applied for study of our samples. We registered the electrical current relaxation through the structure (the current DLTS) using usual procedure of DLTS measurement in all other respects.

3. Results and discussion

CL spectra of SQW structures contained weak narrow emission lines from free and bound exciton emissions near band edge region of ZnTe (ZnSe) and also an intense $I_{\text{qw}}$ line due to quantum well (QW) emission. Energy standing of the $I_{\text{qw}}$ line depended on QW
Table 1. Parameters of studied structures with single Zn$_{1-x}$Cd$_x$Te QW (samples 195–197) and Zn$_{1-x}$Cd$_x$Se QW (samples 207, 209–211).

<table>
<thead>
<tr>
<th>Sample num</th>
<th>%Cd in QW</th>
<th>$L_w$ (nm)</th>
<th>$E_{CL}$ (eV)</th>
<th>$\Delta E_{CL}$ (meV)</th>
<th>$E_1$ (meV)</th>
<th>$E_{e1}$ (meV)</th>
<th>$E_{hh1}$ (meV)</th>
<th>$Q_C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>195</td>
<td>22</td>
<td>3.5</td>
<td>2.246</td>
<td>134</td>
<td>101</td>
<td>80</td>
<td>16</td>
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<tr>
<td>196</td>
<td>22</td>
<td>6.5</td>
<td>2.206</td>
<td>174</td>
<td>133</td>
<td>38</td>
<td>7.5</td>
<td>0.816±0.048</td>
</tr>
<tr>
<td>197</td>
<td>20</td>
<td>5.5</td>
<td>2.226</td>
<td>154</td>
<td>116</td>
<td>46</td>
<td>9</td>
<td>0.814±0.05</td>
</tr>
<tr>
<td>207</td>
<td>21</td>
<td>9</td>
<td>2.562</td>
<td>241</td>
<td>155</td>
<td>19</td>
<td>5.3</td>
<td>0.656±0.038</td>
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<tr>
<td>209</td>
<td>37</td>
<td>5</td>
<td>2.403</td>
<td>400</td>
<td>255</td>
<td>54</td>
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<tr>
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<td>5</td>
<td>2.583</td>
<td>220</td>
<td>140</td>
<td>44.5</td>
<td>12.9</td>
<td>0.695±0.038</td>
</tr>
<tr>
<td>211</td>
<td>34</td>
<td>5</td>
<td>2.427</td>
<td>376</td>
<td>220</td>
<td>53</td>
<td>15</td>
<td>0.632±0.035</td>
</tr>
</tbody>
</table>

parameters. The $I_{qw}$ intensity was 2–3 orders of magnitude more than intensity of near band gap emission lines. Full width on half maximum of $I_{qw}$ lines was equal to 8–10 meV. This fact testifies to high enough quality of structures. Energy standings, $E_{CL}$, of the $I_{qw}$ line maximums for all tested structures are listed in Table 1.

The DLTS spectra contained a peak from deep level with an activation energy $E_2$ (samples 195–197, Fig. 1) or $E_3$ (samples 207, 209–211, Fig. 2) and an intense peak $E_1$ in low temperature region. At used minimal temperature of measurements (77 K) the recording of this peak was possible in emission rate windows providing relaxation time constant less than 5 · $10^{-4}$ s.

We consider that more intense $E_1$ peak is due to emission of electrons from the ground quantified level in the conduction band of the QW. The activation energy of the $E_1$ in this case corresponds to the energy interval between the conduction band bottom in the barrier layer and ground quantified level in the QW. The activation energy of the traps bound with the low temperature $E_1$ peak changed from 101 up to 255 meV for various structures (Tables 1, 2).

The values of the effective bulk carrier concentrations $N_t$, at $E_1$ levels calculated by traditional method, are listed in Table 2. Taking into account that the capture of carriers occurs in the QW, it is possible to estimate the sheet concentration $n_s$ of carriers captured on the ground quantified level as: $n_s = N_t \cdot L_w$, where $L_w$ is the well width. The estimates show that in studied samples $n_s$ does not exceed $1 \times 10^{10}$ cm$^{-2}$. It means that in the QW the ground quantified level is filled only.

The CL and DLTS experimental results were used for the estimation of conduction band offset parameter:

$$Q_C = \frac{\Delta E_C}{\Delta E_G} = \frac{(E_1 + E_{e1})}{(\Delta E_{CL} - \Delta E_h + E_{e1} + E_{hh1})},$$

(1)

where $E_1$ is the DLTS activation energy of electron emission from the ground state $e_1$ in QW, $\Delta E_{CL}$ is the energy shift of the QW emission line from the free exciton emission line in the barrier ZnTe (ZnSe); $E_{e1}$, $E_{hh1}$ is the energy of the ground state for electrons in the conduction band and holes in the valence band, respectively; $\Delta E_h$ is changing of heavy hole exciton binding energies in the barrier layer and QW (see Table 1).

The energies $E_{e1}$ and $E_{hh1}$ were calculated by using known parameters of the QW (its width, $E_1$, electron and hole effective masses in the QW and the barriers, Cd content) and taking the rectangular model of the QW with wide barriers and finite depth [5]. So, $E_{e1}$ and $E_{hh1}$ can be written as:

$$E_{e1} = F(L_w, \Delta E_C) = F(L_w, E_{add} + E_{e1}),$$

(2)
Fig. 1. DLTS spectra of the ZnTe/Zn$_{1-x}$Cd$_x$Te structures No 195 (curves 1–1'), 196 (2–2'), 197 (3–3') measured at the reverse voltage $V_r = -1$ V and the filling voltage $V_f = +1$ V.

Fig. 2. DLTS spectra of the ZnSe/Zn$_{1-x}$Cd$_x$Se structures No 207 (curves 1–1'), 209 (2–2'), 210 (3–3'), and 211 (4–4') measured at the reverse voltage $V_r = -1$ V and the filling voltage $V_f = +1$ V.

\[ E_{hh1} = G\{L_w, \Delta E_V\}G\{L_w, \Delta E_C(1 - Q_C)/Q_C\} \]
\[ = G\{L_w, (E_{add} + E_{e1})(1 - Q_C)/Q_C\}. \quad (3) \]

where $F$ and $G$ are the transcendental functions.

Band gap of the QW material and its change respect to band gap of the barrier material can be expressed through the energy standing, $E_{CL}$, of the QW emission line and its energy shift, $\Delta E_{CL}$, from the free exciton emission line in the barrier ZnTe (ZnSe) (see Table 1) as:

\[ E_G = E_{CL} + E_{hh}^b - E_{e1} - E_{hh1}, \quad (4) \]
\[ \Delta E_G = \Delta E_V + \Delta E_C = \Delta E_V + E_{e1} + E_{e1} = \Delta E_{CL} - \Delta E_{hh}^b + E_{e1} + E_{hh1}. \quad (5) \]

where $E_{hh1}$ is the heavy hole exciton binding energy. Thus, it is supposed that the free exciton emission line formed by the heavy hole. Further, in calculations of $E_G$, it has been taken into account that the exciton binding energy, $E_{hh}^b$, is incremented approximately on 10 meV in the narrow QW when Bohr radius of the exciton ($\sim 5.5$ nm for ZnCdTe and $\sim 4$ nm for ZnCdSe [7]) becomes comparable with well width. In case of the wide QW (9 nm, sample 207) the exciton binding energy is close to value 18 meV observed in the bulk ZnSe [5].

The numerical solution of combined equations (1–5) allows to calculate values $E_{e1}$, $E_{hh1}$ and parameter $Q_C$ using the experimental data from the CL and DLTS spectra without calculation of the built-in elastic strains. Observed such manner values $Q_C$ are listed in
Table 2. Parameters of the deep levels detected by DLTS. $E_t$, $N_t$ — activation energy and bulk concentration of the trap, $\sigma = \sigma_\infty \exp(\Delta E_b/kT)$ is the capture cross section, $\Delta E_b$ is the activation energy for capture of electrons, $\sigma_\infty$ is the temperature independent prefactor of the capture cross-section.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>DLTS level</th>
<th>$\Delta E_t$ (eV)</th>
<th>Cross section, $\sigma_\infty \exp(\Delta E_b/kT)$ (cm$^2$)</th>
<th>$N_t$ (cm$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>195</td>
<td>$E_1$</td>
<td>0.101±0.01</td>
<td>$6 \cdot 10^{-16}$</td>
<td>$3.3 \cdot 10^{15}$</td>
</tr>
<tr>
<td></td>
<td>$E_2$</td>
<td>0.58±0.02</td>
<td>$1.3 \cdot 10^{-16}$</td>
<td>$3.6 \cdot 10^{13}$</td>
</tr>
<tr>
<td>196</td>
<td>$E_1$</td>
<td>0.133±0.01</td>
<td>$2.6 \cdot 10^{-14}$</td>
<td>$2.6 \cdot 10^{15}$</td>
</tr>
<tr>
<td></td>
<td>$E_2$</td>
<td>0.58±0.02</td>
<td>$3.4 \cdot 10^{-16}$</td>
<td>$3.4 \cdot 10^{13}$</td>
</tr>
<tr>
<td>197</td>
<td>$E_1$</td>
<td>0.116±0.01</td>
<td>$3.5 \cdot 10^{-14}$</td>
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<tr>
<td></td>
<td>$E_2$</td>
<td>0.58±0.02</td>
<td>$5 \cdot 10^{-16}$</td>
<td>$2.1 \cdot 10^{13}$</td>
</tr>
<tr>
<td>207</td>
<td>$E_1$</td>
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<td>$8 \cdot 10^{-14}$</td>
<td>$3.5 \cdot 10^{13}$</td>
</tr>
<tr>
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<td>$E_1$</td>
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<td>$E_3$</td>
<td>0.72±0.03</td>
<td>$6.3 \cdot 10^{-14}$</td>
<td>$2 \cdot 10^{12}$</td>
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<tr>
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<td>211</td>
<td>$E_1$</td>
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<tr>
<td></td>
<td>$E_3$</td>
<td>0.72±0.03</td>
<td>$4.1 \cdot 10^{-14}$</td>
<td>$3.6 \cdot 10^{12}$</td>
</tr>
</tbody>
</table>

Table 1. We have found that $Q_C \approx 0.82$ for ZnTe/Zn$_{1-x}$Cd$_x$Te structures at cadmium concentration of 0.2–0.22 in the QW and $Q_C = 0.66$ as an average value for ZnSe/Zn$_{1-x}$Cd$_x$Se structures with $x = 0.2–0.37$.

4. Conclusion

Thus, in this work the signal caused by electrons emission from the ground quantified level in the conduction band ZnCdTe/ZnTe and ZnSe/ZnCdSe SQW structures was found by DLTS method. Activation energy of this level correlates with the energy standing of the QW emission line on CL spectra. The calculation procedure of the conduction band offset parameter $Q_C$ based on experimental data from DLTS and CL at known QW width is offered.

Acknowledgements

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References

A giant shot of radiation intensity of space indirect exciton line in double quantum wells in GaAs/AlGaAs

V. V. Krivolapchuk, E. S. Moskalenko and A. L. Zhmodikov
Ioffe Physico-Technical Institute, St Petersburg, Russia

Investigation of properties of double quantum wells (DQW) attracts nowadays a great interest of scientists both from theoretical and experimental points of view. This interest is caused by existence of space indirect excitons (IX) in a DQW that are formed of an electron (e) and a hole (h) localized in different quantum wells of a DQW. Due to the fact that e and h in an IX are separated in real space this IX has a significantly larger radiative time than a direct exciton (DX) formed of an electron and a hole in the same quantum well. The latter fact allows us to obtain practically a gas of IX of rather large density even at small densities of excitation and, as a consequence, to expect different collective properties of a system of separated electron–hole pairs (excitons) of large density to display experimentally.

One of the interesting collective properties following from the Bose-gas statistics is a possibility of appearance in such a system of Bose–Einstein condensation (BEC) when a macroscopic number of particles occupies the lowest energetic state in the system. Though it is well known that no BEC exists for free bosons in exactly two dimensions, the situation drastically changes if in addition to the extended (free) states excitons can occupy discrete (localized) states, which are positioned lower in energy with respect to the extended states. Really, as it was shown theoretically [1], in this case chemical potential is not allowed to approach bottom of the boson free states and hence even at nonzero temperatures \( T \) there is a finite value of bosons \( n_c(T) \) which can be accommodated by the extended states. The existence of the upper limit \( n_c(T) \) means that whenever actual boson density \( n \) exceeds this limit, the extra amount \( n - n_c(T) \) will spill over to the localized states — the BEC effect.

We present systematic study of the IX luminescence line from the samples with GaAs/AlGaAs DQWs (detailed descriptions of sample and experimental setup are given elsewhere [2]) in the wide range of experimental parameters such as bath temperature, excitation power and value of the external electric field applied to the sample. At certain values of external parameters we detected the giant (up to three times) shot in luminescence intensity within some part of the IX spectral profile. We discuss the observed phenomenon in the frame of theoretical model [1] of BEC which in our case is determined by the existence of localized states formed by the heterointerface potential fluctuations. In this sense the occupation of localized states by the macroscopic number of excitons which takes place within the limited space regions restricted by the heterointerface potential fluctuations resembles an experimentally discovered phenomenon of BEC on alkali atoms in space-limited traps produced by the magnetic field (for a detailed review see [3]).

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References

Multiphonon relaxation in ZnSe thin films and ZnSe/ZnCdSe superlattice

P. N. Lebedev Physical Institute, RAS, 117924, Moscow, Russia

Abstract. The photoluminescence (PL) and resonant Raman scattering (RRS) spectra of thin films and multiple quantum well (MQW) structures were investigated at room temperature. The light scattering spectra of MQW consist of intensive band of the quantum wells luminescence and additional narrow bands. The energy shift of these additional bands was equal to the multiple value of longitudinal oscillation phonon of the strained ZnSe barriers. The intensity of these bands is resonance amplified at approaching the excitation radiation frequency to MQW luminescence bands frequency. We suppose, that this process consists of several stages: absorption of stimulating light by quantum wells; exchange of energy between hot electrons and barriers; relaxation of system including longitudinal phonon generations in barriers.

Introduction

The structures with single and multiple quantum wells are objects of intensive experimental and theoretical investigations. The interest for II–VI low dimensional structures is caused by possibility of creation of effective light-emitting devices with spontaneous or coherent radiation completely overlapping visible region of spectrum. In this work the optical properties of thin ZnSe films and MQW ZnSe/ZnCdSe structures were researched by methods of a PL and RRS.

1. Experimental procedure

All investigated samples were grown by molecular beam epitaxy (MBE) on GaAs (100) substrates. Homogeneous ZnSe films were of various width: from 100 Å up to 9000 Å. The MQW structures configuration included a ZnSe buffer layer with thickness 1 µm and periodical sequence of Cd$_x$Zn$_{1-x}$Se quantum wells separated by ZnSe potential barriers. The total quantum-size region thickness equals $\approx 1.5$ µm for all structures. The thickness of the ZnSe cap layer on the surface of structures is 300 Å. The additional information on a quantum well composition was received by measuring Auger-spectra of Cd$_x$Zn$_{1-x}$Se films of the greater thickness in the analytical chamber of MBE unit and from energy standing of the low-temperature photoluminescence lines of the MQW structures. All used methods gave the coinciding results. The main structure parameters are given in the Table 1.

The PL and RRS spectra are registered at room temperature on a U-1000 spectrometer in

### Table 1. List of structure parameters.

<table>
<thead>
<tr>
<th>MQW sample number</th>
<th>Widths of quantum wells (Å)</th>
<th>Molar concen. of cadmium, $x$</th>
<th>Width of ZnSe potential barriers (Å)</th>
<th>Quantity of quantum wells</th>
<th>RS band frequency (cm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>178</td>
<td>66</td>
<td>0.4</td>
<td>100</td>
<td>100</td>
<td>246</td>
</tr>
<tr>
<td>182</td>
<td>80</td>
<td>0.5</td>
<td>500</td>
<td>30</td>
<td>248</td>
</tr>
<tr>
<td>183</td>
<td>55</td>
<td>0.27</td>
<td>1000</td>
<td>15</td>
<td>250</td>
</tr>
<tr>
<td>184</td>
<td>61</td>
<td>0.3</td>
<td>5000</td>
<td>4</td>
<td>250</td>
</tr>
</tbody>
</table>
geometry of “backscattering”. RRS and PL spectrums were excited by 4416 Å, 4880 Å and 5145.3 Å laser lines for all samples. The resolution was 1–5 cm$^{-1}$.

2. Results and discussion

In Fig. 1 the RRS spectrums both films and MQW structures at 4416 Å laser line excitation are shown. The energy of quantum of this line is higher than energy of a ZnSe forbidden region. The spectrums consist of the narrow bands and the wide bands of a ZnSe edge luminescence. On the diagram the spectrums were shifted upwards in accordance with increasing of film width or MQW barrier width. It is easy to see that the spectrums have common features. The changes in a spectrum (varying distribution of band intensity) occur in accordance with increasing of ZnSe layer width. The narrow bands with shifted on the multiple value frequencies are observed in RRS spectrums of polar semiconductors. This shift is featured by expression $n\omega_{LO}$, where $\omega_{LO}$ is energy of longitudinal oscillation of a semiconductor lattice, $n \sim 1, 2, 3 \ldots$ [1]. The smoothly varying distribution of these band intensity in RRS spectrums ZnSe films at change of film thickness from 1 µm up to 0.1 µm was observed in [2]. Results of experiments was explain on the basis of cascade model. In this model the electrons, excited highly in a conduction band, can relax, letting out one behind other LO-phonons.

Apparently in our case there is a same process of an excited electron relaxation including of ZnSe LO-phonons both in ZnSe films and in MQW barriers. The basic change of a spectrum (see Fig. 1) occurs at thickness 100–2000 Å. In paper [2] basic changes of a spectrum occurred at thickness 1000–10000 Å. It is possible to explain by higher quality of ZnSe films structure (in [2] the films obtained by cathode sputtering were explored).

![Resonant Raman scattering spectra of thin films and MQW structures at room temperature.](image_url)

**Fig. 1.** Resonant Raman scattering spectra of thin films and MQW structures at room temperature. (a) MQW sample number. (b) ZnSe film width (Å).
Fig. 2. PL spectra of various ZnSe/ZnCdSe MQW structures at excitation by various lines of the Ar laser: (a) 4880 Å and (b) 5145.3 Å.

PL spectra for MQW samples at excitation by 4880 and 5145.3 Å laser lines are shown in Fig. 2. The energies of quantum of these lines have an intermediate value between energy of a ZnSe-barrier forbidden region and energies of electron-hole pairs recombination in quantum wells. Each of samples has a luminescence band with frequency position determined by properties of quantum wells. Since the quantum well widths in different structures was varied in small limit, the frequency position of a luminescence band is determined mainly by concentration of Cd in the quantum wells. There is a set of narrow bands in a spectrum of each sample alongside with QW luminescence band. The frequency position of these narrow bands are shifted from excitation light energy on the value multiple energies 246–251 cm\(^{-1}\) for various samples (Table 1). The intensity of these additional bands is being resonant increased at approaching of excitation light energy to luminescent bands energy of the quantum wells.

In our samples the ZnSe buffer layer and barriers of the MQW structures are transparent for 4880 and 5145.3 Å exciting lines and, therefore, LO-phonon repetitions can not be excited in ZnSe. The availability of LO-phonon repetitions and absence of TO-phonons in spectrums testify that these spectra are obtained in RRS conditions. It would seem, these phonon repetitions could be related to solid solutions Cd\(_x\)Zn\(_{1-x}\)Se oscillations of a well. Nevertheless these bands in our sample spectrums can be connected with ZnSe because of their range frequencies. Really, the frequency dependence of longitudinal phonon in a solid solution Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3]. When the value \(x\) in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions Cd\(_x\)Zn\(_{1-x}\)Se on a cadmium concentration \(x\) is known [3].
of the measured LO-modes frequencies contradicts results of definition of the quantum wells parameters by all listed above methods \textit{in situ} and \textit{ex situ}. For example, the cadmium molar concentrations for samples N183 and N182, was calculated on the PL band frequency positions (Fig. 2), differs on 0.23, but concentrations calculated on the RS band frequency positions (Table 1) gives the differs in cadmium concentrations about 0.02 \cite{3}. In addition, for each sample, energy shift of additional bands does not depend on a wavelength of exciting light. Therefore it is more reasonable to assign these bands to a material of a barrier. The small shifts of these band frequencies can be explained by internal strains in structure caused by mismatch of crystalline lattice periods of ZnSe barriers and Cd$_x$Zn$_{1-x}$Se quantum wells \cite{4}.

3. Conclusion

At a photoexcitation of MQW structures with exited light quantum energy higher than energy of a ZnSe forbidden region, a cascade relaxation of hot electrons in barriers including of longitudinal phonons of the ZnSe barrier occurs. If the exited light quantum energy have an intermediate value between energy of a ZnSe-barrier forbidden region and energies of electron-hole pairs recombination in quantum wells, this process occurs in some stages:

(a) the light quantum is absorbing by a quantum well. This process is especially effective if the energy of exciting light is closer to the luminescense line energy of the quantum wells;
(b) there is an exchange of energy between hot electrons and barriers because QW thickness is close to Bohr radius of excitons in QWs;
(c) there is a relaxation of energy of an electron through sequential interaction to longitudinal oscillations of the barrier material.

Acknowledgements

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References


Theoretical investigation of intraband absorption of electromagnetic radiation by holes in quantum wells

N. A. Nezlobin, A. S. Polkovnikov and G. G. Zegrya
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Using four-band Kane model we calculate intraband absorption coefficient of light by holes followed by their transition to the spin-orbital split-off band for AIIIIV semiconductor quantum wells. It is shown that such an absorption mechanism is possible without any participation of a third particle like a phonon or an impurity and this mechanism can give the major contribution to the absorption of light in semiconductor quantum well lasers.

Introduction

It is common to believe that light absorption in semiconductor heterostructures by free carriers is weak, being possible only if together with electron-photon interaction a simultaneous scattering of an electron or hole on some other particle like a phonon or an impurity is considered. Nevertheless strong intraband damping of radiation is observed in long wavelength laser structures, based on narrow band semiconductors at high temperatures [1, 2]. This damping results in eventual collapse of lasing. As the width of the active region decreases, intraband absorption becomes enhanced and in laser structures with quantum wells absorption coefficient can be up to tens of inverse centimeters. At the same time the estimates done for the absorption coefficient accompanied by scattering on phonons or impurities give smaller values then those experimentally observed [3–5].

In this paper it will be shown that photon annihilation can be due to pure interaction with a hole and this process doesn’t require participation of a third particle. Restrictions on intraband light absorption by a hole imposed by quasimomentum conservation are lifted if the hole interacts with hetero boundaries. Due to breaking of translation invariance symmetry the hole wave function appears to be a wave packet including various values of longitudinal wave vector $k_x$. In this way quasimomentum conservation can be satisfied for an arbitrary excited-hole state.

1. Wave functions of SO-holes in a rectangular quantum well

For calculation of intraband absorption coefficient we will use four-band Kane model, which proved to reproduce electron and hole spectrum in AIIIIV semiconductors with a sufficient accuracy. Such a calculation for a homogeneous semiconductor has been done in particular in [8]. Boundary conditions and wave functions for the heavy holes are given in [9, 10]. Spin-orbital split-off wave functions are to be constructed from two basis spinors. Below we give $x$-coordinate dependence of one of them:

$$
\begin{bmatrix}
\frac{i\hbar \gamma (q^2 + k^2)}{E_g + \delta - E} \cos k_x \eta \\
k \sin k_x \eta - \lambda q \cos k_x \xi \\
- i q \cos k_x \eta + i \lambda k \sin k_x \xi \\
- \lambda k \sin k_x \xi + \lambda q \cos k_x \eta
\end{bmatrix}
$$

(1)
The origin of the hole energy \( E \) is at \( \delta = \Delta/3 \) below then the valence band edge inside the quantum well; \( k \) and \( q \) are the \( x \)- and lateral quasimomentum components, \( \lambda = \delta/(E + \delta + \hbar^2(q^2 + k^2)/2m_h) \).

With a good approximation we can neglect by mixing of heavy and light hole branches with large wave vectors to the spin-orbital holes. Notice, that this approximation is different from using \( 2 \times 2 \) model for description of SO-holes. The basis functions given above contain components with both \( j = 1/2 \) and \( j = 3/2 \), where \( j \) being the total angular momentum, see for example [11]. In this way we can considerably simplify procedure of finding both discrete and continuous spectrum and wave functions of SO-holes. Wave functions of the holes situating on such quasidiscrete energy levels exponentially decay away from the interface with the characteristic wave vector \( \kappa \). The dispersion equation for these holes is as follows:

\[
(\alpha \kappa \cot \frac{ka}{2} + k) \left( \alpha \kappa \tan \frac{ka}{2} - k \right) = q^2 (\alpha \bar{\lambda} + \lambda)^2, \tag{2}
\]

where \( a \) is the quantum well width, \( U_c \) is the electron barrier height, \( \bar{\lambda} \) corresponds to \( \lambda \) in a wide band semiconductor and

\[
\alpha = \frac{E_g + \delta + U_c - E}{E_g + \delta - E} \frac{q^2 + k^2}{q^2 - k^2}.
\]

States with different parity don’t separate and as the analysis shows wave spectrum is not parabolic. It is important to take this non-parabolicity into account when calculating intraband absorption coefficient [8]. The spectrum calculated without parabolic approximation is quite simple, curves \( k(q) \) are almost the straight lines and the approximation \( k(q) = \text{const} \) is nearly exact. This approximation considerably simplifies calculation of absorption coefficient.

2. Matrix element of optical transition

The matrix element of the optical transition between he states \( \Psi_h \) of a heavy hole and \( \Psi \) of a SO-hole is equal to:

\[
M = \frac{\hbar e}{c} \gamma \int (\mathcal{A}_0, \Psi_p, h^+ \Psi) \, dx \tag{3}
\]

We assume that the light absorption mostly occurs at narrow band region of the heterostructure, i.e. in a quantum well. Therefore in (3) we restrict integration to the region from \(-a/2\) to \(a/2\). As an example we explicitly provide one non-zero matrix element for a transition to discrete and continuous spectra:

\[
M_{sed,x} = \frac{\hbar e}{c} \gamma H_1 q D_1 S_k \left( \frac{\sin (k_h - k)a/2}{k_h - k} + \frac{\sin (k_h + k)a/2}{k_h + k} \right)
\]
\[
M_{osc,z} = -\frac{\hbar e}{c} \gamma H_2 k_h C_1 S_k \left( \frac{\sin (k_h - k)a/2}{k_h - k} - \frac{\sin (k_h + k)a/2}{k_h + k} \right)
\]

Here \( S_k = [\hbar \gamma(q^2 + k^2)]/(E_g + \delta - E) \) is the quantity proportional to \( s \)-component of the wave function of a spin-orbital hole in the quantum well, \( H_1 = H_2 = 1/\sqrt{a(q^2 + k^2)} \) are the normalizing constants for heavy holes, \( D_2 \) and \( C_1 \) are those for a SO-hole. The
Fig. 1. Frequency dependence of absorption coefficients $\alpha_x$ and $\alpha_{yz}$, $a = 50$ Å, $T = 250$ K.

Fig. 2. Frequency dependence of absorption coefficient $\alpha_{yz}$ at different quantum well widths, $T = 100$ K.

The number of transitions per unit time and unit length is equal to:

$$Q = \frac{2\pi}{\hbar} \sum_{k_0} \int_{0}^{\infty} q dq \int_{0}^{2\pi} d\varphi \left( \left| \sum_{k(q)} M_{q}^2 \right| + \int_{0}^{\infty} \left( \left| M_{z1}^2 \right| + \left| M_{z2}^2 \right| \right) \frac{d\tilde{k}}{2\pi} 2A \right)$$

$$\times f(E_h) \delta(E_h - E - \hbar\omega).$$

(4)

The multiplier of 2 reflects double degeneracy of energy levels. The rates $Q_x$ and $Q_{yz}$ corresponding to different initial polarization of the light are to be calculated separately. The corresponding rate for an arbitrarily polarized light can be obtained from

$$Q = Q_x \cos^2 \beta + Q_{yz} \sin^2 \beta,$$

(5)

where $\beta$ is the angle between the vector potential $\vec{A}_0$ and $x$-axis. Direct calculations show that $Q_x$ is less then $Q_{yz}$ almost at all frequencies due to proportionality of $M_x$ to the lateral
component of the quasimomenta $q$. By the same reason dependence of $Q_x$ on temperature at constant density of holes in a well is stronger than that of $Q_{yz}$.

Figure 1 shows frequency dependence of absorption coefficients $\alpha_x$ and $\alpha_{yz}$ (other words of the rates $Q_x$ and $Q_{yz}$ divided by the quantum well width $a$ and photon flux density) for different polarizations of the light at temperature of 250 K.

In numerical calculation we used the following parameters for the InGaAsP/InP quantum well: $E_g = 0.83$ ev, $\Delta = 0.32$ ev, $U_{\nu} = 0.213$ ev, $U_c = 0.136$ ev, $m_b = 0.45m$, $m_e = 0.041m$, $m_{so} = 0.15m$, $p = 10^{12}$ cm$^{-2}$.

For wide quantum wells, absorption coefficient stipulated by heavy hole transition into discrete SO states has an explicit resonant nature and considerably exceeds coefficient caused by the transitions into continuous spectrum. In narrow quantum wells absorption is mainly due to transitions into the continuous spectrum and frequency dependence of the absorption coefficient is smooth (Fig. 2). Also it was found that resonant absorption is strongly affected by the quasistationarity of SO energy levels and by the scattering of phonons, which cause decrease and broadening of the peaks.

References

Built-in electric fields and electronic structure of GaN/AlN QDs

A. D. Andreev† and E. P. O’Reilly‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physics Department, University of Surrey, Guildford, GU2 5XH, UK

Abstract. We present a theory of the electronic structure of GaN/AlN QDs including built-in strain and electric field effects. A simple analytical formula is derived for the Fourier transform of the built-in electrostatic potential. The QD carrier spectra and wave functions are calculated using an original method we have developed based on an 8-band \( k \cdot P \) model. It is shown that due to the strong built-in electric field, the holes are localized in the wetting layer just below the QD bottom, while electrons are pushed up to the pyramid top. The energy of the ground optical transition is found to be in good agreement with available experimental data.

Introduction

Wide-bandgap nitride-based quantum well (QW) and dot structures have significantly different properties compared to the “classical” GaAs-based QW and QD structures. Whereas GaAs and most other III–V compounds have a cubic (zinc-blende) crystal structure, GaN and related nitride alloys generally have a hexagonal (wurtzite) structure, which leads to strong built-in piezoelectric fields in heterostructures, of the order of MV/cm. As a consequence, self-organized GaN/AlN QDs can exhibit a large red shift in the energy of the photoluminescence maximum, with interband emission reported about 0.5 eV below the GaN bulk bandgap [1].

The main aim of this paper is to study theoretically the influence of built-in electric fields on the electronic structure of hexagonal GaN/AlN QD structures and to compare the results with available experimental data. To the best of our knowledge, we present the first theoretical investigation of the carrier states in GaN-based QDs, which we carry out in the framework of a multi-band \( k \cdot P \) model, including the effects of the 3D strain and built-in electric field distributions.

1. Built-in strain and electric fields

The calculation of the spatial strain distribution in a QD structure requires the solution of a 3D problem in elasticity theory, often for a non-trivial quantum dot shape. In this paper we introduce a novel approach based on a Green’s function tensor formalism to calculate the 3D strain distribution in QD structures of arbitrary shape, and with hexagonal crystal symmetry. We derive an analytical expression for the Fourier transform of the QD strain tensor, valid for the case when the elastic constants of the QD and matrix materials are equal. We then obtain a compact analytical expression for the Fourier transform of the strain tensor. The 3D spatial distribution of the strain tensor is then found easily as the sum of the Fourier series.

A schematic 3D view of the GaN/AlN QD considered in this paper is shown in Fig. 1(a), with cross-section shown in Fig. 1(b) (view in \( x-z \) plane). In the calculations below, the shapes of the QDs are as presented in Fig. 1, with the sizes and vertical repeat distances...
Fig. 1. Showing schematic diagrams of GaN/AlN QDs shaped as truncated hexagonal pyramids (a) 3D view of a single QD standing on a wetting layer; and (b) view of the QD structure in the x-z plane. (c) Position dependence of the strain tensor components along the (01\(\overline{1}0\)) direction, \(x = z = 0\).

Fig. 2. (a) Calculated variation of the built-in electrostatic potential components, \(\varphi\), and total electric field, \(E\), along the (0001) direction \((x = y = 0)\). Solid line: Total built-in electrostatic potential, \(\varphi_{\text{total}}\), found as the sum of the strain-induced piezoelectric potential, \(\varphi_{\text{strain}}\) (dot-dashed line) and the spontaneous polarisation term, \(\varphi_{\text{spon}}\) (dashed line). (b) Contour plot of the variation in the total built-in electrostatic potential, \(\varphi_{\text{total}}\) in the x-z plane \((y = 0)\). The darkest areas show regions of low potential (where holes are trapped), and the brighter areas regions of higher potential (where electrons are trapped). The numbers in boxes show the magnitude of the potential (in eV) along the different contour lines.

used taken from experimental data [2]. Figure 1(c) shows the calculated variation of the strain tensor components along the (01\(\overline{1}0\)) direction. The magnitudes of the the strain fields peak near the pyramid edges, with smoother variations away from the edges. We have used a similar technique to calculate the Fourier transform of the built-in electric field, including the strain-induced piezoelectric field and the contribution due to the spontaneous polarization. The two sets of field terms give approximately equal contributions to the calculated built-in electric field in GaN/AlN QD structures, where the overall electric field magnitude can be of the order of several MV/cm. For example, using the piezoelectric
constants and spontaneous polarization values predicted by Bernardini et al. [3], we find the electric field in a GaN/AlN QD of height 4.1 nm to be around 6 MV/cm at the QD base and 4 MV/cm at the QD top (see Fig. 2(a)). Such giant built-in electric fields are characteristic for GaN/AlN QD structures.

2. Electronic structure

We use an efficient technique to calculate the carrier energy spectrum and wave functions in a semiconductor heterostructure containing QDs of arbitrary shape. The method is a natural combination of the plane-wave expansion and Fourier transform techniques used to derive the built-in strain and electric field distributions. Each carrier wave function is expressed in a series expansion based on a suitable set of bulk states. The coefficients of the series and the carrier energy levels in any QD are then found as the eigenvectors and eigenvalues of a Hamiltonian matrix, all of whose matrix elements can be found analytically. The proposed technique does not require explicit calculation of the 3D spatial distribution of the built-in strain and electric fields. This makes the method effective and fast not only for spectrum calculations, but also for further modeling of the optical properties of the QD structures.

The built-in electric field has a crucial influence on the carrier states in GaN/AlN QDs. The electrons are pushed up to the QD top and holes are pushed down into the wetting layer below the QD. In addition, both electrons and holes experience a significant lateral confinement due to the built-in electrostatic potential (see Fig. 2(b)). This creates an effective 3D potential for electrons and holes, with flat, circular symmetry. The form of the electron and hole wavefunctions in GaN/AlN QDs is therefore very similar to what would be expected for an infinitely deep flat cylinder (see Fig. 3(a,b)).

Finally, the calculated dependence of the energy position of the first PL maximum on QD size is found to be in good agreement with two experimental points from [1] for “large” and “small” GaN/AlN QDs (heights respectively ~4 nm and 2 nm), if we assume that the difference between the spontaneous polarization values of GaN and AlN, \( \Delta P_{\text{spont}} \approx 0.032 \text{ C/m}^2 \) (see Fig. 3(c)). This value of \( \Delta P_{\text{spont}} \) is ~40% smaller than the one of 0.052 C/m\(^2\) calculated by Bernardini, et al. [3], but is consistent with other experimental data and analysis on GaN/AlGaN heterostructures [4, 5, 6]. We conclude
that the method introduced here gives valuable information on the electronic structure of GaN/AlN quantum dots, and should also be particularly convenient for a range of future studies, including modeling of optical transition rates, and laser gain characteristics of realistic quantum dot structures.

Conclusion

In conclusion we note that the technique which we present here for calculation of the built-in electric and strain fields in semiconductor structures with QDs of arbitrary shape can also be applied to other structures based on a variety of compounds like GaAs/InAs, InP and ZnSe/CdSe.

Acknowledgements

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References

Abstract. It is shown that bulk reactive-ion-etched MOVPE-grown GaN can exhibit dramatically increased near-band-edge and below-band-gap emission when compared with unetched samples. A detailed spectroscopic study reveals, firstly, that donor bound excitons are the dominant species in the near-band-edge emission and, secondly, that previous models of the yellow luminescence band must be revised due to the clear linkage between the yellow emission and a little studied blue emission band. UV illumination over a period of time enhances the yellow emission by a factor of 20 at room temperature, and it is shown that this effect can be employed for optical data storage and retrieval.

Introduction

Reactive ion etching (RIE) is an important device processing technique for many semiconductors. This is especially true of Gallium Nitride (GaN), which is the material for a wide variety of opto-electronic devices in the blue-green part of the electromagnetic spectrum [1]. RIE is also a vital tool for the fabrication of GaN-based nanostructures due to its potential for precise control over etch profiles.

Despite a number of advantages over wet chemical etching methods, RIE has the potential disadvantage that it can introduce damage and/or defects, with subsequent deleterious effects on device performance. It is therefore important that RIE-induced damage in GaN is understood and controlled [2].

One of the key features of the RIE process is the interplay between chemical and physical etching mechanisms. In order to elucidate the different effects of these mechanisms we have used optical spectroscopy to examine both SF₆-etched samples (mainly chemical etching) and Ar-etched samples (physical bombardment). The SF₆-etched samples have been discussed extensively elsewhere [3]: in this paper we focus on new and surprising results from Ar-etched samples.

Experimental

Our samples are 2.5 µm thick nominally undoped GaN, grown by metal organic vapour phase epitaxy on c-plane sapphire substrates. Previous studies [3] focussed on the use of SF₆ as an etching gas with a power density of 0.45 W/cm², pressure 15 mtorr and flow rate 40 sccm, resulting in a DC bias of −440 V. In this work samples from two wafers were etched in an Ar-plasma (typically for 2.5 min) under conditions designed to achieve a similar DC bias (i.e. −440 V). Photoluminescence (PL) experiments were carried out on both etched and unetched samples using an argon-ion laser operating at a wavelength of 333.8 nm and a typical spot size ~0.25 mm. At this wavelength, the penetration depth of the laser beam is estimated to be 100 nm [3]. Samples were mounted in a closed cycle helium cryostat and variable temperature experiments were performed in the range 24 K to 300 K.
Figure 1 shows the band edge PL at 24 K in detail. The unetched sample shows the $A_1$ exciton at 3.477 eV, $B_1$ exciton at 3.483 eV, and negligible luminescence from bound excitons \[3\]. The temperature dependence of the spectra from the etched samples \[3\] allows us to identify the peak at 3.471 eV with the donor bound exciton (D0X). Spectra from these Ar-etched samples are more than an order of magnitude more intense than from unetched samples and spectra are dominated by the D0X peak, indicating an enormous increase in the number of donors in these samples. The free excitons $A_1$ and $B_1$ have not been eliminated: they are masked by the D0X peak at low temperatures and are revealed again at elevated temperatures \[4\].

The increase in near-band-edge emission in the Ar-etched sample is accompanied by a dramatic increase in the strength of the defect-related yellow emission (compared to the unetched sample) \[4\]. However, even this effect is dominated by a dramatic time dependence of the below band gap emission that is clearly visible to the naked eye: the illuminated spot is initially bright blue and becomes bright yellow after a few minutes. Figure 2 shows a very clear transfer of intensity from blue luminescence (BL) at $\sim$3.0 eV to yellow luminescence (YL) at $\sim$2.2 eV as a function of time. Similar time-dependent behaviour is observed in other samples etched with an Ar-plasma using different DC biases.

In the remainder of this paper we focus on results from a single sample with dimensions $\sim$6×6 mm \[5\]. One half was reactive-ion-etched with an argon plasma (as described...
Fig. 3. Normalised YL at room temperature as a function of time while exposing etched and unetched parts of the sample to the 50 mW UV write beam.

above) for 2.5 minutes while the other half of the sample was masked and thus remained unetched. The sample was mounted on a motorised stage so that it could be moved horizontally through the illuminating laser beam while simultaneously measuring the BL or YL (c.f. Ref [6]).

By illuminating various parts of the sample, the transfer of intensity from the BL to the YL has been used to record ‘data’. Data writing was achieved with high-intensity (usually 8 mW) 333.8 nm UV light while a low-intensity spot (100 µW, same diameter) was used to read the data. At low temperature, the written data were visible to the naked eye (yellow emission from previously exposed regions of the sample, blue emission elsewhere) or by setting the spectrometer to record either the BL or YL as a function of position on the sample [5]. We focus here on the results at room temperature (where no BL is observed).

Figure 3 shows that the YL from the etched sample can increase by more than a factor of 20 over 1000 seconds, but that there is very little change in the YL from the unetched sample. Figure 4 shows the YL recorded during spatial scans across the etched part of the sample before (top) and immediately after (second trace) writing at various points. Enormous peaks in YL are clearly visible from the exposed parts of the sample. At room temperature the enhanced YL from the exposed parts of the sample decays more quickly than at low temperature [5] but the features are still clearly observable more than 16 hours after writing (Fig. 4 bottom).

Discussion and conclusions

The results clearly show that reactive ion etching with an Ar-plasma can actually increase the near-band-edge emission from GaN, in contrast with the expectation that etch-induced damage will decrease the band edge PL. It is possible that Ar-plasma etching, or post-fabrication exposure of devices to an Ar-plasma, could be used to increase band edge emission.

The transition from the BL to YL is clearly due to the introduction of metastable defects during the etching process. In fact, a BL band has commonly been observed in unetched GaN [7] in conjunction with the YL but its possible importance seems not to have been widely recognised. Any model of the YL must also be able to account for the existence of the BL and allow for the possibility of a transfer of intensity from the BL to YL. This is an important result because, despite intensive investigation over the past 30 years, no existing model of the YL [8] appears capable of explaining the key features of the data reported here.

Finally, we have demonstrated that the metastable defects created in Ar-etched samples
Fig. 4. YL at room temperature read with $P = 100 \mu W$, as a function of horizontal position across the etched half of the sample. Top: before writing; second trace: immediately after writing with $P = 5 \text{ mW}$ for 15 min at positions 1.0, 1.5 and 2.3 mm, with $P = 50 \text{ mW}$ for 15 minutes at 3.1 mm, and $P = 100 \mu W$ for 2 min at 3.8 mm; third trace: after about 2 hours and additional writing with $P = 100 \mu W$ for 10 min at 4.7 mm and $P = 15 \text{ mW}$ for 37 min at 5.3 mm; bottom: after 16 hours it is still possible to read the data. Arrows indicate the edges of the sample.

can be employed in an all-optical data storage and retrieval scheme, even at room temperature. Some alternative schemes have been reported elsewhere, but in those cases the optical memory effect relies on either InGaN heterostructures [9] or near-band-edge emission [6]. The crucial feature of the present data is the dramatic sensitisation of the material to UV exposure after etching.

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GaN uniformity control on multiple 3 inch wafer grown in planetary reactors

AIXTRON AG, Kackerstr. 15-17, D-52072 Aachen, Germany
Tel: +49 (241) 8909-162, fax: +49 (241) 8909-40, e-mail: heu@aixtron.com

We report on recent results obtained using an AIX 2400G3HT production type Planetary Reactor® in the 5×3 inch configuration for growth of typical group-III nitride layer structures consisting of GaN, InGaN and AlGaN. The optimum reactor geometry has been found by extensive modeling of the reactor design. Increased thermal management allows maximum reactor temperatures above 1400°C. The temperature uniformity could be improved to less than 1°C over the satellite and from satellite to satellite. As a consequence of extensive reactor modeling, the process transfer from 6×2 inch to 5×3 inch configuration was carried out by simple scaling of the corresponding process parameters of the 6×2 inch configuration. The scaling factor is calculated with respect to the changed reactor geometry. We used optical reflectrometry for in situ growth control during this process development and could confirm the theoretical scaling requirements for obtaining identical growth conditions as compared to the 6×2 inch reactor configuration. This is verified by the generation of identical reflectance spectrum features, leading to identical growth results as shown in Fig. 1. This important issue of in situ control will be discussed in detail. The TMGa efficiency could be kept at about 17%. Switching to the 8×3 inch configuration the efficiency increases up to about 27%, which is an improvement of 63% as compared to the 6×2 inch configuration.

The obtained thickness uniformity on three inch wafers is around 1% standard deviation without rim exclusion. Typical photoluminescence emission wavelengths of 480 nm with 6 nm standard deviation could be achieved as shown in Fig. 2. For these results a run to run reproducibility of less than 1% standard deviation was proven by growth of several identical consecutive runs. The full width at half maximum (FWHM) of the 300 K emission is in the range of 35 nm at 475 nm as depicted in Fig. 3. We also report on doping uniformity data of single layer growth. The n-doping uniformity is better than 5% standard deviation on 3 inch. Electroluminescence test structures with emission wavelengths of 480 nm show average forward voltages of less than 4 V at 20 mA operation current indicating a uniform and highly conductive p-type cap layer.

Additionally we present results of AlGaN bulk layers. The growth rate of AlGaN has been reduced to about 1 µm/h to obtain good layer quality with mirror like surfaces. The Al distribution uniformity is in the rage of about 1% without rim exclusion for 10% Al content. The full width at half maximum (FWHM) of 300 K photoluminescence is about 5 nm at 340 nm emission wavelength. To maintain the low cycle time the successful etch back of the AlGaN residuals at 1400°C using our standard HCl etching procedure is an important step.

All of these results demonstrate that the up-scaling of the high temperature production reactor to larger wafer diameter applications is just a question of scaling the corresponding process parameters.
Fig. 1. Comparison of reflectance spectra of 5 period InGaN/GaN MQW structures grown in 6×2″ and 5×3″ planetary reactor configuration, respectively.

Fig. 2. Photoluminescence mapping of a 5 period InGaN/GaN quantum well structure grown on 3 inch sapphire. Standard deviation of wavelength using 3 mm rim exclusion is 5.6 nm which is 1.2%.

Fig. 3. Typical photoluminescence spectrum of a 5 period InGaN/GaN MQW structure. Wavelength is 480 nm, full width of half maximum (FWHM) is about 35 nm.
Optical phonons in hexagonal GaN/Al\(_x\)Ga\(_{1-x}\)N multilayered structures

V. Yu. Davydov†, A. A. Klochikhin‡, S. V. Goupalov†, I. N. Goncharuk†, A. N. Smirnov†, W. V. Lundin†, A. S. Usikov†, E. E. Zavarin†, A. V. Sakharov†, M. V. Baidakova†, J. Stemmer§, H. Klausing§, D. Mistele§ and O. Semchinova§

† Ioffe Physico-Technical Institute, St. Petersburg, Russia
‡ Petersburg Institute for Nuclear Physics, RAS, 188350, St. Petersburg
§ LFI Universit"at Hannover, Schneiderberg 32, 30167 Hannover, Germany

Abstract. The results of the phonon behavior in multilayered periodic structures GaN/Al\(_x\)Ga\(_{1-x}\)N are presented. It is shown that the A\(_1\) (TO), E\(_1\) (TO), and E\(_2\) (high) phonons can be considered as the vibrations propagating through the multilayered structure.

Introduction

Recently GaN/Al\(_x\)Ga\(_{1-x}\)N MQW structures have been grown by MBE, and now great efforts are directed towards fabrication of GaN/Al\(_x\)Ga\(_{1-x}\)N superlattices. At present there is a growing need for precise non-destructive characterization of these nanostructures. One of the most efficient, sensitive, and direct techniques which can be used for the quantitative characterization of semiconductors is Raman spectroscopy. Nevertheless, there are few works devoted to studies of the phonon behavior in low-dimensional GaN/Al\(_x\)Ga\(_{1-x}\)N structures. Theoretical treatment of phonons in such structures is restricted to consideration of superlattices (SL) consisting of several monolayers. However, it can be expected that in thicker structures with a period of tens nanometers the phonon behavior will also have specific features due hexagonal symmetry of GaN and AlN and crossing of their optical bands. The goal of our work was to reveal the peculiarities in the behavior of optical phonons in hexagonal multilayered GaN/Al\(_x\)Ga\(_{1-x}\)N structures grown by the MOCVD and MBE techniques.

1. Samples and experimental procedure

The objects were three multilayered structures grown on sapphire substrate. Two structures were grown by MOCVD and one structure was grown by MBE. The MOCVD-grown samples consisted of 37 pairs of GaN and Al\(_x\)Ga\(_{1-x}\)N layers grown on a thin Al\(_x\)Ga\(_{1-x}\)N buffer layer. The Al content in the first sample was \(x = 0.24\), and the thicknesses of GaN and Al\(_x\)Ga\(_{1-x}\)N layers were 31.3 nm and 46.2 nm, respectively. The Al content in the second sample was \(x = 0.29\), and the GaN and Al\(_x\)Ga\(_{1-x}\)N layers were 37.4 nm and 53.1 nm thick, respectively. The details of the growth can be found in Ref. [1]. The third sample consisting of 10 pairs of Al\(_{0.53}\)Ga\(_{0.47}\)N/GaN layers was grown by plasma-assisted molecular beam epitaxy (PAMBE) in a Riber 32 MBE system. The thicknesses of the GaN and the Al\(_{0.53}\)Ga\(_{0.47}\)N layers were 36.9 nm and 43.9 nm, respectively. All three structures were characterized by X-ray diffraction (XRD) and electron probe microanalysis (EPMA). Raman spectra of the multilayered structures were measured in a backscattering
configuration at room temperature. An Ar$^+$ laser ($\lambda = 488$ nm) was used as a source of excitation.

2. Experimental results and discussion

There are six optical modes $1A_1$ (TO) + $1A_1$ (LO) + $1E_1$ (TO) + $1E_1$ (LO) + $E_2$ (low) + $E_2$ (high) active in the first-order Raman scattering in hexagonal GaN and Al$_x$Ga$_{(1-x)}$N. The $\Gamma$-point phonon frequencies are well studied for both compounds [2–4]. The results described below refer to phonons of the $A_1$ (TO), $E_1$ (TO) and $E_2$ (high) symmetry which have the highest intensity in the Raman spectrum, and therefore their behavior can be traced in detail.

Figure 1 shows Raman spectra of the multilayered structures for the scattering geometry corresponding to the $A_1$ (TO) phonon. In addition, Raman spectra of bulk GaN and Al$_x$Ga$_{(1-x)}$N layers grown on a sapphire substrate are given. Note that the Al content in bulk Al$_x$Ga$_{(1-x)}$N layers was the same as in the structures studied. One can see that in the multilayered structures the $A_1$ (TO) phonon is detected only as a single line occupying an intermediate position between the frequencies of the $A_1$ (TO) phonons in bulk GaN and Al$_x$Ga$_{(1-x)}$N layers. We have also found that the situation is the same for the $E_1$ (TO) and $E_2$ (high) phonons in multilayered structures.

![Raman spectra](image)

**Fig. 1.** Raman spectra in the scattering configuration corresponding $A_1$ (TO) phonon mode for different samples.

To our knowledge, only one paper concerned with Raman studies of hexagonal GaN/AlN SL has been published [5]. The authors assigned the observed $E_1$ (TO) and $E_2$ (high) lines to the phonons of GaN layer and explained the blue shift of these lines with respect to their positions for bulk GaN by the built-in strain. However, the position of the line corresponding to the $A_1$ (TO) phonon was not consistent with calculations which included only the SL deformation. As an alternative, the interface origin of this line was suggested in this paper. The absence of lines corresponding to phonons in an AlN layer was attributed to a considerable difference in the Raman cross sections.

In our case the Raman cross sections for the layers comprising the multilayered structures were comparable, and two lines should be observed for a confined phonon of GaN and alloy. Nevertheless, the $A_1$ (TO), $E_1$ (TO), and $E_2$ (high) phonons were observed as single lines, which contradicts the hypothesis of phonon confinement. According to theoretical predictions [6], optical phonons in the AlN/GaN low dimensional structure can be considered as propagating due to the energy overlap between optical phonon regions of GaN and AlN. Our results indicate that propagating phonons rather than confined modes were
observed in Raman scattering from multilayered GaN/Al\textsubscript{x}Ga\textsubscript{1-x}N structures.

Large wavelengths of the phonons participating in the Raman scattering suggest that the lattice dynamics characteristics averaged over the multilayered structure period should be used to describe phonons in the GaN/Al\textsubscript{x}Ga\textsubscript{1-x}N structure. The averaging procedure can be performed by using one of the variants of the isodisplacement approach, which for many solid solutions describes well the phonon modes at the \Gamma-point. To check the validity of this approach, we compared our experimental data and the data obtained for bulk Al\textsubscript{x}Ga\textsubscript{1-x}N with the Al content equal to the averaged Al content in the multilayered structure $\mathcal{T} = a x/(a + b)$, where $a$ and $b$ are the thicknesses of the Al\textsubscript{x}Ga\textsubscript{1-x}N and GaN layers, respectively. Positions of the experimental maxima for $A\!{}_{1}(\text{TO})$, $E\!{}_{2}$\textsubscript{(high)} and $E\!{}_{1}(\text{TO})$ phonons in GaN/Al\textsubscript{x}Ga\textsubscript{1-x}N structures at corresponding $\mathcal{T}$ (shown by open circles), and experimental compositional dependence of these phonons for bulk Al\textsubscript{x}Ga\textsubscript{1-x}N are plotted in Fig. 2. Additionally, Fig. 2 presents the positions of the phonon frequencies corresponding to the averaged Al content for the AlN/GaN structure studied in [6]. The agreement between experimental data and the data corresponding to the averaged Al content looks acceptable. The propagating phonons are supposed to be insensitive to the strain due to the different signs of strains in GaN and Al\textsubscript{x}Ga\textsubscript{1-x}N layers.

We have also estimated the $A\!{}_{1}(\text{TO})$ phonon position in our multilayered structures using the approach developed in [7, 8]. For the $A\!{}_{1}(\text{TO})$ phonon in the GaN/Al\textsubscript{x}Ga\textsubscript{1-x}N structure this approach gives the dielectric function in the form

$$\left[\frac{1}{\varepsilon(\omega)}\right]^{-1} = \frac{\varepsilon_a(\omega)\varepsilon_b(\omega)(a + b)}{a\varepsilon_b + b\varepsilon_a},$$

where $\varepsilon_a$ and $\varepsilon_b$ are the low-frequency dielectric functions of Al\textsubscript{x}Ga\textsubscript{1-x}N and GaN, respectively. This approach gives two TO-modes; the low-frequency TO-mode is presented in Fig. 2 (triangle). Its position is seen to coincide well with the position of the corresponding mode of bulk Al\textsubscript{x}Ga\textsubscript{1-x}N with the Al content equal to that averaged over the structure $\mathcal{T}$.

3. Conclusion

To summarize, Raman spectroscopic studies of phonon modes behavior in multilayered GaN/Al\textsubscript{x}Ga\textsubscript{1-x}N structures grown by the MBE and MOCVD techniques have been carried out for the first time. It has been found that the observed $A\!{}_{1}(\text{TO})$, $E\!{}_{1}(\text{TO})$, and $E\!{}_{2}$\textsubscript{(high)} phonons can be considered as propagating over the multilayer structure. We interpret the $A\!{}_{1}(\text{TO})$ mode in the multilayered structure as the vibration modified by interaction
between phonon modes in GaN and Al$_x$Ga$_{(1-x)}$N in this structure through a macroscopic electric field.

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References


Symmetry-induced effects on the band structure of wurzite III–V nitride-based quantum wells

Yu. E. Kitaev†, M. F. Kokorev‡ and P. Tronc§
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Radioengineering and Electronics Department, State Electrotechnical University, 197367 St. Petersburg, Russia
§ Laboratoire d’Optique Physique, ESPCI, 10 rue Vauquelin, 75005 Paris, France

Abstract. The symmetry of wurzite III–V nitride-based quantum wells (QW) is shown to be described by the layer group P3m1 (DG 69) and does not depend on the number of atomic monolayers constituting the QW. The modification of bulk electron state symmetries and optical transition selection rules when inserting a QW is established.

Introduction

The III–V nitrides (GaN, AlN, InN) and heterostructures based on these materials have been intensively studying during the last years both theoretically and experimentally [1]. The fast progress in MBE and MOCVD technologies allowed to fabricate heterostructures such as superlattices and quantum wells (QW’s) with layer widths comparable with the GaN lattice constant [2].

Usually, when studying QW’s one speaks about two-dimensional (2D) systems that is not completely correct. Indeed, due to confinement, the carriers are allowed to move only in directions parallel to the layer, i.e. their \( k \)-vectors have only \( k_x \) and \( k_y \) components and the corresponding Brillouin zone (BZ) is 2D. However, the electron wave functions are still 3D. The same is true also for lattice vibrations which can have atomic displacement components along all three directions. Next, when analyzing optical selection rules, the vector representation according to which the electric field is transformed should be taken also 3D. So, the system is still 3D though with 2D periodicity and needs a special symmetry description.

1. Layer symmetry of a single GaN/AlN QW

The insertion of a single GaN QW into the AlN bulk crystal destroys the translational symmetry along the direction perpendicular to the QW layer. (Below we choose this direction to be the sixfold rotational axis of the bulk crystal). The translational symmetry of such a system turns out to be 2D whereas the point symmetry remains 3D though can be different from that in the bulk crystal. The space group symmetry of the system is described by one of the 80 diperiodic groups in three dimensions which are called layer groups [3].

Thus, the Bravais lattice of the system (bulk AlN with a GaN QW) is 2D whereas the unit cell in the \( z \)-direction becomes an infinite column of atoms. However, though the system as a whole acquires layer symmetry, it is obvious from physical reasons, that beyond the QW region all the excitations are still “bulk-like” ones. Therefore, the layer symmetry determines mainly the excitations localized within the QW and adjacent parent crystal layers. Within the framework of symmetry, it is impossible to determine how many Al-N atomic planes adjacent to the QW one should take into account. However, it is possible
to understand how to separate the contribution of the QW spectrum from the bulk one basing on numerical calculations. In the bulk crystal, the number of degrees of freedom of the system (that determines the sets of electron states at each point of the BZ) equals to the number of atoms in the primitive cell multiplied by the number of localized orbitals per atom taken into account. For example, in the GaN bulk crystal there are 4 atoms per primitive cell (2Ga and 2N). If we take into account only \( s \), \( p_x \), \( p_y \), and \( p_z \)-orbitals then the number of degrees of freedom of the system will be 16. In this case, at the \( \Gamma \) point, \( s \)- and \( p_z \)-orbitals induce \( 4\Gamma_1 + 4\Gamma_4 \) states (non-degenerate) whereas \( p_x \) - and \( p_y \)-orbitals induce \( 2\Gamma_3 + 2\Gamma_6 \) states (doubly degenerate) [4]. When introducing a single QW, the number of degrees of freedom becomes infinite. If we begin with the minimal unit cell comprising only atomic planes of the QW and then add one Al-N monolayer from the adjacent layers we increase the number of electron states step-by-step. At last, new states added will not be differed from that of the bulk. Then one can say that the states obtained in the previous steps are originated from the QW.

We have determined that the symmetry of a single GaN QW in wurtzite AlN crystal does not depend on the number \( n \) of GaN monolayers constituing the QW. Its symmetry is described by \( \text{P3m1} \) (DG 69) layer group with the atomic arrangement over the Wyckoff positions given in columns 1–3 of Table 1. However, there is a variation of occupation numbers of Wyckoff positions \( b \) and \( c \) with the same site symmetry which affects the sets of electron and phonon states at the BZ points other than \( \Gamma \). Thus, the insertion of a QW results in lowering the point group symmetry from \( \text{C}_6 \) to \( \text{C}_3 \) since the sixfold screw axis is replaced by the threefold rotation one.

2. Electron state symmetry

The symmetry of electron states in the QW can be obtained by constructing the band representations of its layer group induced by those irreducible representations (irreps) of site symmetry group according to which localized Wannier-type atomic functions transform. The induced representations of a layer group DG can be obtained from the induced representations of the corresponding Fedorov space group \( G \) that can be expressed as a semi-direct product \( G = T_3 \rtimes DG \) where \( T_3 \) is a group of one-dimensional translations [5]. In the table of induced representations of the Fedorov group \( G \) one should take the lines corresponding to the Wyckoff positions of the diperiodic group DG, i.e., the positions having no coordinates expressed in fractions of translations along the \( z \)-axis. The 2D BZ of the DG is a section of the 3D BZ of the corresponding group \( G \). The symmetry of electron states in the GaN/AlN QW obtained by the method of induced band representations [5] is given in Table 1. The irreps of the QW space group describing the symmetry of electron band states are generated by the single and double-valued (marked with a bar over an irrep symbol) irreps \( \beta \) of the groups \( G_q \) of the sites \( q \) where the atoms are located. The localized atomic orbitals of \( s -, p -, \) and \( d \)-types are transformed according to these site-group irreps. The space group irreps are labeled according to [6] and the labeling of site group irreps follows [7]. It is seen that the symmetry types of electron states at the BZ points do not depend on the dimensions of the QW unit cell. However, the number of times each irrep enters into each set of electron states does depend.

The QW can be considered as a limited case of a SL with infinitely large period and thin GaN regions. Thus, when increasing a SL period keeping the GaN region fixed, the SL BZ decreases in the \( k_z \) direction and transforms, at last, into a sheet being a cross-section of the bulk-crystal BZ. As a result, all the states from \( \Gamma-\text{A}, K-H, \) and \( M-L \) symmetry lines will be projected into \( \Gamma, K, \) and \( M \) points, respectively.
Table 1. Electron state symmetries in GaN/AlN QW’s (n is the number of Ga atomic planes in the QW, m = 2s is the number of adjacent Al atomic planes at each side of the QW. N_I and N_{II} are nitrogen atoms in the QW and adjacent layers, respectively).

<table>
<thead>
<tr>
<th>Atomic arrangement</th>
<th>q</th>
<th>β</th>
<th>( \Gamma )</th>
<th>K</th>
<th>M</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{n}{2} ) Ga</td>
<td>( \frac{n}{2} ) Ga</td>
<td>( \frac{n}{2} ) Ga</td>
<td>1b</td>
<td>( a_1(s; p_z) )</td>
<td>1</td>
</tr>
<tr>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{1}{2} ) z</td>
<td>( e(p_x, p_y) )</td>
<td>3</td>
</tr>
<tr>
<td>mAl</td>
<td>mAl</td>
<td>mAl</td>
<td>C_{3v}</td>
<td>( \bar{e}_1^{(1)} )</td>
<td>4</td>
</tr>
<tr>
<td>mNi_{II}</td>
<td>mNi_{II}</td>
<td>mNi_{II}</td>
<td></td>
<td>( \bar{e}_2^{(2)} )</td>
<td>5</td>
</tr>
<tr>
<td>( \frac{n}{2} ) Ga</td>
<td>( \frac{n}{2} ) Ga</td>
<td>( \frac{n}{2} ) Ga</td>
<td>1c</td>
<td>( a_1(s; p_z) )</td>
<td>1</td>
</tr>
<tr>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{n}{2} ) N_I</td>
<td>( \frac{1}{2} ) z</td>
<td>( e(p_x, p_y) )</td>
<td>3</td>
</tr>
<tr>
<td>mAl</td>
<td>mAl</td>
<td>mAl</td>
<td>C_{3v}</td>
<td>( \bar{e}_1^{(1)} )</td>
<td>4</td>
</tr>
<tr>
<td>mNi_{II}</td>
<td>mNi_{II}</td>
<td>mNi_{II}</td>
<td></td>
<td>( \bar{e}_2^{(2)} )</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>( \bar{e}_3 )</td>
<td>6</td>
</tr>
</tbody>
</table>

The correspondence between the bulk and QW states is as follows:

\[ \Gamma_{1,4}(A_{1,4}) \rightarrow \Gamma_1, \quad \Gamma_{5,6}(A_{5,6}) \rightarrow \Gamma_3, \quad \Gamma_{7,8}(A_{7,8}) \rightarrow \Gamma_6, \quad \Gamma_9(A_9) \rightarrow \Gamma_4+\Gamma_5, \]
\[ K_{1,2}(H_{1,2}) \rightarrow K_1, \quad K_3(H_3) \rightarrow K_2+K_3, \quad K_{4,5}(H_{4,5}) \rightarrow K_5, \quad K_6(H_6) \rightarrow K_4+K_6, \]
\[ M_{1,4}(L_{1,4}) \rightarrow M_1, \quad M_{2,3}(L_{2,3}) \rightarrow M_2, \quad M_5(L_5) \rightarrow M_3+M_4. \]

It is seen that the bulk states \( \Gamma_9, A_9, \text{ and } M_5, L_5 \) split into complex-conjugated irreps \( \Gamma_4+\Gamma_5 \) and \( M_5+M_4 \), respectively. These irreps form doubly-degenerate cores and correspond to the same energy. The degeneracy is connected with time inversion and may be lifted when applying a magnetic field along the QW symmetry axis which does not modify the QW symmetry.

On the other hand, the states \( K_2 \) and \( K_3 \) (as well as \( K_4 \) and \( K_6 \)) of the QW though described by complex-conjugated irreps do not form a corep and hence correspond to the states with different energies. This means that doubly-degenerate \( K_3 \) and \( K_6 \) bulk states split in the QW into pairs of states \( K_2+K_3 \) and \( K_4+K_6 \), respectively.

The correspondence between the conduction and valence band levels at the BZ centre of bulk GaN crystal and GaN/AlN QW as well as optical transitions involving these states are shown in Fig. 1. Note that, according to [8], the hierarchy of A,B,C exciton series in thin (less than 25 Å) QW’s is different from that of the bulk. That is, in the QW, the A exciton formed by \( \Gamma_4+\Gamma_5 \) level has higher energy than the B exciton formed by \( \Gamma_6 \) valence band level. It means that the \( \Gamma_4+\Gamma_5 \) level originating from the uppermost valence-band level \( \Gamma_9 \) of the GaN bulk crystal is lower in energy than the \( \Gamma_6 \) level originating from the \( \Gamma_7 \) bulk level.

From Fig. 1, we see that in the QW the A, B and C exciton series obey the same selection rules as in bulk GaN [4]. The only difference is connected with A excitons. Since the A exciton is formed by the \( \Gamma_4+\Gamma_5 \) corep, it splits when applying the magnetic field along the z-axis.
Fig. 1. Energy level diagrams at the BZ centre and allowed optical transitions for the GaN bulk crystal and GaN/AlN QW’s. The allowed polarizations for the case when spin-orbit interaction is taken into account are shown in parentheses.

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Effect of annealing on phase separation in ternary III–N alloys

A. V. Sakharov†, W. V. Lundin†, I. L. Krestnikov†, E. E. Zavarin†, A. S. Usikov†, A. F. Tsatsul’nikov†, N. N. Ledentsov‡, A. Hoffmann‡, D. Bimberg‡ and Zh. I. Alferov†

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany

Major developments in wide band gap III–N semiconductors have led to commercial production of high brightness light-emitting diodes and to demonstration of room temperature laser light emission under pulsed and continuous-wave operation in violet [1], and, recently, in blue spectral region [2]. However, the internal structure of InGaN and AlGaN alloys indispensable for fabrication of high-quality light emitting devices has not been properly investigated. There exist many reports on optical properties of thick ternary alloys and GaN/InGaN and GaN/AlGaN heterostructures. Most of papers are deduced to determination of compositional dependence of band gap of “bulk” AlGaN and InGaN alloys. Values reported for bowing parameters of AlGaN range from $b = 0$ [3] to $b = 1.0$ eV and $b = 1.3$ eV [4, 5]. For InGaN alloys reported range of bowing parameter is much wider: from $b = 1$ eV to $b = 4.8$ eV [6, 7]. Nearly all authors use approximation of strained, but homogenous layer determining In composition from X-ray diffractometry. However, it was shown recently that at least for InGaN ternary alloys compositional fluctuations (phase separation) can play a very important role [8] and must be taken into account calculating In composition from optical transitions energy.

In this paper we report investigation of phase separation in ternary III–N alloys (AlGaN and InGaN) and influence of annealing on optical and structural properties of these samples.

The samples studied in this work were grown by low pressure metalorganic chemical vapor deposition (MOCVD) employing an AlGaN nucleation layer deposited at 530°C on (0001) sapphire substrates. Ammonia, trimethylindium (TMI), trimethylgallium (TMG) and trimethylaluminum (TMA) were applied as component precursors. Purified hydrogen and/or argon [10] were used as carrier gases. Three types of samples were grown: thick (2–3 µm) AlGaN layers grown directly on sapphire substrate (sample A); structures with thin (50 nm) InGaN layer sandwiched between 3 µm GaN buffer layer and 100 nm thick GaN cap layer (sample B); structures with multiple ultrathin (3–5 nm) InGaN insertions in GaN matrix sandwiched between thick GaN buffer layer and 100 nm GaN cap layer (sample C). The details of growth were reported elsewhere [9, 10].

The photoluminescence (PL) study was performed in the temperature range 4–300 K using a continuous wave He-Cd laser (excitation density 25 W/cm²) or a pulsed excimer laser for excitation.

Temperature dependencies of photoluminescence peak position for sample A and the same sample after rapid thermal annealing (RTA) are shown in Fig. 1. Temperature-dependent PL study of as-grown sample revealed so-called “S-shaped” temperature-dependent emission shift. This feature indicates presence of some kind of localization centers in the layer. After RTA at 1100°C for 120 sec this feature practically disappears and the near-band-edge PL peak position shifts to the high-energy side more than 20 meV. This high-energy
Fig. 1. Photoluminescence peak position vs temperature for as grown and annealed AlGaN layer. GaN PL peak position is shown for reference.

Fig. 2. Photoluminescence and stimulated emission (at threshold) peak position vs temperature for InGaN layer. GaN PL peak position is shown for reference.

Shift of near-band-edge emission after RTA was also observed in PL spectra of thin AlGaN layers grown on GaN epilayers. RTA also strongly affects electronic properties of the structure. Room temperature Hall mobility after the RTA increased to $200 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ from $30–40 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ in the as-grown structure indicating efficient decrease in compositional fluctuations for annealed structure.

Figure 2 shows temperature dependence of PL peak for sample B at low excitation density (25 W/cm²) (Fig. 2, solid curve) and energies of stimulated emission at edge geometry (Fig. 2, dashed curve). (For stimulated emission energies were taken at threshold excitation density). It is clearly seen that stimulated emission follows the band gap of GaN, while PL at low excitation density reveals “S-shaped” emission shift. This behavior points to low density of localized states formed by phase separation of InGaN layer.

In our previous work [10] it was shown that growth of multiple ultrathin (3–5 nm) InGaN insertions in GaN matrix (structure C) leads to formation of dense array of nanoislands with high In composition (quantum dots). The density of these localization centers is so high that at low temperatures these quantum dots (QD) can produce gain necessary even for lasing in vertical direction. Temperature dependencies of photoluminescence peak position for as-grown sample C and the same sample after rapid thermal annealing (RTA) are shown in Fig. 3. After RTA at 1300°C for 30 sec QD-related PL peak shifts to low-energy side.
more than 20 meV. This shift is accompanied by increasing width of PL spectra from 100 to 130 meV indicating increase both in maximum depth of localization potential and in its nonuniformity.

In conclusion, we investigated temperature dependence of photoluminescence for different types of structures with ternary III–N alloys. It is shown that optical properties can be strongly affected by composition fluctuations in AlGaN or InGaN alloys. For AlGaN layers rapid thermal annealing leads to effective decrease in nonuniformity, while for In-GaN multilayer structure with dense arrays of quantum dots RTA leads to increase in phase separation.

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References

Light confinement in quantum dots

S. A. Maksimenko†, G. Ya. Slepyan†, N. N. Ledentsov‡§, V. P. Kalosha†, A. Hoffmann§ and D. Bimberg§
† Institute for Nuclear Problems, Belarus State University, Bobruiskaya 11, 220050 Minsk, Belarus
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia
§ Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

Abstract. A concept of light confinement in quantum dots due to diffraction of electromagnetic waves at the dot boundary, is introduced. Possible manifestations of the phenomenon, such as depolarization shift of the exciton frequency, polarization-dependent splitting of the gain band, asymmetry of the absorption and gain spectra, induced magnetization of quantum dots, contribution to radiative lifetime, are discussed both for isolated quantum dots and quantum dot ensembles. We propose that the effect of light confinement should be properly addressed to optimize the design of optoelectronic devices involving quantum dots.

Introduction

A fundamental breakthrough in semiconductor device physics is connected with the recent progress in the synthesis of sheets of nano-scale 3D confined narrow-gap insertions in a host semiconductor, quantum dots (QDs). The large body of recent results on physical properties of QDs and their utilization for the QD laser design has been accumulated in a monograph [1]. The key peculiarity of QDs emerges from the 3D confinement of the charge carriers determined by QD size and shape. However, there exists a class of effects governed by the QD size and shape, which have not received much attention so far. These effects are related to resonant nature of the exciton which provides a dramatic resonant discontinuity of the permittivity at the QD boundary and, consequently, gives rise inhomogeneity of the electromagnetic field both inside and outside QD. By analogy with charge carrier confinement, redistribution of the electromagnetic field energy between the QD interior and exterior under effect of the QD boundary can be referred to as light confinement. In many cases the role of light confinement can properly be accounted for the formation in QD of depolarization electromagnetic field, e.g., in dipole approximation of the diffraction theory.

To our knowledge, some physical consequences of the light confinement in an individual QD first time were considered by Schmitt-Rink et al. [2]. Manifestation of this phenomenon in relation to the scanning near-field optical microscopy was discussed by Martin et al. [3] for geometrically complex mesoscopic systems and by Hanewinkel et al. [4] for QDs. An asymmetry of optical absorption and gain spectra in single QD because of depolarization field has been mentioned in Ref. [4]. Recently it has been predicted and experimentally verified that the light confinement in QD arrays constituted by anisotropically shaped QDs manifests itself as polarization splitting of the gain band [5] and, in more general case, as the fine structure of this band [6]. Such a splitting was first experimentally observed by Gammon et al. [7], where the depolarization field effect has been proposed as possible
explanation of the splitting. Some new effects related to the light confinement in QDs are considered in Ref. [8].

In our paper we introduce sequential concept of light confinement in 3D-confined resonant nanoinsertions and discuss some general consequences of this phenomenon in an isolated QD and in QD arrays. Our consideration is based on classical electrodynamics of inhomogeneous media.

1. Depolarization shift of the exciton resonance

1.1. Polarizability of a single QD

Conventional phenomenological model of the gain in a QD is based on semi-classical theory of two-level systems which gives the equation of motion for the mean polarization $\mathcal{P}$ caused by transitions between the levels:

$$\left(\frac{\partial^2}{\partial t^2} + \frac{2}{\tau} \frac{\partial}{\partial t} + \omega_0^2\right) \mathcal{P} = -\frac{\omega_0}{2\pi \varepsilon_0} \tilde{g}_0 \mathcal{E}. \quad (1)$$

Here $\omega_0$ is the exciton resonant frequency and $\tau$ is the exciton dephasing time in QD. The phenomenological parameter $\tilde{g}_0$ is proportional to the oscillator strength of the transition. In anisotropically shaped QDs this parameter is tensorial owing to anisotropy of the charge carrier confinement [10]. In an inverted medium $(\tilde{g}_0)_{ij} > 0$. The field $\mathcal{E}$ stands for the field inside the QD, different from the external acting field $E$. This difference is determined by the depolarization field which is as follows [9]: $\mathcal{E} = E - 4\pi \hat{N} \mathcal{P}$, with $\hat{N}$ as the depolarization tensor. This tensor is symmetrical and depends only on the QD shape. If we neglect the contribution of the depolarization field putting $\mathcal{E} = E$ into Eq. (1), solution of this equation in the vicinity of resonance for time-periodic fields and isotropic $\tilde{g}_0 = g_0 \hat{I}$ gives the well-known Lorentz contribution to the medium polarizability: $\alpha(\omega) = (g_0/\varepsilon_0)e^{-i/\tau}[\omega - \omega_0 + i/\tau]^{-1}$, which is commonly used as phenomenological model of the dispersion and the gain of a single QD: $\varepsilon_d(\omega) = \varepsilon_0[1 + \alpha(\omega)]$. Otherwise, taking into account the contribution of the depolarization field, we obtain the tensorial polarizability of QD in the vicinity of resonance:

$$\tilde{\alpha}(\omega) = \frac{1}{\varepsilon_0} \left[ \omega \hat{I} - \left( \omega_0 - \frac{i}{\tau} \right) \left( \hat{I} - \frac{1}{\varepsilon_0 \omega_0 \tilde{g}_0 \hat{N}} \right) \right]^{-1} \tilde{g}_0. \quad (2)$$

Thus, the QD’s shape reflects itself as fine structure of the resonance which itself is a superposition of three bands with frequencies $\omega^{(i)}_j = \omega_0 - \nu_j$, $i, j = 1, 2, 3$, where $\nu_j$ are the eigenvalues of the inner tensorial product $\tilde{g}_0 \hat{N}/\varepsilon_0$. For spherical inclusions the tensors $\hat{N}$ and $\tilde{g}_0$ are isotropic and the fine structure manifests itself as a polarization-independent shift of the gain line depicted in Fig. 1. If the energy splittings are much less than the bandwidth, which means the inequality for energy spacings $\max |\Delta \omega_{ij}| = \omega^{(i)}_j - \omega^{(j)}_i \ll 2/\tau$ to be true, the depolarization field will lead to a distortion of the gain band similar to the inhomogeneous broadening. Otherwise, when $|\Delta \omega_{ij}| \sim 2/\tau$, three separate bands will appear in the gain spectrum of a QD array.

1.2. Birefringence in QD arrays

Since the QD linear extension is much smaller than the resonance wavelength, electromagnetic properties of such ensembles – composite materials – can be modeled in the framework
of the effective-medium approach. Thus, a homogeneous medium with effective constitutive parameters instead a composite is being considered. We restrict ourselves to a regular array of QDs arranged in a tetragonal lattice. Assuming QD to have a symmetry axis aligned with the lattice vector \( e_z \), the effective permittivity tensor of the composite can be expressed in terms of a Cartesian basis diadics by

\[
\hat{\varepsilon}_{\text{eff}}(\omega) = \varepsilon_H(\omega)\left(e_x e_x + e_y e_y\right) + \varepsilon_E(\omega)e_z e_z,
\]

where

\[
\varepsilon_\sigma(\omega) = \varepsilon_h + \frac{f_V \alpha_\sigma(\omega)}{1 + f_V \delta_\sigma(\omega)},
\]

and \( \sigma = (E,H) \) refers to light polarized along (E-polarization) or normal (H-polarization) to the \( z \)-axis; \( f_V \) is the volume fraction of QDs. The polarizability components \( \alpha_\sigma \) follow from Eq. (2). The depolarization factors \( N_\sigma \) and the geometrical coefficients \( \delta_\sigma \) (see [5, 6]) correspond to two different mechanisms responsible for modification of the gain in arrays. The first mechanism is related to the light confinement at individual QDs. The second one is a collective effect defined by electromagnetic interaction between QDs in the ensemble. The combined effect of both mechanisms is given by

\[
\omega_N^\sigma = \omega_0 - \frac{g_0^\sigma}{\varepsilon_h}(N_\sigma + f_V \delta_\sigma), \quad \Gamma(\sigma) = \frac{1}{\tau} \left(1 - \frac{g_0^\sigma}{\varepsilon_h \omega_0}\right).
\]

The phenomenological temporal parameter \( \tau \) in these equations is the collective characteristics of the QD array which must be extracted from the experiment.

In the language of crystal optics, the QD composite being considered is effectively a uniaxial dielectric medium with the \( z \) axis as its preferred axis. The phenomenon of birefringence is characteristic for this medium: Both ordinary and extraordinary planewave propagation can occur in it. The refractive indices of these waves, \( n_H \) and \( n_E \), respectively, are given by

\[
n_H = \sqrt{\varepsilon_H}, \quad n_E = \left[\frac{\varepsilon_E \varepsilon_H}{\varepsilon_H + (\varepsilon_E - \varepsilon_H) \cos^2 \theta}\right]^{1/2},
\]

where \( \theta \) is the angle between the \( z \) axis and the propagation direction. Eq. (5) shows that \( n_H = n_E \) when the propagation direction coincides with the \( z \) axis (\( \theta = 0 \)), and \( n_E = \sqrt{\varepsilon_E} \) when the propagation occurs in the x0y plane (\( \theta = \pi/2 \)). Distinction between \( n_H \) and \( n_E \) in this geometry is responsible for the polarization splitting of the gain band described in details in Refs. [5, 6].
2. Radiative lifetime of spherical QD

Let an isolated spherical QD of the radius $R$ be exposed to an external time-harmonic electromagnetic field. Well-known exact solution of the diffraction problem for a sphere is essentially simplified [11] in view of the condition $kR\sqrt{\epsilon_h} \ll 1$, which is valid for any realistic QDs. This solution and presents the field outside the sphere in terms of its electric and magnetic polarizabilities:

$$\alpha_e(\omega) = \frac{3[\epsilon_d(\omega)F(\kappa) - \epsilon_h]}{[\epsilon_d(\omega)F(\kappa) + 2\epsilon_h][1 - i kR\sqrt{\epsilon_h}] + i(kR)^2 \epsilon_h^2 F(\kappa)}$$  \hspace{1cm} (6)

$$\alpha_m(\omega) = \frac{3[F(\kappa) - 1]}{[F(\kappa) + 2][1 - i kR] + i(kR)^2 F(\kappa)}.$$  \hspace{1cm} (7)

Here $\kappa = kR\sqrt{\epsilon_d(\omega)}$, $F(\kappa) = (\sin \kappa - \kappa \cos \kappa)/[(\kappa^2 - 1) \sin \kappa + \kappa \cos \kappa]$; $F(\kappa) = 1$ in dipole approximation. In QWs, the problem of the radiative lifetime evaluation is solved by finding of frequency poles of the reflection and transmission coefficients for TE- and TM-polarized plane waves (see, e.g., [12]). Real parts of these poles determine resonant frequencies while imaginary parts give the homogeneous linewidths, which are sums of the dephasing broadening and the radiative broadening. For QDs, we must evaluate the poles of the electric and magnetic polarizabilities. In dipole approximation, simple manipulations lead to

$$\tau_{rad}^{QD} \simeq -\frac{9}{4\pi^2 g_0} \left(\frac{\lambda}{R}\right)^2.$$  \hspace{1cm} (8)

We note that the material gain $g_0$ in QD is incorporated as phenomenological parameter in this equation. It is, indeed, a function of the QD size, shape, strain distribution and an effective coefficient of light confinement in a QD. Depending on particular situation $g_0$ can either increase or decrease with $R$. In the case when the light confinement is not relevant and the overlap integral is not a function of QD size (the case which is shown to be not correct for real QDs) $g_0 \sim R^{-3}$. In any case, Eq. (8) shows additional radial dependence of the radiative lifetime as compared to the conventional dependencies (see, e.g., [13, 1, 14]). Figure 2 presents radiative lifetime numbers obtained from different theoretical approaches and experiments. Conventional model including the realistic overlap integral [14] is shown in Fig. 2 as open squares while this model with unit electron and hole wavefunction overlap integral [13] is presenter by dashed line. The experimental results for radiative lifetimes

**Fig. 2.** Radiative lifetime of an isolated QD as a function of the photon energy
in InAs QDs having different size (pyramid base length between 10 and 20 nm) derived at low temperatures using both resonant and non-resonant excitation is shown in the figure by solid squares. One can see that these results agree only in the case of smaller QDs, where the role of light confinement on radiative lifetime seems to be relatively weak. As opposite, as the QD size increases, the theoretical dependence and experimental values differ qualitatively. As the structural quality and luminescence efficiency of larger QDs remain high allowing high-efficiency high-power device applications [15], the reason for such a discrepancy can be only related to the discussed light confinement effect at QD.

3. Conclusion

We have introduced a concept of light confinement and investigated its role in electromagnetic response of QDs. We calculated significant diffraction-induced shift of the main QD exciton line. Evaluation of the radiative lifetime for spherical QD and its correlation to the QW radiative lifetime shows the origin of the fascinating light-amplifying properties of QDs as compared to QWs and creates a basis for solving of a large number of electrodynamic problems of QDs and QD ensembles. We show that the experimentally measured radiative lifetime qualitatively disagrees with theoretical predictions arising from models neglecting the light confinement effect at QD. Thus one needs to consider redistribution of the electromagnetic wave caused by QD to reach optimized device geometry. This is particularly true for a QD inserted in a microcavity, where cavity modes may interfere with intrinsic photon modes of a single QD. In our paper we mainly restricted ourselves to the spherical model of QD. Different QD configurations like disks or pyramids can be investigated using direct computation on the basis of the well-developed method of classical electrodynamics.

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References

Thin film photonic crystals

Clivia M. Sotomayor Torres†, Torsten Maka‡, Manfred Müller‡, Rudolf Zentel‡ and Sergei G. Romanov†§
† Institute of Materials Science and Dept. of Electrical Engineering, University of Wuppertal, Gauss-str. 20, 42097 Wuppertal, Germany
‡ Institute of Materials Science and Dept. Chemistry, University of Wuppertal, Gauss-str. 20, 42097 Wuppertal, Germany
§ Ioffe Physico-Technical Institute, St Petersburg, Russia

Introduction

The fabrication of 3-dimensional photonic bandgap (PBG) based light moulding and emitting devices requires (i) highly crystalline periodical structures, (ii) emitters incorporated in a PBG environment and (iii) emission band tuned to the optical stop-band of the photonic crystal. The combination of the physically separated (a) light source with a high emission yield and an emission band narrower than the PBG bandwidth and (b) photonic crystal with the refractive index contrast (RIC) large enough to open an omnidirectional bandgap is possible to approach using 3D photonic templates, particularly, opals filled with a light emitting material [1]. We have extended this approach by preparing opal-like structures from polymers. Due to the low RI and the incomplete PBG in polymer-made structures, polymeric photonic crystals possess strongly anisotropic optical properties. From the other hand, crystalline quality packages of polymeric balls can be processed further to prepare semiconductor replicas of a higher refractive index contrast (RIC) and the optimised filling factor. Organic dyes embedded in polymers are known as effective luminescent materials [2–4]. Moreover, they offer excellent flexibility to match the stop-band with the emission band of the dye without raising dramatically the imaginary part of the dielectric function. We discuss here the optical properties of several polymeric photonic materials and their replicas. Taken the anisotropy into account, angular resolved spectroscopy [5–6] has been used to find the PBG effect in a particular direction of the photonic crystal and to study the change of the spontaneous emission.

1. Opal-like thin film crystals

PMMA balls for polymer photonic crystals were prepared by a modified emulsion polymerisation from the monomer-solution. In some cases a small amount of fluorescent dye (Coumarin 6) has been added to the solution to study the PBG effect upon emission from photonic crystal. The ball diameter was controlled by varying the polymerisation time. Particles of larger size were separated by filtration and centrifugation. Films of several cm² area were deposited on hydrophilized microscope slides. Balls are self-assembled in the face-centered cubic fcc package with (111) plane along the substrate. These thin film polymeric crystals consist of ca. 30–50 monolayers and possess domains extending over hundreds of micrometers. Dye molecules are distributed homogeneously in the balls and their concentration can be varied in a controlled manner. The resulting crystal offers the advantage of containing light emitting molecules inside the photonic structure while preserving the RIC of the host polymer. In Fig. 1 an SEM micrograph of the initial photonic structure is shown. The monodispersity of the spheres is less than 10% but shows smaller
variations on short length scales. The vapour phase chemical deposition was applied to fill the polymeric opal-like film with SnS$_2$. Polymeric balls were dissolved after completing the impregnation thus leaving the semiconductor replica film on the glass substrate (Fig. 2). The filling factor of the PMMA-air structure $f_{\text{ball}} = 0.74$ is far from being optimal and the RIC is about 1.5 to 1. In the inverse structure the fraction of high RI material $1 - f_{\text{ball}}$ is closer to the optimum value, whereas the RIC depends on the actual density of a semiconductor and varies from 1.4–1.9 to 1.

2. Diffraction

The transmission and reflectance spectra were measured by illuminating the sample with white light. The angular dispersion of the Bragg diffracted light was studied by measuring reflectance spectra at different angles $\theta$ between the (111) axis of fcc crystal and the beam axis by collecting the scattered light within a solid angle of $2^\circ$. Changes in the light diffraction have been quantified using the Bragg law $\lambda_{(111)} = 2n_{\text{eff}} \cdot 0.81D$, with $n_{\text{eff}}$ being the effective RI of the composite and $D$ the diameter of opal balls. The $n_{\text{eff}}$ was determined using the effective medium approximation $n_{\text{eff}} = n_1 f_{\text{ball}} + n_2 (1 - f_{\text{ball}})$, $n_i$ is RIs of crystal components [7, 8]. It is known, that the angular dispersion of diffraction peaks mimics the dispersion of the stop-band in the $E - k$ space [9]. Ball diameters extracted from the Bragg diffraction are in good agreement with SEM data, if the RI for PMMA $n = 1.4893$ applies [10].

Both changes of the effective RI and the filling factor of the high-RI material ($f_{\text{IR}}$) lead to a shift of the Bragg resonance and the increase of the width of the photonic bandgap in semiconductor replica as compared with polymeric template. A comparison of angle-resolved reflectance spectra is shown in Fig. 3. The reflectance of the polymeric opal shows a relative stop-bandwidth of $\Delta E/E_0 \approx 5\%$. The “blue” shift of the Bragg resonance in the replica due to the decrease of the filling factor is balanced by the “red” shift due to the increase of the RIC. Simultaneously, both factors serve to increase the photonic bandwidth to $\Delta\lambda/\lambda_0 = 13\%$. It is worth mentioning that the distortion of the lattice, which is bigger in replica, also contributes to the broadening of the Bragg peaks.

The rate of the angular dispersion of the stop-band is governed by the effective RI. Correspondingly, it is stronger for replica, but decreases with the increase of the density of SnS$_2$. However, the total improvement of the PBG in the replica as compared with the polymeric template is the result of the much wider stop-band, that allows to overlap them.
Fig. 3. Angle resolved reflectance spectra of PMMA opal film (top) and inverted SnS$_2$ opal film (bottom). Spectra for angles 5, 20, 30, 40 and 50 degrees from left to right.

Fig. 4. Relative dip in density of photonic states obtained from PL measurements.

Fig. 5. Directionality diagram of the emission from polymer-dye reference sample.

Fig. 6. Directionality diagram of the emission from dye-polymer-opal film.

more effectively for different angles. Expressing this overlap in terms of the ratio of the stop-bandwidth $\Delta E$ to the shift of the resonance frequency $E_{\text{shift}}$ as $\delta = \Delta E / E_{\text{shift}}$, the $\delta$ factor improves from 0.5 to 1.3.

3. Photoluminescence

Bragg configuration was used to study the photoluminescence (PL) as the angle was varied between $\theta = 0^\circ$ and $\theta = 50^\circ$ the PL was collected within a 6° solid angle. PBG reduces the probability of the radiative recombination within the stop-band and, consequently, the PL spectrum shows the dip, which spectral position depends on the angle of the emission detection. To demonstrate the relative changes in the density of photonic states in the PBG region PL spectra are shown normalised to the PL spectrum on unstructured Coumarin-PMMA film (Fig. 4).

The anisotropy of the photonic band structure leads to the self-focusing of the emission (Fig. 5). The fingerprints of the PBG are reproduced at different angles for different
frequencies as a dip in the directionality diagram. In contrast the similar diagram of the dye-polymer film shows no obvious wavelength dependence (Fig. 6). The similar effect was mention previously for dye-polymer loaded opals [11]. By analogy with the superprism effect [12], the emission self-focusing in the incomplete photonic crystal is related to the topology of dispersion planes for electromagnetic waves with the frequency near the stop-band, because the vector of the group velocity of propagating wave is always normal to the dispersion plane. Accordingly, the focusing rate depends on the energy.

4. Conclusions

Two photonic crystals have been prepared based on polymer opal-like films. Both polymer and semiconductor structures demonstrate similar photonic behaviour. The width and dispersion of the stop-band has been greatly improved by decreasing the filling factor of a high RI component and increasing the RIC. An antenna like effect — the focusing of emission from incomplete photonic crystals — has been observed and explained as the result of specific dispersion of the electromagnetic waves in the frequency range of PBG.

References

Enhancement and extraction of spontaneous emission from 2-d thin film photonic crystals

Misha Boroditsky†, Rutger Vrijen‡, Thomas F. Krauss§, Roberto Coccioli‡, Raj Bhat¶ and Eli Yablonovitch‡
† AT&T Labs, Red Bank, NJ 07701, USA
‡ Electrical Engineering Dept., UCLA, Los Angeles, CA 90095-1594
§ Electrical Engineering Dept., Glasgow University, Glasgow G128LT U.K.
¶ Corning Inc., Corning NY 14831 USA

Abstract. The results of photoluminescence measurements on thin slab InGaAs/InP photonic crystals are presented, demonstrating a possibility of spontaneous emission engineering at room temperature using 2-d periodic thin film photonic crystals. The angle dependence of the PL spectral peaks is shown to track the photonic band dispersion. This revealed the band structure of the leaky conduction bands within the optical escape cone. Up to a 15-fold emission intensity enhancement was observed and explained in terms of a combination of Purcell enhancement, and Bragg extraction of high wave vector photons. Different design concepts for improving LED performance were demonstrated.

Introduction

Photonic crystals, artificially created, multi-dimensionally periodic structures are known for a forbidden electromagnetic bandgap. For that reason, they can be used to modify spontaneous emission. Initially, it was proposed to use photonic crystals to inhibit spontaneous emission [1], but they can be employed to enhance it as well, with significant implications for light-emitting diode structures. We show that thin slab 2-d photonic crystals can provide a spontaneous emission enhancement up to $F_p = 2.5$ and an overall extraction enhancement up of 15 times.

Spontaneous emission from photonic crystals

We used a thin-film InGaAs/InP 2-d photonic crystal at ambient temperature, but the results would apply equally to InGaN thin films for example. An MOCVD-grown In$_{0.47}$Ga$_{0.53}$As/InP single quantum well double hetero-structure was used for these experiments. Thin films for the photo-pumped LED’s were fabricated using substrate removal, and bonded to a glass slide. A triangular array of holes is defined by electron-beam lithography, using a LEICA EBPG-5 Beamwriter. The semiconductor slab was etched through by reactive ion etching (RIE) using SiCl$_4$ at the elevated temperature of 200°C. The InP film thickness is 240 nm and the InGaAs quantum well active region thickness is 60 nm. Each sample had numerous triangular lattice structures spanning a photonic lattice constant range sufficient for optimization of the external efficiency. In our case of emission wavelength centered at vacuum wavelength $\lambda = 1650$ nm, the photonic crystal’s lattice constant was made to vary from $a = 550$ nm to $a = 910$ nm. Correspondingly, the center of the photonic band gap varied from $\lambda = 1300$ nm to $\lambda = 1900$ nm. The thin-film LED photonic crystal structures are shown in Fig. 1.
Fig. 1. (a) A triangular array of holes in the thin film on InGaAs/InP double hetero-structure; (b) Prospective view.

The LED emission was collected in a solid angle from normal up to 45° angle away from normal, in air. The calibrated photoluminescence signal versus photonic lattice spacing is shown in Fig. 2. As can be seen from the graph, the efficiency is optimized at a photonic lattice constant $a = 900$ nm, where conduction band modes match the InGaAs emission frequency. That gives the external efficiency for that LED structure 48%.

The angular resolved spontaneous emission allows for measurements of the dispersion diagram of a photonic crystal’s leaky conduction band modes, that is modes with frequencies lying above the light cone in the glass substrate. We used the evolution of spontaneous emission spectral peaks versus angle to study the band structure of the photonic crystal. The dispersion diagram (solid lines in Fig. 3) is computed using the Finite Difference in Time Domain technique. Angular resolved spectra on thin film photonic crystals reveal some very sharp peaks in the spectrum compared to the reference emission linewidth of InGaAs. These are signatures of a new type of the Purcell enhancement [2], that can be realized in the spatially extended photonic bands, without a cavity [4].

Leaky conduction band modes bring a two-fold advantage to LED’s. First, by using them, we increase dramatically the light extraction efficiency, bringing it close to 100%. Second, we can speed up the radiative recombination to make it more competitive with the non-radiative recombination on exposed surfaces. Increase in recombination rates drives faster device operation as well.

Photonic crystals as passive outcouplers

Leaky photonic crystal modes can also be used as a passive out-coupling mechanism. These leaky modes are in the shaded area of Fig. 3 and have measured $Q$ between 30 and 100. The periodic structure is in effect an efficient, coherent scatterer of light from the semiconductor film.

In other words, the proposed strategy is to separate the regions where the light is generated from those where the light is extracted. The two respective regions are the hexagonal area of unpatterned thin film for light generation, surrounded by a few periods of the pho-
Fig. 2. Photoluminescence efficiency calibrated with respect to the reference sample, and corrected for the fractional sample absorption.

Fig. 3. Theoretical (lines) and experimental (circles) bands for the thin slab photonic crystal. The modes in the shaded area are leaky.

tonic crystal for light extraction. If the light is generated in the center region, a small $1/2n^2$ fraction corresponding to top and bottom escape cones is emitted directly from the central part of the unpatterned hexagonal area, and the rest is trapped in the thin film waveguide. When the guided light reaches the surrounding patterned region, it scatters or reflects at the interface, or couples to the leaky modes of the photonic crystal, and then scatters into the air or into the glass substrate.

In these experiments, we measured spontaneous emission from a 20 µm diameter unpatterned optically pumped region surrounded by a few rows of photonic crystal. The lattice spacing, $a$, of the photonic crystal was 600, 760 or 900 nm. According to our band structure calculations, in the $a = 600$ nm sample, only guided TM modes overlap with the emission band. For the $a = 760$ nm sample the spontaneous emission band overlaps with both guided TM and leaking conduction band modes of the photonic crystal. For the $a = 900$ nm sample all spontaneous emission should couple to the leaky modes, and thus to free space.

The photoluminescence acceptance angle was 0 to 45°C in the air. The spectrum of
the $a = 600$ nm sample resembles that of an unetched thin film and has almost the same intensity. Indeed, even though the guided waves can couple to the guided TM modes inside the photonic crystal, there is no way for the light to escape. The integrated PL signal from the sample with $a = 760$ nm is about 4 times larger. The shape of the spectrum is also different, there are two distinct shoulders. Finally, the sample with the $a = 900$ nm hole spacing showed even higher overall efficiency, 6.25 times the efficiency of the unpatterned reference sample, which translates into more than 70% external quantum efficiency [3].

**Summary**

The results of photoluminescence measurements on thin slab InGaAs/InP photonic crystals were presented in this paper, demonstrating a possibility of spontaneous emission engineering at room temperature using 2-d periodic thin film photonic crystals. The angle dependence of the PL spectral peaks was shown to track the photonic band dispersion. This revealed the band structure of the leaky conduction bands within the optical escape cone. Up to a 15-fold emission intensity enhancement was observed and explained in terms of a combination of Purcell enhancement, and Bragg extraction of high wave vector photons. The Purcell enhancement factor was probably no more than 2 under our conditions, with most of the efficiency increase associated with Bragg extraction improvement. Different design concepts for improving LED performance were demonstrated.

**References**


Apparent microcavity effect in the near-field photoluminescence of a single quantum dot

A. M. Mintairov†‡, O. V. Kovalenkov†, J. L. Merz‡, S. V. Osinski‡, J. P. Reynolds‡, I. S. Tarasov†, D. A. Vinokurov† and A. S. Vlasov†

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Dept. of Electrical Engineering, University of Notre Dame, Notre Dame, IN 46556, USA

Abstract. We present low-temperature near-field scanning optical microscopy (NSOM) measurements of self-organized InP quantum dots (QD) embedded in a $3\lambda/2$ GaInP layer. We observed an anomalously strong increase of the emission energy (~270 meV) of a single QD by changing the tip-to-surface distance over ~100 nm in the near-field region. The effect indicates formation of a near-field microcavity having extremely high photon-exciton mixing.

Introduction

Near-field optical scanning microscopy (NSOM) allows one to extend spatial resolution of optical experiments far beyond the light diffraction limit. The method opens new possibilities to study photon-matter interaction and allows the manipulation of the non-radiative components of electromagnetic waves. In the present paper we report an apparent microcavity behavior of the NSOM spectra of a $3\lambda/2$ GaInP layer containing InP QDs. The near-field microcavity effect was manifested by anomalously strong dependence of the emission energy of a single QD on the tip-to-surface distance.

1. Experiment

InP QDs were grown at 700°C by low pressure MOVPE on exactly oriented GaAs (100) substrates. Growth was started with the deposition of a 350 nm-thick Ga$_{0.5}$In$_{0.5}$P lower barrier followed by nominally 3 monolayers of InP island deposition. After 5 s growth interruption the islands were overgrown by a 35 nm-thick upper Ga$_{0.5}$In$_{0.5}$P. The plan-view transmission electron microscopy measurements (see Fig. 1(a)) clearly reveal strain-induced contrast characteristic of QDs and point to a typical dot base ~80 nm and density ~$2 \times 10^9$ cm$^{-2}$. The thickness of the GaInP layer was equal ~$3\lambda/2$ for the QD emission wavelength ~730 nm.

The low-temperature near-field scanning optical microscope (NSOM) was built using a cryogenic positioning system CryoSXM by Topac, Inc. The NSOM photo-luminescence (PL) spectra were taken in collection-illumination mode at 10 K under excitation of 514.5 nm line with a power 5 μW. We use tapered fiber tips with a diameter ~0.25 μm and with a thickness of the Al coating of 20 (tips of type I) and 60 nm (tips of type II).

2. Results and discussion

2.1. Spatially resolved near-field PL spectra of InP QDs

Using the tips of type I we measured the PL of our structure in a standard NSOM regime (Fig. 1(b) and (c)). In this regime the intensity of the emission spectra gradually increases
Fig. 1. (a) (upper left) plan-view TEM image of InP QDs; (b) (upper right) spatial evolution of the low-temperature near-field PL spectra InP QDs. Arrows show tip position for spectra presented in (c); (c) (under) low-temperature near-field PL spectra for the tip located on individual InP QDs. Insert shows spectra taken for two different distances from the surface.

when the tip-to-sample surface distance (z) decreases until the tip reaches the sample surface. No change in the energy position of the QDs emission lines are observed in this case (see insert in Fig. 1(c)).

Figure 1(b) shows spatially and spectrally resolved near-field PL intensity obtained in this regime, allowing the resolution of individual QDs (Dot1-Dot7, Dot1e-Dot4e). The QD density estimated from these data is \(\sim 2 \times 10^9 \text{ cm}^{-2}\), which agrees well with TEM data (Fig. 1(a)). In Fig. 1(c) we present the near-field emission spectra taken in lateral positions in which the tip was located on Dot2, Dot3 and Dot4. We can see that the typical emission spectra of a single InP QD consists of a triplet multiexcitonic manifold [1, 2] having energy 1.69–1.76 eV. It has an energy splitting of 1–6 meV and a halfwidth (\(\gamma\)) of the components, conditionally denoted as A, B, C, of 0.2–5 meV. We attribute the difference in the energy splitting and the line halfwidth to the charging of the QDs due to the presence of an impurity.
inside the dot or in the nearby region.

In addition to the lines related to the spatially selected (central) QD, the spectra contain an emission band from the GaInP matrix at \( \gamma = 1.97 \text{ eV} \) (\( \gamma = 15 \text{ meV} \)), ultranarrow lines (\( \gamma \leq 0.2 \text{ meV} \)) from a wetting layer (WL) centered at 1.95 eV, and very weak multiexcitonic manifolds from the QDs located close to the tip edge (Dot1e, Dot2e, and Dot3e).

2.2. Strong shift of the QD emission line in the near-field

For the tips of type II the standard NSOM regime described before takes place for our samples only for \( z \) values higher than a certain critical value of \( z_0 \sim 100 \text{ nm} \). For the lower tip-to-surface distances the energy of the multiexciton manifold of the central QD strongly increases as \( z \) decreases.

The PL spectra taken at different tip vertical positions below \( z_0 \) are presented in Fig. 2. We can see that line A, related to the main emission component of the central QD for \( z > z_0 \), shifts approximately 270 meV in the range \( z = 7 \text{ nm} \) to \( z_0 \) and when the tip approaches the surface it has energy higher than that of GaInP matrix. In this detuning region the manifold components broaden and their shape changes. Simultaneously the intensity is decreased as \( z \) goes to 0. From spectra in Fig. 2 we can see that much smaller energy shifts (1–5 meV) take place for the weak emission lines QD1e, QD2e, WL and GaInP. For the QD1e, QD2e and WL lines the energy increases with \( z \) decreasing, while for the GaInP line the energy decreases.

**Fig. 2.** Evolution of the low-temperature near-field PL spectra of InP QD under approach of the type II tip to the surface. Insert shows the shift of the emission lines as a function of tip-to-surface position.
2.3. Discussion

The observed behavior of the central QD emission for $z < z_0$ is similar to Fabry–Perot detuning in planar microcavities [3, 4]. As the detuning effect is absent for tips of type I, we can suppose that in our experiments the microcavity is formed by the GaAs/GaInP interface, $3\lambda/2$ GaInP layer and an “aperture” created by the subwavelength ring of the Al coating of the NSOM tip. The strongly shifted emission line thus can be interpreted as a resonant photon mode of this cavity. However, as a QD has a discrete energy spectrum, the continuous detuning of the cavity emission in the range where no spontaneous photons are emitted (Fig. 1(c)) suggests a strong coupling of the central QD exciton with a resonant cavity photon [5]. The resonant cavity photon also couples with the excitons localized in the edge QDs, WL and GaInP which is evident from the energy shifts of the corresponding lines, but here the coupling is much smaller. In NSOM emission spectra we did not observe any evidence of the Rabi splitting for the central QD, which may be due to inhibiting of the emission of the lower polariton branch, or even due to quite a different mechanism of QD emission detuning. We expect to detect the Rabi splitting in NSOM reflectivity experiments which are now in progress.

In conclusion our experiments reveal an apparent microcavity effect in the near-field spectra of a single QD embedded in $3\lambda/2$ layer. The strong detuning of the QD emission energy observed opens new possibilities for photonic device engineering.

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References

Two-dimensional photonic crystal fabrication using fullerene films

Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The two-dimensional rectangular network has been fabricated firstly on the base of fullerene films using electron lithography. Such system shows the optical properties of the two-dimensional photonic crystal. The formed periodical structure may be transmitted easy into the semiconductor substrate by plasma etching due to the resistive properties of solid state fullerenes.

The aim of the work was the development of new electron lithography method of photon crystal structures fabrication by using fullerene films. Fullerenes are a perspective material for this purpose due to the possibility of their easy structure modification and therefore simple technological treatment. Moreover this particularity is typical for a lot of fullerene based composite materials possessing setting optical properties.

Initial objects to fabricate investigating structures are the films from fullerene $C_{60}$. Fullerene films were deposited on GaAs (100) substrates by vacuum thermal sublimation of 99.9% pure $C_{60}$-powder from a Knudsen-cell at 350°C. During the deposition at a rate of 0.2 nm/s the substrates were held at room temperature under residual pressure $10^{-5}$ Torr. The thickness of the films was varied from 300 to 500 nm.

For modeling the photonic crystal geometry we fabricated the regular $C_{60}$ structures in the shape of rectangular network with the period 300–600 nm (see Fig. 1(b) with the best lattice). The total network area with uniform parameters exceeded 1 mm$^2$ that was suitable for the optical measurements. An electron lithography technique was employed to prepare the nanostructures using scanning electron microscope CamScan Series 4-88 DV100 with the acceleration voltage $U = 10–40$ kV and beam current $I = 10^{-10}–10^{-8}$ A. The external scan control was realized with industrial two-channel 14 bit digital-analog converter card. The scan step was less than 50 nm. The time of single dot exposure did not exceed 5 ms that corresponded to the reasonable exposure time for the entire sample. In the case of $C_{60}$-films the main contribution to the polymerization process was made by primary electrons of the beam and also by the back-scattered electrons (BSE) from GaAs substrate. A proximity effect led to the depending of the irradiation dose on structure geometry, and scan step. To take into account the contribution to the total irradiation dose from neighboring dot the BSE generation function for GaAs [1] was used. It was shown that in the case of lined structure one should use almost twice shorter exposure than for the array of dots. To get the proper shape for the crossed lines exposure time should be varied with distance to the crossing point according the generation function. The strong structure performance dependence on beam energy density was observed. At accelerating voltage higher than 30 kV one should use the beam current higher than 5 nA because the most part of beam energy dissipated into the substrate. So high density of the beam energy in the incident point led to $C_{60}$ destruction and formation carbon clusters which lateral dimension is close to the period of the structure (see Fig. 1(a) with the worst lattice). The part of beam energy...
Fig. 1. Fullerene based networks made on GaAs substrate at two different regimes of electron exposure: (a) $U = 30$ kV, $I = 6$ nA; (b) $U = 15$ kV, $I = 1$ nA.

Fig. 2. The luminescence spectrum of $C_{60}$ films at $T = 2$ K: (a) initial, (b) after electron beam exposure. Excitation wavelength is 441.6 nm.

dissipated in the fullerene film rose with accelerating voltage decreasing. The lithography performance was improved by using $U = 15–20$ kV.

The process of fullerene film electron modification was investigated and controlled with help of spectroscopic measurements. The studies of photoluminescence $C_{60}$ film have shown the luminescence line shape modification depending on the character of interaction between electron excitation in solid state fullerene and their phonon system and also depending on features of exciton–polaron recombination [2] (see Fig. 2). Moreover it has observed the significant decreasing of luminescence intensity of fullerene film after electron exposure. This fact is due to arising number of polymerizing fullerene molecule complexes playing a part of non-radiate recombination centers in fullerene film.

The polymerized parts of film are characterized by very low solubility as compared with initial film. The exposed films were developed with toluene during 5–60 s. The developing procedure followed immediately after the exposure to avoid depolymerization effect as well as to prevent sample from environmental impact. If the irradiation dose was
Fig. 3. Reciprocal reflection spectrum of fullerene based network on GaAs substrate normalized by reflection of film initial for fabrication of network.

enough for complete polymerization, irradiated part of the film remains on the substrate after developing while the regions with pristine fullerene were completely dissolved in toluene.

The regular structures with line height up to 500 nm fabricated by this means possess optical properties of two-dimensional photonic crystal that have been verified by reflection studies. It is significant for opto-electronic applications that the reflectivity of structures may be changed by many times with help of similar regular coating. This fact is demonstrated in Fig. 3.

The formed regular fullerene based structure may be easy transmitted into semiconductor substrate by ion etching due to high aspect ratio of polymerized fullerenes as a resist for plasma etching. The ion beam etching process was carried out in a specialized Rocappa machine. This is a standard ion beam milling system with 5 cm filamentless ion beam source (Anatech Ltd). The chamber at the milling machine pumped down to low vacuum by oil free Drytec system and turbomolecular pump (less than $2 \times 10^{-7}$ mbar). The flexible mechanical design of the gun holder allows us to tilt the substrate with respect to the beam axis. Total gas pressure (Ar) in the gun is adjusted to $(3.0-4.0) \times 10^{-4}$ mbar, and the ion beam acceleration voltage 300 V, current density 0.5 mA/cm$^2$ and sample temperature 25°C.

With help of described method the GaAs substrate was etched through regular fullerene based mask on depth up to 400 nm. This simple method allows us to fabricate the semiconductor photon type structure with required geometry. The example of structure obtained in such a way is shown in Fig. 4.

Thus, the great potential possibility to fabricate the photonic structures using fullerene films is demonstrated in this work.

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Fig. 4. The images of submicron regular network based on: (a) fullerenes, (b) made by ion etching of GaAs substrate trough fullerene mask.

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References


Two-dimensional Penrose-tiled photonic quasicrystals: is there a pure photonic band gap?

R. A. Abram†, S. Brand†, M. A. Kaliteevski‡, T. F. Krauss§, R. DeLa Rue§ and P. Millar‡

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Department of Physics, University of Durham, South Road, Durham, DH1 3LE, UK
§ Department of Electronics and Electrical Engineering, Glasgow University, Glasgow, G12 8LT, Scotland

Abstract. We report measurements of the diffraction pattern of a two-dimensional photonic quasicrystal structure and use the set of plane-waves defined by the diffraction pattern as the basis of a theoretical approach to calculate the photonic bandstructure of the system. An important feature of the model is that it retains the essence of the rotational and inflational properties of the quasicrystal at all levels of approximation; properties lost in approximate models which artificially introduce elements of periodicity. The calculated density of modes of the quasicrystals is found to display a weakly depleted region analogous to the band gap that occurs in a periodic system.

Introduction

The properties of quasicrystals [1, 2] have become of considerable interest since Shechtman et al. [3] observed the long-range aperiodic arrangement of atoms in AlMn alloys. The concept of quasicrystals can be extended to other physical systems such as dielectric structures with feature size at the length scale of a micrometer or less, which are often called photonic microstructures. Photonic quasicrystals are of interest because a pseudogap, or near vanishing minimum in the density of all modes, might be more readily achieved for all propagation directions in such structures as a result of their high degree of rotational “symmetry”. Indeed, a “sizeable spectral gap” has been predicted theoretically for a two-dimensional photonic structure defined by the periodic repetition of a “supercell” with local octagonal quasisymmetry [4]. Photonic microstructures can provide a number of advantages in the investigation of the physical properties of quasicrystal-like system because the structure can be designed to study particular phenomena and can be investigated by well developed optical techniques.

Here we report a study of the optical diffraction properties, optical eigenmode spectrum.

The structure considered is essentially a two-dimensional Penrose-tiled [1] dielectric slab, where the tiles are two kinds of rhombus: a thin tile (with vertex angles of 36° and 144°) and a fat tile (72° and 108°) as illustrated in Fig. 1(a).

The experimental and model structures are formed by air cylinders, which are positioned at the vertices of the tiles. The experimental sample (quartz substrate with 121 air cylinders, shown in Fig. 2(b)) was fabricated using electron-beam lithography. The tile side \( d = 10 \mu m \) and the hole diameter is about 3 \( \mu m \), the depth of the cylinders is about 700 nm.
1. Diffraction pattern

A He–Ne laser with a gaussian shaped beam and wavelength 633 nm was collimated to provide a beam diameter of about 130 \( \mu \text{m} \), matching the size of the patterned area. The laser beam was incident normal to the sample, thus leading to the appearance of the diffraction pattern on the screen, placed 24 cm behind the sample as shown in Fig. 2(a). Note that although the experiment is carried out on a finite system with only 121 cylinders the pattern is remarkably similar to that reported for essentially infinite atomic quasicrystals [1].

The pattern possesses 10-fold rotational symmetry, and contains a series of spots of different intensity, surrounding the central undiffracted beam. These spots can be associated with vectors in the reciprocal space which we will call “reciprocal vectors” (RVs). Contrary to the case of a periodic crystal, the indexing of the diffraction pattern of aperiodic quasicrystals is not a trivial task, due to the self-similarity of the structure. The RVs of a periodic structure form the periodic reciprocal lattice and we can always find a set of primitive RVs of minimal magnitude whose linear combinations generate the entire reciprocal lattice. However, in aperiodic quasicrystals the RVs densely fill all reciprocal space, and it is not possible to choose any RVs of minimal magnitude. Nevertheless, it turns out to be convenient to choose some basic RVs that correspond to relatively intense peaks in the diffraction pattern, and have magnitudes that are related to the inverse of certain lengths in the structure, such as a tile side [2]. Figure 2(b) shows a photograph of the diffraction pattern of the experimental structure, with only three series of the most intensive peaks visible. The magnitude and orientation of the RVs of the internal series correspond to the inverse of half the long diagonal of the thin tile, and choose these RVs as basic set.
Similarly, the middle series corresponds to half the long diagonal of the thick tile and the external series corresponds to half of that tile side length.

2. Band structure

Our approach is based on the belief that the spatial distribution of the electromagnetic field of any mode of the quasicrystal will reflect the distribution of the dielectric constant, and can be represented by a Fourier expansion based on the set of plane waves defined by diffraction pattern, see [5] for details. With Fourier expansions for the inverse dielectric constant and electromagnetic field the allowed modes can then be found by solving the usual matrix eigenvalue problem [6]. For our calculation we have considered the model structure with the following parameters: the dielectric constants of the cylinders and surrounding media are 1.0 and 9.0 respectively, the radius of the cylinders is 0.309 of a tile side. The bands have been obtained in the decagon in reciprocal space defined by the lines bisecting the basic RVs — the pseudo-Jones zone [2], shown in Fig. 3 (inset). In a crystal the pseudo-Jones zone becomes the first Brillouin zone. The solid lines in Fig. 3 show the calculated (neglecting the RVs with which magnitude is less then magnitude of the basic RV, since we do not see it in the diffraction pattern) bands along symmetry directions in the pseudo-Jones zone, together with the associated density of modes, derived from the entire area of the zone. The results presented are for H-polarized modes (magnetic field parallel to the cylinders), which exhibit the largest gap. Note the complete gap in the band structure near a frequency of 0.3 (in units of \(c/d\), where \(c\) is the light velocity), which originates from the Bragg reflection of electromagnetic waves associated with basic RVs. In quasicrystals, RV densely fill all reciprocal space. Therefore we have to take into account RVs of magnitude less than basic RV. The dotted line in Fig. 3 shows the band structure, calculated taking into account additionally the RVs of the type (10001), of the magnitude 0.618 \(\ldots\). The inclusion of these RVs results in the formation of a “minigap” in the bands, and related dips and spikes in the density of modes. However, the most significant feature is the appearance of allowed states inside the gap. These modes are characterized by small wavevector and can be considered to be due to diffraction from the inflated basic elements of the (self-similar) quasicrystals.

![Fig. 3. Band structure for H-polarization calculated using 301 Fourier coefficients and plane waves and the related density of modes (solid line). Corresponding results calculated using an additional 10 plane waves of smaller magnitude such as (10001) (dotted line). Inset: symmetry points of the pseudo-Jones zone.](image-url)
3. Conclusions

The distribution of matter in quasicrystals can be described using a rather small number of Fourier coefficients; The most substantial Fourier coefficients are related to RVs obtained by taking the sum of a few basic RVs; Quasicrystals possess a strongly depleted density of modes around the frequency defined by the strongest Fourier coefficients; The modes in that part of the spectrum are characterized by wavevectors, corresponding to the self-similar inflated elements of the basic structure, but at all frequencies, these modes experience Bragg reflection; Thus, the band structure and density of states of quasicrystals exhibit fractal behavior.

References

Elastic scattering of light from fluctuating exciton polarization of a quantum well in a semiconductor microcavity

V. A. Kosobukin and A. V. Sel’kin
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. A model of exciton polarization fluctuations due to randomly rough interfaces of a quantum well (QW) is proposed. On this basis, a theory of the resonant elastic scattering of light is developed for a random-thickness QW placed in a semiconductor microcavity. Numerical study of the cross-section shows that the light scattering induced by roughness with standard deviation height as small as 0.2 nm could be detected experimentally. An enhancement of backscattering intensity is predicted.

Introduction

Resonant splitting and enhancement of exciton polaritons in semiconductor microcavities (MC) [1] is realized assuming quantum wells (QW) to possess perfectly flat interfaces. Typical of real QWs are various imperfections whose most significant representative is random roughness of interfaces. The inhomogeneity-induced optic effects and light scattering, in particular, are expected to be of great importance in MCs. This work is aimed at treating the resonant elastic scattering of light from fluctuations of excitonic polarization in QWs, especially as it could occur in a MC arrangement.

1. Model

The basic element of the proposed model is a QW centered at \( z = z_0 \) inside a Fabry–Perot resonator (FPR) (0 < \( z < d \)) whose background dielectric constant is \( \varepsilon_2 \). Following [2], the QW interfaces \( z = z_0 - \bar{L}/2 + \zeta_1(x) \) and \( z = z_0 + \bar{L}/2 + \zeta_2(x) \) are assumed to be randomly rough. For random profile functions \( \zeta_n(x) \) of the \( n \)th interface the mean value over the ensemble \( \{ \zeta_n \} \) is \( \langle \zeta_n \rangle = 0 \), with \( \bar{L} \) and \( \delta L(x) = \zeta_2(x) - \zeta_1(x) \) being an average and a fluctuation of the QW thickness \( L(x) = \bar{L} + \delta L(x) \). At a given frequency \( \omega \) of light, an induced polarization associated with quasi-2D excitons of the QW is of the form

\[
P(r, \omega) = \hat{y} \left[ P^0(z; Q, \omega) + \delta P(z; Q, \omega) \right] \cdot \exp(iQx),
\]

same being true for the other electromagnetic field characteristics, whose tangential wavevector component \( Q \) conserves in passing a flat interface. In Eq. (1) \( P^0 \) corresponds to an “average” QW with the interfaces \( z = 0 \) and \( z = \bar{L} \), and \( \delta P \) is due to the fluctuation \( \delta L(x) \). When the random thickness \( L(x) \) keeps nearly constant within an area whose lateral sizes exceed the exciton Bohr radius \( a_B \), it defines locally the exciton transition energy \( \hbar \omega_0[L(x)] \), which is mainly defined by the energies of electron and hole confinements in the QW. If \( \sqrt{\langle \delta L^2 \rangle} \ll \bar{L} \), or \( \sqrt{\langle \delta \omega_0^2 \rangle} \ll \bar{\omega}_0 \), a random variation \( \delta \omega_0(L) = \omega_0(L) - \bar{\omega}_0 \) relative to the average \( \bar{\omega}_0 = \langle \omega_0(L) \rangle \) is

\[
\delta \omega_0(x) \simeq -\Omega \cdot \delta L(x) / \bar{L} \simeq -2\pi^2 \cdot \eta \cdot (R_e / \hbar) \cdot (a_B / \bar{L})^2 \cdot \delta L(x) / \bar{L},
\]
where \( \eta \sim 1 \), and \( R_\xi \) is the exciton Rydberg energy. In turn, if \( \sqrt{\langle \delta \omega_0^2 \rangle} < \gamma \), with \( \gamma \) being the nonradiative decay rate of the exciton, the term \( \delta P \) is written as a linear function of \( \delta L(x) \) [2].

Next, we assume the FPR to be sandwiched in between distributed Bragg reflectors (DBR), the dielectric constants of the outer half-spaces \( z < -l_1 \) and \( z > d + l_2 \) to be \( \varepsilon_1 \) and \( \varepsilon_3 \), respectively, \( l_1 \) and \( l_2 \) being the DBR thicknesses. The amplitudes of electromagnetic waves in the media, specified by \( \varepsilon_m \), are related to each other with the transfer matrices of the DBRs. For the waves of type (1) \( Q = \sqrt{\varepsilon_m k_0} \sin \theta_m \), where \( \theta_m \) is the propagation angle in the \( m \)th medium, with the \( z \)-component of the wavevector being \( k_m \equiv k_m(Q) = \sqrt{\varepsilon_m k_0^2 - Q^2} \), and \( k_0 = \omega/c \).

2. Quasi-2D excitons in an “average” QW

Given the regular term \( P^0 \) in Eq. (1) and the coefficients \( r_1(Q) \) and \( r_2(Q) \) of light reflection back to the FPR from its interfaces \( z = 0 \) and \( z = d \), respectively, the complex frequency of the exciton transition in the “average” QW is expressed as

\[
\omega_0(Q) = \tilde{\omega}_0 + \Gamma \cdot \text{Im} F(z_0) - i \left[ \gamma + \Gamma \cdot [1 + \text{Re} F(z_0)] \right],
\]

where

\[
\Gamma(Q) = \Gamma_0 \cdot \sqrt{\frac{\varepsilon_2}{\varepsilon_2 k_0^2}}.
\]

\[
F(z_0, Q) = \frac{r_1 \exp(2ik_2z_0) + r_2 \exp(2ik_2(d-z_0)) + 2r_1r_2 \exp(2ik_2d)}{D},
\]

\[
D(Q) = 1 - r_1 \cdot r_2 \cdot \exp(2ik_2d).
\]

The difference \( \omega_0(Q) - (\tilde{\omega}_0 - i\gamma) \) from Eq. (3) gives the excitonic radiative energy shift and damping rate, the former tends to zero and the latter to the value (4), if \( r_1 = r_2 = 0 \), i.e. the QW is placed in a homogeneous semiconductor.

3. Reflectivity

When a light wave with the wavenumber \( Q \) is incident along the \( z \)-axis, the reflection coefficient \( r \) of the whole system (a MC with a QW) is

\[
r(Q) = \rho_1 + \tau_1 t_1 \left[ i \Gamma \Phi(z_0) \Delta^{-1} D^{-1} + r_2 \exp(2ik_2d) \right] D^{-1},
\]

\[
\Delta(Q) = \omega_0(Q) - \omega,
\]

\[
\Phi(z, Q) = \exp(ik_2z) + r_2 \exp(2ik_2d) \exp(-ik_2z).
\]

In Eq. (5) \( \rho_1 \) is the reflection coefficient of a single left DBR \((-l_1 < z < 0)\) surrounded by media \( \varepsilon_1 \) and \( \varepsilon_2 \), when light is incident along the \( z \)-axis. For the same arrangement, \( \tau_1 \) and \( t_1 \) are the transmission coefficients of light propagating along the \( z \)-axis and against it, respectively.

4. Scattering of light

The scattering problem originating in a random polarization \( \delta P \), Eq. (1), is solved perturbatively [2]. When a photon is incident at an angle \( \theta_1 \) on the MC from the left, the
Fig. 1. Reflectance $|\tau|^2$ (a) and the cross-section of elastic scattering $d\sigma/d\theta_1'$ (b) calculated as functions of photon energy $\hbar\omega$ from Eqs. (5) and (7) with the following parameters of a GaAs-based heterostructure: $\hbar\omega_0 = 1.56\text{ eV}$, $h\gamma = 0.25\text{ meV}$, $h\gamma' = 1\text{ meV}$, $\hbar\omega_{0x} = 1.553\text{ eV}$ (MC resonance energy), $\hbar\Omega = 50\text{ meV}$, $L = 14\text{ nm}$, $h = 0.2\text{ nm}$, $\Lambda = 150\text{ nm}$. Solid curve in (b) corresponds to the angles $\theta_1 = -\theta_1' = 18^\circ$ (backscattering), and dashed one to $\theta_1 = 18^\circ$, $\theta_1' = 9^\circ$, for the corresponding curves in Fig. 1(a) the incidence angles coincide with the above $|\theta_1'|$.

dimensionless cross-section for its backward scattering at an angle $\theta_1'$ can be obtained to the lowest order in $\xi_n$ (Born’s approximation) as follows

$$
\frac{d\sigma}{d\theta_1'} = W(Q' - Q) \frac{\cos^2 \theta_1'}{\cos \theta_1} |SS'\Phi(\zeta_0)\Phi'(\zeta_0)|^2 |D'|^{-2} |M_II|^{2}.
$$

Here, prime denotes the values for scattered light with $Q'$, in contrast to those for incident light with $Q$, and $W(Q' - Q)$ is proportional to the Fourier transform of a correlation function $\langle \delta L (x - x') \delta L (0) \rangle$. For Gaussian correlator

$$
\langle \xi_n (x) \xi_{n'} (x') \rangle = \delta_{nn'} h^2 \exp \left( - |x - x'|^2 / \Lambda^2 \right)
$$

$$
W(Q' - Q) = \frac{\sqrt{\pi} k_0 \Lambda h^2}{\sqrt{\pi} / L^2} \exp \left( - |Q' - Q|^2 \Lambda^2 / 4 \right),
$$

where $\Lambda$ is the transverse correlation length and $h$ is the r.m.s. height of roughness. The excitonic resonant features are described by the spectral function

$$
S(Q, \omega) = \sqrt{\Gamma(Q) \Omega / \Delta(Q)}
$$

with (2), (4), (6) inserted, and the transformation coefficients $M_l (Q)$ are defined by the transfer matrices of the DBRs.

5. Calculation and summary

The calculated spectra of reflection and elastic scattering of light are shown in Fig. 1 for the light-hole quasi-2D exciton of a GaAs/AlGaAs QW in a MC. Reflectance (Fig. 1(a)) reveals a strong resonant coupling between an electromagnetic mode of the MC and a quasi-2D exciton of the QW depending on the incidence angle. Typical of the manifestation of
exciton-photon interaction is sharing the mode intensities, which become equal just at the resonance (compare the solid and dashed curves in Fig. 1(a)). Figure 1(b) gives evidence that this effect could result in an enhancement of the maximum intensity of scattered field by about an order of magnitude: compare the solid curve related to strict backward (anti-specular) scattering under the resonance conditions with the dashed one corresponding to different incidence and scattering angles. The latter angle difference is responsible for a doublet spectral structure of radiated field near each of the resonances, whose two components are given by the spectral factors (9) of one incident and one scattered waves. In general, the scattering probability at a resonance is defined by the parameter \( \left( \frac{h}{\bar{L}} \right)^2 \ll 1 \) and is estimated here as \( \sim 10^{-2} \) under the usual conditions and \( k_0 \Lambda \sim 1 \), similarly to a single QW \( (r_1 = r_2 = 0) \) [2]. Comparison of this result with the corresponding experimental cross-sections available for rough semiconductor surfaces [3] allows to conclude the resonant scattering of light to be detectable for QW roughness with the standard deviation height \( h \sim 0.2 \) nm.

References


Spectroscopy of negatively charged excitons interacting with 2DEG in CdTe/(Cd,Mg)Te QWs

G. V. Astakhov†, V. P. Kochereshko†, D. R. Yakovlev†‡, R. A. Suris†, W. Ossau‡, G. Landwehr‡, T. Wojtowicz§, G. Karczewski§ and J. Kossut§

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physikalisches Institut der Universität Würzburg, 97074 Würzburg, Germany
§ Institute of Physics, Polish Academy of Sciences, PL-02608 Warsaw, Poland

Abstract. We report on experimental observation of a combined exciton-electron process in which an incident photon creates a trion (negatively charged exciton-electron complex) and promotes an inter-Landau level transition of an additional electron.

Introduction

Until recently the prevailing opinion has been that the exciton-electron interactions in the presence of a two dimensional electron gas (2DEG) reduce solely to the screening of excitons by free electrons or to the band filling. Thus, the system of excitons and 2DEG was viewed as a homogeneous system without any internal structure. Here we show that in a 2DEG of low and moderate density, i.e., when $E_F \ll R_y$ ($E_F$ is the Fermi energy of the 2DEG, and $R_y$ is the exciton Rydberg), the internal structure of the system of excitons coexisting with electrons is quite rich.

A new type of electron-hole bound state, namely, negatively charged exciton or trion has been experimentally found in 1993 in quantum well (QW) structures with a 2DEG [1]. Since that many papers devoted to trions have been published. Trions were observed in a number of QW structures based on different semiconductor compounds, such as CdTe/CdZnTe, CdTe/CdMgTe, CdTe/CdMnTe, GaAs/AlGaAs, ZnSe/ZnMgSSe [2–4]. Negatively charged trions and positively charged trions, related to heavy holes as well as to the light holes were found and studied experimentally [5, 6]. Trion triplet states, in addition to the singlet ground states, were observed in high magnetic fields [5].

In addition to bound exciton-electron states, resonances involving three-particles have been observed in the presence of magnetic fields [7]. Such states were classified as Combined Exciton-Cyclotron Resonances (ExCR), meaning that in an external magnetic field an incident photon creates an exciton in its ground state and simultaneously excites one of the resident electrons from the lowest to one of higher Landau levels. The energy of these transitions is equal to the sum of the exciton energy and a multiple of cyclotron energy.

In the present paper we report the first observation of another type of combined exciton-electron process. This process involves four particles — one hole and three electrons and is similar to the ExCR. In this new process an incident photon creates a trion and simultaneously excites one of the resident electrons from the lowest to one of higher Landau levels. It appears as a pronounced resonance line in reflectivity and photoluminescence excitation spectra. By the analogy with ExCR we call this resonance as Combined Trion-Cyclotron Resonance (TrCR).
1. Experiment

For this study we used modulation-doped CdTe/(Cd$_{0.85}$Mg$_{0.15}$)Te QW structures with a 2DEG of low and moderate density (in this paper we report data for two structures with the electron density in the QW $8 \times 10^{10}$ cm$^{-2}$ and $1.8 \times 10^{11}$ cm$^{-2}$). The structures contain a 120 Å single QW and are modulation doped in the barriers at 100 Å distance from the QW. A special design of the structures made it possible to control the electron concentration keeping all other QW parameters (QW width, barrier height, background impurity concentration, etc.) constant with a high accuracy [8]. We studied reflectivity and photoluminescence excitation (PLE) spectra in magnetic fields applied in the Faraday geometry in $\sigma^+$ and $\sigma^-$ circular polarizations. Electron concentration in the QW has been found from magnetoreflectivity spectra. The details of method for determination of 2DEG density are presented in Ref. [9].

Figure 1(a) shows a reflectivity spectrum taken from 120 Å thick CdTe/Cd$_{0.85}$Mg$_{0.15}$Te single QW with electron concentration $8 \times 10^{10}$ cm$^{-2}$ at 1.6 K. Only the trion line $X^-$ reveals in this spectrum. Strong modifications of this spectrum are observed in magnetic fields [Fig. 1(b)]. In addition to the trion ($X^-$) and the weak exciton ($X$) reflectivity lines two new lines appear in magnetic fields, namely, the Combined Exciton-Cyclotron Resonance (ExCR) line and the Combined Trion-Cyclotron Resonance (TrCR) line. The ExCR line appears at energies higher than that of the exciton ground state and the TrCR line is observed between the exciton and the trion reflectivity lines.

Photoluminescence excitation spectra show even more remarkable modifications in the presence of magnetic fields. Figure 1(c) shows the PLE spectra for the structure with the electron concentration $8 \times 10^{10}$ cm$^{-2}$ at zero magnetic field. Two lines (of about equal intensities) were observed in the PLE spectrum — the exciton line ($X$) and the trion line ($X^-$). In the presence of magnetic fields again two new lines ExCR and TrCR appear in the PLE spectra [see Fig. 1(d)]. The behavior of these lines in PLE spectra is similar to that in reflectivity spectra.

![Fig. 1. Reflectivity (a),(b) and PLE (c),(d) spectra taken in a CdTe/Cd$_{0.85}$Mg$_{0.15}$Te SQW with an electron concentration of $8 \times 10^{10}$ cm$^{-2}$ in zero magnetic field [(a),(c)] and in a magnetic field of 3 T [(b), (d)] in $\sigma^+$ (dotted) and $\sigma^-$ (solid) circular polarizations.](image-url)
2. Results and discussion

Figures 2(a) and 2(b) show magnetic field dependencies of the energy position of the exciton (X), trion (X$^-$), and combined resonance (ExCR) and (TrCR) lines for two electron concentrations in the QWs. The line corresponding to the Combined Exciton-Cyclotron Resonance (ExCR) shows a linear shift to higher energies with increasing magnetic fields. Its position approximated to the exciton ground state when the field goes to zero. The slope of this dependence is described by the relation $E_{\text{ExCR}} = (1 + m_e/M) \hbar \omega_c$ [7], here $m_e$ is the electron effective mass, $M$ is the exciton mass, $\omega_c$ — cyclotron frequency. This process could be explained in the following way: an incident photon creates an exciton in its ground state and promotes an excitation of an additional electron from the lowest to higher Landau levels.

The combined trion-cyclotron resonance line (TrCR) appearances different from the ExCR ones. This line can only be observed in the magnetic fields corresponding to the filling factors ($\nu$) between 3 and 1. The fact that the line disappears at the filling factors $\nu < 1$ supports the idea that the process requires two additional electrons, as at filling factors higher than one can find two electrons at the same point of the sample only. The position of this line tends to the trion energy as the magnetic field vanishes. The energy of the line position depends linearly on magnetic fields. The slope of this dependence is described by the relation $E_{\text{TrCR}} = 1/2 \hbar \omega_c$.

We assign this line to four-particle processes, which involve three electrons and one hole. Incident photon creates a trion and causes another electron to be excited between the Landau levels. In Fig. 2(b) one can see two transitions of this type: TrCR1 and TrCR2. So, in the initial stage we have two electrons in 2DEG and one photon, then the photon creates an exciton, which binds one electron, and in the final stage we have one trion and one electron in the one of higher Landau levels. The photon energy of this combined transition is:

$$1/2 \hbar \omega_c + 1/2 \hbar \omega_c + ph \rightarrow Tr + 3/2 \hbar \omega_c.$$ (1)

The expected energy of the line position is, thus, $E_{\text{TrCR}} = E_{\text{Tr}} + 1/2 \hbar \omega_c$.

In conclusion, in CdTe-based QW structures, we found a new type of combined exciton processes, which involve four particles: one hole and three electrons. These combined processes were studied in magnetic fields up to 7.5 T in reflectivity and PLE spectra.
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References


Energy level diagram of $X^-$ in high magnetic fields

T. Vanhoucke†, M. Hayne†, V. V. Moshchalkov† and M. Henini‡

† Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Leuven
‡ School of Physics and Astronomy, University of Nottingham, University Park, Nottingham NG7 2RD, UK

Abstract. We have studied the photoluminescence energy of the negatively charged exciton in a 100 Å GaAs/AlGaAs quantum well using magnetic fields up to 50 T. By observing recombination from all of the optically active singlet and triplet states of the charged exciton, we have determined its energy level diagram.

Introduction

The recombination spectra of semiconductors are dominated by excitons. A neutral exciton $X_0$ consists of a hole bound to an electron by the Coulomb interaction. If $X_0$ binds a second electron, then we have a negatively-charged exciton $X^-$, or trion. The neutral exciton is the solid-state analog of the hydrogen atom $H$, while $X^-$ is the analog of the negatively-charged hydrogen ion $H^-$. Since $X^-$ is a three-body system, its physics is more complicated than the $X_0$. Indeed, $X^-$ is currently the subject of much theoretical [1] and experimental [2–4] investigation. Here we present photoluminescence (PL) experiments in magnetic fields, $B$, up to 50 T on a 100 Å GaAs QW. We have measured all the experimentally observable transitions of the singlet and triplet states of $X^-$, and so reconstructed the level diagram.

1. Experimental details

The 100 Å GaAs/AlGaAs QW sample was grown by molecular beam epitaxy. Our experiments were undertaken at 1.2 K with $B$ parallel to the growth direction of the QW. Photon counting times between 0.4 ms and 2 ms during the 25 ms field pulse resulted in a field resolution of $\pm 1\%$ and $\pm 3\%$. The spectral resolution was better than 0.5 meV. A solid-state laser (wavelength 532 nm) was used to excite the sample and reduce the electron density in the QW. Optical access to the sample was provided by a fibre bundle. An in-situ polariser was used in combination with reversing the field direction to distinguish between the right-(\(\sigma^+\)) and left-handed (\(\sigma^-\)) circularly polarised light.

2. Experimental results

Figure 1 shows the energy of the excitonic recombination as a function of $B$. The four lines, labelled as $S_1$, $S_2$, $T_1$ and $T_2$, are due to the luminescence of the different states of $X^-$. The open and closed symbols present $\sigma^+$ and $\sigma^-$ respectively. At low fields ($B < 10$ T) we observe a splitting between $S_1$ and $S_2$ with polarisation $\sigma^-$ and $\sigma^+$. A third line, $T_1$, appears at $B = 4.2$ T and is the highest energy line at these low fields. At higher fields ($B > 10$ T) the recombination energy of $T_1$ and $S_2$ is the same between 14 T and 26 T. However, we can distinguish the different lines via the polarisation which is positive for $S_2$ and negative for $T_1$. Above 26 T, $T_1$ becomes lower in energy than $S_2$ and parallel with $S_1$. 252
Fig. 1. Field dependence of the PL peak energy. The upper inset shows the difference between \( S_2 \) and \( S_1 \), which is identified as the spin splitting of the singlet state. The lower inset shows a spectrum at \( B = 50 \text{ T} \) with \( \sigma^- \).

The fourth excitonic line, \( T_2 \), appears at 10 T and is the highest PL energy for fields up to 43 T. The intensity of \( S_2 \) and \( T_2 \) decreases above 4 T, and these lines disappear at 43 T. \( T_1 \) and \( S_1 \) remain visible at all fields.

3. Discussion

We use our experimental data to construct an energy diagram for the \( X^- \). We start by considering the two electrons, which must have an antisymmetrical total wave function. One possibility is by an antisymmetrical spin wave function and a symmetrical spatial wave function, which we refer to as the singlet state. One can also make three combinations of a symmetrical spin wave function and an anti-symmetrical spatial wave function to get the triplet state. If we then include the hole, every level splits into two sublevels with a total \( z \) component of the spin \( S_z \). The degeneracy of the splitting is lifted by the Zeeman interaction. A schematic diagram showing the previously used level structure for the \( X^- \) is shown in the inset of Fig. 2 [2].

Our experimental results have shown that because the splitting of these levels is sensitive to the electron and hole gyromagnetic ratio (\( g_e \) and \( g_h \) respectively), it is necessary to redraw the energy level diagram as shown in Fig. 2.

We first start with the two possible singlet transitions 1 and 2, which have a different polarisation, \( \sigma^- \) and \( \sigma^+ \) respectively. These transitions (corresponding to \( S_1 \) and \( S_2 \) in Fig. 1) are the lowest in energy at low fields, as can be found in the literature [2]. When taking the difference in PL energy between \( S_1 \) and \( S_2 \) of our data, we observe a linear behaviour as a function of field (inset Fig. 1), with a slope of 0.11 meV/T. Using the equation for the Zeeman splitting of the PL

\[
\Delta E = (g_e + 3g_h)\mu_B B, \tag{1}
\]

where \( \mu_B \) is the Bohr-magneton and \( g_e = -0.2 \) [5], we find that \( g_h = 0.7 \). This value is consistent with that found by Snelling for the exciton in a 100 Å QW [5, 6]. Notice that
because the total electron spin is zero in the singlet, this splitting is only caused by the hole. For the triplet states, there are four optically allowed transitions, two $\sigma^-$ (transitions 3 and 4) and two $\sigma^+$ (transitions 5 and 6). Transitions 3 and 4 have the same PL energy, while they connect different energy levels. The same is true for transitions 5 and 6. This means we cannot make a distinction between transitions 3 and 4 (5 and 6) optically. The levels triplet(−5/2) and triplet(+5/2) are optically dark. We identify transitions 3 and 5 with $T_1$ and $T_2$ respectively in our experimental data in Fig. 1. The Zeeman splitting for the triplet is not as straightforward as for the singlet, and needs more explanation. For a start, the triplet is not bound at low fields [1–2]. Above 25 T, the difference between $T_1$ and $T_2$ is linear in field with the same slope as the singlet. This is to be expected from the level structure in Fig. 2. The linearity becomes very bad at fields between 16 T and 25 T, where we observe $T_1$ to have the same PL energy as the positive component of the singlet, $S_2$. We do not believe that the absence of a clear crossing has any physical significance, and attribute it to a lack of resolution in the data. At the field where the singlet(−3/2) and triplet(+3/2) coincide, the spin splitting of the singlet state must be equal to the separation of the triplet and singlet states in the absence of spin ($T_0$ and $S_0$ in Fig. 2). We can use this level coincidence to determine the hole $g$ factors as we did with the singlet splitting. We find $T_0 - S_0$ to be constant in field and equal to $2.2 \pm 0.3$ meV. Locating the midpoint of the crossing between $T_1$ and $S_2$ at $B_0=20$ T, we can construct the following equation

$$(g_e + 3g_h) \mu_B B_0 = T_0 - S_0. \quad (2)$$

This gives us $g_h = 0.7$ using $g_e = -0.2$. This calculation gives essentially the same value for $g_h$ quoted in the literature [5], thereby confirming the revised energy level diagram of Fig. 2. Although level triplet(+5/2) is optically dark, it becomes the ground state at very high fields. This is the case when it is lower in energy than singlet(+3/2) which, if
we assume $T_0 - S_0$ remains constant, is at about 190 T. This field was predicted by theory to be 30 T [1]. Finally, we note that, in contrast to other studies [2, 4], we do not observe the neutral exciton, except at fields below 15 T, where it is expected to merge with $T_1$. Indeed, we can confirm our assignment of $T_2$ as the triplet($-3/2$) from the polarisation, which should be $\sigma^-$ for the lowest energy splitting for $X_0$ [4]. We believe that the lack of $X_0$ recombination is due to an excessive electron density in the QW.

4. Conclusion

We have studied the PL of the singlet and triplet states of $X^-$ in a 100 Å GaAs QW. Although we do not observe $X_0$, we are able to determine all possible transitions of the singlet as well as the triplet state. This allows us to construct an energy level diagram for $X^-$. The value of the hole $g$ factor we determine for the $X^-$ is the same as that for the $X_0$ in the literature.

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Regular trends in fine structure and localization of excitons in type II GaAs/AlAs superlattices with a gradient of composition.

P. G. Baranov†, N. G. Romanov†, A. Hofstaetter‡, B. K. Meyer‡, A. Scharmann‡, W. von Foerster‡, F. J. Ahlers# and K. Pierz#
†Ioffe Physico-Technical Institute, St Petersburg, Russia
‡1. Physics Institute, University of Giessen, Heinrich-Buff-Ring 16, D-35392 Giessen, Germany
#Physikalische-Technische Bundesanstalt, Braunschweig, Germany

Abstract. Optically detected magnetic resonance and level anticrossing spectroscopy were used to reveal regular trends in the behavior of the fine structure of excitons, their dynamic properties and localization at the opposite interfaces in type II GaAs/AlAs superlattices grown by MBE with a gradient of layer thicknesses in the SL plane.

Introduction

Optical and electronic properties of quantum well heterostructures are particularly sensitive to disorder at the interface between the different compounds forming the well and the barrier. In type II structures electrons and holes are confined in the adjacent layers and the spatially indirect interband optical transitions arise due to the electron-hole overlap within a very narrow region containing the interface. Excitons in such structures are localized at the interfaces and can be used as probes sensitive to the interface microstructure. In type II GaAs/AlAs SL’s radiative lifetimes lie in the µs range which makes possible to use optically detected magnetic resonance (ODMR) for direct measurements of electron and hole g-factors and the exciton exchange (fine structure) splittings with a radiospectroscopy precision [1,2]. Level anticrossing (LAC) spectroscopy of SL’s was developed on the basis of ODMR and provided important complementary information [3, 4]. Due to the lowered point symmetry C2v of the interface all four levels of heavy-hole excitons are split. One to one correspondence was established between the order of the exciton radiative levels and the type of interface at which exciton is localized: the lowest radiative levels are [110] polarized for excitons at the normal (AlAs on GaAs) interface and [110] polarized for the inverted (GaAs on AlAs) interface [5, 6]. In addition, a difference in the fine structure splittings caused by the asymmetry in the interface composition profiles was found for excitons localized at the opposite interfaces [7]. Application of ODMR for a study of exciton dynamic properties was reported in [8]. In all GaAs/AlAs superlattices in-plane linear polarization of luminescence of some per cent was found which correlated with the exciton preferential localization [9].

In the present work we study regular trends and variations of the exciton localization and fine structure splitting in type II GaAs/AlAs SL grown with a gradient of GaAs/AlAs composition.

1. Results and discussion

GaAs SL were grown by MBE with 30 s. interruptions after GaAs layers on the 2 inch (001) GaAs substrate kept at 620°C. A smooth variation of GaAs/AlAs composition in the
SL plane from 18 Å/18 Å to 23 Å/23 Å was obtained. It was controlled by X-ray diagnostics (XRD). Luminescence was excited far above the band gap with an Ar⁺ laser. ODMR at 24 and 35 GHz and LAC was measured by monitoring circular and linear polarization of luminescence, respectively. 2 mm wide samples were cut along the substrate diameters parallel to [110] and [1\bar{1}0] and could be shifted along the axis of the microwave cavity allowing spatially selective measurements.

Figure 1(a) shows variations of luminescence spectra as a function of a position of the excitation spot (x = 0 corresponds to the centre of the wafer). According to XRD data GaAs/AlAs composition changed from 18.6 Å/17.8 Å (x = −22 mm) to 21.8 Å/23.5 Å (x = 22 mm).

LAC signals measured for at x = 10 mm where a doublet luminescence is observed, are shown in Fig. 1(b). Insert shows the luminescence spectrum and the linear polarization signals at B = 0 (dashed line) and B = 0.1 T (dotted line). Analysis of LAC allows to follow variations of the exciton localization at the opposite interfaces. This is illustrated in Fig. 2(a) which shows a decomposition of the observed LAC signals. Excitons at the opposite interfaces have inverted order of the radiative levels (different signs of LAC) and different exchange splittings (different resonance fields). Figure 2(b) shows ODMR measured as variations of σ⁺ and σ⁻ light caused by resonant microwaves chopped at 1 and 100 kHz. Such measurements allow to reveal different exciton dynamic properties similar to [8].

In the ODMR spectra shown in Fig. 3 resonance signals corresponding to spin-flips of exciton holes and exciton electrons are observed. Their positions and splittings are different for the two emission lines. This is connected with the exciton localization in the regions
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Fig. 2. (a) Decomposition of level anticrossing signals (for $x = 10$ mm, $h\nu = 1.85$ eV) into LAC of excitons localized at the normal (AlAs on GaAs) and inverted (GaAs on AlAs) interface. (b) 24 GHz ODMR measured on the two circularly polarized components of emission with 1 kHz and 100 kHz chopping of microwaves.

Fig. 3. 24 GHz ODMR spectra taken at the two extrema of emission (for the point $x = 10$ mm). $T = 1.6$ K. $B \parallel [001]$.

with different local periods and GaAs layer widths. On the basis of the dependencies of the isotropic exchange splitting on the SL period and the hole $g$-factor on the GaAs thickness [2, 4] we can tell that the high energy emission line is due to the excitons localized in the regions $17.5 \, \text{Å}/20.4 \, \text{Å}$ and the low energy line originates from excitons in the region $19.5 \, \text{Å}/20.7 \, \text{Å}$, i.e. low-energy emission comes from c.a. monolayer-high GaAs islands. From LAC (Fig. 2(b)) we can conclude that emission in high-energy line is produced by excitons localized at both normal and inverted interface whereas emission in low-energy line is due to excitons localized at the inverted interface. Excitons localized at the normal and inverted interfaces in the regions with the same local period show different response
time to the applied resonant microwaves (Fig. 2(b)). In addition, effective lifetime of excitons in the high-energy line is shorter as compared to that in the low-energy line.

Similar measurements carried out at different positions on the sample where the layer thicknesses change by more than a monolayer, allowed to reveal regular trends in the exciton fine structure, dynamic behavior and localization at the opposite interfaces. For example, in the regions with close GaAs layer thickness excitons at the inverted interface have larger exchange splitting and shorter effective lifetime, the appearance of low energy line in doublet emission spectra is due to the excitons localized mainly at the inverted interface in the regions of monolayer high GaAs islands, and so on. These regular trends will be discussed in detail.

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References

Reflectivity studies of trion (X\(^-\)) and exciton (X) states in ZnSe/(Zn,Mg)(S,Se) QWs

G. V. Astakhov†, V. P. Kochereshko†, D. R. Yakovlev†‡, R. A. Suris†, W. Ossau‡, J. Nürnberg‡, W. Faschinger‡ and G. Landwehr‡

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Physikalisches Institut der Universität Würzburg, 97074 Würzburg, Germany

Abstract. The oscillator strength of negatively charged exciton (trion) in ZnSe/(Zn,Mg)(S,Se) quantum-well structures with \(n\)-type modulation doping is studied by reflection spectroscopy as a function of electron concentration and temperature. The trion oscillator strength is found to increase linearly with increasing electron concentration up to \(6 \times 10^{10} \text{ cm}^{-2}\). The effect of oscillator strength “shearing” between the exciton and trion states is observed. The value of the oscillator strength shearing is found to be not more than 20%.

Introduction

The existence of negatively charged exciton complexes (trions) in semiconductors, consisting of an exciton attracting an additional electron was predicted theoretically in 1958 [1]. These states have been under intensive study since 1993, when a first experimental proof of the trion existence in quantum well (QW) structures was published [2]. Trions have been observed in QW structures based on different semiconductor compounds, such as CdTe [2], GaAs [3] and ZnSe [4, 5]. However, quite a number of features of the trion states are not understood yet and/or are the subject for controversy.

This paper is concerned with the problem of the trion and exciton oscillator strengths in QW structures with high electron concentration, when the Fermi energy of 2DEG is comparable with the trion binding energy. A possibility for a redistribution of the oscillator strength between the exciton and trion with increase of the 2DEG density is mentioned in the papers [6, 7]. Authors call this redistribution as oscillator strength “shearing”. However, any quantitative analysis of the observed evolution of the exciton and trion oscillator strengths as well as an understanding of the effect is still missing.

1. Modification of X\(^-\) and X states with electron concentration

We chose modulation doped ZnSe/(Zn,Mg)(S,Se) single quantum well structures as a model system for the present study. The choice of ZnSe-based QWs with extremely strong Coulomb interaction, as compared with GaAs- or CdTe-based QWs, allows us to get very pronounced exciton and trion resonances and to evaluate parameters with high experimental accuracy.

The structures with ZnSe/Zn\(_{0.90}\)Mg\(_{0.11}\)S\(_{0.18}\)Se\(_{0.82}\) single quantum well (SQW) of 80 Å width were grown by molecular-beam epitaxy on (100) GaAs substrates (for details of growth and optical properties see Ref. [5]). One structure was nominally undoped and had a residual electron concentration of \(n_e < 10^{10} \text{ cm}^{-2}\) in the SQW. A set of modulation-doped structures with 2DEG concentration in the range from \(n_e = 3 \times 10^{10} \text{ cm}^{-2}\) up to
1.2 × 10^{11} \text{ cm}^{-2} were grown with a Cl doped layer separated from the QW by a 100 Å-thick spacer.

Figure 1(a) shows reflectivity spectra in the vicinity of exciton and trion resonances taken from QW structures with different 2D electron concentrations. Details of identification of the trion transitions and basic trion properties in ZnSe-based QWs were published in Refs. [4,5]. For the nominally undoped sample (upper spectrum) there is only one resonance line which corresponds to a heavy-hole exciton (X) in the QW. In the doped sample (middle spectrum), a new resonance line of a negatively charged exciton (X\(^{-}\)) appears at about 5 meV to low-energy side from the exciton one. The amplitude of the trion resonance line grows with increasing of electron concentration. At the same time this is accompanied by the reduction of exciton-resonance amplitude. At high electron concentration the exciton line disappears from the reflectivity spectra at all.

Parameters of the exciton- and trion resonances (i.e. resonance frequency, radiative damping (\(\Gamma_0\)) and nonradiative damping (\(\Gamma\))), which govern their contribution to the dielectric function, were deduced from the best fit of experimental reflectivity spectra with the calculated ones in the framework of the non-local dielectric response theory [8]. An example of such fit is shown by the dotted line in Fig. 1(a). The electron concentration in the QW was determined from magnetoreflectivity spectra. The details of method for determination of 2DEG density are presented in Ref. [9]. The dependence of the deduced parameters as radiative (\(\Gamma_0\)) and nonradiative (\(\Gamma\)) damping of the excitons and trions are plotted against the 2DEG concentration in Figs. 1(b) and 1(c).

One can see in Fig. 1(b) that the radiative damping of the trion (\(\Gamma_0^T\)) increases linearly with electron concentration increases up to \(n_e = 6 \times 10^{10} \text{ cm}^{-2}\). Note (following book [8]) that the exciton radiative damping \(\Gamma_0\) in the QW is proportional to the exciton oscillator strength (\(F_X\)), i.e. \(\Gamma_0 \propto F_X\). We can introduce the trion oscillator strength per one electron (\(F_X^T/N_e\)), and the exciton oscillator strength per unit cell (\(F_X^N/N\)). Here \(N_e\) is the number of electrons in the QW, \(N\) is a number of unit cells in the square of the QW.
Following the approach developed in Ref. [10] we obtain the relation:

$$\frac{\Gamma^T}{N_e A_T} = \frac{\Gamma^X}{N_a}$$

or

$$\frac{\Gamma^T}{\Gamma^X} = \frac{\pi a_T^2}{N_e}.$$  \hspace{1cm} (1)

Here $A_T = \pi a_T^2$ is the area with trion radius $a_T$, $a$ is the area of the unit cell, $N_a = A$ is the area of the QW and $N_e / A = n_e$ is the two dimensional electron concentration. Putting in Eq. (1) the experimentally determined parameters $\Gamma^X_0 (n_e = 0) = 210 \mu eV$ and $\Gamma^T / n_e = 1.16 \times 10^{-9} \mu eV \cdot cm^2$ we evaluate the radius of the trion as $a_T = 132 \AA$.

The exciton radiative damping decreases in 20% at low electron concentrations in the interval from $10^{10} \text{ cm}^{-2}$ to $3 \times 10^{10} \text{ cm}^{-2}$ and conserves at further concentration increases. This decrease of radiative damping could be attributed to the “intensity sharing” between exciton and trion resonances suggested in Ref. [6].

Let us now turn to the nonradiative damping, which is contributed by homogeneous and inhomogeneous broadening of resonant states. For the trions the nonradiative damping $\Gamma^T$ shows no concentration dependence in the range studied [Fig. 1(c)]. The possible explanation is that the trions are created predominantly in localized states and that $\Gamma^T$ is dominated by inhomogeneous broadening.

Fig. 2(b) shows that the nonradiative damping of excitons increases strongly with electron concentration increase. We suggest that the exciton–electron scattering with pair excitations (when an electron is excited above the Fermi energy and hole remains inside the Fermi sea) is responsible for this effect. In that case a homogeneous contribution to the exciton line-width should be either of the order or higher than of $\varepsilon_F$. Namely, due to this very large exciton-line-broadening at high electron concentration the exciton line goes out from the reflectivity spectra.

2. Modification of $X^-$ and $X$ states with temperature

A set of reflectivity spectra registered from SQW with $n_e = 6 \times 10^{10} \text{ cm}^{-2}$ is shown in Fig. 2(a) for different temperatures in the interval 1.6—35 K. As temperature increases the trion reflectivity line disappears and the amplitude of the exciton line, on the contrary, increases. Radiative and nonradiative damping for exciton and trion states are plotted in Figs. 2(b), (c) as a function of temperatures. There is no temperature effect on the exciton...
nonradiative damping, at the same time a remarkable broadening of the trion line is observed in these dependencies [Fig. 2(c)]. This temperature induced broadening could be attributed to the temperature delocalization of trions.

The trion radiative damping decreases with the temperature increase [Fig. 2(b)]. That is quite understandable because the corresponding matrix element of the optical transition is maximal for small electron energy [11] and falls down rapidly with the electron energy increase. However, the temperature increasing of the exciton radiative damping is very surprising. The value of this increase is just the same as we have in the Fig. 1(b) for the exciton oscillator strength suppression by 2DEG. It could mean that the effect of the oscillator strength “shearing” is present and the value of this “shearing” is about 20% of the initial exciton oscillator strength. The physical reason for such “shearing” could be connected with an interaction between exciton and trion states via emission and absorption of an electron. A trion can lose an electron transforming into an exciton; an exciton can trap an electron transforming into a trion and so on. Such “exchange” by an electron means the interaction between these two states that could lead to some redistribution between the exciton and trion oscillator strengths.

3. Conclusions

Reflectivity spectra have been analyzed in detail for \( n \)-type modulation-doped ZnSe/ (Zn,Mg)(S,Se) quantum well structures. Exciton and trion parameters were determined as a function of 2DEG density and temperature. A relation between exciton and trion oscillator strengths has been established. A linear dependence of the trion radiative damping on 2D-electron concentration has been found. The effect of oscillator strength “shearing” is directly measured.

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References

Dimensionality transformation of exciton state in quantum well with asymmetrical barriers

V. V. Kapaev†, Yu. V. Kopaev and A. E. Tyurin
P. N. Lebedev Physical Institute, RAS, 117924 Moscow, Russia
† e-mail: kapaev@sci.lebedev.ru

Abstract. In the conditions of 2D–3D dimensionality transformation of one-particle states, the dependence of exciton binding energy $E_{ex}$ on asymmetrical well width $h$ has been theoretically studied. It is shown that the contribution of one-particle continuous spectrum to exciton becomes considerable when the inverse exciton Bohr radius $1/a_B$ turns out to be comparable with the critical for 2D–3D transformation value of in-plane wave vector $k_c$. The lack of coincidence between electron and hole $k_c$ values leads to the complicated shape of $E_{ex}(h)$ dependence, which has a minimum at intermediate values of $h$. The minimum value of $E_{ex}$ appears to be less than 3D exciton binding energy.

In quantum wells with asymmetrical in height barriers and different effective masses in structure layers, the localized 2D states exist only within the limited range of in-plane wave vector $k$. The critical $k_c$ value depends on the well width, barrier height ratio $U_2/U_1$ and the ratio of effective masses in the confining barriers. The $k_c$ value can be quite small giving rise to some rather interesting effects [1]. One can control $k_c$ value by the external electric and magnetic fields. The 2D–3D transformation of the exciton states in external electric field was experimentally observed in GaAs/AlGaAs quantum well structures [2]. As it was shown in [1], the drastic change of exciton binding energy occurs when the localized state in asymmetrical well still exists but the critical value of $k_c$ becomes comparable to the inverse exciton Bohr radius $1/a_B$. In this case, the exciton state is constructed from the localized states of electron and hole as well as from the states of continuum.

In the conditions of dimensionality transformation the dispersion law becomes essentially non-parabolic for electron (hole) states. The wave functions depend on $k$, which doesn’t allow one to describe exciton states within the usual variational approach [3].

In this paper, we present the method for description of exciton states in systems with complicated valence zone dispersion law [4] caused by heavy–light holes interaction [5]. In this method, the exciton wave function is represented as an expansion in terms of products of electron and hole wave functions with different in-plane motion momenta, taking into account the dependence of one-particle wave functions on $k$. The additional complication in our case is a necessity to consider the continuum states contribution to exciton state when $k > k_c$.

To simplify computational technique, we accept the following procedure: let us introduce the infinitely high barrier in the region of lower barrier $U_2$ of the structure on rather large distance $L$ from the right edge of the well. In this case, the continuous spectrum is represented by a set of discrete states with very small distance between appropriate subbands. There is the change of wave function localization (from the well region $(0, h)$ to the region $(h, L + h)$) with the variation of wave vector $k$, instead of 2D–3D transformation. We have to achieve the convergence by varying the parameter $L$ for adequate description.
of continuous spectrum. Solving one-particle Schrödinger equations in described structure for different values of $k$, we obtain the energy spectrum $E_{c,v}^{n,h}(k)$ and the set of appropriate electron and hole wave functions.

Let us consider 1-S exciton state. We represent the exciton wave function as a following linear combination:

$$
\Psi_{ex}(r_e, r_h) = \frac{1}{4\pi^2} \sum_{n_e, n_h} \int dk_e \int dk_h \Phi_{k_e, k_h}^{n_e, n_h} \exp(i (k_e \rho_e - k_h \rho_h)) \psi_{n_e, k_e}(z_e) \psi_{n_h, k_h}^*(z_h),
$$

(1)

where $\Phi_{k_e, k_h}^{n_e, n_h}$ are the Fourier-expansion coefficients. It is convenient to use coordinates for center-of-mass motion $(Q, R)$ and relative motion $(k, \rho)$. By multiplying the Schrödinger equation by $\Psi_{ex}^*$ and integrating over $R, \rho, z_e, z_h$ we obtain for the expansion coefficients the following set of coupled integral equations:

$$
\left[ E_{c}^{n_e}(k + Q/2) - E_{v}^{n_h}(k - Q/2) \right] \Phi_{kQ}^{n_e, n_h} - C \sum_{n'_e, n'_h} \int dk' \Phi_{k'Q}^{n'_e, n'_h} \int dz_e \int dz_h \exp(-|k - k'| |z_e - z_h|) \psi_{n'_e, k+Q/2}(z_e) \psi_{n'_h, k'-Q/2}(z_h) \psi_{n_e, k}(z_e) \psi_{n_h, k}(z_h),
$$

(2)

where $C = e^2/2\pi\varepsilon$.

The calculations were performed in the assumption that $Q = 0$ for optically active exciton [6]. Let us make the values $k, z_e, z_h$ uniformly discrete in the considered regions and represent all the integrals in (2) as appropriate sums. Thus, we obtain a system of linear equations for $\Phi_{kQ}^{n_e, n_h}$ instead of set of coupled integral equations. Avoiding the point of integrable singularity at $k = k'$, we solve the usual eigenvalue problem [5].

The choice of integration interval for $k$ is rather important in the method described. At the large $h$ values in formation of 3D-exciton the quasi-continuous spectrum states that have equal degree of wave functions localization participate. The dispersion law is parabolic for every subband. One has to choose in (1) rather small integration interval over $k$ ($\Delta k \sim 1/a_B$) and take into account a considerable number of subbands (variational approach) to achieve the convergence.

The situation is the same for the small $h$ values, when the electron localized state is already vanished.

The situation is rather different for intermediate values of $h$ when the electron energy corresponds to that of 2D-exciton. The parameter $1/a_B$ is so large that in a considerable range of $h$ it can exceed the critical value $k_c$ for dimensionality 2D–3D transformation of electron states. In this case the first subband states $E_1^c(k)$ provide the most considerable contribution to the expansion (1). In the conditions of 2D–3D transformation the appropriate wave functions can be considered as a full set of functions necessary for the exciton formation.

The calculations were made for the structure on the basis of GaAs/Al$_x$Ga$_{1-x}$As with aluminium concentration: $x = 0.4$ in the left barrier, $x = 0$ in the well, and $x = 0.06$ in the right barrier. The distance $L$ necessary for continuum simulation was settled as 100 nm and was chosen from the condition of the convergence of results.

The described procedure allows one to consider exciton states in the whole range of structure parameters, i.e., in the bulk GaAs (with large $h$), bulk AlGaAs ($h = 0$), and the well widths corresponding to 2D–3D transformations of the hole ($h = 1.7$ nm) and electron ($h = 3.8$ nm) states.
The dependence of exciton binding energy $E_{\text{ex}}$ on the width $h$ of asymmetrical well (obtained by the method described) is represented in Fig. 1. The dependence has a complicated shape with two extrema: minimum $E_{\text{ex}} = 3.5$ meV at $h_1 = 2.4$ nm and maximum $E_{\text{ex}} = 10$ meV at $h_2 = 7$ nm. The value $E_{\text{ex}}$ obtained at $h = 0$ equals to 3D-exciton binding energy in bulk Al$_x$Ga$_{1-x}$As with aluminium concentration $x = 0.06$. This confirms the correctness of chosen computational technique.

As the well width $h$ increases the conditions of 3D–2D transformation for the hole state are first met. The hole wave function localizes in the well region while the electron remains delocalized. This leads to the minimum of exciton binding energy at $h_1 = 2.4$ nm. It is essential that the minimum value $E_{\text{ex}}$ appears to be less than the 3D-exciton binding energy in GaAs. The further increase in $h$ leads to the electron 3D–2D transformation and, consequently, it's localization in the well. As a result, the oscillator strength of electron–hole transition rises, reaching maximum at $h = 7$ nm. The decrease in $E_{\text{ex}}$ with the further growth of $h$ is due to the same reasons as in symmetric wells, i.e., a simultaneous decrease in the localization of electron and hole wave functions.

In Fig. 2 the inverse Bohr radius $1/a_B$ dependence on the well width $h$ is shown (solid curve). The dashed curve represents the $h$ dependence of the critical value of wave vector $k_c$ where the 2D–3D transformation of electron state occurs. The curves intersect at $h = 5$ nm. From this moment the less $h$ is (until the electron state vanishes) the more important role
in excitation formation play the quasi-continuous states $E^1_c(k) \ (k > k_c)$. For example, at $h = 4.1$ nm the integration interval in (1) $\Delta k \sim 1/a_B$ is approximately two times greater than $k_c$. As a result there are the equal contributions of electron localized and continuous spectrum states to exciton. When $h = 3.8$ nm the electron state is almost vanished ($1/a_B \gg k_c$) and the exciton is formed by localized hole and electron continuum states.

To describe the exciton states the method of fractional dimension was proposed [7]. In this method, the dimensionality is represented by value $\alpha = 2\sqrt{\text{Ry}}/E_b + 1$. Let us use this formula to estimate dimensionality in our case. At $h_0 = 5$ nm ($1/a_B$ and $k_c$ crossing) the value $\alpha \sim 2.5$. The decrease of well width from $h_0$ to $\sim 4$ leads to $\alpha$ increase up to $\sim 3$. This illustrates essential influence of 2D–3D electron spectrum transformation on exciton dimensionality [1].

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Fluctuation-trapped exciton states in 2D-semiconductor solid solutions

A. Klochikhin†‡, A. Reznitsky‡, L. Tenishev‡, S. Permogorov‡, S. Verbin¶, S. Ivanov‡, S. Sorokin‡, R. Seisyan‡ and C. Klingshirn§

† Petersburg Institute for Nuclear Physics, RAS, 188350, St Petersburg,
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia
¶ St Petersburg State University, 198840 St Petersburg, Russia
§ Institut für Angewandte Physik, Universität Karlsruhe 76128, Germany

Abstract. Exciton localization in quantum wells formed by solid solutions has been studied both theoretically and experimentally. The method for calculation of the density of fluctuation states below the edge of two-dimensional exciton band and the spectral density of exciton transitions is developed. The classification of states in respect to its migration properties and contribution to the luminescence processes has been carried out using the continual percolation theory. The shape of emission and absorption bands, as well as the position of mobility edge are calculated. The results of calculations are in good agreement with the optical spectra of ZnCdSe- and GaInAs-based QW structures.

Introduction

Light emission in most of low dimensional heterostructures is governed by localized exciton states. An inhomogeneous character of exciton emission bands with a strong variation of exciton migration properties across the band contour has been first established in GaAs/AlGaAs QW’s [1] and has been attributed to exciton localization by potential fluctuations in AlGaAs solid solutions forming the barriers. In this connection a concept of mobility edge for the fluctuation states in the tail of exciton states has been introduced. It is a common point of view that localized exciton states in QW’s are due to interface roughness and fluctuation of composition in the case of QW’s formed by solid solutions. A specific feature of the localization is an occurrence of fluctuation effects even for solid solutions with a substitution in the cation sublattice unlike the bulk solid solutions, where cation substitution does not lead to a pronounced tailing [2].

1. Theoretical model

In present paper we shortly communicate the results of our studies of the effect of compositional fluctuations on exciton states in solid solution forming the quantum well. As a first step, we have calculated the value and energy dependence of the density of fluctuation states \( \rho(\omega) \) and the spectral density of optical transitions \( \alpha_0^0(\omega) \) of the ground exciton state. The calculations have shown that the confinement of exciton states in 2D structures strongly enhances the effectiveness of perturbing potential due to compositional fluctuations in comparison with the case of three dimensional localization [3].

For calculation of the emission spectra we have analyzed the properties of fluctuation states at different energies in respect to their mobility followed by energy transfer. Qualitatively, the exciton states be considered either as (i) extended over a whole crystal or (ii) within a restricted region, with the ability to transfer the energy to the states with lower
energy, or (iii) as isolated, for which the probability of energy transfer supposed to be negligible. Both extended and isolated states contribute to absorption processes, whereas the optical emission comes mostly from spatially isolated localized exciton states. Using the Taylor series expansion for the averaged number of clusters in continual classical percolation over the overlapping circles, obtained in Ref. [4], we have calculated quantitatively the relative part of isolated states

\[ P(\omega) = \left[ \exp \left\{ -2P(\omega) \right\} + P(\omega) \exp \left\{ -2.841 P(\omega) \right\} + \ldots \right] , \]

where

\[ P(\omega) = \frac{1}{2} \left[ \frac{1}{\pi} \frac{1}{\mathcal{N}(\omega)} \right]^{1/2} \]

The integral density of states \( \mathcal{N}(\omega) \) gives the concentration of the potential wells with the localization energy restricted from one side by the \( \omega \) and from the other side by Lifshitz border [5]. The first term in the right hand side of Eq. (1) gives the relative number of spatially isolated potential wells, while the second term presents number of complexes consisting of pairs of “interacting” wells [4].

As a next step, the emission spectrum has been calculated, as a product of the relative part of isolated states \( P(\omega) \), and the emission probability, directly related to the absorption coefficient \( \alpha_{1s}^0(\omega) \). Taking into account also the contribution to the emission of the extended (i) and (ii) states we have

\[ I_{1s}^0(\omega) \sim \alpha_{1s}^0(\omega) \tau_{\text{rad}} \left\{ P(\omega) + \frac{\tau_{\text{rel}}}{\tau_{\text{rad}}} \left[ 1 - P(\omega) \right] \right\} , \]

where the first term in the brackets on the right hand side \( P(\omega) \) presents the fraction of the states with localization energy \( \omega \) belonging to the isolated clusters and to the ground states of the finite size complexes of clusters (superclusters)(iii). The second term is the part of the states(ii) which are mobile partially (higher states of superclusters) or completely (states (i) of percolation cluster) and have the possibility for relaxation. As a consequence, the last term is proportional to the ratio of the relaxation lifetime to the lifetime in respect to annihilation \( \tau_{\text{rel}}/\tau_{\text{rad}} \). All other condition being equal, the \( \tau_{\text{rel}} \) depends on the averaged energy which can be transferred in relaxation processes, therefore, it depends on the width of the inhomogeneous broadening of the exciton band due to fluctuations.

2. Experimental details, results and discussion

We have studied the absorption, photoluminescence (PL) and excitation of luminescence (PLE) spectra of three different sets of MBE-grown samples:

(i) MQW’s based on Ga\(_{1-c}\)In\(_c\)As/GaAs \( (c = 0.03 - 0.16) \) with wellwidth \( L_w = 6 - 9 \) nm, separated by barriers \( L_b = 30 - 90 \) nm;

(ii) ZnSe superlattices with 0.5 monolayer insertions of CdSe and 5 nm period;

(iii) single Zn\(_{0.92}\)Cd\(_{0.08}\)Se/ZnSe QW-structures with different wellwidth \( L_w = 5 - 40 \) nm.

Absorption and luminescence spectra of some of the (Ga-In)As/GaAs samples with different In concentration have been shown in semilogarithmic plot in the Fig. 1(a-c). The developed model has been applied for the analysis of the experimental spectra, and the results of model fitting are also presented in Fig. 1. The energy dependence of the spectral density for the \( n = 1s \) exciton state is shown by the curve 1, whereas the curve 2 shows the calculated luminescence spectrum. Curves 3 and 4 show the contribution into emission of isolated and extended states, respectively. The vertical dotted line corresponds to the
Fig. 1. (a)–(c): Absorption and luminescence spectra (open and solid symbols, respectively) of Ga$_{1-x}$In$_{x}$As/GaAs QW’s for $x = 0.03$, 0.06 and 0.16, respectively. Lines — the results of theoretical calculations (see text). Mobility edge positions (vertical dashed lines) correspond to the photon energies 1.4905 eV, 1.4629 eV and 1.4256 eV for $x = 0.03$, 0.06 and 0.16, respectively.

Fig. 2. Comparison of PL and PLE spectra of CdSe/ZnSe submonolayer superlattices (open and solid symbols, respectively) with the results of theoretical calculations (see text). Calculated position of the mobility edge. The energy scale shows the localization energy, measured from the expected position of the $n = 1$s exciton band bottom, not perturbed by fluctuations.

Experimental PL and PLE spectra of ZnSe superlattices with the submonolayer insertions of CdSe have been presented in Fig. 2. The growth conditions were corresponding to the insertion of submonolayers with 0.5 coverage, however the Cd atoms has been spreading over 4 to 6 lattice periods producing the average concentration of Cd in QW’s of order of 10% [6]. The results of comparison are shown in Fig. 2 with the meaning of curves similar to that in Fig. 1.

As it can be seen from the figures, the experimental results can be well described by presented model, assuming the existence of exciton mobility edge near of the main absorption band maximum in accordance with the data presented in [1] and [7]. Similar results have been obtained for the studied set of single ZnCdSe/ZnSe QW’s structures.
Summary

We have shown that the shape and Stockes shift of absorption and luminescence spectra of excitons in QW’s formed by different solid solutions can be describe within the uniform theoretical approach. It has been shown that due to the two-dimensional character of exciton motion in quantum wells the effect of compositional fluctuation on exciton states is much stronger than in the bulk solid solutions of the same composition.

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Time-resolved studies of exciton recombination in direct-gap GaAs/AlAs superlattices

V. G. Litovchenko†, D. V. Korbutyak†, S. G. Krylyuk†, H. T. Grahn‡ and K. Ploog‡
† Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, Prospect Nauki 45, 03028, Kiev 28, Ukraine
‡ Paul-Drude-Institut für Festkörperlelektronik, Hausvogteiplatz 5–7, D-10117 Berlin, Germany

Abstract. The temperature dependence of the recombination dynamics of excitons was investigated by time-resolved photoluminescence spectroscopy in direct-gap GaAs/AlAs superlattices. Peculiarities of excitonic photoluminescence decay were established in the dependence of the well width. In particular, it was shown that a reduction of the well width results in faster exciton recombination due to an enhancement of interface influence.

Introduction

GaAs/AlAs superlattices (SL’s) are considered to be a very perspective system for device applications, including light diodes and low-threshold lasers. For these purposes, detailed knowledge of the recombination processes which take place in SL’s is very important. Previously, it was shown that the measured radiative lifetime of free excitons in such structures lies in the nanosecond time regime, which significantly exceeds the expected value [1, 2]. Exciton scattering on alloy disorder, interface corrugations, etc. results in a decrease of the exciton coherence length so that the radiative lifetime is enhanced. A reduction of the well width leads to a strong increase of the exciton binding energy. At the same time, the influence of the interfaces on exciton recombination is significantly enhanced.

In this paper, we report a detailed study of the photoluminescence (PL) dynamics in several direct-gap GaAs/AlAs SL’s with different well thickness. In the SL with a layer thickness of the order of the exciton Bohr radius, the carrier dynamics was found to be dominated by the transfer from light-hole to heavy-hole excitons. When the layer thickness becomes smaller than the exciton radius, the dynamics of free excitons at low temperatures is controlled by localization.

1. Experimental

The (GaAs)$_n$/AlAs$_m$, where $n$ and $m$ denote the respective layer thickness in monolayers, were grown by molecular beam epitaxy on semi-insulating (100)-GaAs substrates. The well and barrier thicknesses were, respectively, $n = 9, 14, 21, 36$ ML’s and $m = 4, 7, 21, 36$ ML’s. We will label the samples as $n/m$ SL.

The time-resolved PL experiments were performed in a He-flow cryostat allowing a temperature variation between 4.2 K and room temperature. The samples were excited by an Ar$^+$-ion laser pumped, frequency-doubled Ti:sapphire laser operating at 3.37 eV with a pulse width of 150 fs. The PL signal was dispersed by a 0.22 m monochromator...
Fig. 1. (a) Time evolution of the PL spectrum in the 36/36 SL at 5 K. The two peaks at 1.555 and 1.541 eV are labelled A and B, respectively. (b) PL intensity vs time for two detection energies, which correspond to the two maxima of the PL spectrum.

using gratings with 1200 and 600 lines/mm. The temporal resolution of the PL signal was achieved by a streak camera system (Hamamatsu C5680) operating in synchro-scan and single-shot modes. The minimum time resolution is 2 ps for synchro-scan and 50 ps for single-shot operation. The spectral resolution is 0.2 nm for a 1200 lines/mm grating. The excitation power of the laser was adjusted to 70 µW.

2. Results and discussion

For direct-gap SL’s, the recombination takes places between electrons and holes at the \( \Gamma \)-point of the GaAs wells. A typical PL spectrum of a direct-gap SL consists of one narrow line due to the recombination of direct excitons, i.e., electrons and heavy holes are confined within the same well. However, if the splitting between the heavy- and light-hole subbands is less than LO-phonon energy, thermalization of unequilibrium holes into the heavy-hole subband is slowed and an additional PL line due to light-hole exciton recombination can be observed [3]. For the 36/36 SL, the PL spectra shown in Fig. 1(a) consist of two peaks at 1.555 and 1.541 eV labelled A and B, respectively. The energy separation of 14 meV is close to the calculated splitting of 18 meV between the heavy- and light-hole states. Since a typical variation of the confinement energy due to monolayer fluctuations is about 3 meV for one monolayer, we cannot explain the observed splitting by monolayer fluctuations.

The temporal evolution of the PL intensities of lines A and B at 5 K, which are shown in Fig. 1(b), is very different. While line A exhibits a biexponential decay with time constants of 100 ps and about 1.6 ns, line B first grows with a time constant of 230 ps and then decays with a time constant of about 2.5 ns. The initial decay of line A and corresponding increase of line B can be understood by the transfer from light-hole to heavy-hole excitons. The second time constants are then indicative of the radiative recombination lifetimes of both excitons. The transfer from light- to heavy-hole excitons has been observed previously for a 30/27 SL [4]. The transfer time was determined to be 90 ps, which is in a good agreement with our result. In order to identify the type of exciton, we have investigated the temperature dependence of the PL intensities of peaks A and B. An increase of the lifetime with increasing temperature would indicate that the recombination is dominated by free excitons.

A different situation occurs for the 21/21 SL. The PL spectrum of this SL shown in
Fig. 2. (a) Time evolution of the PL spectrum for the 21/21 SL at 5 K. (b) PL intensity vs time measured at 5, 40, and 70 K.

Fig. 2(a) consists of a single peak at 1.631 eV. Within the first 150 ps, a spectral red-shift of 3 meV is observed, which is typical for exciton localization, indicating a transfer from free to localized excitons. This conclusion is in agreement with results reported by Gurioli et al. [5] who observed a Stokes shift of 3 meV at 5 K for a similar sample.

The PL decay of this line can be well described by a biexponential decay as shown in Fig. 2(b). The fast decay is assigned to the transfer of free to localized excitons, while the slow component is determined by the recombination of localized excitons. The decay time due to free excitons increases somewhat from 250 to 500 ps, when the temperature is increased from 5 to 80 K. At the same time, the decay time assigned to localized excitons remains almost constant (about 1.4 ns) in the low-temperature region and decreases to about 1 ns for \( T > 30 \) K. If the temperature is larger then 70 K, the PL decay is well described by a single exponential with a shorter time constant. This observation implies that localized excitons become delocalized above this temperature.

Let us consider now the recombination dynamics in the SL’s with thinner wells (samples 14/7 and 9/4). Their PL spectra also consist of one narrow line due to heavy-hole exciton recombination. Note, that the direct bandgap in the 9/4 SL is caused by very thin barrier layers which leads to pushing up the AlAs-related \( X \)-level in the conduction band above the \( \Gamma \)-level of the well [6].

In contrast to the 21/21 SL, no temporal red-shift of the PL maxima is observed in the spectra of the 14/7 and 9/4 SL’s at low temperatures. PL intensities decay exponentially with time constants of 300 and 200 ps, respectively. It implies that the excitons in these SL’s are localized. However, starting from \( T = 80 \) K, a small temporal shift by 2–3 meV of the PL maximum towards lower energies takes place in the spectrum of the 14/7 SL. At the same time, the PL decay curves exhibit a biexponential behavior, indicating on progressive delocalization of excitons in this sample. On the other hand, in the 9/4 SL the PL intensity decays exponentially up to \( T = 100 \) K, which implies that excitons in this SL remain localized even at rather high temperatures.

3. Conclusions

Thus, if the well width is comparable with the exciton radius such as in the 36/36 SL, two separate PL lines are observed in the PL spectra, which are connected with heavy- and light-hole excitons. In SL’s with thinner quantum wells such as the 21/21 SL, the excitons
are already confined by the fluctuating potential of the hetero-interfaces. However, in the
time-resolved PL spectra, two lines are still observed indicating the rapid transition from
free to localized excitons. In SL’s with even thinner wells the influence of lateral potential
fluctuations due to interface roughness completely governs exciton recombination.

Another peculiarity is a rapid decrease (from about 2.5 ns to 200 ps for the 36/36
and 9/4 SL’s, respectively) of the PL decay time with the decrease of the well thickness,
which could be explained by a progressive influence of the interface centers of nonradiative
recombination.

References

Spectroscopy of the high energy quantum confined excitonic states in the thick GaAs quantum wells

I. A. Yugova†, V. G. Davydov‡§, Yu. K. Dolgikh‡, Yu. P. Efimov†, S. A. Eliseev‡, A. V. Fedorov‡£, I. Ya. Gerlovin‡, I. V. Ignatiev†§, I. E. Kozin†, V. V. Petrov†, V. V. Ovsyankin‡, K. Nishi§, H.-W. Ren§, S. Sugou§ and Y. Masumoto§¶

† Institute of Physics, St Petersburg State University, St Petersburg, Russia
‡ Vavilov State Optical Institute, St Petersburg, 190034, Russia
§ Single Quantum Dot Project, ERATO, JST, Tsukuba 300–2635, Japan
¶ Institute of Physics, University of Tsukuba, Tsukuba 305–8571, Japan
£ Venture Business Laboratory, University of Tsukuba, Tsukuba 305–8571, Japan

Abstract. Strong oscillations were observed in a wide spectral region in photo- and electroreflection spectra of heterostructures with the thick GaAs quantum wells. Different physical mechanisms of these oscillation are discussed. It is proved experimentally and theoretically that the observed phenomenon is due to the quantum size effect of the exciton in the GaAs quantum wells.

It is usually assumed that an epitaxial layer of GaAs with a thickness of a few tens nanometers can be considered as a bulk material. This conclusion based on the fact that the energy of the lowest excitonic state in the thick epitaxial layer coincides with bulk exciton energy. However, carriers in high quality epitaxial layers may have a free path larger than the layer thickness. In this case remoted heterointerfaces can become an important part of potential configuration as shown in [1].

In present paper we demonstrate that the quantum size effect is observable in epitaxial layers with the thickness up to 150 nm. We observed a large number of the quantum confined excitonic states in the photoreflectance (PR) and electroreflectance (ER) spectra of three samples with the thick GaAs quantum wells (QW’s). The studied heterostructures were grown by molecular beam epitaxy on n⁺ GaAs substrate (sample QDP1779) and on semiinsulating GaAs substrates (samples e187 and e188). QW’s in e187 and e188 with the thickness 50 and 100 nm, respectively, were embedded between AlₐGa₁₋ₐAs barrier layers. The QW in QDP1779 with the thickness 150 nm was embedded between In₀.₅₁Ga₀.₄₉P barrier layer and AlAs/GaAs superlattice.

The PR and ER spectra were measured using a radiation of a continuous wave tunable Ti:sapphire laser or dye lasers as a probe beam. The laser radiation with wavelength 530 nm or 800 nm was used as a pump beam in the PR experiments. The amplitude modulation of the pump and probe beams at different frequencies (1 MHz and 2 kHz, respectively) and a double lock-in detection of the signal modulated at the differential frequency allowed us to avoid noises from the scattered light and to detect fractional reflection changes as low as 10⁻⁷. In the case of ER, we applied a small ac voltage to the sample at a frequency of about 3 MHz. All experiments were done at the sample temperature 2 or 5 K.

PR spectrum of the sample QDP1779 is shown in Fig. 1. One can see the large number of quasi-regular oscillations which start from the resonance energy of the GaAs bulk exciton and continue up to the end of the studied spectral region. Energy positions of maxima and
minima of these oscillations versus their number $m$ are shown in Fig. 1(b). They can be well fitted by the phenomenological power law $m^{5/3}$. PR spectrum of the sample does not depend strongly on the wavelength of pump beam.

Fig. 2. ER spectrum of the sample QDP1779 measured at bias $-1.7$ V applied to the sample surface. The other is as in Fig. 1.

ER spectra reveal very similar behavior as seen in Fig. 2. We applied bias to the sample surface and found that the period oscillations does not depend on the external electric field. The amplitude of the oscillations drops at positive and at the strong negative bias. The oscillations amplitude in PR and ER spectra drops down at the elevated temperatures. The thermal shift of the energy positions of the oscillations appears to be almost twice the thermal bandgap shift of the GaAs.

Generally, a few possible physical mechanisms may be responsible for the oscillations observed in the PR and ER spectra. The simplest mechanism like the light interference in thin film can be easily excluded by the presented above data. The second possible mechanism could be a Franz–Keldysh effect in built-in electric field [2]. The electric field was really found in the studied sample as shown in the paper [3] where all observed oscillations were ascribed to the Franz–Keldysh effect. However, further experiments showed that only the oscillations near bulk GaAs exciton are due to the effect. Clear argument against this effect is given by the energy dependence of oscillation maxima $E_m$ (see Figs. 1(b) and 2(b)). This dependence in uniform electric field should be $E_m = E_g + m^{5/3}$ ($E_g$ is the band gap of GaAs) [2] that strongly contradicts to the observed dependence. Of course, inhomogeneity of electric field can result in some another energy dependence.
We theoretically studied this possibility for the case when electric field changes linearly across QW. We found that the inhomogeneity field may cause only a small deviation from “power law 2/3”.

We assume that the observed oscillations in the PR and ER spectra are caused by the quantum confined excitonic states with large quantum numbers. The observation of such large number of the excitonic states is possible if the free path of carriers is larger than the thickness of QW. Therefore our observation of many excitonic states is the evidence of the high structural quality of the sample. In the framework of this assumption, the energy dependence of maxima in the spectra is related with the energy dispersion for electron and hole in the bulk material. It is well known that the dispersion is quadratic for a small energy and linear for higher energy. Our fit by “power law 5/3” is a good approximation of these dependencies in a wide energy region.

To check this assumption we have grown and studied the samples e187 and e188 with QW’s that are thinner than in the sample discussed above. PR spectra of these samples are presented in Fig. 3. As seen the energy distance between the neighboring maxima depends strongly on the QW thickness. The features in the PR spectrum of the sample with the thinnest QW have a complicate shape.

In order to describe the experimental data, we developed a simple model. The dielectric function of quantum well \( \epsilon = \epsilon_1 + i\epsilon_2 \) has been calculated with regard to the width of
optical transitions, where

\[
\begin{align*}
\varepsilon_1 &= \varepsilon_\infty + \frac{2\mu |d_{cv}|^2}{\hbar^2 L} \sum_n \ln \left( \frac{\tilde{E}_n^2}{\Delta_n^2 + \gamma_n^2} \right), \\
\varepsilon_2 &= \frac{4\mu |d_{cv}|^2}{\hbar^2 L} \sum_n \left[ \pi^2 + \arctan \left( \frac{\Delta_n}{\gamma_n} \right) \right],
\end{align*}
\] (1) (2)

\(\varepsilon_\infty\) is the background dielectric constant, \(\mu\) is the reduced mass of electron and hole, \(L\) is the well thickness, \(d_{cv}\) is the dipole moment of interband transition, \(\tilde{E}\) is the energy parameter taking into account the finite widths of valence and conduction band, \(n\) and \(\gamma_n\) are the number and width of optical transition, \(\Delta_n = \hbar\omega - E_g - (\hbar^2 \pi^2 n^2 / 2\mu L^2)\), \(\hbar\omega\) is the photon energy. As is well-known[2], the response of modulation spectroscopy is proportional to the some derivative of the dielectric function with respect to the modulated parameter. Let us consider the case when the modulated parameter is \(\gamma_n\) and the change in the reflectivity (\(\Delta R\)) is determined by the imaginary part (\(\varepsilon_2\)) of dielectric constant; then

\[
\Delta R \sim \frac{d\varepsilon_2}{d\gamma} = -\frac{4\mu |d_{cv}|^2}{\hbar^2 L} \sum_n \frac{\Delta_n}{\Delta_n^2 + \gamma_n^2}. \] (3)

It is easily to see that this function oscillates with respect to \(\hbar\omega\) (Fig. 3(c)). The right zeros of \(\Delta R\) are determined by the conditions, \(\Delta_n = 0\), i.e. \(\hbar\omega_{0,n} = E_g + (\hbar^2 \pi^2 n^2 / 2\mu L^2)\). The same result is obtained in the case when the modulated parameter is \(\Delta_n\) and the change in the reflectivity (\(\Delta R\)) is determined by the real part (\(\varepsilon_1\)) of dielectric constant. For comparison of theoretical prediction and experimental data (Fig. 3(b)), the measured right zeros (solid squares) as a function of its number and the fitting curve have been plotted in inset in Fig. 3(c). It has been found that \(\hbar\omega_{0,n} - E_g = 3.6n^{1.966} \text{[meV]}\) for GaAs quantum well \((L = 50 \text{ nm})\). Despite the fact that theoretical model is very simple, the agreement with experimental data is satisfactory.

In conclusion, many quantum confined excitonic states are observed in the PR and ER spectra of the thick QW’s. The observed phenomenon opens up the wide possibilities for precise study of the physical properties of bulk GaAs material.

References
Ferroelectric-paraelectric phase transitions in P(VDF-TrFE) Langmuir-Blodgett films studied by optical second harmonic generation

Yu. G. Fokin†, T. V. Misuryaev†, T. V. Murzina†, V. M. Fridkin‡, S. P. Palto‡, L. M. Blinov‡ and O. A. Aktsipetrov†

† Department of Physics, Moscow State University, Moscow 119899, Russia
‡ Institute of Crystallography, Russian Academy of Sciences, Moscow 117333, Russia

Abstract. Ferroelectric-paraelectric phase transitions are studied in Langmuir–Blodgett (LB) films of vinylidene fluoride with trifluoroethylene (P(VDF-TrFE)) by an electrode-free method of optical second harmonic generation (SHG). The temperature dependences of the SHG intensity are studied for the LB films of the thickness down to a monolayer and two peculiarities attributed to two different ferroelectric-paraelectric phase transitions are observed. A qualitative correlation between the temperature dependences of the static dielectric constant and the SHG intensity is underlined. The results are interpreted in terms of the spontaneous polarization induced lack of the initially centrosymmetric structure of the film which gives rise to an electro-dipole SHG.
Fig. 1. The temperature dependence of the SHG intensity for a 60-monolayer-thick LB film. Filled and open symbols — the cooling and heating branches, respectively.

by bandpass filters and detected by a PMT and gated electronics. The p-in, p-plus-s-out (no analyzer) combination of polarization of the fundamental and second harmonic waves were measured in order to collect the depolarized and diffuse SHG radiation. The angular aperture of the detection system was approximately $10^{-1}$ sr. The temperature of the LB films placed into the optical cryostat was varied in the range from $-30^\circ$C up to $120^\circ$C at a rate of 0.5 deg./min.

The main panel in the Figure shows the temperature dependence of the SHG intensity under the cooling and heating for a 60 monolayer thick P(VDF-TrFE) LB film. A clear hysteresis in the SHG intensity can be seen as the direction of temperature variation is changed. A sharp peak was observed under the heating of the film in the vicinity of 10–15$^\circ$C, which can be related to a low-temperature ferroelectric-paraelectric phase transition observed previously by the static dielectric measurements. In [1] this low-temperature phase transition was attributed to the ferroelectric properties of the surface layer of the P(VDF-TrFE) film. The second specific point in the SHG temperature dependences was observed at 30–40$^\circ$C, which was determined as a maximum of the cooling branch of the SHG intensity temperature dependence. It corresponds probably to a ‘bulk’ ferroelectric-paraelectric phase transition [1]. Similar dependences were observed for the films of various thickness. For a qualitative description of the temperature dependence of the SHG intensity in P(VDF-TrFE) LB films, we consider a nonlinear-optical model of the ferroelectric LB structure which involves the non-linear optical sources located at the film interfaces and in the bulk of the LB film. In the paraelectric phase and in the electro-dipole approximation, the SHG is determined by the surface contribution to the nonlinear polarization which originates from the discontinuity of the structure of the film in the normal direction. For the ferroelectric phase, additional electro-dipole contributions from both the surface and bulk should appear. Thus the SHG intensity, $I_{2\omega}(T)$, which is proportional to the square of
the nonlinear polarization, $\vec{P}_{2\omega}(T)$, $I_{2\omega}(T) \propto |\vec{P}_{2\omega}(T)|^2$, is determined by the vector sum of the following contributions:

$$
\vec{P}_{2\omega}(T) = \hat{\chi}^{(2)}(T)\vec{E}_{\omega}\vec{E}_{\omega} + \hat{\chi}^{(3)}(T)\vec{E}_{\omega}\vec{E}_{\omega}\vec{P}_{sp}(T) \\
+ \hat{\chi}^{(2)b}(T)\vec{E}_{\omega}\vec{E}_{\omega} + \hat{\chi}^{(3)f}(T)\vec{E}_{\omega}\vec{E}_{\omega}\vec{P}_b(T),
$$

(1)

where the superscripts $s$ and $b$ are related to the surface and the bulk contributions, respectively, $\vec{E}_{\omega}$ is the electric field of the fundamental beam, $\vec{P}_{sp}(T)$ is the spontaneous polarization. Here the two types of the quadratic susceptibility tensors are described. First is the dipole second-order susceptibility tensor, which is always present at the surface of the film and which appears in the bulk of the film only in the ferroelectric state below the Curie temperature. This bulk term appears due to the modification of the crystalline structure of the film under the transition to the ferroelectric phase. Second, dipole third-order susceptibility tensors, which describe SP-induced contributions to $P_{2\omega}(T)$.

As the static dielectric constant $\varepsilon(T)$ is a linear function of SP, the comparison of the temperature dependences of the SHG intensity and of the static dielectric constant can allow to determine the role of the SP-induced terms in the SHG. The inset in the Figure shows the $\varepsilon(T)$ dependence (after [1]) measured for the 30 monolayer-thick P(VDF-TrFE) LB films. A clear qualitative correlation between these dependences shows that the SP-induced components to the nonlinear polarization play the main role in the temperature dependence of the SHG intensity. The differences in the values of temperature corresponding to the two types of the paraelectric-ferroelectric phase transitions and observed by the dielectric [1] and nonlinear-optical measurements, can be caused, first, by the interference of the terms of the nonlinear polarization in (1), second, by the different temperature scanning rate in the SHG experiments. It should be mentioned that a strong temperature dependence and hysteresis of the SHG intensity can not be explained by the variations of optical refractive index and the corresponding changes in the Fresnel coefficients caused by the temperature-induced variations of the static dielectric constant.

In summary, optical SHG is used as an electrode-free method to study the 2D ferroelectric-paraelectric phase transitions in LB films of P(VDF-TrFE). Temperature dependence of the SP-induced SHG intensity reveals two peculiarities which can be attributed to a 2D ferroelectric phase transition in the interfacial layers of the LB films and a thickness independent (almost 2D) ferroelectric phase transition in the bulk of the LB films.

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References


Anharmonic lifetime of H and D on the Si surface

I. P. Ipatova, O. P. Chikalova-Luzina, and K. Hess†
Ioffe Physico-Technical Institute, St Petersburg, Russia
† Beckman Institute, The University of Illinois, USA

Abstract. The study of H and D adatoms on the surface of Si by STM technique and by optical measurements indicate that anharmonic interaction of adatom localized vibrations with surface phonons of Si defines both the absorption line width and the yield of the desorption. The anharmonic lifetime of H and D adatoms is calculated.

STM studies of the Si surface terminated by H or D demonstrated that the lifetime of D-adatom is much shorter than the lifetime of H-adatom. At the low level of the excitation, the desorption yield demonstrates the temperature dependence which indicates on the essential role of the H or D vibrational anharmonic lifetime in the process of desorption [1].

The study of IR absorption by stretching localized vibrations of H adatoms on the surface Si (111) [2] has demonstrated the anomalous temperature dependence of the integrated absorption. The line intensity (integrated area) decreases by 20% as the temperature is raised above the temperature 130 K to 560 K. Authors of the paper [2] believe that the anomalous temperature dependence follows from the strong anharmonic interaction of H–Si stretching modes with H–Si bending mode.

All the facts of the case have something in common with those which were widely discussed in 60-es in connection with optical properties of U-center localized vibrations in alcali halides. It had been shown by Hughes [3] and Ipatova et al. [4] that anomalous temperature dependence of the integrated intensity can be understood to be due to the large difference of the localized frequency and the frequencies of the bulk modes of alcali halides (adiabatic approximation). It had been shown that the strength of the absorption by the localized mode has the exponential temperature dependence of the Debye–Waller type.

This paper deals with the theory of anharmonic line width and integrated absorption by localized vibrations of H- or D-adatoms on the surface of Si (111). The anharmonic interaction of Si–H stretching localized mode with the substrate vibrational Si modes is taken into account. Since the Si–H stretching mode frequency is \( \omega_{loc}(H) = 2085 \text{ cm}^{-1} \), stretching mode frequency of Si–D is 1516 cm\(^{-1} \) [5], and the maximum phonon frequency of Si is \( \omega_L(Si) = 514 \text{ cm}^{-1} \), the adiabatic parameter,

\[
\lambda = \frac{\omega_L}{\omega_{loc}} < 1, \tag{1}
\]

holds for both H–Si and D–Si adatoms. It means that due to anharmonic interaction the slow motion of Si-atoms occur in an averaged field of fast vibrations of light adatom. It is shown that there appear static displacements of Si atom equilibrium sites. Vibrations of the Si atoms occur about new equilibrium positions.

The shift of equilibrium positions results in the exponential temperature dependent factor, Debye–Waller factor, in both the integrated absorption and in the spectral line width of the light absorption by localized vibrations of the H or D adatoms. This exponential
factor depends on the temperature and on the mass of adatom. It is different for the localized vibrations of isotope complexes Si–H or Si–D.

We assume that the adatom (H or D) of the mass \( M' \) is situated on the (111) surface of Si. Adatom is supposed to be bound by the force constant in the “on-site” configuration to the surface atom of Si at the lattice site \( l = (l_x, l_y, l_z) = (0, 0, 0) \equiv 0 \). The position of adatom is \( l = (0, 0, 1) \equiv 1 \). The mass of the Si atom is \( M \).

The coefficient of the absorption of IR light at frequencies \( \omega \approx \omega_{\text{loc}} \) has the form [6]

\[
K(\omega) = -c_d \frac{e^2}{\hbar v_0} \text{Im} D_{\alpha\beta}^{R}(1, 1; \omega) \frac{E_\alpha E_\beta}{E^2},
\]

where \( c_d \) is the surface concentration of adatoms, \( v_0 \) is the volume of the crystal primitive cell and \( D_{\alpha\beta}^{R}(1, 1; \omega) \) is the one-particle retarded Green’s function of the adatom localized vibrations.

The retarded Green’s function \( D_{\alpha\beta}^{R}(1, 1; \omega) \) can be calculated by the temperature Green’s function technique. The temperature Green’s function \( D_{\alpha\beta}(1, 1; i\omega_n) \) with \( \omega_n = 4\pi n T/\hbar \) satisfies Dyson equation

\[
D_{\alpha\beta}(1, 1; i\omega_n) = D_{\alpha\beta}^{(0)}(1, 1; i\omega_n) + \sum_{l, l'} D_{\alpha\beta}^{(0)}(1, ls; i\omega_n) \Pi_{\gamma\delta}(ls, l'l'; i\omega_n) D_{\delta\beta}(l'l', 1; i\omega_n),
\]

where \( l, l' \) are positions of the crystal lattice sites; \( \Pi_{\gamma\delta}(ls, l'l'; i\omega_n) \) is the polarization operator produced by anharmonic terms in the potential energy of the system. Taking into account the square symmetry of the adatom position on the surface, one can find \( D_{\alpha\beta}(1, 1; i\omega_n) = D(1, 1; i\omega_n) \delta_{\alpha\beta} \) and \( \Pi_{\alpha\beta}(1, 1; i\omega_n) = \Pi(1, 1; i\omega_n) \delta_{\alpha\beta} \).

Adiabatic parameter, Eq. (1), allows to neglect in the sum over \( ls, l' \) in Eq. (3) non diagonal terms \( D(1, ls; i\omega_n) \) and to find \( D(1, 1; i\omega_n) \) from algebraic equation. The retarded Green’s function \( D(1, 1; \omega) \) is the analytic continuation in \( \omega \) of temperature Green’s function \( D(1, 1; i\omega_n) \). One can separate in \( \Pi(1, 1; \omega) \) the real and the imaginary parts, \( \Pi(1, 1; \omega) = \Delta(1, 1; \omega) - i\Gamma(1, 1; \omega) \), which are in charge of the anharmonic shift \( \Delta(1, 1; \omega) \) of localized frequency and the decay of the localized vibration \( \Gamma(1, 1; \omega) \).

The quantity \( \Gamma(1, 1; \omega) \) is defined by the processes which satisfy the conservation laws of the energy and the momentum. When the third order anharmonic term is taken into account, in the first approximation of the perturbation expansion, \( \Gamma(1, 1; \omega_{\text{loc}}) \) does not vanish only when \( \omega_{\text{loc}} < \omega_{\text{loc}} < 2\omega_{\text{loc}} \). The condition holds neither for H- nor for D-adatoms. In order to find \( \Gamma(1, 1; \omega) \) the anharmonic terms of the 4th order should also be taken into account. There are no limitations of this type for the shift \( \Delta(1, 1; \omega) \); all possible virtual anharmonic phonon processes contribute to \( \Delta \). As a result, \( \Delta(\omega) > \Gamma(\omega) \) and the shift \( \Delta \) is defined by the third order anharmonisity. The expansion of \( \Pi(1, 1; \omega) \) near \( \omega \approx \omega_{\text{loc}} \) gives

\[
D^R(1, 1; \omega) = \frac{1}{1 - \Delta'(\omega_{\text{loc}})} \left( \frac{\hbar}{M'} \right) \frac{1}{\omega - \omega_c(T) + i\gamma(\omega)},
\]

where

\[
\omega_{\text{loc}}^2(T) = \omega_{\text{loc}}^2 + \frac{\Delta(\omega_{\text{loc}})}{1 - \Delta(\omega_{\text{loc}})} \quad \text{and} \quad \gamma(T) = \frac{\Gamma(\omega_{\text{loc}})}{1 - \Delta(\omega_{\text{loc}})}.
\]
Here $\Delta'(\omega_{loc}) = \left( \frac{d}{d\omega} \right)_{\omega=\omega_{loc}}^2$.

Calculateds of polarization operator $\Pi(1,1;\omega)$ and corresponding $\omega_{c0}$ and $\gamma$ with the help of the temperature Green’s function technique allow to use the parameter Eq. (1) and to find the main contribution to $K(\omega)$

$$K(\omega) = c_d \frac{e^2}{\hbar v_0} \left( \frac{\hbar}{2M'\omega_{loc}} \right) e^{-2W(T)} \frac{e^{-2W(T)} \Gamma}{(\omega - \omega_{c0})^2 + (e^{-2W(T)} \Gamma)^2}.$$  \hspace{1cm} (6)

Here the quantity $W(T)$ is defined by the 3rd order anharmonic terms $\Phi^{(3)}$

$$2W(T) = \sum_{\lambda} \frac{\hbar}{2M'\omega_{loc}} \left( \frac{\hbar}{2M'\omega_{loc}} \right)^2 \Phi^{(3)} \left( 2N_\lambda + 1 \right) \frac{(\omega_{c0})^2}{(\hbar\omega_{c0})^2},$$  \hspace{1cm} (7)

where $\omega_{c0}$, $N_\lambda$ are the normal mode frequencies and the occupation phonon numbers of vibrations in semi-infinite Si, $\Phi^{(3)}$ is the closure of the anharmonic force constants with the phonon polarization vectors.

It follows from Eq. (6) that the quantity $e^{-2W(T)} \Gamma(\omega_{loc})$ is the line width of optical absorption by high frequency localized mode. At low temperatures $T < \Theta_D$, $\Theta_D$ being Debye temperature, the factor $e^{-2W}$ does not depend on temperature: $e^{-2W(0)} = \text{const}$. At high temperatures $T > \Theta_D$ the Debye–Waller factor is the linear function of temperature.

Two contributions to the quantity $\Gamma$ are known: the relaxation, $\Gamma_1$, and the dephasing, $\Gamma_2$. At low temperatures $T < \Theta_D$, the main contribution to the line width for H adatom $\Gamma(H)$ is shown to come from dephasing mechanism of broadening. $\Gamma_2(H)$ is vanishing as $e^{-(\hbar\omega_{c0}/T)}$ at $T \to 0$. In case of D-adatom, the dephasing $\Gamma_2$ defines the line width in the temperature interval $T > T_1$ only. At $T = T_1 \approx 60$ K, the relaxation broadening $\Gamma_1(D)$ becomes more important.

At low temperatures $T < \Theta_D$, the comparison of the line width for H- and D-adatoms shows that at $T > T_2 \approx 30$ K the main mechanism of broadening is dephasing $\Gamma_2$ for both H- and D-adatoms. Order of magnitude evaluation shows that $\Gamma_2(H) > \Gamma_2(D)$. At $T < T_2$ the relaxation broadening $\Gamma_1(D)$ becomes more important for D-line width and H-line width becomes less than D-line width: $\Gamma_2(H) < \Gamma_1(D)$. At high temperatures $T > \Theta_D$, the temperature dependence of the broadening is defined by $\Gamma(T) e^{-2W(T)}$ where $\Gamma(T)$ is dephasing process for both H- and D-adatoms. Numerical evaluations shows that at $T > 250$ K, the relaxation between the line widths, $\Gamma(D) e^{-2W(D)} > \Gamma(H) e^{-2W(H)}$, holds due to exponential factor.

The integrated absorption is equal to

$$J = \int_0^\infty d\omega K(\omega) = c_d \frac{e^2}{\hbar v_0} \left( \frac{\hbar}{2M'\omega_{loc}} \right) e^{-2W(T)}.$$  \hspace{1cm} (8)

It follows from Eq. (8) that the integrated intensity $J$ has exponential temperature dependence which is stronger in case of H-adatom than for D-adatom. Order of magnitude evaluation has shown that $J(H) \approx 2J(D)$ what is in the good agreement with experimental data from [6] where $J(H) = 1.9J(D)$ is obtained.

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Comparison of phenomenological models with a microscopic theory for semiconductor optical nonlinearities

I. Rumyantsev†, N. H. Kwong†‡, R. Takayama†‡ and R. Binder†
† Optical Sciences Center, University of Arizona, Tucson, AZ 85721, USA
‡ Cooperative Excitation Project, ERATO, Japan Science and Technology Corporation

Abstract. A microscopic theory of the third order coherent semiconductor optical response is used for the investigation of four-wave mixing signals. The theory is based on the dynamics-controlled truncation formalism and evaluated for the case of resonant excitation of heavy-hole excitons. This approach allows for a direct comparison with other theories (microscopic or phenomenological), and yields a detailed understanding of important many-body effects such as excitation-induced dephasing.

Introduction

The nonlinear optical response of semiconductors quantum wells has long been an important aspect of both theoretical and experimental semiconductor physics [1, 3]. The understanding of many-body effects such as excitonic effects and charge carrier-correlations is important for a correct interpretation of nonlinear optical response of quantum wells. Important examples of recent experiments include the observation of biexcitonic beatings in four-wave mixing (FWM) experiments [2] and strong polarization dependence of the FWM signal in semiconductor microcavities [4].

The theoretical description of optically excited semiconductors is usually based on various modifications and extensions of the so-called semiconductor Bloch equations, which are equations of motion for the wavevector-dependent interband polarization function and the charge carrier (i.e. electron and hole) distribution functions. The simplest approximation is the Hartree–Fock (HF) approximation, in which all correlation functions are factorized in terms of one-particle functions.

However, there exist several theoretical approaches for dealing with many-body processes beyond the HF approximation. One of them is the dynamics-controlled truncation (DCT) formalism, which is a rigorous approach in, for example, the $\chi^{(3)}$ nonlinear regime if one restricts the analysis to the coherent limit with zero electron and hole density initial conditions [5]. These equations can be written in terms of the interband polarization function and the two-exciton correlation function. If consistent with the excitation conditions, the equations can be restricted to contain only optically excited 1s-excitons. In that case it is advantageous to expand all functions in terms of exciton functions and keep only the smallest set of excitons, that is consistent with the excitation conditions [7].

While the numerical complications only allow for an inclusion of a few eigenstates (1s in our case), this is still well justified approximation for a resonant excitation, especially in semiconductor with large exciton binding energies are used or spectrally narrow pulses (e.g. picosecond pulses). From this approach one can extract and calculate the nonlinear optical response function and extract different quantities related to many-body interaction effects, e.g. phase-space filling, Hartree–Fock terms, coherent biexciton contributions and exciton continuum scattering contributions. These quantities can be directly compared with
the phenomenological parameters entering other models such as weakly interacting Boson (WIB) model (see e.g. [6]) or various few-level models. The comparison with the WIB model is done elsewhere [8], while the comparison with few-level models is discussed in this papers.

1. Theory and discussion

In this section we discuss both the microscopic theory based on the DCT formalism as well as the 3rd-order nonlinear optical response based on a commonly used phenomenological five-level model.

The five-level system corresponds to two two-level systems representing the heavy holes in the angular momentum states $|\pm 3/2\rangle$, the electrons in the spin states $|\pm 1/2\rangle$, and a phenomenologically included biexciton state.

Using an atomic density matrix approach one can write the general equation of motion for the density operator in the form

$$i\hbar \frac{d}{dt} \rho_{mn} = (\hbar \omega_{mn} - i\gamma) \rho_{mn} + \sum_{i\geq g} \rho_{ni} \rho_{im} + \sum_i (H'_{ni} \rho_{im} - \rho_{ni} H'_{im}),$$

(1)

where the interaction hamiltonian $H'_{mn} = -\vec{\mu}_{mn} \cdot (\vec{E} + L \vec{P})$, $\vec{\mu}_{mn}$ is the dipole matrix element, $\vec{E}$ is the optical field, and $\vec{P}$ is the spin-resolved polarization function. The many-body effects are accounted for by the inclusion of the biexciton level, the excitation-induced dephasing (EID) associated with the parameter $\gamma^i$, and the local field factor $L$. Iterating this equation to the third order one can derive the expression for the total polarization using $\vec{p} = e \text{Tr}(\vec{r} \rho) = \sum_{nm} \vec{\mu}_{nm} \rho_{mn}$. For the spin +1 polarization component one obtains

$$P^{(3)}_+ \propto -i \frac{2}{\hbar} \int_0^t dt' e^{-i\omega_0(t-t')} \left[ -i \gamma' (t') \rho_+(t') \rho_-(t') \right] + E_+(t')^2 \rho_+(t')^2 + \vec{L} \rho_+(t')^2 \\
- \frac{i|\beta|^2}{\hbar} \int_0^t dt'' e^{-i(2\alpha_0-\delta_0)(t-t'')} \left[ E^+_{\tau}(t')(E_+(t'')\rho_-(t'') + E_-(t'')\rho_+(t'')) \right] + \vec{L} \rho^+_{\tau}(t') \rho_+(t'') \rho_-(t'') \right] + \vec{L} \rho^+_{\tau}(t'') \rho_+(t'') \rho_-(t'') \right] + \vec{L} E^+_{\tau}(t') \rho_+(t'') \rho_-(t'') \right] + \vec{L} E^+_{\tau}(t'') \rho_+(t'') \rho_-(t'') \right] + \vec{L} E^+_{\tau}(t') \rho_+(t') \rho_-(t') \right] + \vec{L} E^+_{\tau}(t'') \rho_+(t') \rho_-(t'') \right] + \vec{L} E^+_{\tau}(t') \rho_+(t'') \rho_-(t'') \right].$$

(2)

A similar expression can be derived for the spin −1 polarization component. We use the notation $\rho_{\pm}$ to denote the first order density matrix (linear optical response) $E_{\pm} = \vec{\mu}_{\pm} \cdot \vec{E}$, $\vec{L} = L |\mu|^2 / V$, where $V$ is the volume or, as in our case of a quasi-two dimensional system, the total area of the system.
Four-wave mixing signal vs pump-probe delay time in co-polarized (dashed lines) and cross-polarized (solid line) configuration computed with the microscopic (a) and phenomenological (b) theory.

We now turn to the microscopic formulation. The equation for the third-order polarization that one can be obtain from the DCT equations in terms of exciton wave functions can be written in the following form:

\[
p^{(3)}(t) = \frac{-i}{\hbar} \int_{t_0}^{t} dt' e^{-i\omega_{xx}(t-t')} \left[ (2C_{1s}E_+(t')|p_+(t')|^2 + V^{(3)}|p_+(t')|^2 \right] \\
- \frac{i}{\hbar} \int_{t_0}^{t'} dt'' e^{-i(2\omega_{xx}-\delta_{xx})(t'-t'')} f_0 p_-^*(t') p_+(t'') p_-(t'') \\
+ 2 f^+(t'-t'') p_+^*(t') p_+(t'') p_+(t'') \\
+ [f^+(t'-t'') + f^-(t'-t'')|p_-^*(t') p_+(t'') p_-(t'')]. \tag{3}
\]

Here, \( p \) is the first order coherence function. The two terms on the first line of this equation represent the phase space filling (PSF) and Hartree–Fock (HF) contributions. This is followed by the bound biexciton term on the 2nd line, (restricted to the singlet channel) and the rest of the equation contains the continuum scattering terms, with contributions in both triplet and singlet channels. Here “singlet” \((f_0 and f^-)\) and “triplet” \((f^+)\) refer to the electron spin states. The definitions of the parameters \(C_{1s}\) and \(V^{(3)}\) (see, e.g., [10]), as well as \(f_0\) and \(f^\pm(t'-t'')\) are lengthy and not given here, they include multiple integrals with exciton eigenfunctions, the Coulomb potential. The biexciton contribution and continuum contribution in the singlet channel contain also the biexciton wave function (which can be found in Ref. [11]).

Equations (3) and (2) are a suitable basis for a detailed comparison of parameter entering the phenomenological model with the interaction terms derived from the microscopic theory. Whereas a strict one-to-one correspondence does not exist, one can indeed identify approximate correspondences and, thus, validate to a certain extent the phenomenological model. As far as numerical agreement is concerned, we show in Fig. 1 results for time-integrated four-wave mixing signal in a ZnSe semiconductor quantum well.

In conclusion, we present a theoretical analysis of ultrafast optical nonlinearities in semiconductors. We believe that our detailed comparison of phenomenological model
with a microscopic theory is important in order to validate conclusions about physical processes obtained from the widely used phenomenological models.

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References


Carrier accumulation due to insertion of nanoscale As clusters into n- and p-type GaAs

P. N. Brunkov†, V. V. Chaldyshev†, A. V. Chernigovskii†,
A. A. Suvorova†, N. A. Bert†, S. G. Konnikov†,
V. V. Preobrazhenskii‡, M. A. Putyato‡ and B. R. Semyagin‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institute of Semiconductor Physics, SB RAS, 630090 Novosibirsk, Russia

Abstract. Accumulation of electrons and holes has been revealed by capacitance-voltage technique in As-cluster containing GaAs layers sandwiched between n-type or p-type GaAs buffers. As a result of the majority carrier accumulation, a large depletion region forms in adjacent buffers. Simulation of the capacitance-voltage characteristics based on numerical solution of the Poisson equation showed the concentration of accumulated charge carriers to be as high as $\simeq 1 \times 10^{12} \text{ cm}^{-2}$ which is comparable with concentration of As clusters determined from transmission electron microscopy study.

Introduction

Gallium arsenide containing nanoscale arsenic clusters (so called LT-GaAs) has attracted much attention in the past few years due to its unique properties, such as a very high electrical resistivity and extremely short (femtoseconds) carrier lifetime [1–4]. While this material has found several device applications, the nature of its electronic properties is still under discussion. Our previous investigations showed that the As-cluster containing material can accumulate electrons from adjacent n-GaAs layers [5, 6]. In this paper we report on capacitance-voltage (CV) study of Schottky barrier structures where LT-GaAs layer is sandwiched between p- or n-type GaAs buffers. We elucidate fundamental difference in the electron and hole accumulation in LT-GaAs layers.

1. Experimental details

N(P)-type structures consisted of three layers 0.5 $\mu$m–thick n(p)–GaAs/0.1 $\mu$m–thick LT–GaAs/0.5 $\mu$m–thick n(p)–GaAs and were grown by MBE on n$^+$ (p$^+$)–substrate with (100) orientation, where n(p)-type buffers were grown at 580$^\circ$C and doped with Si(Be) up to concentration of $2 \times 10^{16} \text{ cm}^{-3}$. The LT-GaAs layer was grown at 200$^\circ$C and doped with the same concentration of shallow impurities. These growth conditions result in a high arsenic excess ($\simeq 1.5 \text{ at.}\%$) in the LT-GaAs layer. Arsenic clusters were produced in LT-GaAs matrix during the growth of the upper buffer layer. The P-type structure was capped by a 38 nm–thick GaAs/AlAs (1 nm/1 nm) short-period superlattice (SPSL). Alloy of AuGe (AuZn) was evaporated and alloyed on the n$^+$ (p$^+$)–substrate to produce ohmic contact. The Au circular Schottky contacts had a diameter of 0.4 mm and 0.5 mm for the N- and P-type structures, respectively.
Ordered Arrays of Nanoparticles

2. Results and discussion

Figure 1 shows a cross-sectional transmission electron-microscope (TEM) image of the P-type structure. It can be seen that the LT-GaAs layer containing arsenic clusters is as thick as \( \approx 0.1 \mu m \) and is sandwiched in between two cluster-free p-layers. The sheet As cluster density \( N_{cl} \) is \( \approx 6 \times 10^{11} \text{ cm}^{-2} \), and their averaged diameter is 5–7 nm. TEM image of N-type structure reveals As clusters with \( N_{cl} \approx 4 \times 10^{11} \text{ cm}^{-2} \) and a diameter of 6–8 nm \[6\]. It should be noted that no extended defects have been observed by TEM in the both P- and N-type structures.

CV-characteristics of N- and P-type structures are shown in Figs. 2(a) and 3(a), respectively. Apparent concentration profiles (Figs. 2(b) and 3(b)) were calculated from the CV characteristics using the depletion region approximation:

\[
N_{CV}(W) = \frac{C^3}{q \varepsilon \varepsilon_0 (dC/dV)} W = A \frac{\varepsilon \varepsilon_0}{C}
\]  

where \( q \) is the charge of electron, \( \varepsilon_0 \) is the dielectric constant of the vacuum, \( \varepsilon \) is the
Fig. 3. CV characteristics (a) and apparent concentration profiles $N_{CV}(W)$ (b) of P-type structure: experimental data ($T = 290$ K (○) and $T = 77$ K (●), measurement frequency 10 kHz) and model simulations with different concentration of the charge carriers in LT-GaAs layer $N_{LT}^Q$: $6.0 \times 10^{11} \text{ cm}^{-2}$ (dotted), $8.0 \times 10^{11} \text{ cm}^{-2}$ (full), $1.0 \times 10^{12} \text{ cm}^{-2}$ (dashed). The insert in Fig. 3(b) shows the position of LT-GaAs layer.

dielectric constant of the semiconductor, $W$ is the depth from the surface of the structure, and $A$ is the area of the Schottky barrier contact.

One can see from Figs. 2(a) and 3(a) that CV characteristics of both structures are similar at low temperature. The specific feature of the CV characteristics is an abrupt drop of the capacitance when depletion region from the surface of the structure reaches a wide built-in depletion region induced by the cluster containing LT-GaAs layer. Using numerical solution of the one-dimensional Poisson equation [7], we have simulated the CV and $N_{CV}(W)$ characteristics of both N- and P-type structures at low temperatures (Figs. 2 and 3). It was found that the depletion around LT-GaAs layer arises from the accumulation of the majority carriers. The best fit of the experimental curves was obtained with a concentration of accumulated electrons of $N_{LT}^Q \approx 1 \times 10^{12} \text{ cm}^{-2}$ in N-type structure and a concentration of accumulated holes of $N_{LT}^Q \approx 8 \times 10^{11} \text{ cm}^{-2}$ in P-type structure. The concentrations of the accumulated carriers in the LT-GaAs layers are comparable with the concentrations of the As clusters obtained from TEM study.

With elevating temperature a pronounced difference appears between CV characteristics of the N- and P-type structures. In the case of N-type structure (Fig. 2(a)) after an abrupt drop of capacitance at 1.0 V one can see a wide plateau in the range from 2 V to 12 V. The plateau on the CV characteristic (Fig. 2(a)) relates to electron emission from the LT-GaAs layer [5, 6]. In contrast, in the case of the P-type structure (Fig. 3(a)) no hole emission can be observed at temperatures $\leq 300$ K. From the analysis of the frequency and temperature dependencies of the CV characteristics the electron emission rate is found to be higher than the hole emission rate by at least several orders.

3. Conclusions

We studied CV characteristics of N- and P-type sandwich structures where As-cluster containing LT-GaAs layer was inserted between n-type or p-type GaAs buffers. Majority carrier accumulation was revealed in the LT-layers. Accumulated carrier concentration was found to be comparable with arsenic cluster density determined by TEM. A strong
difference between electron and hole emission rates from cluster-containing LT-GaAs layers was observed.

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References


Fabrication and structural studies of “opal–III nitrides” nanocomposites

V. Yu. Davydov†, V. G. Golubev†, N. F. Kartenko†, D. A. Kurdyukov†,
A. B. Pevtsov†, S. M. Samoilovich‡ and N. V. Sharenkova†
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Almaztechnocrystal Ltd., Aleksandrov, Russia

Abstract. Three-dimensional system of GaN and InN nanoclusters have been synthesized for the first time in a void sublattice of artificial opal. Structural studies of the samples by X-ray diffraction phase analysis and Raman spectroscopy were carried out.

Introduction

It was recently [1] proposed to use synthetic opals as matrices for obtaining three-dimensional (3D) arrays of electronic nanodevices. In [2–4] 3D regular systems of Si and Si-Pt nanostructures have been fabricated in a void sublattice of synthetic opal. The results obtained demonstrated an ability of creating 3D multilayer p-n junctions and Schottky barriers on the inner surface of opal voids. This approach is expected to yield a higher (by six orders of magnitude, up to $10^{14} \text{ cm}^{-3}$) density of active elements as compared to that produced by state-of-the-art planar technology.

In this work the synthetic opals were infiltrated by GaN and InN for the first time. Group-III nitrides based structures are highly attractive materials because of their great potential for development of optoelectronic devices [5]. Use of opal matrices enable to reach the working area of the junctions per unit volume in III nitride-based LED as high as $10 \text{ m}^2/\text{cm}^3$. This will allow reducing the current density in such nanodevices by 3–4 orders of magnitude as compared with the conventional planar systems. It is also to be expected that the photonic-crystal electromagnetic structures of the opal matrices, characterized by the presence of an stop band, will lead to novel photonic effects in these nanodevices [6, 7].

1. Experimental

The perfect “monocrystals” of synthetic opals consist of 230 nm diameter close packed amorphous silica spheres and have a regular sublattice of voids (45–90 nm) up to 26% accessible to filling by other substances [1].

To incorporate III nitrides into opal voids we used chemical heterogeneous reaction of solid precursors containing group III elements preliminary embedded inside opal voids with gaseous nitrogen hydrides.

The microstructure of substances was determined by X-ray diffraction (XRD) and Raman measurements. The XRD patterns were obtained by using Cu$K_a$ radiation (Ni filter). Polycrystalline germanium was used as a standard. The Raman spectra were measured at room temperature under Ar$^+$ laser (488 nm) excitation.

2. Results and discussion

Figure 1 shows XRD patterns of an “opal-GaN” nanocomposite (a) and a hexagonal bulk GaN (b). As evident from these diffractograms the material synthesized in the opal is the
hexagonal GaN. The lattice parameters were found to be 3.18(1) Å and 5.19(1) Å, which are in good agreement with data available for bulk GaN [8]. The average size of GaN crystallites were determined by the approximation method. It was found from the angular dependence of the “half-width” of reflections that the physical broadening of the diffraction lines is only caused by a small size of the crystallites. The crystallite average size calculated from the measurements of the “half-width” of 101 and 100 GaN reflections is 180(20) Å.

Figure 2(a) shows the Raman spectra of a “opal-GaN” composite (1) and a hexagonal polycrystalline GaN (2). Experimental data are compared with the one-phonon density-of-states (DOS) function of the hexagonal GaN crystal (Fig. 2(b)) [9]. There are the two features in Raman spectrum of the polycrystalline sample: the high-frequency line centered at 740 cm$^{-1}$ results from the longitudinal optical phonons, the low-frequency feature ranging in 530–570 cm$^{-1}$ is attributed to the transverse optical phonons. The phonon modes of the opal-GaN sample are shifted and broadened with respect to the lines of the polycrystalline GaN. Such behavior is due to a spatial confinement of optical phonons when the crystal has a finite dimension. As a result phonons with the wave vector $q \neq 0$ add a contribution to the first-order Raman-scattering spectrum at energies $\hbar\omega$ that are determined by the dispersion relations $\omega(q)$. Based on the above reasoning the Raman spectra of nanocrystalline materials are to reflect a phonon DOS which is similar to the one-phonon DOS in a bulk crystal. Thus, the observed low-frequency shift of the LO phonon line towards the phonon DOS maximum and broadening of this mode in the opal-GaN sample as well as the transformation of TO mode correspond to a nanocrystalline structure of GaN formed in the opal voids.

Figure 3(a) shows a XRD pattern of hexagonal InN in opal voids. The determined lattice constants of InN ($a = 3.54(1)$ Å, $c = 5.68(3)$ Å) are in agreement with those of bulk InN [8]. The Raman spectra of the “opal-InN” sample is not detected because Raman-scattering cross-section of InN is essentially (at least on one order of magnitude) less than that of GaN.

3. Conclusion

It has been shown that hexagonal nanocrystalline GaN and InN can be synthesized by chemical methods directly within a 3D regular void sublattice of artificial opals. We hope
that this work is the first step to create 3D systems of efficient optoelectronic nanodevices with a high density of elements and a large working area per unit volume based on artificial opal matrices.

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Copper and cobalt nanoclusters embedded in hydrogenated amorphous carbon: an X-ray absorption study

A. V. Kolobov†‡§, H. Oyanagi‡, H. Akinaga†, T. K. Zvonaryova§ and V. I. Ivanov-Omskii§

† Joint Research Center for Atom Technology - National Institute for Advanced Interdisciplinary Research, 1-1-4 Higashi, Tsukuba, Ibaraki 305, Japan
‡ Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki 305, Japan
§ Ioffe Physico-Technical Institute, St Petersburg, Russia

1. Introduction

Investigation of metallic clusters in insulating media, also called discontinuous or granular metals has recently attracted much interest. This interest was further stimulated by possible applications in single-electron devices. Recently, confinement of copper nanoclusters in hydrogenated amorphous carbon (α-C:H) was reported [1] The structure of α-C:H is a mixture of sp2 and sp3 bonded carbon species [2]. The presence of sp3 bonding results is high hardness and optical transparency while incorporation of a metal ensures conductivity. Confinement of magnetic species in a dielectric matrix is of special interest for spin-electronic applications [3].

In this paper we report the results of X-ray absorption study (EXAFS and XANES) of the dependence of the structure of clusters on the concentration of the metal and on annealing.

2. Experimental

The α-C:H(Cu, Co) films were grown on fused quartz and/or crystalline silicon substrates by ion (magnetron) co-sputtering of graphite and copper targets in argon-hydrogen (80% Ar and 20% H2) plasma. The substrate temperature, gas pressure in the growth cell and average magnetron power were 500 K, 10 mtorr, and 0.4–0.5 kW, respectively. The energy of Ar+ ions was 350–450 eV. The metal concentration was varied from 0 to 30%. For more details on preparation see [1].

The measurements were performed at BL13B station at the Photon Factory using a 27-pole wiggler in a fluorescence mode. An array of 19-element high-purity Ge solid-state detectors was used to detect the fluorescence. Details of the equipment are given in [4]. Cu K-edge EXAFS and XANES spectra of α-C:H(Cu) were measured at 300 K. As reference samples we have also measured the spectra for bulk metallic copper and for Cu2O.

3. Results

The EXAFS oscillations, after subtraction of smooth backgrounds due to the atomic absorption from the fluorescence yield spectra, were multiplied by \( k^2 \chi(k) \) and Fourier-transformed (Fig. 1) using the region extending from 4.5 to 15 Å\(^{-1}\). A spectrum for bulk Cu is shown for comparison. One can see that for the as-made samples there is basically no peak corresponding to Cu–Cu interaction. Peaks at \( R \sim 1 \) Å are most likely due to poor background subtraction for a very dilute sample. A broad peak is observed in these samples at distances smaller than the Cu–Cu bond length. Curve-fitting analysis gives the
average Cu–Cu coordination number of $\sim0.5$ for those samples. Figure 2 shows XANES spectra for the measured samples and for the standards (bulk Cu and Cu$_2$O).

Upon annealing, a peak situated at $\sim2.1$ Å which corresponds to the Cu–Cu first shell appears and grows. Further annealing results in appearing of peaks corresponding to higher shells. The XANES spectra are also modified and approach that for bulk copper.

Figure 3 shows XANES spectra for as-made cobalt-containing samples. The change in the spectra with the Co concentration is similar to that reported earlier for Cu. We have not found any magnetic-field effect in the as-made 25% Co sample. The conductivity, however, was found to drop by several orders of magnitude when $\sim40$ V were applied to the sample which we believe was due to modification of the contact region.

4. Discussion

The observed coordination number of $<1.0$ in as-made samples can indicate either a complete absence of Cu–Cu correlations or probably also the formation of small portion of Cu dimers. The accuracy in determination of the coordination number does not allow us to unambiguously distinguish between single atoms and dimers.

It is not quite clear from the present study whether copper in this geometry mainly interacts with carbon or oxygen species of the surrounding matrix. The fact that the XANES spectra resemble that of Cu$_2$O and non-existence of Cu–C compounds support the latter possibility. The presence of Cu$_2$O has also been confirmed by IR spectroscopy.

Upon annealing, Cu–Cu correlations are clearly seen as a peak at 2.1 Å corresponding to the first-nearest and more distant shells are also seen. The obtained coordination number for a cluster in a sample annealed for 1 h is 4$\pm$2 and the Debye–Waller (DW) factor is 0.08 Å (compared to 0.045 Å in bulk copper). The Cu–Cu bond length equals 2.54 Å which is 0.02 Å shorter than in the bulk metal. There are various reasons which may account for a coordination number in clusters being smaller than that in the bulk. They include cluster size dependence of photoelectron mean free path, anharmonicity of the atomic potential, and pure geometrical factor (atoms which are at the surface of the cluster only have copper neighbours on one side), the latter being probably the most important. With the obtained

![Graph](image_url)

**Fig. 1.** Fourier transforms of EXAFS spectra for as-made and annealed samples with 12% Cu and that for bulk Cu.
coordination number the cluster size can be estimated as being 5 to 20 Å assuming the spherical shape of the cluster [5, 6].

For cobalt, only as-made samples have been studied so far and the observed change in the spectra suggests increased Co–Co correlations in samples with higher Co concentrations. There is a systematic change in the spectrum indicating a change in the density of states. This change is similar to that for copper. Further studies or annealed samples are needed and are currently underway.

5. Conclusion

Application of EXAFS and XANES spectroscopies have revealed that in as-prepared samples containing up to 24% copper the metal atoms is randomly distributed throughout the film with possible formation of a small number of dimers. Upon annealing copper clusters are formed.

For cobalt, we observed also Co–Co correlations in as-made samples but it is not yet clear whether nanoclusters are formed upon annealing.
Fig. 3. Normalized XANES spectra for as-made samples containing copper (left) and cobalt (right)
in different concentrations.

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References
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Magnetic force microscopy of Fe nanoparticles buried into SiO₂

D. V. Ovchinnikov†, A. A. Bukharaev† and R. Wiesendanger‡
† Kazan Physical Technical Institute, RAS, Russia
‡ Institute of Applied Physics of Hamburg University, Germany

Abstract. The MFM-images of the array of separately placed α-Fe nanoparticles buried into SiO₂ formed by ion bombardment have been obtained for the first time. The method of the computer analysis of MFM-images with consideration of the shape and magnetic properties of MFM-tip apex has been developed and applied for analyzing of the obtained magnetic images.

Introduction

Nanostructured surface magnetic materials have been being objects of close interest for investigating for several last years due to the perspective of creation of quantized magnetic disks based on such media. The magnetic force microscopy is a powerful instrument to study for such objects due to its high potential possibilities in investigating of magnetic properties of surface structures with nanometer resolution. However the images obtained with magnetic force microscope (MFM) consist of the complicated convolution of the magnetic and shape features of the MFM tip apex with magnetic field created on tip by the sample magnetic nanostructures. That is why it is necessary to hold further analysis based on computer simulation of MFM-images to interpretate the obtained results.

In this paper there are presented for the first time the results of MFM investigations of SiO₂ stripes on silicon containing ultrafine ferromagnetic α-Fe particles formed by ion bombardment. Previously [1] we have studied for magnetic and optic properties of such samples with the following methods: FMR, Mössbauer spectroscopy, optic spectroscopy, magnetooptics, X-ray analysis and electron microscopy. In order to interpretate the obtained MFM-images, the computer method of analysis has been developed, which was firstly presented for analysis of the MFM-images of single domain Ni nanoparticles [2]. The consideration of shape and magnetic features of MFM-tip close to the real ones has been made as the improvement of the computer method of analysis mentioned above.

Experiments and discussions

In spite of good magnetic properties of Fe the formation of ferromagnetic materials based on it meets some difficulties connected to bad chemical features of Fe which is quickly oxidized in the presence of Oxygen. In this work the investigated magnetic samples based on Fe were formed in the following way. The stripes of SiO₂ formed on silicon with about 100 nm in height and 1500 nm in width (Fig. 1(a,c)) were bombarded by Fe⁺ ions with energy of 40 keV and doze of 1.4 × 10¹⁷ ions/cm², afterwards α-Fe ferromagnetic nanoparticles with typical sizes from 60 to 80 nm and disk-like shapes with diameter-to-height ratio from 1.5 to 2.5 were formed on the depth up to 60 nm (Fig. 1(b,d)).

Nanoscope IIIa scanning probe microscope working in the oscillating mode [3] has used to investigate the topography and magnetic properties of the described sample. The MFM-tips were magnetized along their axis of symmetry beforehand. Fig. 1(e) presents the MFM-image of single stripe after ex situ magnetizing along X direction with the external magnetic...
Fig. 1. The topography, structure and MFM-images of the sample with α-Fe nanoparticles formed by ion bombardment buried into SiO₂ stripes. (a) The topography of a sample surface area; (b) the structure of a stripe and (c) topography of the stripe (top view); (d) the image of α-Fe nanoparticles array into SiO₂ obtained with electron microscopy; (e), (f) the MFM-images of the stripe after ex situ magnetizing of the sample in the directions marked by arrows. Insets present the MFM-signal profiles along the accentuated lines.

field of about 3000 Oe. Such strong MFM-signal is a result of rather high magnetization of ferromagnetic nanoparticles and can be explained by the fact that the most of the particles are in single domain state that is confirmed by the previous investigations [1]. After applying of the magnetic field in opposite direction the MFM-signal has inverted (Fig. 1(f)), that is caused by the reverse of total magnetic moments of every Fe nanoparticle. Such MFM-signal reverse was also observed when the external magnetic field of the smaller magnitudes (up to 300 Oe) was applied, so one we can say that the magnitude of the sample coercivity is not more than 300 Oe at least, which is also in agreement with data obtained in [1].

From the obtained MFM-images of the single stripe it is clear that the signals of separate particles cannot be distinguished, moreover the profile of the MFM-signal has rather large
width. The computer program for simulation of MFM-images with taking into account the shape and magnetic features of MFM-tip apex has been developed to explain these facts.

From the theory of MFM-response [3] it is known that MFM-image presents the distribution of $z$-component of gradient of magnetic force of interaction between tip and sample which is determined by the equation (in case of absence of currents):

$$F'_z = \int \int \int_{\text{tip}} \frac{\partial}{\partial z} \left[ M_{\text{tip}}(\mathbf{r}) \cdot \nabla \right] H_z(\mathbf{r}) dV_{\text{tip}}. \quad (1)$$

Here $M_{\text{tip}}(\mathbf{r})$ — tip magnetization in $\mathbf{r}$ point, $H_z(\mathbf{r})$ — $z$-component of the magnetic field created by the sample on the tip in $\mathbf{r}$ point. The model of dipole-dipole interaction [2] when magnetic volumes of the tip and the sample are divided into physically small fragments of cubic shape and each fragment is replaced by the point magnetic dipole has been used. So the Eq. (1) can be rewritten by sum of interactions between the separate dipoles:

$$F'_z = \sum_{i-\text{tip}} \sum_{j-\text{sample}} \frac{\partial}{\partial z} \left[ M_{\text{tip}}^{ij}(\mathbf{r}^{ij}) \cdot \nabla \right] H_z^{ij}(\mathbf{r}^{ij}). \quad (2)$$

Here the summarizing is held among $i$-th cubs of the tip and $j$-th cubs of the sample.

The held computer simulation was based on Eq. (2) with the approximation of the nanoparticles array by thin ferromagnetic film which was divided into small cubic fragments with 50 nm in size. Figure 2 presents the results of the simulation of the profiles of the MFM-images across the single stripe held in the case of that the tip height in lifting mode was 100 nm and it was uniformly magnetized along $Z$-axis and all of the particles were in single domain state and their total magnetic moments were ordered along $X$ direction. Figure 2(b) presents the MFM-image profile in case of tip approximation by point magnetic dipole. It is clear that magnetic signals from the separate particles are not distinguished that correlates to experimental data (Fig. 2(a)) but there is no agreement in width of the corresponding curves of the magnetic signals. To explain the observed experimental wideness the MFM-tip has been approximated by the cone covered with magnetic coating and then by the truncated cone with magnetic coating. We can observe the consequent slight increase of wideness in both cases (Fig. 2(c, d)) but it not enough to explain the experimental results. We think that some not correspondence of simulated MFM-images to experimental results may be due to that we haven’t yet considered the fact that MFM-tip can have complicated non-uniform structure of magnetization. The further additional investigations are necessary to check this affirmation.
Conclusion

Thus, in this work the MFM-images of the array of separately placed $\alpha$-Fe nanoparticles buried into $\text{SiO}_2$ formed by ion bombardment have been obtained for the first time. The method of the computer analysis of MFM-images with consideration of the shape and magnetic properties of MFM-tip apex has been developed and improved. It has been applied to analyze the obtained magnetic images. Some not correspondence of the results of the computer simulation to the experimental data may be explained by necessity of the further consideration of internal nonuniform magnetization structure of the MFM-tip.

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References


Oriented silver halide nanocrystals embedded in crystalline alkali halide matrix as studied by EPR and ODMR

N. G. Romanov, R. A. Babunts, A. G. Badalyan, V. A. Khramtsov
and P. G. Baranov
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Shallow electron centers, self-trapped holes and self-trapped excitons in small AgCl crystals embedded in KCl matrix were investigated by optical spectroscopy, optically detected magnetic resonance (ODMR), and electron paramagnetic resonance (EPR) techniques. These investigations showed that single AgCl microcrystals are formed in KCl crystalline matrix with their lattice symmetry axes oriented parallel to the axes of the host lattice. Multiquantum ODMR spectroscopy was used for the investigation of the effect of confinement on singlet-to-triplet splitting of self-trapped excitons and on the parameters of shallow electron centers.

Introduction

Semiconductor and solid-state physics nowadays appears to be the physics of systems with the reduced dimensionality. The fabrication of single or periodic potential wells by simply combining two materials of different bandgap energies and having spatial dimensions confining the motion of electrons and holes, results in many impressive possibilities for engineering of the new properties of solid-state structures.

It has long been known that in alkalihalide crystals doped with silver halides the silver ions Ag\(^+\) substitute for alkali ions and both single Ag\(^+\) ions and pairs of silver ions can be observed. Under ultra-violet (UV) light or x-ray irradiation a number of different silver-related defects can be formed: Ag atoms in cationic and anionic positions, Ag\(^-\) in anionic position, Ag\(^{2+}\) ions, Ag atoms near anionic vacancies (laser active A\(_F\) centers), silver pairs Ag\(^{+}\)\(_2\) [1, 2]. Along with Ag-related point defects oriented silver halide nanocrystals (down to 5 nm) embedded in crystalline alkali halides matrix can be produced in the process of the growth of single crystals of alkalihalides heavily doped with silver halides [3]. The bandgap energies of KCl and AgCl are 7.8 eV and 3.1 eV, respectively. Thus AgCl nanocrystals can be considered as a system of quantum dots (QD).

Silver halides have some unique features and occupy a particular position in solid state physics because their properties can be considered as of borderline nature between ionic and covalent bonding, that is they are located close to the boundary corresponding to the critical value of ionicity 0.785, namely 0.856 for AgCl and 0.850 for AgBr [5]. Silver halides play an important role in the photographic industry, since it is one of the few materials in which the process of latent image formation takes place. In order to get better understanding of this process, it is important to learn more about the intrinsic properties of the material particularly when one goes from the present micrometer (\(~10 \text{ mm}\)) world to the nanometer (\(~10 \text{ nm}\)) world where materials are known to behave quite differently because of quantum effects.

Under UV light irradiation of AgCl an electron is excited from the valence band into the conduction band and a hole is left in the valence band, which is subjected to self-trap, to form the self-trapped hole (STH) Ag\(^{2+}\). Free electrons can be captured by some
Ordered Arrays of Nanoparticles

Coulombic core and form shallow electron (SE) centers. A STH can capture an electron from the conduction band in a very delocalized hydrogen-like 1s orbital forming the self-trapped exciton (STE). STE, STH and SE were successfully studied by ODMR (see [5] and references therein). It was shown that STE in AgCl consist of a very diffuse electron (with the Bohr radius of 15.1 ± 0.6 Å) attracted by a strongly localized STH [6].

One of the basic problems which could not be solved during last 50 years, in spite of a big progress in application of silver halides in photography, was the identification of the structure of the intrinsic shallow electron centers which are believed to play an important role in the latent image formation process. The first direct determination of the wave function distribution of the intrinsic shallow electron center in AgCl was reported in [7]. It was shown that the wavefunction of SE is very diffuse and is spread over a radius of nearly 4 nm. A microscopic model was suggested in which an electron was shallowly trapped by two adjacent silver ions on a single cationic site (split-interstitial silver pair).

In the present paper, we report the study of shallow electron centers, self-trapped holes and self-trapped excitons in small AgCl crystals embedded in KCl matrix applying optical spectroscopy, optically detected magnetic resonance (ODMR), and electron paramagnetic resonance (EPR) techniques.

1. Results and discussion

KCl: AgCl monocrystals were grown by the Stockbarger method and contained 1 to 3% of AgCl. Two types of samples were available: crystals with a natural abundance of the Ag isotopes and 109Ag-enriched crystals. Optical absorption and EPR were studied with commercial apparatus. 35 GHz ODMR was detected at 1.7 K by monitoring luminescence excited with a Deuterium arc lamp.

Figure 1 shows ODMR spectra recorded by monitoring the emission intensity in two samples which were cut from different parts of KCl: AgCl (2%) crystals. The microwaves were chopped at 80 Hz and a lock-in amplifier was used to detect microwave-induced variations of the luminescence intensity. The upper spectrum (1), which was measured in a sample with higher Ag concentration, is similar to the ODMR of bulk AgCl. Anisotropic resonance signals of STH and STE and an isotropic line of SE centers are observed as indicated by arrows. Since STH’s are Jahn-Teller distorted along one of three ⟨100⟩ axes, three type of centers exist. The lines indicated by belong to the centers with the distortion axes oriented parallel to the magnetic fields \( B \), whereas the lines indicated by result from the two centers with the axes perpendicular to \( B \). The same applies for the triplet state of STE, because the central hole of the STE is virtually identical to the isolated STH. In the latter case two lines are observed for each of possible center orientations because of zero-field (fine structure) splitting which is characterized by a parameter \( D \). \( D = −710 \text{ MHz} \) for bulk AgCl. The \( g \)-factors of SE centers and STH and also the fine structure splitting \( D \) of STE determined from the ODMR spectrum 1 of KCl:AgCl (Fig. 1) are close to those of bulk AgCl but the ODMR lines are broadened. This proves that AgCl crystals are formed inside KCl matrix.

The spectrum 2 in Fig. 1 was measured in a part with lower Ag concentration. It corresponds partly to the ODMR ascribed to STE in the AgCl nanocrystals as reported in [3]. The parameters of this spectrum are considerably different from those of STE in bulk AgCl. In our measurements additional lines which probably belong to hole and electron centers in nanocrystals were also observed with different spectral dependencies.

In bulk AgCl crystals multiquantum transitions with the absorption of up to seven microwave quanta (total energy \( 7 \times 35 = 245 \text{ GHz} \)) were found which allowed to measure
Fig. 1. ODMR of two samples cut from different parts of a KCl:AgCl single crystal. The spectra were recorded by monitoring the luminescence intensity at 500 nm (1) and 550 nm (2).

the singlet-to-triplet splitting of STE for with a very high accuracy: $J = -161.0 \pm 0.1$ GHz [7].

In the sample 1 we have observed multiquantum transitions in ODMR of AgCl in KCl only within the triplet state of STE, which may imply a different (larger) value of the singlet-triplet splitting. A rather strong non-resonant background signal which had a weak dependence on magnetic fields and corresponded to an increase of the emission intensity by microwaves was also observed. In the sample 2 the background signal was much smaller.

SE centers are believed to play an important role in the latent image formation process, which occurs in the silver halides, when they are irradiated with light. Since STE in AgCl consist of a very diffuse electron attracted by a strongly localized STH, the exchange splitting between the singlet and triplet states is small. It should be noted that a value of the singlet-to-triplet splitting can be a measure of the space distribution of the electron wave function and therefore can give information about the effect of confinement of SE centers and shallow electron in STE due to the nanocrystals size. The confinement of the wave function distribution will affect the $g$-factor shift of SE centers.

Thus ODMR investigations have shown that there exist AgCl microcrystals of different size embedded in KCl crystalline matrix and that the AgCl lattice symmetry axes are oriented parallel to the axes of the surrounding KCl lattice. This conclusion was unambiguously proved by the observation of angular dependencies of ODMR spectra of the STH and STE in AgCl and EPR of Ag$^{2+}$ centers in KCl. The Ag$^{2+}$ centers in KCl were produced by UV irradiation at absorption bands of Ag$^+$ ions in KCl:Ag as reported in [2].

ODMR in such complicated KCl:AgCl and KBr:AgBr systems was also investigated by monitoring the tunneling afterglow, which is observed after irradiation of the crystals by ionizing radiation. In our previous papers we investigated the tunneling recombination afterglow in crystals with small concentration of Ag (less then 0.5%) [9, 10].

In conclusion, self-trapped excitons, self-trapped holes and shallow electron centers in AgCl microcrystals embedded in KCl monocrystals and maintaining the orientation of the host lattice were studied by ODMR and EPR. This new system seems to be very promising for the investigation of spatial confinement effects in nanocrystals since it provides a possibility to study oriented nanocrystals in a transparent matrix and to apply such a pow-
erful techniques as ODMR to study variations of the parameters of STE, STH and shallow electron centers which are well known for bulk silver halides.

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References


Critical magnetic field in regularly nanostructured indium

D. V. Shamshur†, A. V. Chernyaev†, A. V. Fokin† and S. G. Romanov†‡
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institute of Materials Science and Department of Electrical Engineering, University of Wuppertal, 42097 Wuppertal, Germany

Introduction

The behavior of nanostructured superconductors, being well documented in 2-dimensional case, is far less studied for 3D regular arrays of superconductor nanostructures because it is much more difficult to prepare the high-quality 3D lattices. To achieve the uniformity of intergrain contacts and the crystalline quality of the array we applied the template method, which is based on the impregnation of the crystalline porous dielectric matrix with the metal. The aim of this report is to discuss the critical magnetic field of the indium-opal nanocomposite.

1. Materials and measurement techniques

Opals consist of identical silica balls assembled in face centred cubic (fcc) lattice. There are alternated voids in opal: O-voids ($d_O = 0.41D$, $D = 234$ nm — ball diameter in the sample) each connected with eight T-voids ($d_T = 0.23D$ via channels of characteristic diameter $d_b = 0.15D$ [1]). In order to reduce the free volume the opal was impregnated with amorphous silica. Further reduction of porosity was achieved by coating the opal voids with 34 monolayers of TiO$_2$. Effectively, the overall reduction of the void size was about two times as compared with ideal package for the same lattice parameter. Impregnation with molten In has been performed and metal forms the precise 3D replica formation of opal voids [2]. Detailed investigation of the composition of In-opal structures is given elsewhere [3].

2. Experimental results and discussion

The $R(T)$ curve (Fig. 1(a)) shows a peak at $T = 4.24$ K followed by the broadened decline of resistance to 0 at $T_{c0} = 3.7$ K. The critical temperature of superconducting (SC) transition $T_c$ exceeds sufficiently that of the bulk indium (3.41 K). The $R(T)$ curve shows three steps, temperatures of which correspond to maxima of the $dR/dT$ curve (Fig. 1(b)).

The SC transition is spread broadly by a field (Fig. 2) and demonstrates several steps: $H_0$ as the critical magnetic field of transition from the SC to the resistive state, $H_1$ and $H_2$ — characteristic fields for steps. Remarkably, the $H_0$ field exceeds dramatically the critical field for the bulk In (280 Oe at $T = 0$). Temperature dependences of $H_1$ and $H_2$ show nearly the same slope as $H_0(T)$, moreover the field values follow roughly the 1:2:3 sequence.

Confining the SC condensate in nanostructures with characteristic dimension $d < \xi_{bulk}$ implies the involvement of the whole volume of grains in the supercurrent transfer. Obviously, the increase of the $T_c$ above the bulk value correlates with the reduced dimensionality of the In network. The presence of different parts possessing the well-defined dimensions is reflected by the steps on $R(T)$ curve. Assuming that the SC nucleates at the narrowest part, the onset of the resistance drop corresponds to the SC transition in intergrain bridges.
The empirical expression $T_c = 3.41 + 5.1/d$ [d in nm] [4] gives the $d$ estimate as 7 nm, which is the bridge diameter.

In In-opal every current path consists of symmetrical arms, moreover both arms joined and separated in periodical manner. In other words, the current encircles the loop circumference. The Ginsburg–Landau estimate for the lower critical field of the mesoscopic loop varies between $\mu(0)H_{c1}^{\text{loop}} = \Phi_0/(2\pi r^2)$, where $r = \sqrt{r_{\text{out}}r_{\text{in}}}$ is the average between inner and outer ring radii, for the ring with $r_{\text{out}} - r_{\text{in}} \ll r$ and up to 4 times higher value for $r_{\text{out}} - r_{\text{in}} \leq r$ [5]. The projection of the smallest loop to (111) plane is about $2r = 2D/\sqrt{6} = 98$ nm, correspondingly, the $H_{c1}^{\text{loop}} = 1.4 \cdash 5.5$ kOe applies. This estimate matches the critical fields observed in In-opal. In contrast, if one accounts for the size-dependent enhancement of the critical magnetic field only $H_{d,c}^{\text{loop}}(T)/H_{c}^{\text{bulk}}(T) = 4\sqrt{5}\lambda(T)/d$, where $\lambda(T) = \lambda(0)(1 - (T/T_c)^4)^{-0.5}$, $\lambda(0)$ — the penetration depth in the bulk superconductor, the particle size $d$ has to be less than 5 nm.

In a loop with a fixed circumference the screening supercurrent must flow to fulfil the fluxoid quantization conditions if the external field corresponds to the non-integer number of flux quanta per loop. These currents can be described in the same manner as magnetic vortices. In the loop lattice there is the possibility of mutual cancellation of screening currents in common arms of adjacent loops, i.e. the actual distribution of the current depends on the lattice symmetry. Magnetic field penetrates deeply $\Lambda = \lambda^2(T)/d$ in the nanostructured sample, i.e. the field around the single vortex is weakly screened and the repulsive interaction is of longer range as compared with the type II superconductor. Thus, loops within the cluster of $\xi$, $\Lambda$ dimensions are strongly coupled. Correspondingly, the behaviour of the lattice as the whole is definitely different from that of a collection of loops with the smallest areas. Due to these complications the observed characteristics deviate sufficiently from estimates made for the single loops and grains.
Let us suppose that the critical field is the field when each loop has caught one flux line. At higher field the transport plus screening current exceeds the critical current of the intergrain bridge. In this case the connectivity changes from multiply connected to single connected and a flux line can move easily in and out the loop. This state of the network is obviously the resistive state because the dissipative motion of flux lines across the field/current direction causes the voltage $V \propto (H - H_0)$ and the onset of the magnetoresistance. With further increase of the field it becomes energetically favourable to add the second quantum in each loop. This field $H_1$ is roughly twice as $H_0$. The next kink can be assigned to the accumulation of the third flux quantum per each loop, because its field $H_2 \approx 3H_0$. With the use of the expression for the flux quantization and value for $\Delta H = 2.7 \text{ kOe}$ one can estimate the effective loop diameter as 88 nm, which is appealingly
close to the size of the smallest loop in the lattice.

3. Conclusions

The giant increase of the critical magnetic field was observed in the regular lattice of nanosize In loops. Two effects — the reduction of the loop arm cross-section and the regularity of the ensemble, are believed to contribute mainly to the observed effect.

Acknowledgements

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References

The enhancement of luminescence in ion implanted Si quantum dots in SiO\textsubscript{2} matrix by means of dose alignment and doping

D. I. Tetelbaum†, O. N. Gorshkov†, S. A. Trushin†, D. G. Revin‡, D. M. Gaponova‡ and W. Eckstein§

† Physico-Technical Research Institute of Nizhnii Novgorod State University, Gagarin prosp. 23/3, 603600, Nizhnii Novgorod, Russia
‡ Institute of Physics for Microstructure, GSP-105, 603600, Nizhnii Novgorod, Russia
§ Max-Planck Institute of Plasmaphysics, Boltzmannstr. 2, D-85748, Garshing, Germany

The system of Si quantum dots imbedded into SiO\textsubscript{2} matrix (SiO\textsubscript{2}:Si) is very promising one for many modern micro- and optoelectronic employments due to its compatibility with common silicon technology. The ion implantation is one of the most suitable and common method for the production of such a system [1–3]. Because of small thickness of implanted layers, the problem of the luminescence enhancement for the SiO\textsubscript{2}:Si system is important. There is one apparent way to resolve this problem — the increase of excess concentration \(C_{Si}\) of Si by the increase of Si\textsuperscript{+} dose. But this way is not a simple one. Indeed, the luminescence intensity must increase with areal density \(N_S\) of Si nanoinclusions (NI) with appropriate for quantum confinement sizes. Generally speaking, it is not clear if the \(N_S\) would increase proportionally to \(C_{Si}\): with increase of \(C_{Si}\) not only \(N_S\) but also average sizes of NI may grow, as well. Besides, the relation between dose and \(C_{Si}\) value is not simple due to complicated processes that must be taken into account at high doses of ion implantation (sputtering, swelling, ion mixing, change of composition, etc). It is evident that at sufficiently high doses, the sizes of NI will be so large that quantum confinement (and, hence luminescence related with quantum dots) reduction will occur. Therefore, one should look forward that, even at favourable conditions, the dose dependence of luminescence should not be monotonous one.

Recently, we have established once more factor that enhances the photoluminescence (PL) in SiO\textsubscript{2}:Si system, namely, the additional doping of system by phosphorous implantation [4, 5]. The mechanisms of phosphorous influence on PL properties are unknown, although some assumptions were suggested in [4, 5].

In this work, we investigated, both theoretically and experimentally, the relation between Si\textsuperscript{+} dose and PL intensity and carried out some experiments to clear out the mechanisms of phosphorous doping influence on PL.

The thermal oxidized samples of Si with oxide thickness of 300 nm were used. The room temperature ion implantation was carried out at energy \(E = 150\) keV and at doses \(\phi = 1 \times 10^{17}\) to \(1 \times 10^{18}\) cm\(^{-2}\). After implantation, the samples were annealed at 1000 °C, 2 h. Part of the samples was additionally implanted by P\textsuperscript{+} with the same \(E\) at \(\phi = 1 \times 10^{16}\) cm\(^{-2}\). The PL was measured at room temperature at \(\lambda = 488\) nm (Ar laser). The Raman spectra were determine at oblique beam incidence to decrease the substrate signal. The PL peak at approximately \(\sim 800\) nm that is characteristic one for the SiO\textsubscript{2}:Si system [1–3] is observed for all the implanted samples. The position of the peak is about the same with expection of the largest doses \((\phi > 4 \times 10^{17}\) cm\(^{-2}\)), for which it is shifted towards the red site. As it was expected, the PL intensity nonmonotoniously changes with dose.
In order to interpret these data, we have carried out the calculation of \( N_S \) according the following assumption. The concentration of NI at any depth is changed proportionally to the volume occupied by the excess (superstoichiometric) concentration of Si atoms until NI is overlapped at this depth. The overlapping means that volume \( \alpha \) occupied by excessive Si atoms becomes equal to \( \alpha^* \), where \( \alpha^* \) is varied parameter, \( \alpha^* \) is near 0.5. This assumption must be fulfilled if mean size of NI does not change and if all (or constant part of) the excess Si atoms enter into NI during annealing. Then the relative value of \( N_S \) at any dose is determined by the integration over the depths for which \( \alpha < \alpha^* \). The \( \alpha \) values versus depths were computed by Monte Carlo method (TRIDIN code \([6]\)) taking into account pointed out processes.

In Fig. 1 the calculated dose dependence of \( N_S \) (curve) is presented together with experimental values of integral PL peak intensify (points). Good accordance between experimental and calculated data speaks in favour of that above pointed assumption is right. This means that NI average size, indeed, is not changed with dose at \( \phi < 4 \times 10^{17} \text{ cm}^{-2} \). The data of Raman spectra show that Raman shift is not altered until \( \phi > 4 \times 10^{17} \text{ cm}^{-2} \). This fact serves as additional evidence of NI size constancy. As it is follows from PL peak position and from Raman shift, average size of NI is about 4–5 nm.

The theoretical estimation shows that at sufficiently high supersaturation and providing that the complexes composed of two Si atoms serve as nucleus of NI, the concentration of NI should be approximately proportional to the excess Si atom density at not too large doses.

Other factor that enhances the PL is phosphorous doping (Fig. 2). Three mechanisms can provide this enhancement. First, the influence of P on processes of NI nucleation and growth. Second, the passivation of defects (dangling bonds) in NI or on their surfaces. (This defects may serve as nonradiative centres of recombination reducing the PL.) The third mechanism is related with the change of radiative interband transitions between quantum dot levels when the electron of donor atom fills in the level in conduction band of quantum dot. (Even at lowest Si dose, the concentration of P atoms at \( \Phi_p = 1 \times 10^{16} \text{ cm}^{-2} \) is enough to provide more than one donor atom into each NI).

It is shown that intermediate annealing at 1000 °C, 2 h does not influence on the degree of PL enhancement by the phosphorous doping. It permits to deny the first mechanism of PL.
enhancement. In order to check the rest two mechanisms, we have measured the relations of PL intensities with/without phosphorous implantation in dependence of excitation light power. The relation occur to be lower at small powers and then it goes to constant value. This may be explained as follows. At low excitations, the stationary density of excited electrons on the first quantum level is small, and therefore the role of doping is large. As the steady-state density of excited electron grows, the role of additional electrons related with doping is decreased. At high light power, the relation of intensities does not tend to unity. Hence, the third mechanism (passivation) becomes apparently dominating one at high excitation power.

Thus, the PL intensity can be enhanced both by Si dose alignment and by additional phosphorous doping. The influence of acceptor doping is now under investigation.

References

In-situ STM analysis and photoluminescence of C-induced Ge dots

A. Beyer, O. Leifeld, S. Stutz, E. Müller and D. Grützmacher
Laboratory for Micro- and Nanotechnology, Paul Scherrer Institute,
CH-5232 Villigen PSI, Switzerland

Abstract. Molecular beam epitaxy has been utilised to grow small C induced Ge islands in silicon. Using in-situ STM analysis, it is shown that the amount of C deposited on the Si(100) surface prior to Ge growth permits the control of lateral size and height of Ge quantum dots. The Ge grows in a Volmer-Weber growth mode in areas between the C-rich patches on the Si surface. Thus laterally smaller but higher dots are found with increasing C coverage. Accordingly, intense photoluminescence (PL) with a stronger confinement shift in dependence on the Ge coverage is observed for samples prepared with large C concentrations. The impact of the Si spacer layer width on the dot size has been studied by TEM and compared to PL data.

Introduction

The search for paths to integrate optoelectronic devices in the mature Si technology has lead to various techniques for the fabrication of nanostructures in Si. The one taken up in this study is the formation of small Ge dots on Si(100) surfaces covered with sub-monolayers of C [1]. It has been shown that using this technique the lateral island size can be reduced below 20 nm at growth temperatures in the range of 500° C. These growth temperatures provide excellent crystalline quality and consequently pronounced photoluminescence (PL) was observed for these C-induced Ge quantum dots [2]. However, even smaller and more compact islands are required to further tailor the PL intensity. Previously study we showed using in-situ STM analysis that the C forms patches exhibiting a c(4×4) reconstruction, whereas the areas between these C rich patches remain free of C and exhibit buckled Si dimer rows [3]. Here we explore the Ge nucleation on the C alloyed Si surface. It is found that C can be used to limit the area available for the nucleation of the Ge dots. Using the amount of deposited C and Ge as growth parameters, samples containing multiple layers of C-induced Ge dots have been fabricated by MBE at 460° C. Additionally the Si spacer layer has been varied to study its impact on the structural and optical properties of the dots. Furthermore, transmission electron microscopy (TEM) and PL has been used to investigate the dots embedded in Si. A sensible correlation of the gathered features yields insights into the peculiarities of C-induced Ge dots, with respect to the carrier confinement.

Results and discussion

Figure 1(a) shows the evolution of 3-dimensional (3D) islands on the C-alloyed Si surface. The STM image is taken after the deposition of 0.1 monolayer (ML) of C and 1 ML of Ge at 550° C and 350° C, respectively. The Ge does not cover the whole surface but instead grows in 3D piles. In areas between the islands the unperturbed c(4×4) reconstruction is visible [3]. The density of Ge islands amounts to 8 × 10¹¹ cm⁻², the diameter to 4–5 nm and the height up to 6 ML. The line scan in Fig. 1(a) is taken at the position indicated by the white line in the STM image. The islands show an irregular shape of the basis, in contrast to lower Ge coverages (0.1–0.5 ML), where rectangular island shapes are most often observed. No
distinct facet reconstruction can be seen, because the side walls are irregular. Nevertheless a certain affinity to side facet formation either in \( \langle 100 \rangle \) or \( \langle 110 \rangle \) directions is visible. Due to the freedom to expand in 3 dimensions, the islands are presumably relaxed towards their apex. The height to base ratio is approximately 1/10, which is similar to relaxed pure Ge hut clusters [4].

It is important to notice that no Ge wetting layer is formed. We assume that no Ge is incorporated into the C rich \( c(4 \times 4) \) reconstructed areas, since this would almost certainly alter the appearance of these regions in STM. Most likely the C patches are highly strained due to the big lattice mismatch between Si and C, thus the lattice constant in the C rich region is smaller than that of Si. Ge expands the Si lattice and is therefore easier accommodated on the Si surface free of C.

At higher deposition temperatures for the Ge, up to 550°C the islands tend to become flatter. But the general feature, that no Ge wetting layer is formed and that the island start to nucleate in regions free of C, persists. STM images taken for Ge islands formed by 2 ML indicate that finally the \( c(4 \times 4) \) reconstructed areas vanish [5]. Apparently the C rich areas are overgrown by Ge, however, it is unknown how the C is incorporated into the crystal at this stage. The C deposition in Fig. 1(a) is only 0.1 ML, at higher C coverages, up to 0.3 ML, the area available for Ge nucleation becomes even smaller leading to a smaller lateral size and to an increase in the height to base ratio.

Figure 1(b) shows the dependence of PL spectra of C-induced Ge dots (0.2 ML C) on the amount of Ge deposited. The samples, grown at 460°C, contained 10 layers of dots separated by 8 nm wide Si barrier layers. All samples were annealed at 650°C for 4 min subsequent to growth, leading to an intensity increase by a factor of 2 to 4, whereas the change in the energetic peak positions is minor. The increase from 1.4 to 3.4 ML Ge leads to an energy shift of approximately 250 meV. The peak intensity drops significantly for Ge depositions exceeding 2.5 ML, indicating a weaker 0-dimensional localisation in
larger dots.

In Fig. 2(a) the energetic position of the peak maximum of the dot related PL is plotted in dependence on the number of Ge monolayers deposited on top of the 0.2 ML of C. A strong confinement shift is observed in the region between 3 and 1.8 ML, for smaller dots the shift saturates. Since the dots have the shape of platelets, and their dimensions in the xy plane are much larger than their heights, the height of the islands determines predominantly the confinement shift.

The saturation in the confinement shift for islands formed by less than 1.8 ML might be attributed to the heavy hole state in the Ge dot area approaching the Si band edge. Also annealing at 800°C has only little impact on the peak position for these very small dots, which fits to the latter conclusions. C on lattice sites does not diffuse in the temperature range of 800°C. Annealing leads to an intermixing of Si and Ge, thus to bigger dots with a lower Ge content, which explains the strong shift towards higher PL energies collected from the large dots. For small dots, where the subband level of the holes is shifted close to the band edge by the confinement, the annealing has only a minor effect. The difference in the peak energy between the dot PL for very small dots and the Si NP peak at 1150 meV is only in the range of 50–60 meV, which might be the confinement energy of the electrons in the C rich Si. The highest peak as well as integrated intensity is found for dots deposited with 1.8–2.2 ML of Ge. For smaller dots the confinement shift saturates; the localisation of the wavefunction is weak when the quantum state in the well approaches the band edge of Si, thus the matrix element decreases.

As found by TEM analysis, also the shape of the Ge dots is affected by the amount of C deposited. They grow in height at the expense of their diameter at higher C coverages. The C-covered area increases for higher amounts of C deposited, whereas the local density of C within the C covered areas is constant, as indicated by the c(4 × 4) reconstruction detected by STM. The PL lines shift significantly to lower energies with increasing C contents. The peak positions of a series of samples grown with 0.3 ML of C are also indicated in Fig. 2(a) (closed triangles). They are significantly shifted to lower energies for the same amount of Ge deposited, when compared to those of dots induced by 0.2 ML C. This is attributed to the increase in dot height.

Figure 2(b) shows four PL spectra of Ge dots fabricated by depositing 2 ML Ge on top of 0.2 ML C, each comprising 10 layers separated by 8, 16, 24 and 32 nm Si spacer
layers. Surprisingly, the PL shifts by 35 meV to higher energies, if the spacer thickness is increased from 8 to 16 nm. At the same time the intensity increases. This can be understood by assuming an overlap of the wavefunctions confined in adjacent dot layers, leading to a system of weakly coupled dots. A further increase of the Si spacer thickness leads to a red shift and a broadening of the lines.

The latter effect is difficult to explain. To gather more detailed insights into these samples, they have been carefully analysed by TEM. Surprisingly, the dot size is smaller for the 16 nm wide Si buffer layers as for layers grown with 24 and 32 nm wide barriers.

Conclusions

In conclusion, clear indications of a zero dimensional confinement were detected. It increases, when the C coverage of the surface is increased, thus the C free area shrinks, leading to a more compact shape of the Ge islands. They grow in height and reduce their lateral dimensions. The results indicate that the electrons are confined in the C-rich areas between the dots and that the holes are confined in the Ge dots. The recombination is a spatial indirect process. This makes the design for optimised PL efficiency quite complex. On the one hand side a strong localisation of the wavefunction in local space is desired to overcome the indirect bandgap in k-space. On the other hand side an overlap of the electron and hole wavefunctions in local space is required to increase the possibility of the spatial indirect recombination.

References

Optical properties and structure of Si/InAs/Si layers grown by MBE on Si substrate

N. D. Zakharov†, P. Werner†, U. Gösele†, V. M. Ustinov‡, G. E. Cirlin‡, B. V. Volovik‡, N. K. Polyakov‡, V. N. Petrov‡, V. A. Egorov‡, N. N. Ledentsov‡, Zh. I. Alferov‡, R. Heitz§ and D. Bimberg§

† Max-Planck Institute of Microstructure Physics, Halle/Saale, Germany
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia
§ Technical University of Berlin, Germany

Abstract. Epitaxial Si/InAs/Si heterostructure grown on (001) Si substrate by molecular beam epitaxy (MBE) and annealed at 800°C was investigated by High Resolution Transmission Electron Microscopy (HRTEM). Extensive interdiffusion leads to the formation of an InAs solid solution in the Si cap layer. Additionally, InAs-enriched regions with extensions of 6nm, which exhibit two kinds of ordering are observed. The ordering of InAs molecules has occurred. The sample show photoluminescence in the 1.3 µm region, which is tentatively attributed to the recombination of excitons localised in the ordered regions.

Introduction

The potential benefit from combining the advantageous optical properties and flexibility of III–V semiconductors with silicon technology widely used in microelectronics has attracted great interest for decades. Up to now, researchers have focussed on the growth of continuos layers of III–V materials on silicon [1]. The large misfit between Si and, e.g., InAs (ε = 10.6%) renders the growth of electronic or optical quality material practically resolvable problem. More recently, the possibility to exploit the formation of narrow-gap III–V islands on Si substrates has been pointed out [2]. Indeed, such small InAs islands on Si(100) surface have been observed by scanning tunnelling microscopy and high-resolution transmission electron microscopy (HRTEM) [3, 4]. HRTEM investigations of capped InAs/Si structures revealed a high density of coherent InAs clusters with typical dimensions in the 3 nm region at the InAs/Si interface for optimised growth conditions [5, 6]. Such samples exhibit a broad photoluminescence (PL) peak in the 1.3 µm region at 10 K [3]. Detailed optical investigations of this PL line indicated a k-indirect type II transition, which has been tentatively attributed to excitons localised in the small coherent InAs clusters [7]. The extreme small size (< 3 nm) of these clusters might, however, prevent sufficient carrier localisation. The present work present a detailed structural characterisation of such InAs-Si layers providing new insight into the origin of the 1.3 µm PL peak

Experimental

The InAs/Si heterostructure was grown by molecular beam epitaxy (MBE) on p-type Si(100) substrate using an EP 1203 machine. The growth rates for InAs and Si were 0.03 nm/s and 0.017 nm/s, respectively, and the As/In flux ratio was ~ 4. InAs was deposited at a substrate temperature of \( T_s = 350^\circ C \). The nominal thickness of the deposited InAs was 1.6 ML. Immediately after the InAs deposition, a 10 nm Si cap layer was grown at the
same $T_s = 350^\circ C$ followed by a 10 min annealing step at $700^\circ C$. Further 40 nm Si cap layer was grown at $700^\circ C$ with a final 10 min annealing step at $800^\circ C$ to smooth resulting surface. The crystalline quality of the structure and the composition of the grown layers were investigated by techniques of transmission electron microscopy (TEM). PL spectra were measured at a temperature of 7K using Argon laser excitation.

**Results**

Typical cross-section and plan-view images of the investigated structure are shown in Fig. 1(a,b) respectively. The plan-view image shows the good structural quality of the sample with a relatively low density ($10^8 1/cm^2$) of structural defects, marked A in Fig. 1(b). These defects (A) are located at the InAs/Si interface and do not penetrate into Si cap layer. First, the average InAs concentration in the Si cap layer can be estimated from selected area diffraction (SAD) taken at once from substrate and cap layer in cross-sections sample as shown in Fig. 1(a). Such SAD pattern reveal a splitting of reflections in [001] direction perpendicular to the layer surface (Fig. 1(c)). This splitting is attributed to a tetragonal distortion of Si cap layer due to the formation of an InAs solid solution. The magnitude of the observed splitting of the reflection (206) measured in SAD corresponds to a tetragonal distortion $\Delta = \Delta g/g(206) = \Delta a/a_{Si} = 0.007$. The volume of the distorted unit cell is $V_t = a_{Si}(1 + \Delta a/a_{Si})^{1/3}$. It immediately follows that average cubic unit cell parameter of

![Fig. 1.](image)
the solid solution \( a_{ss} \) is given by:

\[
a_{ss} = V_{t}^{1/3} = a_{Si}(1 + \Delta a/a_{Si})^{1/3}.
\]  

(1)

Taking \( a_{ss} \) as to be linearly dependent on the InAs concentration in the Si matrix, it follows:

\[
C_{InAs} = (a_{ss} - a_{Si})/(a_{InAs} - a_{Si})
\]  

(2)

where \( C_{InAs} \)-concentration of InAs in Si cap layer, \( a_{Si}, \ a_{InAs} \)—unit cell parameters of Si and InAs respectively. Substitution of \( \Delta a/a_{Si} = 0.007 \) into Eqs. (1), (2) gives \( C_{InAs} = 0.004 \). Thus the averaged composition of the cap layer can be written as \( Si_{0.996}(InAs)_{0.004} \).

Second, in high resolution cross-sectional images the dark regions marked by B in Fig. 1(a) reveal a doubling of periodicity of \{002\} lattice planes (Fig. 2(a)). It leads to appearance of diffuse maxima situated halfway between \( \pm (220) \) matrix reflections in the Fourier transformed image (FFT) (see insert in Fig. 2(a)). This result can be interpreted as a partial ordering of InAs in Si. A possible idealised model of such an ordering is shown in Fig. 2(b) where InAs occupies every other atomic (101) plane inclined by 45° to the surface. A HREM image (Fig. 2(c)) simulated on the basis of this structural model shows that the darker rows correspond to InAs atomic rows. It is obvious that the contrast calculated for an ideal ordering is in a qualitative agreement with the experimental image. These partially ordered regions can also be observed at low magnification using diffraction contrast technique (see features B in Fig. 1(a)). In this case, the image contrast (dark regions) results from variations of the extinction distance. The size of coherent ordered regions (\( \geq 6 \) nm) is about 2 times larger than the size of coherent InAs clusters formed at the InAs/Si interface described in a former paper [5].

Figure 3 depicts a low temperature PL spectrum of the investigated sample revealing a broad PL peak in the 1.3 \( \mu \)m region. Recently, this luminescence has been studied in detail in a different sample, suggesting a k-indirect type II transition in the epitaxial layer [7]. Such a transition is indeed expected for coherent InAs clusters observed near the InAs/Si interface. However, the small size (\( \sim 3 \) nm) of such clusters might be to small for sufficient carrier localisation. As shown above, the Si-cap layer is actually a Si-InAs solid solution with ordered InAs-rich regions. The ordered regions with a high InAs concentration (and therefore smaller band gap) and a size of \( \sim 6 \) nm, can provide sufficient carrier localization to explain the observed 1.3 \( \mu \)m emission. The incorporation of InAs molecules into the Si
Fig. 3. PL spectrum of the investigated sample showing a broad luminescence at 1.25 µm. The peak at 1.1 µm correlates to emission from the Si bulk.

is expected to shift the relative positions of the conduction and valence bands leading to a quantum structure.

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References


STM investigation of a strong electric field effect on local photocurrent spectra in InAs/GaAs quantum dot heterostructures

Institute for Physics of Microstructures RAS, 603600, Nizhny Novgorod, Russia

Abstract. We demonstrate a possibility of using a scanning tunneling microscope (STM) for a high spatial resolution study of photocurrent spectra of quantum-dimensional structures. The effect produced by an electric field on the position and intensity of the size quantization levels maxima in the local photocurrent spectra for InAs/GaAs structures with quantum dots located near the sample surface is investigated.

Introduction

The probe microscopy methods are being widely used currently for high spatial resolution investigations into the properties of quantum-dimensional structures based on InGaAs/GaAs. Major success in this research has been achieved in the study of local luminescence by the use of near-field optical microscopy [1, 2] and electroluminescence in the STM tunneling contact (the so-called STM-cathodoluminescence) [3–5]. When applied quantum dot structures, these techniques allow observation of narrow lines in the luminescence spectra, which correspond to the transitions between the levels of size quantization in single dots. The drawback common for all of the above methods, however, is in low-temperature requirement to enable observation of local luminescence and, besides, the condition that a quantum well or a quantum dot layer must be outside the region of a GaAs space charge, otherwise the electric field would separate the charges in the near-surface layer thus impending recombination process. This would degrade space resolution, since the actual probing area increases through diffusion of nonequilibrium carriers.

A promising method for detecting the local photocurrent spectra by STM was offered recently in [6]. It allows investigation of the excited states in the energy spectrum of quantum-dimensional objects located immediately near the sample surface. In this case the field of a surface space charge separates the electron-hole pairs generated by photoexcitation so that spatial resolution in this methods depends mainly on the surface rather than volume diffusion of carriers, which is less intensive. For quantum dots located near the sample surface the spatial resolution can, in principle, be brought to the size of the wavefunction of minority carriers localized on a single quantum dot.

In this paper we report the results obtained in the STM studies of the electric field effect on the spectra of local photocurrent in InAs/GaAs quantum dot heterostructures.

1. Experiment

The structures were MOCVD-grown on GaAs substrates [7]. A layer of quantum dots was located near a sample surface. The GaAs covering layer thickness was 1.5–2 nm. To avoid oxidation, the samples were immersed in vacuum oil immediately after growth, so the photocurrent spectra were taken from the tunneling contact realized through an oil film. In experiments we used a scanning tunneling microscope of our own design,
which is combined with an optical system [8]. The structures were of n-type conductivity, with an STM current-voltage characteristic (I–V characteristic) that was typical of a metal-semiconductor tunneling contact. The probe was held over a surface via a feedback system of STM operating at $j_f = \text{const}$ and a voltage corresponding to the forward branch of the I–V characteristic. Photocurrent was defined as the difference between the current in the backward branch of the I–V curve for an illuminated contact and the dark current. A 100 W halogen lamp radiation transmitted through the monochromator and a passive optical filter to cut off the visible part of spectrum was used for an optical pumping of samples. The monochromatic radiation was carried to a semiconductor structure by a multicable lightguide from the back side of the substrate which provided additional filter to cut off the quanta of light with energies higher than the GaAs band gap. Due to this arrangement photocarriers were generated only in the epitaxial layer of InAs/GaAs. The spectral dependence of photocurrent were plotted from the data overaged over 100 measurement results.

2. Results and discussion

As demonstrated in the experiments, the spectral dependence of a local photocurrent contain a number of peaks corresponding to the carrier transitions between different states of the energy spectrum of a quantum-dimensional structure. A characteristic STM spectrum of a photocurrent is shown in Fig. 1(a). The first short-wave peak with the energy of about 1.397 eV appears to result from carbon impurity contained in the GaAs layer of a MOCVD grown structure. Its energy corresponds to that of transition between the acceptor level which is by 26 meV higher than the valence band top, and the conductivity band. As follows from the calculations, the interband transition energy in a wetting layer of InAs (one monolayer thick) is 1.377 eV at room temperature, which is in good agreement with the value for the second peak in the photocurrent spectra. The peaks in the long-wave part of the spectra seem to be related to the transitions from the excited hole states of a quantum dot to the electron levels in the wetting layer and to the levels of the excited electron states in a quantum dot.

A study was carried out on the effect produced by an electric field on the position and intensity of peaks in the STM-yielded spectra of photocurrent, corresponding to size quantization levels. The spectral dependences of a local photocurrent in quantum-dot structures for different values of bias voltage at a tunneling gap contact are shown in Fig. 1(b). They exhibit a few features. In the short-wave part of the spectrum ($\lambda \leq 950$ nm) the peaks become more intensive and broader with a higher voltage. A different situation is observed for the spectral components in the long-wave part of spectrum ($\lambda \geq 950$ nm). An increase in voltage therein lessens intensity of the peaks and simultaneously causes them to broaden. The intensity of the first peak corresponding to the transition of carriers from the acceptor level of carbon to the GaAs conductivity band increases, and the peak shifts towards the long-wave of the spectrum.

A higher local photocurrent in the short-wave region of spectrum can be accounted for by the Franz–Keldysh effect in a bulk GaAs layer lying closest to a tunneling contact, where electric fields are strong. Indeed, the light absorption in a strong electric field gets higher, due to the tunneling of charge carriers, in the region of quantum energies lower than the band gap [9]. It reaches a maximum near the fundamental absorption edge and falls off exponentially with a decreasing photon energy. A similar increase of photocurrent in this spectral region is also observed for structures without quantum objects, where it is caused by the electric field effect on the electron states in bulk GaAs.
A completely different effect is produced by an electric field on the intensity of optical transitions between localized states. In a strong electric field the mean coordinates of the electron and hole localized states of a quantum dot shift in opposite directions, thereby reducing the overlapping integral of the wave functions for the initial and final states and, hence, the probability of optical transition between them. Besides, the electric field delocalizes localized states, since there appears a non-zero probability of tunneling transition to the continuous spectrum states. As a result, the corresponding absorption lines intensity lessens, and the lines become blurred. The blurring of a line depends on the time of an electron tunneling from a localized state to a continuous spectrum state. It is nicely seen in Fig. 1(b) that the peaks in the spectral region corresponding to transitions involving the localized states of a quantum dot become smaller and broader with higher voltage and practically vanish at \( V = 3.5 \) V. The latter is likely to imply that the frequency of tunnel escape from the excited states involved in absorption at this voltage becomes comparable with the value equal to the localization energy divided by the Planck constant, so the states actually become delocalized.

3. Conclusion

STM aided investigations into the effect of a strong electric field on the local photocurrent spectra in InAs/GaAs structures with quantum dots located near a sample surface are reported. It is shown that a higher photoresponse in the short-wave region of spectrum is due to the Franz–Keldysh effect, whereas the intensity lessening for peaks in the long-wave region of spectrum of photocurrent and the peaks broadening may be accounted for by the influence of an electric field on the wavefunction of excited localized states in quantum dots.

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References

Tunneling spectroscopy of nonequilibrium interacting impurity states on semiconductor surface

N. S. Maslova, S. I. Oreshkin, V. I. Panov and S. V. Savinov
Chair of Quantum Radio Physics, Moscow State University, 119899 Moscow, Russia

Abstract. New STM/STS observation method has been used for investigation of interacting nonequilibrium impurity states on semiconductor surface. The dependence of electronic density spatial distribution on applied tunneling voltage is analyzed. Energy values of “switching on and off” of interatomic interactions are determined. The dependence of Cr impurity dangling bond hybrid orbital electron spatial localization on tunneling bias is observed.

In present work we report the results of STM/STS investigations of interacting nonequilibrium impurity states on A₃B₅ semiconductor surface. Images of tunneling conductivity spatial distribution on semiconductor surface have been obtained by means of new observation method. The series of images has been consequently measured step by step changing applied tunneling bias (energy of electronic states) by 5 mV. The dependence of electron density spatial distribution in the vicinity of interacting impurity atoms upon applied bias voltage (the energy of electronic states) has been investigated by STS method.

Monocrystal GaAs Si doped samples and double doped samples with Si and Zn impurities (compensated) have been used in our experiments. All the measurements were performed at \( T \sim 4 \text{ K} \) using low temperature STM with sample cleavage mechanism [1].

The evolution of local electronic density spatial distribution of interacting impurities with energy changing is depicted in Fig. 1. Changes in spatial configuration of different impurity atoms overlapping electronic states with energy variation are quite visible on series of \( dI/dV(V, X, Y) \). Analyzing spatial distribution of tunneling conductivity one can follow symmetry properties of electronic states with different energies and also determine characteristic energy values of “switching on and off” of interatomic interaction.

Electronic density spatial structure measured in the vicinity of two interacting impurities at fixed value of applied bias voltage is sensitive to symmetry properties of localized state with definite energy which can be determined from applied tunneling voltage value. We should mention the main specific features observed in tunneling conductivity spectra:

1. Peaks on tunneling conductivity spectra \( dI/dV(V, X, Y) \) are clearly seen just above interacting impurity atoms and disappear if the distance from impurities exceeds the maximum value of localization radius for interacting impurities localized state (for “shallow” impurities it is about 10 nm). Peaks structure on tunneling conductivity curves strongly depends on type of impurity and interatomic interaction. With increasing of the distance from impurities not only peaks intensity changes but its maximum energy also shifts. This fact can be connected with strong influence of Coulomb interaction of localized impurity charges on electronic energy spectrum.

2. Characteristic energy range of considerable changes in local electronic density distribution is about 0.1 eV. This value is comparable with localized state energy broadening due to manyparticle interaction. Theoretical analysis of energy spectrum and symmetry
Fig. 1. Evolution of local electron density distribution (LEDD) of two interacting impurity Si atoms with tunneling sample bias (energy) changing: (a) STM image Si–Si, $U_t = 1.0$ V, (b) LEDD images Si–Si, $U_t$ from $1.0$ V to $-1.5$ V, (images size $10$ nm × $10$ nm).

Fig. 2. The dependence of Cr impurity hybrid orbital localization on applied tunneling sample bias: (a) $U_t = -1.0$ V, (b) $U_t = -1.1$ V.

properties of different electronic states in two-site extended Hubbard model gives qualitative explanation of main specific features experimentally observed in local tunneling conductivity spectra.

Interacting impurity states of Cr atom on InAs (110) surface have been also investigated by STS/STM methods. Such states appear due to substitution of As site by Cr atom in InAs lattice. Effects of resonant tunneling for these impurities were observed earlier [2]. The dependence of Cr impurity atom dangling hybrid orbital spatial localization on applied tunneling voltage is shown in Fig. 2. It is clearly seen, that Cr atom occupies As site and impurity perturbation potential radius is limited by crystal lattice unit cell. Shape of enhanced electron density near impurity atom corresponds to spatial localization of Cr
dangling bond hybrid orbital unpaired electron when “upper” In atom is absent due to crystal cleavage. Directed shape of hybrid orbital with characteristic size \( \sim 0.5 \text{ nm} \) points to the presence of d-electron state [3]. Estimation of Coulomb repulsion energy \( U \) for localized electrons in Hubbard model yields \( U \sim e^2/\alpha_0 \sim 0.5 \text{ eV} \) if \( \alpha_0 \sim 0.5 \text{ nm} \). This value is comparable with band gap width. So STS spectra shift occurs in the vicinity of impurity state.

Analyzing the results of STS/STM investigations of different impurities we have found out some general similarities in behaviour of their STS/STM images and tunneling conductivity spectra. We have also examined fundamental physical effects responsible for specific features of tunneling spectra and changes in STM/STS images near atomic impurities of different types [4]. So it become possible to identify single atomic impurity in semiconductor matrix.

We should also mention that unique method firstly used here for registration of evolution of electronic density spatial distribution can be also applied for control and modification of information bit (Q-bit) states, based on individual interacting impurity atoms in semiconductor matrix [5].

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Local light polarisation mapping and electromagnetic field imaging by SNOM

A. A. Ejov, D. A. Muzychenko and V. I. Panov
Physical Department, Moscow State University, 119899 Moscow, Russia

Abstract. An influence of the incident light polarization on the aperture probe collection SNOM images have been investigated. Great difference between reflection SNOM images obtained at two orthogonal linear polarizations of the incident light have been observed. The SNOM images of domains and domain walls were studied. Three-dimensional light confinement near small hole in thin metallic film have been mapping by SNOM aperture probe.

Scanning near-field optical microscopy (SNOM) is rapidly emerging as a powerful tool for studying nanometer-scale phenomena such as optical properties of submicro- and nanos- tructures [1, 2]. Wide ranging of SNOM application demands to apply different modes of SNOM operation with different optical image formation mechanisms. This situation induces the great interest on the field of the physical principles of SNOM operation. Most popular aperture type SNOM is based on metallized sharpened optical fiber probe which may used both as nanosource or as nanodetector. The possibilities and limitations of SNOM with aperture type probe used as local electromagnetic field detector were the aim of presented work.

The experimental device was described previously [3, 4]. It is module type scanning probe microscope which was operated in SNOM mode. Nonoptical shear force probe sample distance regulation based on quartz tuning fork was used [5]. Chemical etching techniques [6, 7] were used for preparing sharpened optical fiber. The optical schemes of the device provide to use different optical elements combinations.

An influence of the incident light polarization on the aperture probe collection SNOM images have been investigated. Diffractional gratings which had been preparing by means of laser induced photoelectrochemical etching of doping GaAs and InP were used as the samples for this observation. In Fig. 1 a great difference between reflection SNOM images obtained at two orthogonal linear polarization state of the incident light is demonstrated. Dramatically decreasing of the individual features optical contrast in the case of $s$-polarization of incident light is noticed. Simple explanation of this effect may be the matching of electromagnetic boundary conditions at a dielectric-metal interface [9].

Iron-garnet films are one of the most popular objects for transmission SNOM magnetooptical investigation [8]. Optical images of iron-garnet films were shown in Fig. 2, where three different orientation of polarizers were applied for the differentiation between the optical images of domains and the optical images domain walls. It is seen that the polarization of local electromagnetic field detected by aperture SNOM probe permit us to resolve the distribution of polarization state in three dimensions.

Three-dimensional light confinement near small hole in thin metallic film have been mapping by SNOM aperture probe. The main aim of this part of the work was to observe the spatial distribution of evanescent light components. In Fig. 3 the local electromagnetic field with respect to sample probe distance is demonstrated. It is clear that decreasing of
Fig. 1. Topography and optical images of the same region of 400 nm InP diffractional grating (scan size 4800×4800 nm² for all images): (a) topographic image with 35 nm full grey scale; (b) optical image obtained with $s$-polarization incident light; full grey scale 250 arb. units; (c) optical image obtained with $p$-polarization incident light; full grey scale 480 arb. units. Arrows indicate the place of single defect.

Fig. 2. Optical images of the same region of iron-garnet film (scan size 5700×5700 nm² for all images): (a) and (b) images of magnetic domains with inverse contrast; full grey scale about 1100 arb. units; (c) domain walls image; full grey scale 550 arb. units.

Fig. 3. Topography and optical images of the same region of experimental CD surface after special Al deposition and subwavelength holes formation (scan size 2500×2500 nm² for all images): (a) topographic image with 170 nm full grey scale; (b) optical image obtained at sample probe distance 5 nm ($\lambda/100$); full grey scale 1500 arb. units; (c) optical image obtained at sample probe distance 500 nm ($\lambda$); full grey scale 500 arb. units.
the integral intensity accompanied simultaneous increasing of lateral size of hole optical image. This result demonstrate the localisation of evanescent light components. In conclusion it must be say that the local probing of the electromagnetic field realised by means of aperture type SNOM probe allow to observe several properties of the mapping light. This mode of SNOM operation provide complementary potential in comparison with the conventional SNOM mode which use aperture probe as nanosource of irradiation and detect light in far field. Effects associated with different combinations of incident light properties as it may be seen from the presented results require further investigations.

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References

Resonant second-harmonic phase spectroscopy of the buried interfaces of Column IV semiconductors

A. A. Fedyanin†, T. V. Dolgova†, D. Schuhmacher‡, G. Marowsky‡
and O. A. Aktsipetrov†
† Department of Physics, Moscow State University, 119899 Moscow, Russia
‡ Laser-Laboratorium Göttingen, Hans-Adolf-Krebs-Weg 1,
D-37077 Göttingen, Germany

Abstract. The second-harmonic interferometric spectroscopy (SHIS) is proposed as a new spectroscopic technique to study the resonant electron response of solid state nanostructures. The combination of the second-harmonic (SH) amplitude and phase spectra is shown to be more sensitive to resonant parameters of the electron density of states in comparison with the conventional SH intensity measurements. The resonant anisotropic SH response of buried oxidized Si(111) and Ge(111) surfaces is studied using SHIS in the vicinity of the $E_2$ critical points. Both spectra of the phase and amplitude of the SH field with different lineshapes for Si and Ge surfaces are obtained.

Introduction

The nonlinear-optical response of the solid-state nanostructures is an effective fingerprint of the properties of the quantum-confined electron subsystem of low-dimensional structures. The second-harmonic (SH) generation stands out among other nonlinear-optical techniques as inherently sensitive to nanostructures fabricated from centrosymmetric materials, since the SH generation is forbidden in the bulk of media with inversion symmetry in the dipole approximation. It means that the SH spectroscopy can be used for a study of the features of the electron density of states in nanostructures and in subsurface and interface layers of centrosymmetric semiconductors, where electron motion is also confined, that was successfully demonstrated in numerous works [1]. Recently the technique of a single-beam SH phase measurements [2] was modified in order to measure simultaneously the spectra of the phase and amplitude of the SH radiation of nanostructures, including the buried semiconductor interfaces [3]. The combination of the SH phase and amplitude spectra, extracted from the SH interferometric spectroscopy (SHIS) data, is more sensitive to parameters of resonances and it circumvents the sign uncertainty of the real part of quadratic susceptibility inhering in the conventional spectroscopy of the SH intensity. In this paper, the spectral dependences of the phase and amplitude of the SH field from the buried oxidized Si(111) and Ge(111) interfaces are measured in the vicinity of the $E_2$ critical point (CP) of Si and $E_1$ and $E_2$ CP of Ge using SHIS. The different types of spectral lineshapes of the SH resonances for Si and for Ge are observed.

Results and discussion

The samples are natively oxidized Ge(111) and slightly vicinal Si(111) wafers. The p-polarized output of a tunable nanosecond parametric generator/amplifier laser system operating in the interval of 490–690 nm is used. A 1 mm-thick plate of fused quartz coated with a 30 nm-thick indium-tin oxide (ITO) film is chosen as a reference for SHIS. The total
SH intensity $I_{2\omega}$ detected is a result of the coherent superposition of the SH fields from the reference, $E_{2\omega}^r$, and from the sample, $E_{2\omega}^s$. The dependence of $I_{2\omega}$ on the distance $l$ between the reference and the sample (the SH interferogram) is given as

$$I_{2\omega}(l) = \frac{c}{8\pi} |E_{2\omega}^r + E_{2\omega}^s|^2 = I_{2\omega}^r + I_{2\omega}^s + 2\alpha I_{2\omega}^r I_{2\omega}^s \cos \left( \frac{4\pi \Delta n}{\lambda_{2\omega}} l + \Phi_0 \right),$$  

with $\Delta n = n_{2\omega} - n_{\omega}$ describing the air dispersion and $\alpha < 1$ stands for the laser coherence. Additional position-independent phase shift $\Phi_0(\lambda_{2\omega}, \lambda_\omega)$ between $E_{2\omega}^r$ and $E_{2\omega}^s$ is associated with spectral properties of a quadratic susceptibility $\chi^{(2)}$ of the reference and the sample and Green’s corrections for SH radiation propagation and reflection.

$$I_{2\omega}^s(\psi) = \left| C_0 + C_1 e^{i\phi_1} \cos(\psi - \psi_0) + C_3 e^{i\phi_3} \cos[3(\psi - \psi_0)] \right|^2. \tag{2}$$

The one-fold component, $C_1$, comes entirely from the vicinal surface, while the three-fold, $C_3$, and isotropic, $C_0$, terms are generated by both surface dipole and bulk quadrupole sources. For Si(111)-SiO$_2$ interface $I_{2\omega}^s(\psi)$ dependences measured for different fundamental wavelengths are similar one to each other. This indicates that both anisotropic $C_1$, $C_3$ and isotropic $C_0$ components have almost the same spectral dependences, while their relative phases, $\phi_1$ and $\phi_3$, are spectral-independent. In the case of the Ge(111) surface, the vicinal contribution $C_1$ is negligible and the SH azimuthal dependences comes from the interference of the three-fold anisotropic and isotropic SH contributions. The $I_{2\omega}^s(\psi)$ dependence for Ge(111) surface varies from the almost three-fold shape at the “red” end of

**Fig. 1.** The SH rotational azimuthal dependences a slightly vicinal Si(111) (left panel) and Ge(111) (right panel) oxidized surfaces measured for different SH photon energies in the $p$-in, $p$-out geometry.
the tuning region to the six-fold shape at the “blue” end. It means that not only amplitudes of the isotropic and anisotropic contributions are spectral dependent, but the relative phase $\phi_3$ has also a pronounced spectral dependence, changing approximately on $\pi/2$ on the region of the fundamental wavelength tuning.

![Graph](image)

**Fig. 2.** The SH intensity (top panels) and the SH wave phase (bottom panels) as functions of the SH photon energy measured for Si(111) (left) and Ge(111) (right) buried interfaces in different points of the SH azimuthal dependence.

Figure 2 shows the spectra of intensity (top panel) and the phase (bottom panel) of the SH wave measured in the maximum of the p-in, p-out SH azimuthal anisotropy, where both isotropic and anisotropic SH components contribute to the SH signal (filled circles), and in the position, $30^\circ$ shifted from the maximum, where the SH response originates entirely from the isotropic term (open circles). For the Si(111) surface, both SH intensity spectra peak at approximately 4.3 eV close to the resonance of the $E_2$ CP and have a Lorenz-like line shape. The spectral dependences of the SH phase, extracted from the set of the SH interferograms, increase within the interval of 4.2–4.6 eV, which is typical for near-resonance phase behavior near $E_2$ CP; however outside this energy region the spectral derivative of the SH phase changes the sign, what unambiguously indicates the influence of the lower ($E'_0/E_1$) and higher ($E'_1$) resonances of Si interband transitions. In the case of the Ge(111) surface, the SH intensity spectra show a strong increase in the spectral range of 4.0–4.6 eV. However, the maxima of the spectra are sufficiently shifted from the interval of 4.1–4.3 eV, where both one-photon $E_1$ and two-photon $E_2$ resonances are located and where the SH phase spectra have the maxima of the spectral derivative in this region. Such a nonsymmetric line shape could be attributed with the 2D type of CP.

Summarizing, the general scheme of the SH interferometric spectroscopy is presented. Using this technique, the phase and amplitude of the SH radiation from the buried Si(111) and Ge(111) surfaces are measured in the interval of SH photon energies from 3.6 eV to 5 eV, and contributions from interband transitions located at $E_2$ critical point of Si and Ge are separated.
Acknowledgements

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References


Control of photocurrent relaxation in GaAs/AlGaAs nanostructures

S. P. Grishechkina, I. P. Kazakov, V. T. Trofimov, M. V. Valeyko
and N. A. Volchkov
A. N. Lebedev Physical Institute of RAS, 117924, Moscow, Russia

Abstract. Strong influence of an interband carrier generation on deep states relaxation in GaAs/AlGaAs nanostructures has been found. Considerable variation of relaxation time, sign and amplitude of impurity photoconductivity (PC) signal has been detected at rather low interband excitation. In single quantum well structures the additional interband excitation results in strong (more, than $10^3$ times) decrease of a deep states relaxation time. In modulation doped GaAs structures the interband generation results in long-time negative photoconductivity signal with decay time controlled by the interband generation intensity. The effect may be used for the analysis of deep states distribution in GaAs/AlGaAs nanostructures.

Introduction

The changes of semiconductor conductivity at photoexcitation yield the information on deep centers (DC). In the inhomogeneous structures this process is accompanied by spatial separation of the excited carriers and DC by built-in electric field, that results in relaxation time change along with bulk charge accumulation and band edge lineup distortion. A quantum well is the “ideal” uniform system for DC studies because the uniform photoexcitation in the quantum well is realized both in impurities and fundamental absorption regions provided the excitation is performed by “cold” radiation, which do not excite interband and intracenter transitions in barrier layers.

Solving of the reverse problem, namely the relaxation processes analysis of the known DC in semiconductor structure with built-in electric field, allows one to receive the information about band edge positions and DC spatial localization. The purpose of our activity is to study the relaxation of well known DC — EL2 centre — in the GaAs/Al$_{0.3}$Ga$_{0.7}$As nanostructures for analysis of the band edges behavior in nanostructure.

1. Experimental technique

For PC measurements we have used GaAs epitaxial layers uniformly doped by Si, Be and carbon, n-GaAs/Al$_{0.3}$Ga$_{0.7}$As single quantum well structure with 20 nm wide well and n-GaAs ($\delta$–Si) structures prepared by molecular beam epitaxy on i-GaAs substrates. Sheet carrier concentration and mobility at 77 K were $\sim 10^{12}$ cm$^{-2}$, and $\sim 6 \times 10^{4}$ cm$^2$/V·s and $\sim 6 \times 10^3$ cm$^2$/V·s for quantum well and $\delta$-doped structures respectively.

Optical excitation was made simultaneously by two sources: pulse semiconductor laser of $h\nu = 0.38 – 2$ eV and CW He-Ne laser ($h\nu = 1.97$ eV) or quartz lamp armed by appropriate optical filters, background radiation was completely screened. The kinetics measurements were performed mainly with $h\nu = 0.87$ eV source which causes only intracenter transitions in GaAs and practically does not excite DX centers in Al$_{0.3}$Ga$_{0.7}$As.
2. Results

The characteristics of PC signal mainly depended on conductivity type of the studied structure. Weak signal with onset photon energy $\sim 0.8 \text{ eV}$ and time constant $< 10^{-6} \text{ s}$ was found in p-type samples in the temperature range of 77–300 K. Background radiation had no effect on this signal.

Positive signal of impurity PC was observed for n-type structures at room temperatures. PC onset was found to be $\sim 0.8 \text{ eV}$ and the laser with $h\nu = 0.87 \text{ eV}$ was used further for excitation. As temperature was increased from 323 to 410 K, the relaxation time decreased from $7 \times 10^{-3}$ to $6 \times 10^{-5} \text{ s}$ for all studied structures having a good fit to $\tau(T) = \tau_0 \exp(\Delta E/kT)$ equation with $\tau_0 = 1 \times 10^{-12} \text{ s}$ and $\Delta E = 0.62 \text{ eV}$.

Under background radiation with photon energy larger than band gap $E_g$ both the time constant and the signal amplitude decreased. Even under relatively small intensity $\sim 10^{15} \text{l/(cm}^2 \cdot \text{s)}$ this decrease was about three order of magnitude (Fig. 1(a,b)). Background radiation with $h\nu < E_g$ did not influence PC signal. The correlation between impurity PC signal amplitude and luminescence intensity of carbon ($h\nu = 1.49 \text{ eV}$) was observed for uniformly doped samples.

Photoresponse signal characteristics at 77 K were drastically dependent on the structure configuration. The shape of photoresponse signal was not changed for GaAs/AlGaAs(Si) quantum well structure as compared to that at room temperature, PC threshold being $\sim E_g/2$. For GaAs $(\delta - \text{Si})$ structures the photoresponse shape changed essentially with the increase of CW irradiation with $h\nu > E_g$. First, the positive PC signal was detected, then the time constant started decreasing, and finally the PC signal became negative with the simultaneous sharp increase of time constant. As CW irradiation intensity further increased, time constant of negative PC signal decreased (Fig. 1(c,d)). PC signal sign inversion occurred at 150 K at sufficiently high irradiation intensity. The threshold photon energy of impurity PC was

![Fig. 1. Photoresponse signal for n-GaAs/AlGaAs single quantum well structure (a), (b) and n-GaAs(\delta - \text{Si})/GaAs (c), (d). Laser excitation with $h\nu = 0.87 \text{ eV}$ in darkness (a), (c) and under background CW-illumination with $h\nu = 1.97 \text{ eV}$ (b), (d).](image-url)
3. Discussion

Experimental results are explained in frame of single-electron model of the deep donor level in GaAs. Energy diagram of the n-GaAs/AlGaAs structure is shown in the Fig. 2(a) along with DC configuration diagram, which can be described by [1]:

$$H = \Delta/2\Delta_0 + (\Delta_0 + \Delta)n + F(1 - n),$$  \hspace{1cm} (1)

where $\Delta$ is the configuration coordinate, $\Delta_0$ is the energy of DC level, $F$ is the Fermi energy and $n$ is the number of electrons coupled to DC ($n = 0, 1$).

3.1. Model of positive PC signal

Impurity PC in n-GaAs is excited as a result of ionization of filled DC. Excited electron and DC are not separated spatially, so the time of free electron capture from conductance band to empty DC is determined by the time needed to overcome the potential barrier $\Delta E$ in configuration space which is described by thermoactivation dependence.

Impurity PC in the p-type semiconductor is excited as a result of electron transition from valence band to the empty DC. Time constant of PC for p-GaAs is much less than that for n-GaAs and do not depend on the temperature because the potential barrier for electron to return to the band can be neglected (see Fig. 2).

![Fig. 2. Energy configuration of n-GaAs/AlGaAs quantum well structure combined with configuration diagram of deep center (a) and of n-GaAs(δ−Si)/GaAs structure (b). (1, 2, 3)—possible optical transitions.](image)

Additional illumination of the n-GaAs structure by CW-radiation with $h\nu > E_g$ leads to the electron-hole pairs generation. Nonequilibrium holes are captured by DC’s and cause their partial recharge. While DC relaxation rate is proportional to empty centers concentration this accelerate the PC relaxation. DC’s are fully recharged under very high interband generation rate, so the electron transition from valence band to DC and its fast relaxation become the dominant process for impurity PC.

3.2. Model of negative PC signal

The negative PC arises in the inhomogenous in growth direction structures with built-in electric field, for example in n-GaAs(δ−Si), HEMT-structures etc. (Fig. 2(b)), if some empty DC are present. The transition of electron from the valence band to empty DC leads to negative PC generation. Appeared hole recombines with electron in conducting channel through the contact areas of the structure within a time $\sim 10^{-6}$ s, that results in persistent...
negative PC. The generation of holes by CW-radiation with $h\nu > E_g$ creates the possibility for electrons to return to the valence band in the pauses between pulses of the radiation with $h\nu < E_g$ and makes the negative PC observable.

Empty DC can be formed in undoped buffer due to contamination of epitaxial layers with carbon, which forms deep donor EL2 center in complex with antisite Ga$_{\text{As}}$ defect. When substituting the As atom, carbon acts as shallow acceptor and captures electrons from EL2 center. This is confirmed by correlation between PC amplitude and intensity of luminescence line of free carbon in studied samples.

We have performed numerical simulations of PC processes accounting all possible optical transitions (Fig. 2), recombination and relaxation channels. The comparison of experimental and simulated signals in dependence on excitation conditions allows to determine the charge state and to estimate the concentration of empty DC in the depth of the structure. It was found that the threshold temperature of negative PC signal generation is the most sensitive parameter for determination of DC concentration.

In summary, strong influence of interband carrier generation on the relaxation of impurity photoconductivity in $\text{A}_3\text{B}_5$ nanostructures has been detected. It has been demonstrated that the concentration of deep centers in the depth of structure can be determined by analysis of the photoconductivity signal.

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References

Quantum magnetotransport in the semimetal channel at the type II broken-gap GaInAsSb/InAs heterojunction

Ioffe Physico-Technical Institute, St Petersburg, Russia
e-mail: mkd@iorpt2.ioffe.rssi.ru
† International High Magnetic Fields and Low-Temperature Laboratory, Wroclaw, Poland

We report for the first time the study of quantum magnetotransport in the type II broken gap p-GaIn\(_{0.16}\)As\(_{0.22}\)Sb/p-InAs heterostructures with the self-consistent quantum wells under high magnetic fields up to 16 T. The structures lattice-matched to InAs substrate were grown by the LPE method with the high quality abrupt heterointerface (\(d = 12\) Å). Energy gap of the quaternary solid solution (\(E_g = 0.64\) eV at 77 K) and energy band bending at the interface were determined from photo- and electroluminescent measurements. Recently the electron channel with the high carrier mobility (\(\mu_H = (6-7) \times 10^5\) cm\(^2\)/Vs) was found out in the isotype single p-GaInAsSb/p-InAs heterojunction based on undoped quaternary layer [1].

Shubnikov–de Haas (SdH) oscillations in the Hall effect (\(\rho_{xy}\)) and magnetoresistance (\(\rho_{xx}\)) were studied at low temperatures (1.45–4.2 K) in magnetic fields up to 16 T. To identify the 2D-nature of the electrons in the channel we measured the angular dependence of SdH oscillations on the interface orientation in the magnetic field. Three groups of the SdH oscillations were observed (Fig. 1(a)). Two of them correspond two 2D-electron subbands \(E_1\) and \(E_2\) with the carrier concentrations \(n_1 = 1.1 \times 10^{11}\) cm\(^{-2}\) and \(n_2 = 4.2 \times 10^{11}\) cm\(^{-2}\),
Fig. 2. Sample A. \(a\)—Hall effect voltage at \(T = 1.45\) K, \(I = 0.2\) µA under ac condition in superconducting coil; \(a'\)—Hall effect voltage after the substraction of the non-linear background. 

\(b\)—Hall effect voltage at \(T = 2\) K, \(I = 50\) µA under dc condition in Bitter magnet system. Arrows indicate SdH extremum.

respectively, in the potential well at the p-InAs side of the interface (see Fig. 1(b)). The third period measured in high magnetic field (>10 T) can be ascribed to a hole subband with the carrier concentration \(p \sim 1 \times 10^{12}\) cm\(^{-2}\) at the quaternary solid solution side. Indeed, as shown in Fig. 2, in high magnetic fields the total Hall resistance exhibits a drastic reduction and changes a sign near 15 T. It indicates to a pronounced hole contribution into the parallel conductivity. In these magnetic fields at \(T = 1.45\) K we observed also electron spin-splitting of the SdH extremum \(N = 1.5\). Thus, the semimetal channel at the type II single p-GaIn\(_{0.16}\)As\(_{0.22}\)Sb/p-InAs heterointerface determines the lateral magnetotransport under high magnetic fields.

The most impressed result is the demonstration of integer quantum Hall effect (QHE) plateaus in the Hall conductivity \(\sigma_{xy}\) (Fig. 3) with the total filling factor \(\nu = 2, 3, 6\) in the magnetic field range according to the equations

\[
\sigma_{xy} = \frac{e^2}{h} \nu
\]

\(v = v_eE_1 + v_eE_2 - v_h\)

when \(E_1\) subband moves above the Fermi level (ultraquantum limit for 2D-electrons of \(E_1\)). The transition from the \(v = 3\) state to \(v = 2\) state occurs through the maximum value of \(\sigma_{xy}\) that was not observed for the quantum well structures with one type conductivity carriers. That result is due to the crossing the Fermi level by electron and hole Landau levels simultaneously.
Fig. 3. The conductivity tensor components versus magnetic field for samples with the different doping of GaInAsSb layer (A—$n_{R_0} = 3.4 \times 10^{11}$ cm$^{-2}$, B—$n_{R_0} = 4.2 \times 10^{11}$ cm$^{-2}$, C—$n_{R_0} = 9 \times 10^{11}$ cm$^{-2}$) at $H \rightarrow 0$. (a) Hall conductivity ($\sigma_{xy}$): calculated positions of quantum Hall effect plateaus are represented for several $\nu$ values. $\nu$ is the filling factor (the number of the occupied Landau levels). (b) Dissipative conductivity ($\sigma_{xx}$): arrows indicate the crossing of Landau levels with Fermi level. $1.5^+$ and $1.5^-$ are the result of the electron spin-splitting.

Recently the QHE was observed for the broken-gap GaSb-InAs-GaSb heterostructure grown by MBE with the same $\rho_{xy}$ plateau numbers [2]. It is important to notice, in our case, it is the first observation of QHE in a type II single GaInAsSb/InAs heterostructure with self-consistent quantum wells grown by LPE. Thus, it is established that in the single heterostructure the plateaus on the Hall resistance can be observed under condition when only the $E_2$ electron subband participates in total conductivity.

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References

Current instabilities in negative differential resistance region of a large area resonant tunneling diode

V. G. Popov, Yu. V. Dubrovskii, K. L. Wang†, L. Eaves‡ and J. C. Maan§

Institute of Microelectronics Technology RAS,
142432, Chernogolovka, Moscow District, Russia
† Electrical Engineering Department, University of California, Los-Angeles, USA
‡ University of Nottingham, NG72 RD Nottingham, United Kingdom
§ University of Nijmegen, 6525ED Nijmegen, The Netherlands

Abstract. The current instabilities in the negative differential conductance region of the resonance tunneling diode have been thoroughly studied at different parameters of the external circuit. The instabilities were dealt with an excitation of the current oscillations in the circuit. In a homogeneous approximation two small signal equivalent circuits of the resonant tunneling diode were used to describe the observed instabilities. Some revealed effects were out of the approximation.

Inhomogeneous current instabilities in the semiconductor devices are of the most interest investigated already 50 years. They were observed in a wide range of the devices [1]. As far as we know, a resonant tunneling diode (RTD) has not been experimentally investigated for the inhomogeneous current instabilities yet. Some theoretical works already predict this kind of instabilities in the RTD [2–6]. For RTD local probe method could not be lucky, in spite that it was successfully used to study the inhomogeneous instability previously [7]. Another way to study RTD for the instabilities is a comparison of the experimental data with calculated ones from widely used RTD models derived in the preposition of a homogeneous lateral current distribution along the tunnel junction [8, 9], and search for features which could not be explained by these models. For comparison with calculations we have measured \( R_T \) — the threshold negative differential resistance (NDR), i.e. the \( R_T \) values at which the current instabilities in the measuring circuit arisen.

The investigated resonant tunneling diodes were fabricated from InGaAs-AlAs heterostructure. Mesa structures had 70 μm in diameter and was made by conventional wet etching.

The simplified measurement circuit is shown in Fig. 1(a). The typical current-voltage characteristics of the diodes at room temperatures is shown in Fig. 2(a). The NDR was observed in the voltage range from 0.47 V to 1 V when the diode was shunted with resistance \( R_S = 25 \Omega \) and the inductance of the shunt was very low approximately 10 nH (curve 1 in Fig. 2(a)). To obtain actual I–V curves the current through the shunt was subtracted in situ by means of balance scheme. One can see current steps and jumps on the I–V curves (curve 2, Fig. 2(a)) measured without shunt resistance, but with \( C_S = 15 \text{ pF} \) capacity shunt, and \( R_L = 47 \Omega \), \( L_L = 100 \text{ mH} \) (see Fig. 1(a)). The current steps and jumps are just the manifestations of the current instabilities in the circuit. High frequency oscillations (up to 1 GHz) were observed in the voltage range of the current steps and they were absent out of the range. One can see the NDR region without instabilities at bias voltages between points A and B in Fig. 2(a), curve 2. The current oscillations arisen in the point A,
Fig. 1. (a) The measuring circuit with RTD. $R_L, L_L$ are load resistance and inductance, $U$ is sweep generator. (b) RC — model of the RTD. $R_C$ is contact resistance, $L_C$ is bonding inductance, $r$ is differential resistance of the tunnel junction, $C$ is differential capacity. (c) The ST — model of the RTD. $\delta \psi_i$ — electrochemical potential variations of the emitter ($i = 1$), well ($i = 2$), collector ($i = 3$). $G$ — emitter-well transconductance, $C_1, C_2, C$ — emitter, collector barriers and well capacitances.

Fig. 2. (a) The I–V characteristics of the RTD. Curve 1 is I–V curve measured for the diode shunting with the $R_S = 25 \, \Omega$ and $L_S \approx 10 \, \text{nH}$. Curve 2 is I–V curve measured for the diode shunting with the $C_S = 15 \, \text{pF}$. The threshold resistance $R_T$ is the NDR measured at point A. (b) The $R_T$ dependence upon the load resistance $R_L$. Square symbols are the experimental data. Triangle and circles symbols are calculated values for RC and ST models of the RTD.

When the bias voltage decreased. Thus measuring the NDR value in the point A we get the threshold resistance $R_T$. In the theoretical models $R_T$ can be related to the measuring circuit parameters and internal parameters of the diode. The dependencies of $R_T$ on the different parameters of the measuring circuit were obtained experimentally and then compared with calculated ones.

Figure 2(b) shows the dependence of the threshold resistance $R_T$ on the $R_L$ — load resistance. Other circuit parameters were fixed. Figure 3(a) shows the dependence of the threshold resistance $R_T$ on the $R_S$ — shunt resistance, with $R_L = 9.8 \, \Omega$, and $L_L = 10 \, \text{mH}$ kept constant, in this measurements we did not add any shunt capacitance. Figure 3(b) shows the dependence of the threshold resistance $R_T$ on the $C_S$ — shunt capacitance, with
fixed $R_L = 47 \, \Omega$, and $L_L = 10 \, \text{mH}$, shunt resistance was absent. In all the figures the experimental data are presented by square symbols. To calculate theoretical values of the threshold resistance for different circuit parameters it is necessary to know equivalent RTD circuit which definitely include NDR. Than knowing the full equivalent circuit we found the boundary between circuit stable and unstable conditions. From the boundary conditions relations between $R_T$ other circuits parameters can be found. The equivalent RTD circuit depends on the model used to describe the resonant tunneling diode. We used two equivalent circuits: the first one — RC model (Fig. 1(b)) — is suitable for coherent tunneling [8], and the second one — ST model (Fig. 1(c)) — for sequential tunneling processes [9]. It is necessary to emphasize here, once more, that both equivalent circuits described the uniform distribution of the current along the tunneling structure.

The calculated data are shown in Figs. 2(b)–3(b) by triangles for RC model, and by circles for ST model. The procedure of comparison was following. First of all from comparison of $R_T$ dependencies on $R_L$ (Fig. 2(b)) we obtain best fit parameters for $R_C$, $L_C$ and $C$ for RC model and $R_C$, $L_C$, $C_1$, $C_2$, $v$ for ST model. With found best fit parameters theoretical dependence of $R_T$ on $R_S$ was drawn in Fig. 3(a). The $R_T$ experimental and theoretical values differ two times. If the best fit was done for data in Fig. 3(a), than the strong discrepancies appeared for $R_T$ on $R_S$ experimental and calculated dependencies. In other words it was impossible to make best fit for $R_T$ on $R_L$ and $R_S$ dependencies at the same time. More drastic discrepancies was found for $R_T$ on $C_S$ data (see Fig. 3(b)). For both model in the range of used $C_S$ value the dependence should be constant. The experiment shows quite complicated dependence with clear local minimum around $C_S = 7 \, \text{pF}$. The experiment has demonstrated that the current instabilities of the RTD with 20 nF internal capacity are very sensitive to change of $\Delta C_S \sim 1 \, \text{pF}$. Moreover a frequency of the current oscillations is about 250 MHz that is higher than the cut-off frequency of the diode ($\sim 10 \, \text{MHz}$) calculated for both models. It worth to note that the models have been tested with ultra-high frequency techniques on the diode with quite small lateral sizes ($\sim 20–30 \, \mu\text{m}$) [8, 9] and relatively large negative differential resistance. We used the diode that had larger lateral sizes than in previous studies. From the comparison of the experimental and calculated data we concluded that current models based on the uniform distribution of the tunneling current along the junction cannot explain experimental findings. The theories where inhomogeneous distribution of the current along the junction was considered did not propose any equivalent circuit which in principle would permit one
to make the similar comparison with experimental data. It means that more sophisticated approaches are necessary to develop for proving these theories.

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References


Fine structure of photoresponse spectra in double-barrier resonant tunneling diode

S. A. Vitusevich†1, A. Förster†, W. Reetz†, H. Lüth†, A. E. Belyaev‡ and S. V. Danylyuk‡

† Institut für Schicht- und Ionentechnik, Forschungszentrum Jülich, D-52425 Jülich, Germany
‡ Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, Kiev 252028, Ukraine

Abstract. The photo-responsivity spectra of double-barrier resonant tunneling diodes have been measured in a wide range of light wavelength as well as applied voltage. The complex behaviour of measured spectra is analysed taking into account different channels for electron injection into the quantum well (QW). The results obtained yield evidence for modulated electron–hole (e−h) recombination in the QW provided by direct excitation of e−h pairs in the QW.

Over the last decade resonant tunneling diodes based on double-barrier heterostructures attract an elevated attention due to their electronic and photonic applications. The latters were concentrated on utilization of the unique electronic and optic properties of these devices for lasers, detectors, and modulators in the infrared to visible wavelength range [1, 2], and high-speed optically switched electronic devices [3, 4]. At the same time investigation of such devices under illumination could be successfully used for the understanding and analysis of basic electronic processes occurring in vertical transport devices.

This communication presents photo-responsivity measurements done as a function of excitation wavelength on a GaAs/AlAs double-barrier resonant tunneling diode. The results obtained at various bias voltages help to elucidate the relative contributions to the tunnel current from carriers directly injected into the well from 3D emitter, versus those that tunnel in from an accumulation layer formed in front of the first barrier. Furthermore, they yield evidence for modulated electron–hole (e−h) recombination in the well provided by direct excitation of e−h pairs in the well. The resonant tunneling diodes (RTDs) we have studied were manufactured with the use of a GaAs/AlAs double-barrier heterostructure grown by molecular beam epitaxy (MBE) on n+-(100)GaAs substrate. Active part of RTD studied in this work consists of a 4-nm-thick GaAs quantum well (QW) with 2-nm-thick AlAs barriers on both sides. These three layers were nominally undoped. A 100-nm-thick undoped GaAs spacer layer was grown adjacent to the barrier on the substrate side and was separated from the substrate by a 100-nm-thick highly doped (∼10^{18} \text{ cm}^{-3} \text{ Si}) n⁺-GaAs electrode region. A 100-nm-thick GaAs top layer of similar doping was grown directly on top of the other barrier followed by 0.5-µm-thick heavy doped GaAs contact layer. The 16 × 16 µm² square devices were defined by wet-chemical mesa etching and Au/Ge/Ni alloyed ohmic contacts were fabricated on substrate and on top of the mesa to provide measurements under applied bias.

1On leave from the Institute of Semiconductor Physics, National Academy of Sciences of Ukraine, Kiev 252028, Ukraine
The responsivity spectra of the RTD were measured using a standard spectral response system based on a 450 W Xenon lamp and a grating monochromator. The bandwidth of the monochromatic light is 20 nm and the wavelength is changed in the 400 to 900 nm wavelength range in 10 nm steps. A calibrated single crystalline silicon detector is used for the system calibration which gives the light intensity at each wavelength position. The RTD was illuminated by light incident at 45° on the side-wall facet. With an incident optical power of 30 nW, corresponding to an irradiance of 0.043 mW/cm² we did not observe visible changes in current–voltage characteristics. Thus, the optically generated carriers did not significantly modify the potential distribution within the device. Typical responsivity spectra of the RTD, measured at 77 K and with different applied bias, are shown in Fig. 1(a). These spectra reveal a narrow band (labeled A in Fig. 1(a)) at 1.5 eV and a broad band (labeled B in Fig. 1(a)) at 1.65 eV with an extended short-wavelength tail. The intensities of both bands strongly depend on applied voltage and qualitatively follow the dark current dependence of the I–V curve (Fig. 1(b)). The current–voltage characteristics of the RTD measured in the dark reveals three resonant peaks under forward bias (Fig. 1(b)), instead of the two peaks which actually appeared at reversed bias polarity. In all measurements the forward bias corresponds to a negatively biased substrate. Previously we suggested that the first two peaks observed for both bias polarities arise as a result of tunneling of electrons injected from the 3D emitter through the first and second quasi-bound states of the QW, while the third peak observed at forward bias is stipulated by electrons injected from a 2D level in the accumulation layer formed in the front of the emitter barrier [5]. In the further we will concentrate on results obtained under forward bias.

In the band-to-band excitation case with energy close to GaAs bandgap the $e – h$ pairs are creating throughout the structure outside the QW. The $e – h$ pairs generated in the collector are shifted by the electric field. Electrons move towards the doped layer and recombine, thus do not contribute to the photocurrent, while the holes move to the collector barrier and are accumulated there. These carriers could tunnel throughout the double barrier structure directly or overcome the barrier thermionically. The latter could be neglected

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**Fig. 1.** (a) Photo-responsivity spectra measured at $T = 77$ K on the RTD biased at: 1—0.6 V, 2—0.8 V, 3—1.7 V, 4—2.0 V. (b) Current–voltage characteristics of the RTD measured at $T = 77$ K under forward bias. Solid circles and triangles show the intensity of the photo-response corresponding to maxima of the bands A (circles) and B (triangles). Enlarged dependence of these values within the first resonance is shown on inset.
due to the height of the barriers and the low temperature. The tunneling holes recombine
with electrons on the emitter side and thus contribute to the photocurrent. In contrast, the
electrons photogenerated on the emitter side are injected into the QW. The magnitude of
photo-response increases with applied voltage under resonant conditions while it is low
at biases corresponding to the resonance-off conditions. On the spectral dependence of
the photo-response the processes conditions. On the spectral dependence of the photo-
response the processes mentioned above should reveal itselfs as narrow asymmetric band
with cut-off corresponding to the GaAs gap energy. That is because with increasing energy
of the incident photons we should expect a sharp decrease of the photocurrent due to a
high absorption coefficient and, consequently, of a small amount of photogenerated pairs.
Surprisingly, a deep dip has been observed followed by a wide band, as it is seen in Fig. 1(a).
The shape of the band is strongly affected on the applied voltage. There is a pronounced
maximum at 1.65 eV followed by short-wavelength tail within the first resonance, while the
band becomes more flat with the center of gravity shifted towards higher excitation energy
within the second resonance and, finally, it almost vanishes at voltages corresponding to
passage of carriers above the barrier. The behaviour of the spectral response like that could
be explained with the following arguments. If the energy of the incident photons coinsides
with the energy difference between electronic and hole quasi-bound states in the QW, a
direct generation of $e^-h$ pairs inside the QW is possible. The photo-carriers are removed
out the QW by the electric field remaining behind holes which are filling immediately by
electrically injected electrons from the emitter as well as holes from the accumulation layer
adjacent to the collector barrier thus giving rise to a photocurrent. Such process should be
efficient at resonance, while it is insignificant in the off-resonance regime because of a low
probability to supply carriers into the QW. This implies a strong field dependence of the
photocurrent.

It is worth to note there is an essential limitation of the photocurrent intensity related to
the recombination of the photogenerated carriers. It has been reported earlier [6] that a QW
photoluminescence (PL) in double barrier structures reveals field-dependent behaviour.
Moreover, Vodjdani et al. [7] have shown the radiative recombination does occur in the
QW and roughly follows the current variation with bias either the $e^-h$ pairs electrically
injected or photocreated directly in the QW. At the same time the PL dynamics directly
reflects the source for holes participating in the recombination. The study of time-resolved
photoluminescence from excitons in the QW shows that the net decay time of holes inside
the QW is much shorter if the holes are created directly in the QW [8].

Now, it becomes clear why the peak intensity of the band B (Fig. 1(a)) grows more
quickly that one of the band A under voltage region corresponding to the first resonance.
Indeed, the QW luminescence is sublinear in the electron density and saturates at high
electron injection. Taking into account the difference between the decay times for holes
generated by different processes, we can conclude that a saturation occurs at lower bias
in the case of the $e^-h$ pairs generation directly in the QW. The fact is confirmed by the
$R_{p}^{A,B} = f(V)$ dependences plotted in the inset of Fig. 1(b). On the other hand, in the voltage
region corresponding to the second resonance the recombination will be determined by the
number of electrons which came out from the resonance and relaxed onto the ground quasi-
bound state of the QW. This amount, obviously, is smaller than is necessary for a saturation
and the $R_{p}^{A,B} = f(V)$ dependences almost coincide.

It should be noted that the spectral responsivity data obtained with illumination of the
RTD in the wavelength range corresponding to interband transitions do not allow to distin-
guish directly either carriers injected into the QW from 3D or 2D channels. Nevertheless,
an explanation of the experimental results given above is more consistent with ballistic transport of tunneling electrons.

In conclusion, we have observed considerable transformation of the responsivity spectra in double-barrier resonant tunneling diode resulting on different origin of the photogenerated carriers.

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References
Formation of defect-free InGaAs-GaAs quantum dots for 1.3 µm spectral range grown by metal-organic chemical vapor deposition

I. L. Krestnikov†‡, N. N. Ledentsov†, M. V. Maximov‡, D. Bimberg†, D. A. Bedarev‡, I. V. Kochnev‡, V. M. Lantratov‡, N. A. Cherkashin‡, Yu. G. Musikhin‡ and Zh. I. Alferov‡

† Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Recently, remarkable progress is reported in GaAs-based quantum dot (QD) lasers emitting near 1.3 µm [1–5]. For single-sheet QD lasers ultralow threshold current densities (∼20 A/cm², 300 K) are reported for the case of low mirror losses [3]. To come to practical device applications requesting high power and high-frequency applications one needs, however, to use stacked 1.3 µm-emitting QDs [2] to ensure sufficient modal gain to maintain lasing at requested cavity lengths and facet reflectivities. Due to this approach, high-power (2.7 W CW) operation [5] in broad area devices and single transverse mode (300 mW) operation in narrow (7 µm) stripes [4] were realized. Very recently, modal gain as high as 50 cm⁻¹ in the 1.3 µm range was obtained for injection lasers based on 10-fold stacked InAs QDs in a GaAs matrix [6].

All these results are obtained, however, for long-wavelength QDs grown by molecular beam epitaxy (MBE) [7]. As opposite, despite the first GaAs-based QDs emitting in the 1.3 µm range both under photoexcitation [8] and current injection [9] were reported for MOCVD growth, these structures seemed to be not suitable for laser applications due to low density of QDs and high density of defects [9–10] and only lasers emitting in the range of up to 1.1 µm are developed [11–12].

In this paper we have grown by metal-organic chemical vapor deposition structures with InGaAs QDs and studied the influence of post-deposition treatment on their structural and optical properties. The samples studied in this work are grown by MOCVD using trimethyl gallium (TMG), trimethyl aluminium (TMA), ethyl-dimethyl indium (EDMI) and arsine (AsH₃). Hydrogen is used as a carrier gas. The total pressure in the reactor is kept at 76 Torr and the flux ratio between group III elements and arsine is 75. The structures are grown on semi-insulating GaAs (100) substrates. First, a Al₀.₁₅Ga₀.₈₅As buffer layer (0.8 µm) is grown at 600 °C, followed by a 0.1 µm-thick GaAs layer. The substrate temperature is then reduced to 490 °C and In₀.₅Ga₀.₅As deposition (or In₀.₅Ga₀.₅As/GaAs multilayer deposition) is performed. In the first case 3 monolayers (ML) of In₀.₅Ga₀.₅As are deposited. In the second case the deposition of 3 MLs of In₀.₅Ga₀.₅As is followed by multi-cycle GaAs (140 nm) — In₀.₅Ga₀.₅As (2.5 ML) deposition (two periods). After this 40 nm of GaAs is grown at 490 °C, the substrate temperature is increased to 600 °C, and 40 nm of GaAs is deposited. To avoid surface recombination of nonequilibrium carriers 20 nm of Al₀.₃Ga₀.₇As is grown on the top at the same temperature. As opposite to the case of vertically-coupled MOCVD QDs [13], large separation between QD prevented any wavefunction-coupling effects. Two types of structures were grown. In one case, the InGaAs deposition was completed with GaAs overgrowth at 490 °C. In the other case, for both single and multi-sheet structures, first thin GaAs layer (8 nm) is deposited at 490 °C and,
then the substrate temperature is increased to 600°C for 10 minutes under AsH₃ exposure. During this procedure InAs accumulated in large clusters, which are not covered by thin GaAs cap layer, evaporates and redistributes through the GaAs surface forming the second wetting layer. This procedure was shown using deep level transient spectroscopy (DLTS) to result in complete suppression of the EL2 and EL3 traps, associated with dislocations, and in reduction of the concentration of deep traps associated with point defects by more than one order of magnitude [14].

Transmission electron microscopy (TEM) and high-resolution electron microscopy (HREM) studies are performed by using a high voltage Philips EM-420 (100kV) microscope. Plan-view and cross-section TEM images are taken under (220) and (200) diffraction conditions, respectively. Photoluminescence (PL) is excited by using the 514.5 nm line of a Ar⁺ laser and detected by using a cooled germanium pin-photodetector.

In Fig. 1 we show plan view TEM images of the structures with single QD insertions grown without or with annealing steps (Fig. 1(a) and (b), respectively). One can clearly see coherent InGaAs QDs on both images with areal density of about $2 \times 10^{10}$ cm$^{-2}$. The lateral size of QDs is about 20–25 nm, being similar to reported in [13]. In addition to coherent QDs, the structure grown without the annealing step demonstrates contrast features due to dislocated InGaAs clusters, having a density of about $0.5 \times 10^9$ cm$^{-2}$. No such clusters are revealed in the structure grown with the annealing step.

In the case of the plan-view images, the foil thickness was kept below 100 nm to allow QD imaging. The only upper row of InGaAs QDs is trapped in the TEM foil in this case. As it is clearly seen from Fig. 2, the QDs are present in the upper sheet with InGaAs insertion only in the case of the structure grown with the annealing steps. The QD in-plane density in this case is similar to that for the single-sheet insertion ($\sim 2 \times 10^{10}$ cm$^{-2}$). As opposite, the structure grown without the annealing steps demonstrates complete disappearance of QDs in the upper InGaAs sheet region. One may conclude, that most of the InGaAs material is trapped in the regions in the vicinity of propagating dislocations originating at the clusters in the first row with QDs. These dislocations are clearly visible in the plan-view (Fig. 2(a)) cross-section TEM (Fig. 2(b)) images of the stacked InGaAs insertions grown without the annealing step. Thus, the concept of stacking of QDs does not work without the annealing step.

Cross-section TEM image of the structure grown with the annealing step demonstrates
Fig. 2. Plan view (a, c) and cross-section (b, d) TEM images of the structures with 3-fold stacked InGaAs insertions grown without (a, b) and with (c, d) annealing steps.

complete suppression of defect formation (see Fig. 2 (c),(d)). Higher magnification cross-section TEM images of this sample demonstrate also an additional dark stripe in the contrast due to the second InGaAs wetting layer formed from the InGaAs accumulated in large dislocated clusters, which are not completely covered with 8 nm-thick GaAs cap layer, as opposite to coherent QDs having smaller height [8, 10, 13].

Formation of defects has significant impact on optical properties of the structures studied. The PL spectrum of the sample with single InGaAs QD insertion grown with the annealing step is shown in Fig. 3. At room temperature relatively broad PL emission in the range 1–1.4 µm peaking at around 1.3 µm is observed. At high excitation density PL
due to excited state of quantum dots becomes more pronounced. The peak $I$ is attributed
 to InGaAs two-dimensional islands having several monolayer height similar to discussed in [10, 11]. The single-sheet structure grown without the annealing step shows similar PL spectrum, but the PL intensity is an order of magnitude lower at 300 K.

In the case of the stacked QD structure grown with the annealing step the PL spectrum is
 similar to shown in Fig. 3 in lineshape and intensity both at low temperatures and at 300 K.
 As opposite, the PL spectrum of the stacked InGaAs-GaAs structure grown without the
 annealing step shows only weak emission in the 850–1000 nm range at low temperatures and
 essentially no PL in the long-wavelength range. This indicates that most of the carriers are
 trapped in the wetting layer- or island-induced states dominating in the upper rows of
 the stacked structure in agreement with TEM data. At room temperature, the PL intensity
degrades by 3 orders of magnitude in this case.

To conclude, we have investigated structural and optical properties of MOCVD-grown
 single and stacked InGaAs insertions formed with and without annealing step after thin-
 layer overgrowth. We found that this overgrowth/annealing procedure results in elimina-
 tion of dislocated InGaAs clusters, thus, only the structures grown with such an approach
 are suitable for long-wavelength laser applications for the growth mode chosen in this
 work. Additionally, according to our results, growth of stacked long-wavelength QDs is
 principally possible only when QDs are subjected to the cluster-elimination procedure.

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Optically detected magnetic resonance of semiconductor quantum dots

E. Lifshitz, A. Glozman and I. D. Litvin
Department of Chemistry and Solid State Institute,
Technion – Israel Institute of Technology, Haifa 32000, Israel
e-mail: sefrat@tx.technion.ac.il

Abstract. Semiconductor quantum dots have attracted large scientific and technological interests in the last decade. This document describes our attempts to characterize the localization of carriers in CdSe and CdS quantum dots, utilizing optically detected spin and orbit magnetic resonance spectroscopy. The spin resonance resembled a typical magnetic resonance spectrum and has been analyzed in a similar manner. For example, spin Hamiltonian simulation of the paramagnetic spin interactions enabled the chemical identification of carrier’s trapping sites (e.g., core, surface or interface). On the other hand, the precession motion around the direction of the external magnetic field was restricted by the so-called magnetic length $\lambda_m = \sqrt{\hbar / eB}$. Still, theoretical consideration associated either with ballistic motion of carriers against the boundary, or orbital motion should enable the determination of the effective mass of an electron, hole and additional charged specie (presumably a charged exciton).

Introduction

Nearly two-decades of research have explored the benefits in the optical and opto-electronic properties of semiconductor quantum dots (QDs) [1]. These materials exhibit unique chemical and physical properties, differing substantially from those of the corresponding bulk solids. Evidently, these differences are associated with quantum-sized effect and the relatively large number of atoms at the QD’s surface.

The quantum size effect had been studied extensively in II–VI, III–V, IV–VI and I–VII (e.g., CuCl) randomly dispersed or as single QDs [2]. These studies showed that the aforementioned materials offer the opportunity to tune the electronic and optical properties, with variation in the QD’s size. However, the influence of surface/interface at the current time is not well understood. The quality of the surface becomes important in determining electron and hole trapping together with the chemical reactivity. Trapping of charge carriers may influence the observed emission properties to a large extent. For example, the photoluminescence (PL) of QDs occasionally present red-shifted luminescence band associated with surface recombination [3] or surface states mixing with the band edge [4]. Trapped carriers at the surface may polarize the core exciton, leading to a delayed fluorescence and multi-exponential decay process (in combination with the influence of dark states) [5]. It was shown that light emission from a single QD, under continuous excitation, turns on and off intermittently with a characteristic time-scale of seconds [2]. This phenomenon can be due to trapping or to an Auger process at the surface. Thus, the ultimate goal of the described research is concerned with the identification of the surface/interface sites, and trapping and de-trapping processes of photo-generated carriers at those sites. Furthermore, the influence of the aforementioned issues on the recombination processes is mainly emphasized.

Currently there are three main methods for the fabrication of QDs: (1) Chemical, Electrochemical and Epitaxial deposition of a QD’s compound on top of a substrate with
a different lattice constant leads to strain induced three dimensional islands, known as Stranski–Krastanow (SK) QDs [6–9]; (2) Colloidal chemistry techniques, which enable the growth of QDs with diameters of <10 nm, size distribution of <10%, strain-free dots and the termination of surfaces with organic or inorganic capping [1, 2]; (3) In situ growth of QDs during the preparation of glass or polymer media [10].

The present document discusses the study of representative CdS and CdSe QDs samples, prepared in either one of the synthetic methods discussed above. Optically detected magnetic resonance (ODMR) spectroscopy was used for the study of the localization properties of photo-generated carriers in the studied materials. This method provides the means to chemically identify core or surface/interface states, and correlate them with specific optical transition.

**Experimental**

Colloidal CdSe QDs, capped either with tri-octy-phosphine-oxide [TOPO] or epitaxial layer of CdS were prepared according to the procedure described in [11]. Then these colloidal QDs were embedded in a transparent polymer film [12] for the convenience of the optical measurements. Strain grown chemical deposited QDs were prepared according to the procedure given in Ref. [13], while CdSe and CdS QDs grown in glass media were prepared by the procedure given in [14].

The ODMR spectra were recorded by placing the sample on a special sample probe at the center of a High-Q resonance cavity, coupled to a microwave (mw) source (~10 GHz), and surrounded by a superconducting magnet. These spectra were obtained by measuring the change in luminescence intensity induced either by a spin or orbit magnetic resonance event at the excited state. This change was plotted versus the strength of the external magnetic field, B.

**Results and discussion**

Representative ODMR spectra of colloidal CdSe (capped with CdS monolayer), CdSe deposited on glass, and CdS grown in a phosphate glass, are shown in Fig. 1. Each spectrum consists of two resonance signals (narrow and wide), ranging between 2000–5000 Gauss, super-imposing on a wide background. Recording of the ODMR spectra at various experimental conditions, by varying the modulation frequency of the mw or

![Fig. 1](image_url)

**Fig. 1.** Spin ODMR spectra of following samples: (a) Colloidal CdSe QDs, capped with epitaxial layer of CdS, (b) Chemical deposited CdSe QDs and (c) CdS QDs grown in phosphate glass medium.
laser excitation power, showed in all cases that the distinct resonance signals have different response from that of the background (not shown). In addition, the narrow and wide signals also differ from each other. This suggests that the spectra consist of several resonance events, associated with different chemical sites. Theoretical considerations (e.g., treatment of the external magnetic field as a perturbation of the electron motion in confined dots) may suggest that the background is associated either with an orbit or cyclotron motion of the carriers, while the distinct narrow and wide band, correspond to their spin motion. The latter were simulated by the use of the following phenomenological spin Hamiltonian:

\[ H_s = \beta S_e g_e B + \beta S_h g_h B + S_e D S_h + J S_e S_h \]  

(1)

The first two terms correspond to the effective Zeeman interaction of an electron and hole, the third term corresponds to the zero field splitting (may be overlapped by an anisotropic exchange interaction), while the last term, to the isotropic electron–hole (e–h) exchange interactions. The spin Hamiltonian simulation suggests the trapping of a hole within the core of the QD and trapping of an electron at the surface. Twin boundary or edge dislocations were identified as the carrier’s trapping sites.

The resonance background, ranging between 0–8000 Gauss (after the subtraction of the spin resonance contribution) is plotted in Fig. 2. Theoretical considerations published elsewhere [12] suggest the correlation between the peak maxima and the effective mass parameters. The represented curves consist of three signals with corresponding effective masses, as indicated in the figure. The 0.13\(m_0\) is identical with the bulk electron effective mass, while the 0.41\(m_0\) is close to the hole effective mass of bulk CdSe. The third resonance has an effective mass of 0.74\(m_0\), presumably corresponding to a three-body complex, composed of one hole bounded to two electrons (e.g., charged exciton). This charged exciton is found in various quantum structures recently, while never been discussed before in semiconductor QDs.

References


Properties of InAs quantum dots on silicon(001) and (111)

L. Hansen  A. Ankudinov‡, F. Bensing†, J. Wagner†, G. Ade§, P. Hinze§, V. Wagner†, J. Geurts† and A. Waag†
† Physikalisches Institut EP III, Am Hubland, 97074 Würzburg, Germany
‡ on leave from Ioffe Institute, Polytechnicheskaya 26, St. Petersburg, Russia
§ Pysikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany

Abstract. The self assembled formation of InAs Quantum Dots (QDs) on silicon has been studied by RHEED, AFM, Raman and TEM. On Si(001) up to $10^{11}$ cm$^{-2}$ InAs QDs can either be grown in a conventional growth mode, or by utilizing a postgrowth dewetting transition at decreasing substrate temperatures. On Si(111) instead, QDs only form on an As-passivated surface. Otherwise InAs grows in large 2D clusters.

Introduction

Besides first successes with the (heterovalent) growth of germanium QDs on Si, the most popular material of semiconductor industry, silicon, has remained to be a great challenge as a material for optoelectronic devices due to its indirect bandgap [1]. Recent results on the self-organized growth of InAs QDs embedded in a silicon matrix seem to be very promising. An intense photoluminescence peak at a wavelength of 1.3 $\mu$m could be observed up to room temperature [2]. In a former study on the embedding of the QDs in a silicon matrix we report on the possibility of growing up to $10^{11}$ cm$^{-2}$ InAs QDs and the formation of large Indium clusters between the InAs QDs [3]. Further, we investigated the QD size distribution, ways of improving their uniformity as well as the defect structure in the QDs, information which is essential for a potential application of this system [4]. So far, the problem arises that RHEED, Raman and TEM indicate that after the 2D/3D growth mode transition of InAs on Si, the InAs QDs are fully relaxed with the inclusion of misfit dislocations. It seems that an annealing after the growth of a Si-cap layer finally leads to the formation of strained InAs inclusions in the Si matrix [5]. Besides that, the investigation of QD growth on differently terminated silicon substrates is believed to give new insight into the physics of self-organization in the InAs/Si system. A dewetting transition of a single monolayer (ML) of InAs during the postgrowth lowering of the temperature has been observed and being described on the basis of a thermodynamic model in [4]. For this paper, we compare the properties of InAs growth on Si(001) with Si(111) where growth depends strongly on the surface termination. InAs forms large clusters when grown directly on Si(111), whereas on the arsenic terminated surface QDs show up.

1. Experimental details

Growth experiments were carried out on 2 inch Si-substrates using a Riber 2300 MBE system. (001) substrates were Hydrogen-passivated, details on their preparation and optimized growth conditions can be found in [3, 4]. (111) substrates were either also H-passivated or after an RCA-etch the protective SiO$_2$ layer was desorbed in a separate chamber belonging to the same UHV-MBE-cluster. Growth was monitored in-situ by RHEED, the Raman
measurements were performed without breaking the UHV via an UHV transport-box. TEM images were obtained with a Phillips CM 200 FEG at 200 kV. AFM images, taken with a Rasterscope 4000, were evaluated by SPM image magic software.

2. Results and discussion

On Si(001) the 2D–3D transition in growth mode occurs after depositing 1.7 ML of InAs at a growth temperature of 370°C. The critical thickness has been observed to depend on the growth temperature [2] which qualitatively matches with our experiments, though we always determine a lower critical thickness than reported in [2]. Figure 1(a) shows an AFM image of a sample with nominally 5 ML of InAs on Si(001) leading to approximately $10^{11}$ cm$^{-2}$ QDs. Statistics on radii and heights were evaluated and show a Gaussian distribution with mean values of 14.2 nm and 10 nm, respectively. Cross sectional TEM images let us assume, however, that the real size of the nanostructures is smaller (Fig. 1(b)). Indeed, the shape of the AFM tip has not been accounted for, and thus lateral sizes of QDs are overestimated by AFM. Information on the coherence of the QDs can be achieved by an analysis of RHEED images obtained during the growth which directly shows a projection of the reciprocal space of the epitaxial surface. At the growth-mode change, we observe an abrupt 10% decrease of the lateral distance of the appearing 3D spots compared to the former 2D streak pattern of Si(001). This indicates the full relaxation of the dots. The lattice mismatch between InAs ($a = 6.058$ Å) and Si ($a = 5.431$ Å) makes up 11.5%. As a consequence TEM images (Fig. 1(b)) show the inclusion of misfit dislocations. An averaged information on the whole QD ensemble can be achieved by Raman spectroscopy. Those spectra, taken on a sample with 3 ML InAs, show optical phonon modes of the InAs dots in the range from 216 cm$^{-1}$ to 238 cm$^{-1}$ (TO and LO), which corresponds to relaxed bulk frequencies of InAs (218 cm$^{-1}$ and 243 cm$^{-1}$). In contrast, a fully strained state would result in a shift of 30 cm$^{-1}$ to higher wavenumbers, which therefore can be excluded in these structures.

![Fig. 1. (a) 500 nm$^2$ AFM image of 5 ML InAs/Si. (b) TEM image of the same sample.](image_url)

To avoid nonradiative recombination of electron hole pairs at defects inside the QDs the structural quality of the QDs has to be improved. An interesting phenomenon in this context is the observation of a dewetting transition of a thin (below the critical thickness) InAs-film on Si(001). When reducing the substrate temperature after growth the 2D layer gets thermodynamically unstable and a spotty RHEED image indicates an abrupt transition into 3 to $8 \times 10^{10}$ cm$^{-2}$ islands. Though the average size of those QDs determined by AFM is smaller than for the dots grown in the conventional growth mode, TEM images again show misfit dislocations inside of the dots.

It has always been pointed out that strain is a necessary but not sufficient criterion for the
selforganized formation of QDs. For InAs on GaAs the formation of QDs is only observed for the (001) surface, whereas on GaAs (110) and (111) growth occurs in a 2D way under inclusion of many misfit dislocations [6]. We now have grown InAs on 2 differently treated Si(111) surfaces. The first procedure was that after an anneal in a separate chamber the samples were transferred via UHV into the InAs growth chamber. Figure 2(a) shows the RHEED image of the $(7 \times 7)$ reconstruction of the starting surface. In contrast to a deposition on Si(001) the RHEED image remains streaky and shows even a twofold reconstruction after deposition of 30 ML. In Fig. 3(a) an AFM image shows the structure of large InAs clusters separated by a network of misfit dislocations, but nevertheless consisting of a more or less “flat” surface according to RHEED.

The second procedure is to grow on an inert surface. From [7] it is known that the Si(111) surface can be terminated with Arsenic by heating the sample up to 700°C under As-flux. The uppermost Si-layer is removed and exchanged by an As-layer. The 5 bonding electrons of the As lead to 3 bonds to Si and one fully occupied electron orbital reaching perpendicularly out of the surface. The samples were prepared by an additional anneal at approximately 700°C under As-flux and then cooled to growth temperature for the InAs growth. As soon as the In was opened in addition to As the RHEED image turned spotty. AFM images (Fig. 3(b)) show nanocrystals of approximately 70 nm in diameter and 20 nm in height which formed on Si(111):As after depositing 3 ML of InAs. The morphology on this inert surface, where the InAs is rather van-der-Waals bonded than chemisorbed is very different from the growth modus on the untreated Si(111) surface. Due to the weakness of physisorption we expect the InAs to form crystals with its own lattice constant.

3. Conclusion

A broad range of parameters can be varied to finally achieve a self organized growth of InAs QDs on Si. In this study we investigated the influence of different substrate orientations
and substrate treatments. In contrast to InAs on GaAs, QDs can also be grown on arsenic passivated (111) oriented silicon. So far, the InAs nanostructures all seem to include crystal defects. Further work has to be done to optimize the growth and reduce the dimensions of the QDs.

References

Two-pulse coherent population of quantum states in inhomogeneous ensemble detected by the phonon-assisted resonant luminescence

A. Baranov†‡, V. Davydov†‡, A. Fedorov‡§, H.-W. Ren†, S. Sugou†¶ and Y. Masumoto†§
† ERATO Single Quantum Dot Project, JST, 5–9–9 Tokodai, Tsukuba, Ibaraki 300–2635, Japan
‡ S. I. Vavilov State Optical Institute, St Petersburg, 199034, Russia
¶ Opto-Electronics Research Lab., NEC Corp., Tsukuba, Ibaraki 305–8501, Japan
§ Institute of Physics, University of Tsukuba, Tsukuba, Ibaraki 305–8571, Japan

Abstract. The novel technique of coherent control of quantum states in semiconductor nanostructures was applied to the inhomogeneously broadened ensemble of quantum dots. By taking an advantage of the interaction between electronic states in quantum dots and phonons in the host matrix the inhomogeneous broadening was effectively eliminated. The coherence decay rate was measured.

Introduction

Recent development of ultra-short pulse tunable lasers has opened numerous opportunities in time-domain spectroscopy of quantum systems. Typical example of the great value of this approach in the field of semiconductor quantum dots (QDs) was published recently [1]. That work took advantage of the narrow optical transitions in the single QD selected from ones formed by the natural width fluctuations of the quantum well.

The present work is devoted to testing a different approach based on the spectral narrowing of the response of the inhomogeneously broadened system containing a large number of QSs. The core idea of this technique is to utilize the intrinsic narrow resonances in the III–V semiconductor heterostructures — optical phonons of the host matrix crystal [2]. These resonances can tightly bound the energy positions of levels excited by laser and those whose luminescence is detected.

1. Experimental

1.1. Sample

The sample studied was grown by the gas source molecular beam epitaxy on the GaAs substrate and contains InGaAs quantum well sandwiched between epitaxial GaAs barriers. QDs are formed in the concentrations of local strains caused by the InP stressors grown in Stranski–Krastanow mode on top of the whole structure. Details of the sample structure were published elsewhere [3].

It is the property of that kind of structures, that longitudinal (LO) and transversal (TO) optical phonons of the GaAs possess a narrow spectral width of the order of <1 meV and, on the other hand, are strongly coupled to the electronic states of stress-induces quantum dots. This coupling results in strong quasi-Raman lines in the resonant photoluminescence
Fig. 1. Resonant photoluminescence spectrum of the stress-induced InAs quantum dots. Positions and widths of the TO (peak with lower energy) and LO (higher energy peak) phonons are shown.

Fig. 2. Experimental setup. Notation: Ti:S — laser, LIA — lock-in amplifiers, PD — photodiodes, M — monochromators, S — sample in cryostat, PMT — photomultiplier, F — fiber bundle, C — counter, PC — computer.

(RPL) spectra. An example of such a spectrum is shown in Fig. 1. The similar picture is observed in the photoluminescence excitation spectra.

1.2. Setup

The core parts of the experimental setup (schematically shown in Fig. 2) are the mode-locked tunable Ti:sapphire laser, modified Michelson interferometer, 1 m double monochromator and acquisition electronics.

Time sweep was implemented by precise mechanical displacement of one of the interferometer’s retroreflectors by means of piezo microstepper motor. Both beams of the laser light emerging from the interferometer consist of the pairs of coherent pulses separated by the time delay \( \Delta t \) up to few hundreds picoseconds.

To obtain the exact measure of the \( \Delta t \), we extract single narrow spectral component from one of this light beams by use of the auxiliary “reference” monochromator (M1) tuned to a wavelength \( \lambda_R \) somewhere within the spectrum of the laser pulses. The mean intensity \( I_R \) of this component is detected by the photodiode (PD2) placed behind the output slit and can be described by an equation

\[
I_R = I_{R0} \left[ 1 + r(\Delta t) \cos \left( 2\pi \frac{c}{\lambda_R} \Delta t \right) \right],
\]

(1)

where \( I_{R0} \) is a constant proportional to the laser power at the wavelength \( \lambda_R \) and combined efficiency of the monochromator and photodiode, and the slowly changing envelope \( r \) is determined by the finite bandwidth of the auxiliary monochromator and minor misalignments of the interferometer during the scan. In our experiments the value \( r(0) \) is almost 100% and gradually falls to about 30% at the \( \Delta t \approx 200 \) ps.

The small part of the same beam was split off by the beamsplitter and detected by the broadband photodetector (PD1), that is plain photodiode without any spectral filters (except for the laser spectrum itself, of course). The signal from this photodetector \( I_A \) obeys the formula

\[
I_A = I_{A0} \left[ 1 + a(\Delta t) \cos \left( 2\pi \frac{c}{\lambda_0} \Delta t \right) \right],
\]

(2)
This formula is quite similar to the Eqn. 1, however it’s terms bear a different physical sense. The $\lambda_0$ term is the mean wavelength of the laser pulse (which coincides with the wavelength of the spectral maximum for typical symmetric pulses of Ti:sapphire laser), and the envelope $a(\Delta t)$ describes an amplitude of the autocorrelation function of light electric field. For the transform-limited pulses, this term is equal to the square root of the light intensity autocorrelation function usually obtained by the autocorrelators with nonlinear crystals.

The remaining second laser beam is routed to illuminate the sample (S) immersed in the superfluid helium inside the cryostat. Because this beam has already passed the interferometer, both pulses are filling exactly the same spatial mode, and their mutual coherence does not suffer from the quality of the routing optics, cryostat windows and sample surface. The luminescence of the sample is collected by the spherical mirror and fed to the double monochromator (M2) through the fiber bundle (F). Monochromator is equipped with the cooled infrared photomultiplier tube (PMT) and tuned to the wavelength $\lambda_e$ such, that energy of photon of this wavelength is 1 LO phonon lower than the mean energy of laser photons. It effectively detects the luminescence excited at the wavelength

$$\lambda_e = 1 \left( \frac{1}{\lambda_d} + \frac{\Omega_{LO}}{2\pi c} \right)^{-1}.$$

The intensity of luminescence light detected by the PMT is described by the already familiar formula

$$I_L = I_{L0} \left[ 1 - l(\Delta t) \cos \left( 2\pi \frac{c}{\lambda_e} \Delta t \right) \right], \quad (3)$$

where both the envelope $l(\Delta t)$ and the effective detection wavelength $\lambda_e$ are determined by the combined properties of the sample and monochromator together. Knowing the properties of monochromator (or just taking a limit of series of measurements with different slit widths) we can obtain the desired dephasing time from the decay of the envelope $l$. The minus sign comes from the odd number of reflections of this beam off the interferometer’s beamsplitters.

Data acquisition while continuously scanning the interferometer is triggered by the AC component of the signal (1), which also serves as a reference to the two lock-in amplifiers (LIA 1 and 2). One of them collects luminescence (3) from the PMT, and another is used for simultaneous recording of the pulse autocorrelation (2). Because all of the three wavelengths in this formulae are chosen to almost coincide, the input signals of the LIA are oscillating at the frequency very close to the reference which allows us to use long accumulation time constants in order to improve signal to noise ratio. In this case the good estimations for the envelope functions $a$ and $l$ can be obtained just from the amplitude $\sqrt{x^2 + y^2}$ of the corresponding LIA output.

2. Results and discussion

In Fig. 3 several of traces $l(\Delta t)$ are presented taken with the different values of the slit width of monochromator. Also the autocorrelation $a(\Delta t)$ was recorded.

It can be seen from this data, that in the limit of the narrow slit the envelope takes a simple form of

$$l(\Delta t) = \exp \left( - \left| \frac{\Delta t}{\tau} \right| \right), \quad (4)$$
Fig. 3. Amplitudes of the coherent component seen in the luminescence of the sample and interference of the exciting light. The magnitude of latter was arbitrarily scaled to fit into the graph.

where the only parameter $\tau$ can be determined from the fit (with some small corrections for the finite width of $a$) and appears to be approximately 20 picoseconds, which is fairly close to the radiative lifetime of this kind of semiconductor structures. Note that the form (4) implies Lorentzian shape of the studied spectral transition, which is typical for the homogeneous broadening.

3. Conclusion

The strong interaction between the optical transitions and bulk crystal phonons in the III–V heterostructures provides a ways to perform fluorescence line narrowing spectroscopy on this systems. Using of the Ti:sapphire mode-locked laser allows to do measurements directly in the time domain. We utilize both of this features for study of the dephasing time in quantum dots.

References

Carrier relaxation in InGaAs-GaAs quantum dots formed by activated alloy phase separation

M. V. Maximov†, A. F. Tsatsul’nikov†, A. E. Zhukov†, N. A. Maleev†, V. M. Ustinov†, Zh. I. Alferov†, N. N. Ledentsov†‡, D. Bimberg‡, T. Maka§ and C. M. Sotomayor Torres§

† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Institut für Festkörperphysik, Technische Universität Berlin, D-10623 Berlin, Germany
§ Institute of Materials Science and Dept. of Electrical Engineering, University of Wuppertal, Gauss-Strasse 20, 42097 Wuppertal, Germany

Abstract. Structures with InGaAs-GaAs quantum dots (QDs) formed by activated alloy phase separation have been studied by resonant photoluminescence. Near resonant PL at 20 K reveals a series of broad phonon replicas. This linewidth is explained by multiphonon relaxation processes involving LO phonons in the QDs, the wetting layer, interface, and GaAs as well as by additional scattering by acoustic phonons. The complex distribution of InGaAs in QDs formed by activated alloy phase separation is believed to cause a dispersion of QD phonon energies and to lead to a further broadening of phonon lines. For the sample with high QD confinement energy the photoluminescence lineshape depends on the excitation wavelength for near resonant excitation even at room temperature, which suggests a non-Fermi carrier distribution in the QD array.

Introduction

Carrier relaxation in quantum dots (QDs) has attracted much interest in recent years and been studied by different groups using resonant photoluminescence (PL) and PL excitation (PLE) spectroscopy [1–5]. Multi-phonon relaxation mechanisms [1, 2] as well as excited state absorption [3, 4] have been controversially proposed to explain PLE spectra of QD structures. In general, the relaxation processes are influenced by inhomogeneous broadening of QD array and competition between radiative and non-radiative recombination. Thus, prevalence of a concrete relaxation mechanism may depend on the parameters of a particular QD array. In Ref. [5] it has been shown that peaks due to excited state absorption as well as multiple LO resonance can be registered in the same PLE spectrum.

1. Experiment

InGaAs QDs were formed using the approach we refer to as Activated Alloy Phase Separation. In this growth approach a sheet of small Stranski–Krastanow islands was formed by depositing 2.1–2.7 ML of InAs and then overgrown by a 4–8 nm layer of InₓGa₁₋ₓAl₀.₁₅As (x = 0.10–0.16, y = 0.0–0.15). During the overgrowth, the strain field induced by each island causes migration of In atoms from the InGaAlAs alloy toward the islands leading to an increase in their size (Fig. 1). Details of the QDs growth as well as the structural and optical properties of these QDs have been described elsewhere [6–7]. A remarkable feature of QDs formed by the activated alloy phase separation is that their size and correspondingly confinement energies of the exciton in the ground state with respect to InGa(Al)As alloy and wetting layer can be tuned in a wide region 240–400 meV depending on the growth
Table 1.

<table>
<thead>
<tr>
<th>Number</th>
<th>Initial islands</th>
<th>Overgrowth</th>
<th>QD size† (300 K)</th>
<th>PL maximum (300 K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>QD1</td>
<td>2.1 ML InAs</td>
<td>4 nm In$<em>{0.1}$Ga$</em>{0.9}$As</td>
<td>12.5 nm</td>
<td>1.1 eV</td>
</tr>
<tr>
<td>QD2</td>
<td>2.5 ML InAs</td>
<td>5.5 nm In$<em>{0.13}$Ga$</em>{0.87}$As</td>
<td>15.5 nm</td>
<td>0.95 eV</td>
</tr>
<tr>
<td>QD3</td>
<td>2.5 ML InAs</td>
<td>8 nm In$<em>{0.15}$Ga$</em>{0.75}$Al$_{0.15}$As</td>
<td>17.5 nm</td>
<td>0.958 eV</td>
</tr>
</tbody>
</table>

† As determined by Transmission Electron Microscopy.

Fig. 1. A schematic diagram illustrating the Activated Alloy Phase Separation.

parameters [6–7]. In this paper we study three samples which we refer to as QD1–QD3. The growth parameters for the samples are presented in Table 1.

The photoluminescence (PL) was excited by an Ar$^+$ or Ti-sapphire laser (100 W/cm$^2$) and dispersed by a double-pass monochromator fitted with a Ge photodetector. The wavelength of the Ti-sapphire laser could be tuned in the region 900–1050 nm.

2. Results and discussion

To investigate the relaxation mechanisms in QDs we performed resonant PL studies using a Ti-sapphire laser. Figure 2 shows the resonant PL spectra for the structure QD2 at 10 K. When the energy of exciting photon is close to or above the InGaAs quantum well continuum energy, the shape of the PL spectrum depends only weakly on the excitation wavelength (Fig. 2(b)). By contrast, when the energy of exciting photon approaches the energy of the QD ground state transition (within 100–150 meV range) the shape of the PL spectrum changes and begins to depend significantly on the excitation wavelength. A set of equally-spaced peaks appears in the PL spectrum (Fig. 2(a)). The peaks are separated by a multiple of InAs- and GaAs-related LO-phonons with energies around 31 and 37 meV, respectively, and therefore we attribute the origin of these peaks to a multiple phonon relaxation mechanism. These resonant PL results are in an agreement with our previous PL excitation studies using lamp excitation [6–7].

In spite of the narrow spectral width of the excitation source and high resolution of the detection system, no sharp features were resolved for 2LO and 3LO replica for the sample QD2 pointing to an additional intrinsic broadening mechanism. Several reasons can account for the broadening of phonon lines. The first reason is a contribution of different phonons [2] including those from the InAs wetting layer (29.6 meV), InAs QD phonons (31.9 meV), interface phonons (35.0 meV) and GaAs phonons (36.6 meV). The scattering by acoustic phonons can also contribute to the broadening of phonon replicas. Taking into
account that the InGaAs LO phonon energy is altered by both In composition and strain, an additional reason for the broadening of phonon replicas may be caused by a complex distribution of strain and In concentration in the vicinity of the QDs formed by activated alloy phase separation.

Figure 3 shows the temperature dependence of the resonant PL spectra for the sample QD3. A set of peaks separated by a multiple of InAs- and GaAs-related LO-phonons from the laser line is observed at the resonant PL spectra of this structure, which is similar to the case of the structure QD2. For the non-resonant excitation PL peak shifts towards the low energies with the temperature increase, following the temperature dependence of the InAs bandgap energy. In contrast, the position of the peaks marked by the dashed lines in Fig. 3 is found to be temperature independent, which confirms that these lines are due to the multiple phonon relaxation mechanism.

For the structure QD1 the wavelength of the Ti-sapphire laser can be tuned closer to PL maximum, since the PL line of this sample is blue shifted as compared to those for the samples QD2 and QD3 (see Table 1). When the lasing wavelength is within the 50–30 meV range from the maximum of PL line recorded under non-resonant excitation several qualitatively new lines marked L and E appear in the spectra (Fig. 4). These lines are narrower than phonon replica in Fig. 2 and Fig. 3. The shift between line L and the excitation wavelength is still within the range of typical InGaAs LO phonon energies, whereas for line E this shift is about 43 meV. Thus, peak E might be due to the excited state absorption. Further experiments are in progress to verify the origin of lines L and E.

Depending on the sample temperature and the confinement of electron and hole energy levels with respect to the continuum, the carrier distribution within the QD array can be in equilibrium (Fermi) or non-equilibrium. At high temperatures or (and) low confinement energies the thermal nonequilibrium carrier emission, lateral transport via wetting layer and matrix and then recapture results in the quasi-equilibrium distribution of carriers in the array. At low temperatures or (and) efficient confinement of electron and hole energy levels, the carrier escape from the individual QDs is suppressed and the QD array can not be described by a common quasi-Fermi level.
At 20 K lineshape for the samples QD1–QD3 dramatically depends on the excitation wavelength under resonant excitation (Fig. 2–4), which suggests that at 20 K there is no lateral transport and correspondingly quasi-Fermi level in these QDs arrays. However at room temperature the shapes of resonant PL spectra for the sample QD1 does no longer seem to depend on the excitation wavelength (Fig. 5). Thus, for this sample a common quasi-Fermi level is already established at 300 K. In contrast for the structure QD2 PL line shape depends on the excitation wavelength even at room temperature (Fig. 6). We note that for this sample the exciton confinement energy is higher than that for the sample QD1 (see Table 1). The data shown in Fig. 6 suggest a non-thermal distribution of electrons and holes at room temperature for the sample QD2 at room temperature.

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References

Comparison of hole and electron emission from InAs quantum dots

C. M. A. Kapteyn†, M. Lion†, R. Heitz†, D. Bimberg‡, P. N. Brunkov‡, B. V. Volovik‡, S. G. Konnikov‡, A. R. Kovsh‡ and V. M. Ustinov‡

† Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, D-10623 Berlin, Germany
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Carrier escape processes from self-organized InAs quantum dots (QDs) embedded in GaAs are investigated by time-resolved capacitance spectroscopy. Electron emission is found to be dominated by tunneling processes. In addition to tunneling from the ground state, we find thermally activated tunneling involving excited QD states with an activation energy of 82 meV. For holes, the tunnel contribution is negligible and thermal activation from the QD ground state to the GaAs valence band with an activation energy of 164 meV dominates. Extrapolation to room temperature yields an emission time constant of 5 ps for holes, which is an order of magnitude larger than for electrons. The measured activation energies agree well with theoretically predicted QD levels.

Introduction

The peculiar properties of self-organized quantum dots (QDs) have stimulated rapidly growing research interest during the last few years [1]. Capacitance spectroscopy of self-organized QDs has been used to investigate the electronic level structure and Coulomb charging effects [1–3]. In this paper, we report on study of carrier emission from single-layer InAs QDs in GaAs matrix by means of deep level transient spectroscopy (DLTS). Using very similar QDs in n- and p-doped matrices, it is possible to investigate electron and hole escape separately and to compare the emission mechanisms. We clarify fundamental differences in the emission processes for electrons and holes.

1. Experimental details

Two types of samples were investigated: The first, referred to as ‘P’, is a n+p diode structure with InAs QDs (nominally 3.3 ML, grown at 480 °C) embedded in a Be-doped GaAs matrix ($p = 2.0 \times 10^{16}$ cm$^{-3}$) at a distance of 500 nm from the n$^+$p interface. The QDs are sandwiched between two 10 nm thick undoped GaAs buffers. To form devices, processing based on standard optical lithography was applied. Circular mesas with 200 µm diameter were created by wet chemical etching and ohmic contacts to the p$^+$ substrate and the n$^+$ top layer were formed by evaporation and alloying of AuZn and AuGe, respectively. The second sample, referred to as ‘N’, contains InAs QDs grown under very similar conditions (nominally 4 ML InAs, grown at 485°C) in Si-doped GaAs ($n = 2.0 \times 10^{16}$ cm$^{-3}$) at a distance of 400 nm from AuCr Schottky contacts, also with 10 nm thick undoped buffers. The circular Schottky contacts have a diameter of 350 µm. From photoluminescence experiments (not shown here) we conclude that the structural properties of both sets of QDs are very similar. The ground state transition of the QDs in sample ‘P’ at 1.113 eV at 10 K is only 33 meV lower than the one of the QDs in sample ‘N’. Furthermore the excited state splitting observed in PL under high excitation density of 75 meV as well as the linewidths are almost identical for both samples.
2. Results and discussion

The capacitance–voltage characteristics (CV) of both samples (Fig. 1) show pronounced steps. This behavior is directly related to the QDs [2, 3]. When the reverse bias is increased, the depletion region of the n$^+$p or Schottky interface reaches the QD layer and is pinned there. As a consequence the capacitance remains roughly constant until all charge from the QD layer is removed and the depletion region starts to extend further into the matrix material. This happens for a reverse bias of about $-3$ V in sample ‘N’ (Fig. 1(a)) and a reverse bias of about $0.5$ V in sample ‘P’ (Fig. 1(b)). For higher biases the device capacitance $C$ follows the bulk dependence on external bias $U$ for constant doping concentration: $C \sim U^{-1/2}$. From such steps it is possible to estimate the QD energy levels by comparison with simulations [2]. If the depletion region is sufficiently larger than the distance of the QDs from the interface, the QD ground states are lifted above the Fermi level. Consequently the QDs are releasing their carriers (see inserts of Fig. 1).

![Fig. 1. Capacitance–voltage characteristics of samples ‘P’ (a) and ‘N’ (b) at $T = 160$ K for a measurement frequency of 1 MHz. The schematic valence band for a QD in the depletion region of p-GaAs at finite reverse bias is depicted in the inset of panel (a), the schematic conduction band for a QD in the depletion region of n-GaAs at finite reverse bias is depicted in the inset of panel (b). The QD ground state levels, $H_0$ and $E_0$ respectively, are lifted above the Fermi level $\mu$ of the undepleted GaAs, therefore carriers are emitted.](image)

In order to investigate the carrier emission processes, we used a standard DLTS technique. First, the diodes were biased with a detection bias $U_{low}$ for which the QDs are empty. Then during a forward bias pulse $U_{high}$ the QDs are filled with carriers. After the pulse, carrier emission can be directly monitored via the transient capacitance of the space charge region of the diode. The data is finally converted into DLTS plots using a double-boxcar correlator with a reference time constant $t_{ref}$. The width of the space charge region depends on the amount of charge inside, and thus on the amount of carriers in the QDs.

The DLTS signal of sample ‘N’ (open circles in Fig. 2(a)) is due to electron escape. It exhibits a peak at about $40$ K and a constant contribution towards lower temperatures. Similar behavior has previously been observed for electron emission from vertically coupled QDs [3]. The constant low-temperature signal is due to tunnel emission from the QD ground state to the matrix conduction band and the peak is attributed to thermally activated
The thermal emission rate is usually written:

\[ e_a = \gamma T^2 \sigma_\infty \exp \left( -\frac{\Delta E_a}{kT} \right) \]  

where \( \Delta E_a \) is the activation energy, \( \sigma_\infty \) the capture cross section for \( T = \infty \), and \( \gamma \) a temperature-independent constant. From an Arrhenius plot of the DLTS peak position for varying reference time constants \( t_{\text{ref}} \) (Fig. 2(b)) we obtain \( \Delta E_a^P = (164 \pm 10) \text{ meV} \) and \( \sigma_\infty^P \approx 5 \times 10^{-12} \text{ cm}^2 \). These values depend somewhat on the detection- and pulse-biasing conditions, which reflects the fact, that it is possible to only partly fill or empty the QD ensemble, depending on the position of the Fermi level during the filling pulse and detection, respectively. Also barrier lowering effects due to the vertical electric field, as described previously for electrons [3], may play a role. For the biases applied here, this effect is estimated to lie in the order of 5 meV and is therefore not taken into account. In contrast to the case of electrons, however, the hole emission is purely thermally activated on the time-scale accessible by DLTS.
The measured activation energy $\Delta E_{\text{a}}^{\nu}$ is attributed to thermal activation from the QD hole ground state to the GaAs matrix valence band, i.e. the emission in the case of holes is taken to be a single step process, see inset of Fig. 1(a). This interpretation is supported by predictions for the hole localization based on 8-band $k \cdot p$ calculations for pyramidal InAs/GaAs QDs with $[101]$ side facets, see Ref. [4].

The fundamental difference in the emission processes observed for electrons and holes can be explained by the larger effective mass of the latter, which has a twofold effect. First, tunneling is suppressed for holes, since the tunneling current depends exponentially on the square root of the effective mass. And second, the level spacing in a QD is predicted to be much smaller for holes than for electrons [4], which is also supported by the present data. The effectively higher density of hole states in the QD and provides more effective carrier relaxation. An increasing relaxation rate decreases the efficiency of multi stage escape processes and conclusively increases the effective barrier height. The extrapolated hole confinement time of 5 ps at room temperature in our experiment, is much larger than for electrons with about 0.4 ps.

In conclusion, we have investigated hole and electron escape from a single layer InAs QDs in a GaAs matrix by time-resolved capacitance spectroscopy and found fundamentally different behavior. In the case of electrons, besides direct tunnel emission a thermally activated tunneling process involving excited QD states dominates for high temperatures. For holes, however, only thermal activation from the QD ground state directly to the GaAs valence band contributes and tunneling is not observed. Therefore the hole localization time in the QDs is significantly larger than the one of electrons. This effect can be explained by the larger effective mass having two consequences: a reduced tunneling probability and more effective carrier relaxation mechanisms.

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References


Carrier relaxation dynamics in self-assembled quantum dots

V. Davydov†§, I. V. Ignatiev†‡, I. E. Kozin†‡, S. V. Nair†, J.-S. Lee†, H.-W. Ren†, S. Sugou† and Y. Masumoto†¶
† Single Quantum Dot Project, ERATO, JST, Tsukuba 300–2635, Japan
‡ Institute of Physics, St Petersburg State University, St Petersburg 198904, Russia
§ Vavilov State Optical Institute, St Petersburg, 190034, Russia
¶ Institute of Physics, University of Tsukuba, Tsukuba 305–8571, Japan

Abstract. A novel technique to study carrier relaxation dynamics based on the artificial control of nonradiative losses by an external electric field is proposed. A clear evidence of phonon assisted relaxation as the main relaxation mechanism of hot electron-hole pairs in InP self-assembled quantum dots is found by the proposed method. Efficient one step relaxation processes with emission of acoustic and optical phonons are observed. These findings give new and important insight into the interaction of the electron-hole pairs in quantum dots with the phonon subsystem.

In this work we propose a new powerful method to study the carrier relaxation in self-assembled quantum dots (QDs) exploiting the phenomenon of photoluminescence (PL) suppression when an external electric field is applied to a sample containing the QDs. We found that under quasiresonant excitation, distinct resonances shifted from the excitation line by the LO or acoustic phonon energy appear in the spectra of the partially suppressed PL. We explain this phenomenon as a result of the competition between nonradiative losses and intraband carrier relaxation processes. The rate of nonradiative losses may be intentionally controlled by changing the applied bias. This opens up wide possibilities for the study of carrier relaxation dynamics.

We studied a few heterostructures with one layer of InP or InGaAs self-assembled QDs between Ga_{0.5}In_{0.5}P or GaAs barrier layers, respectively. The samples was grown by gas source molecular beam epitaxy on an n+ GaAs substrate. To study of the optical spectra of the sample in an electric field we provided the samples with a semi-transparent gold Shottky contact (thickness ≈ 20 nm) on the top surface and an ohmic contact on the back surface.

The PL bands of the QDs recorded under quasiresonant excitation at zero bias have a smooth profile without any sharp features as shown in Fig. 1(a). When a negative bias is applied to the sample surface, the intensity of the PL decreases and distinct sharp resonances appear in the PL spectra. Most prominent resonances are shifted from the excitation line approximately by the LO phonon energy of InP bulk crystal (ℏω = 43.5 meV). PL excitation spectra reveal very similar behaviour with prominent 1LO and 2LO resonances at negative bias as is shown in Fig. 1(b). The PL spectra of the other studied samples reveal similar behavior (see Fig. 2).

All experimental data strongly suggest that the observed resonances are caused by fast relaxation of hot carriers with emission of LO or acoustic phonons rather than by phonon sidebands of the resonance PL or by resonant Raman scattering.

Our explanation of the observed phenomenon may be summarized as follows. The applied electric field activates a process of nonradiative losses of the QD excitation. Most likely cause of these losses is the tunneling of the hole from the QD into the barrier layer.
Fig. 1. Dependence of PL (a) and PL excitation (b) spectra on applied bias for the sample QDP1779 with InP QDs. Excitation power density 100 W/cm$^2$, $T = 2$ K. The excitation and detection photon energies are marked by dashed lines. Applied bias is shown near each spectrum.

Fig. 2. PL spectra dependence on applied bias for sample QDP1778 with InP QDs (a) and 1142D with InGaAs QDs (b). Excitation photon energy marked by dashed arrows. Applied bias shown near each spectrum.

because of the depth of the potential well for holes is smaller than for electrons in such heterostructures. The details of this process is discussed elsewhere [1]. The rate of the nonradiative process increases with the increase of negative bias. The tunneling from excited state competes with relaxation to the ground state. This leads to selective PL suppression because of different relaxation rates with emission of LO and acoustic phonons.
To verify the assumptions implicit in the above description, we performed the PL kinetics measurements for thin samples with InP QDs. Kinetics for selected spectral points and their dependence on the applied bias for the LO resonance are shown in Fig. 3(a) and (b). As seen, the kinetics clearly demonstrate a shortening of the decay time with the increase of negative bias. So the nonradiative process forms some kind of time (or optical) gate for the PL. Initial (rise) part of the kinetics contains information about the relaxation rates. As seen from Fig. 3(b), it is very fast for LO resonance and limited by our time resolution of about 6 ps. In the spectral region formed by the acoustic phonon assisted relaxation (hereafter referred to as the acoustic region), the rise time is about 50 ps that is much slower than at the LO resonance.

It is clear from presented data that the manipulation of the nonradiative losses allows us to determine spectral dependence of the relaxation rate. To demonstrate it, we measured the PL kinetics at many spectral points at zero bias, then multiplied them by $e^{-\gamma t}$ to imitate the fast nonradiative losses and integrated. The time resolved PL spectrum reconstructed this way is shown in Fig. 3(c) together with the PL spectrum measured at $U_{bias} = -1.5$ V. As the estimated tunneling time at $-1.5$ V is shorter than the time resolution of our set-up, $\tau_s = 6$ ps, we have used $\gamma_t = 1/\tau_s$. The excellent agreement between the two spectra clearly demonstrates the consistency of our description of the PL process. At the same time, the spectral and effective time resolutions in the PL spectrum of the biased sample are better than that achievable in kinetics experiments.

A few important statements about relaxation mechanisms and relaxation rates can be derived from the experimental data and the analysis presented above.
The main relaxation mechanism of hot carriers in QD’s when only one electron-hole pair is created in the QD is the phonon assisted relaxation. Different relaxation channels are possible. Electron and hole may relax separately or together, mediated by the Coulomb interaction, by emission of phonons. The PL kinetics for the LO resonance shows mainly a fast rise component. From this kinetics it follows that single LO phonon emission contributes more than 70% of the PL at the LO resonance energy.

The relaxation time with emission of LO phonons lies within the range $0.6 \text{ ps} < \tau_{\text{LO}} < 6 \text{ ps}$. The lower bound is derived from fitting the peaks in the LO resonance by Lorentzians. The upper bound is defined by the time resolution of the direct kinetics measurements.

Acoustic phonon resonances are observed in the spectra of the partially suppressed PL. A comparison of the PL spectrum with the phonon density of states of InP crystal (see Fig. 3(d)) allows us to attribute the resonances to the transverse acoustic (TA) and longitudinal acoustic (LA) peaks in the phonon density of states (DOS) of InP and Ga$_{0.5}$In$_{0.5}$P. The rising kinetics of the PL in these peaks are faster than in the rest of the acoustic part of the spectrum. This observation is evidence of efficient relaxation of electron-hole pairs with emission of high frequency acoustic phonons, in stark contrast with theoretical predictions [3].

In conclusion, we developed a novel method for the study of carrier relaxation in QD’s based on the artificial control of the nonradiative losses by an external electric field. The nonradiative process with a controllable rate gives rise to an effective optical gate with variable duration for the PL of the QD’s. This method allows one to study the spectral dependence of the carrier relaxation rates with high spectral and time resolution at a low power density of optical excitation. Our results clearly demonstrate that the main relaxation mechanism in InP self assembled QD’s is the phonon assisted relaxation. Clear evidence of efficient one step relaxation with emission of high frequency acoustic phonons is found. This observation poses a new principal problem for theoretical analysis.

References
Optical properties of semiconductor (InP)–dielectric quantum wires

K. Chernoutsan, V. Dneprovskii, S. Romanov*, O. Shaligina and E. Zhukov
Moscow State University, Department of Physics, 119899 Moscow, Russia
* Institute of Materials Science and Department of Electrical Engineering,
University of Wuppertal, 42097 Wuppertal, Germany and
Ioffe Physico-Technical Institute, St Petersburg, Russia

The absorption and luminescence spectra of InP embedded in chrysotile asbestos nanotubes have been explained in terms of exciton transitions in semiconductor – dielectric quantum wires (S–D QWRs). The features of the time-resolved luminescence of porous InP have been explained by numerous physical processes in S–D QWRs or quantum dots (QDs): slowing down of intraband relaxation in one-dimensional energy band of QWRs or excess energy of holes in QDs, collective exciton–exciton (electron) interaction in nanostructures, Auger recombination etc.

In quantum wires, consisting of semiconductor and dielectric components, Coulomb attraction between electron and hole inside semiconductor filaments may be considerably enhanced. Qualitatively it can be explained resorting to the electric lines of forces that connect electron-hole pair. They propagate partially outside the semiconductor filament in dielectric with \( \varepsilon_d \ll \varepsilon_s \) (\( \varepsilon_d, \varepsilon_s \) — dielectric constants of dielectric and semiconductor). Thus it is possible to change the binding energy and the oscillator strength of QWRs constituting semiconductor filaments and dielectric matrix with different dielectric constants — realizing Coulomb interaction engineering [1].

Chrysotile asbestos is a natural material that consists of the bundles of closely packed parallel transparent dielectric up to a few sm long nanotubes. The internal and external diameters of nanotubes of the samples, measured using the electron microscope, were 4–5 nm and 30 nm². The standard atmospheric pressure MOCVD reactor [2] has been used to grow crystalline InP inside nanotubes³.

The absorption spectrum (Fig. 1) of the InP crystallized in chrysotile asbestos has a well pronounced band with maximum at 1.9eV that we attribute to the absorption of nanostructures. This spectrum has been obtained using optical microscope to choose the 2 \( \mu \)m diameter area of the sample. We account the monotonous increase of absorption at higher energies (the background) to the absorption of the bulk InP that may be crystallized at the surface of the bunches of nanotubes and to the absorption of chrysotile asbestos matrix.

The luminescence of InP embedded in chrysotile asbestos exited by 14ns polarized pulses of the second harmonic (\( h\omega = 2.39 \text{eV} \)) of Al-Y laser has been investigated using polychromator with 0.01 eV spectral resolution. The maximum intensity of excitation was 4 MW/cm². The spectra have two wide bands. We attribute the band with maximum at about 1.9 eV to the luminescence of nanostructures and the broad band (with much weaker intensity) at higher energies to the luminescence of the matrix. The latter is confirmed by the measurement of pure chrysotile asbestos matrix luminescence. The luminescence

³Measurements were done by N. A. Kiselyov and D. N. Zakharov at Institute of Crystallography.
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spectra presented in this paper were obtained by subtracting the luminescence spectra of the pure chrysotile asbestos from the luminescence spectra of InP embedded in chrysotile asbestos. The independence of the halffwidth of the luminescence spectra ($\lesssim0.18$ eV) upon the intensity of the exciting laser beam allows us to suppose that InP has been crystallized in the form of nanostructures inside the chrysotile asbestos nanotubes because the density of QDs and QWRs states differs from that of the bulk semiconductor. The essential difference of the luminescence intensity for parallel and perpendicular polarization of the exciting laser beam (Fig. 2) enables us to suppose that the luminescence of InP S–D QWRs dominates for our samples. This difference in the luminescence intensity may be explained by the anisotropy of absorption - by the modification of the electromagnetic field eigenmode in the vicinity of QWRs [3] In QWRs the exciton transitions play the dominant role [4].
so we attribute the 1.9 eV luminescence and absorption bands to the exciton transitions in InP (semiconductor) – chrysotile asbestos (dielectric) quantum wires. The measured energy of exciton transitions (∼1.9 eV) in 4–5 nm InP S–D QWRs corresponds to that calculated using a variation technique accounting for the effect of dielectric enhancement in the cylindrical QWRs [5].

The samples of porous InP have been obtained by electrochemical etching of n-InP(100) substrate. The time resolved luminescence of porous n-InP excited by picosecond pulses of the second harmonic ($h\omega = 2.34$ eV) of Nd-glass laser (pulse duration about 10–14 ps) has been investigated using streak camera with 5–10 ps resolution. The luminescence spectra of porous InP (broad band 2.07±0.04 eV, Fig. 4) excited by powerful picosecond pulses are shifted to the blue spectral region compared with that of the bulk. This shift allows us to estimate the average size of nanostructures: the diameter is about 5 nm (3.4 nm) if the luminescence of QDs (QWRs) is assumed. In the latter case the dielectric enhancement of excitons in S–D QWRs has been taken into account.

The kinetic properties of the intensity of luminescence differ for low and high levels of laser excitation (Fig. 5). For low intensities of excitation ($I < 250$ MW/cm², Fig. 5(a)) the luminescence is characterized by exponential decay and very short rise time. With increasing of excitation the rise time of luminescence grows up (Fig. 6). When the intensity of excitation exceeds 250 MW/cm² the additional fast decay arises (Fig. 5(b)). For the highest excitation level the decay of luminescence has three typical parts: the relatively slow relaxation is followed by fast fall and then by slow relaxation (Fig. 5(c)). The slow decay of luminescence at low intensities of excitation (Fig. 5(a)) and the final slow part at high intensities (Fig. 5(c)) might be attributed to the linear recombination of excitons. The insignificant narrowing of the luminescence spectrum at high excitation allows us to exclude the contribution of the stimulated emission and explain the fast part of the decay (Fig. 5(b, c)) by exciton–exciton (electron) interaction and Auger nonradiative recombination.

The appearance of the initial relatively slow part of the decay at high excitation (Fig. 5(c)) and the increasing of the luminescence rise time with the increasing of the excitation level may be explained by the slowing down of intraband relaxation in semiconductor QWRs (hole excess energy relaxation in semiconductor QDs) at high density of the excited carriers because of the screening of electron–phonon interactions and the saturation of phonon
modes [6]. Thus the intraband relaxation time becomes comparable with the recombination time, and the relatively slow recombination of excitons may be followed by fast decay due to the exciton–exciton, exciton–electron interaction or Auger nonradiative recombination when considerable quantity of quasiparticles is accumulated.

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References

Emission line instabilities of single quantum dots of InAs in GaAs

N. Panev, M.-E. Pistol, M. P. Persson, L. Samuelson and M. S. Miller§
Solid State Physics, Lund University, Box 118, S-221 00 Lund Sweden
§ Department of Electrical Engineering, University of Virginia,
Charlottesville, VA 22903, USA

Abstract. We have measured the energy levels of quantum dots of InAs in GaAs as a function of time. On a very slow timescale there is a shift in energy for some dots. This shift can be enhanced by annealing the sample at different temperatures up to room temperature. The data are consistent with a model based on defects interacting with the dots.

Introduction

Semiconductor quantum dots grown by the Stranski–Krastanow (SK) technique [1] have recently become the focus of intense interest. The discrete density-of-states in these structures allows detailed spectroscopy to be performed, in particular if individual quantum dots are studied. Recent studies of individual quantum dots have demonstrated the importance of few-particle effects [2, 3], highly non-linear exciton–phonon coupling [4], and random telegraph noise in emission [5]. The most commonly studied system is InAs quantum dots in GaAs. We shall here show that the emission lines of InAs quantum dots shift in energy with time. The shift is temperature dependent. Similar spectral shifts have previously been observed for colloidal quantum dots [6] but have not been reported for SK-dots and not for dots formed by interface fluctuations in quantum wells [7].

1. Experimental

The quantum dots were grown by chemical beam epitaxy [8] and a sample containing low-density dots was selected for measurements. The emission from single dots was selected using a microscope, dispersed using a monochromator and detected by a cooled CCD-camera. Typical integration times were 5 minutes at a temperature of 4 K. In order to exclude possible strain effects, the sample was very lightly glued (in one corner) on the horizontal sample holder. In order to check the stability of the detection system (monochromator and CCD-camera) we repeatedly measured the emission from a spectral lamp, often simultaneously with measurements on dots. A second source of errors comes from the temperature stability at the measurement temperature and from possible strain effects. This was checked by measuring shifts of several quantum dots at the same measurement occasion. These dots were situated within less than about 1 μm from each other. We found that the shifts were uncorrelated between different dots, which shows that temperature differences do not cause these shifts. Since the sample is 300 μm thick we also exclude strain effects, which should be homogeneous over 1 μm as a source of systematic error.

2. Results and discussion

In Figure 1 we show spectra from one quantum dot, both from the ground state, at an energy of 1.329 eV, and from the excited state at an energy of 1.378 eV. After heating the sample to room temperature for one hour the experiment was repeated and the emission lines had
Fig. 1. Spectra of one quantum dot at two different occasions. Inbetween the measurements the sample was heated to room temperature for about one hour. Both the ground state emission at an energy of about 1.33 eV and the excited state emission at an energy of about 1.38 eV shift.

now shifted about 0.4 meV to higher energy. The shift was the same for both the ground state emission and the excited state emission. Such spectral shifts, after heat-treatment to room temperature, have been observed for every dot investigated (ten).

In Figure 2 we show the shift of the emission energy as a function of measurement occasion. Inbetween each measurement different heat-treatments were performed, indicated in the figure. Most dots have no measurable spectral shift at 4 K, but there are exceptions. In the insert of Fig. 2 we show the energy-shifts of a dot which was kept at a constant temperature of 4 K. The spectral shifts of this dot appear not as sudden jumps but rather as a slow shift with time.

In Figure 3 we show histograms of the distribution of shifts with energy for different temperatures and for different times of the heat treatment. It appears from the data that there is only a weak correlation of the shifts with annealing time. The correlation with annealing temperature is also weak, except that annealing at 4 K usually gives no shift. Concerning the cause of the spectral shifts, we can only speculate. One possibility is that the surface of the sample is charged differently at each measurement occasion. We have tested this possibility by measuring several quantum dots which were situated within 1 µm (i.e. within our spatial resolution limit) of each other.

The shifts of each dot are uncorrelated with the shifts of other dots, as shown in Fig. 4. This indicates that surface charging is not responsible, since in that case we would expect shifts in the same direction. We rather attribute the shifts to changes in the crystalline environment of the dots which is a much more local effect than surface charging. In Figure 4 we also show the spectral shifts of two different emission lines originating from one quantum dot. The shifts are in this case highly correlated. An attractive origin of the shifts is defects in the GaAs. Such defects could be mobile and in addition they could be metastable in a variety of ways. In any case they should be activated by high temperatures, in agreement with experiment. For the dot which experienced spectral shifts at 4 K (see Fig. 2) we observed a slow shift in the spectral position, consistent with the expected behaviour of a defect passing by. Due to the absence of a clear activation energy and the
Fig. 2. Energy position of a spectral line as a function of measurement occasion (N). Different annealing temperatures were performed between each measurement, indicated in the figure. In the inset we show the energy shift for a dot at a constant temperature of 4 K.

Fig. 3. A histogram of the energy shifts for different annealing times and temperatures.
non-existent dependence on the annealing time, we believe that we actually observe the effect of several defects passing by. This instability of the emission lines from individual quantum dots may be a serious problem for the use of such structures in advanced devices. Present (conceptual) designs of quantum logic using quantum dots typically involve two coupled dots and the alignment and stability of the energy levels of each dot are critical for the operation of the gate. Other types of devices such as quantum dots in a high-Q microcavity also rely on the stability of the emission lines over time.

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References

Spin-dependent electron behaviour in quantum point contacts and dots

A. M. Bychkov†‡, I. I. Yakymenko† and K.-F. Berggren†
† Department of Physics and Measurement Technology, Linköping University, S-581 83 Linköping, Sweden
‡ Moscow State Engineering Physics Institute, 115409 Moscow, Russia

Abstract. We investigate theoretically the effect of spontaneous electron spin polarisation in quantum point contacts and quantum dots. Self-consistent calculation process has been developed in the framework of Kohn-Sham LSDA scheme. The existence of the detected spin-polarised states in the low-density regime can be used to explain an anomalous 0.7-structure in the conductance of quantum point contacts. The properties of the potential device based on such an effect are briefly discussed.

Introduction

The physics of interacting electrons in reduced dimensions has begun to unfold in ultra clean systems. In short 1D wires (quantum point contacts — QPCs) a major breakthrough in interaction studies has been achieved by the discovery of spontaneous spin polarisation of the 1D electron gas in the low-density regime. In addition to the quantised plateaux at multiples of \((2e^2/h)\) an anomalous structure at \(0.7(2e^2/h)\) has been discovered at zero magnetic field [1, 2]. This structure was found to evolve into the spin-split conductance plateau at \((e^2/h)\) in a strong in-plane magnetic field. More recent measurements on high-mobility double-layers QPCs have revealed that the anomaly in conductance may occur at half-integer values already at zero magnetic field.

1. Theoretical model and calculations

The anomaly in the conductance has been interpreted within the Kohn-Sham local-spin-density approximation (LSDA). For one-channel conduction in a model system, it was found in an earlier work [3] that the electron gas acquires local ferromagnetic order as the electron density is decreased towards pinch-off. By sweeping the gate voltage (i.e. transforming the geometry of the QPC channel) one effectively changes the density of electrons filling a higher energy sub-band. When the density reaches a certain threshold value, an effect of spin-polarisation spontaneously occurs in the channel due to the interplay between electron kinetic and exchange energies. At that very moment, the transmission barrier suddenly becomes different for electrons having up- and down-spin projections. The spin-aligned electrons still conduct current through propagating states in a normal way, while the electrons with opposite spins now have to tunnel from source to drain. Thus an anomaly in the channel conductance repeatedly takes place at certain values of the gate voltage.

We study this problem using proper modelling of the QPC device including a patterned gate, doping, surface states, etc. In this way we consider the case of higher sub-band conduction and predict how the anomalous conduction may depend on the channel geometry. In our approach, the QPC is simulated by a system of two large quantum dots (source and
drain pads) with a smaller dot (the channel) in between, the dots being formed in 2DEG using the split-gate technique (Fig. 1). Thus, modelling a QPC we study magnetic properties of single and coupled quantum dots and how these may be monitored by external means. The topic is currently under active investigation, both theoretically and experimentally [4–7].

The system is described by the Schrödinger equation:

\[
-\frac{\hbar^2}{2m^*} \left( \frac{d^2}{dx^2} + \frac{d^2}{dy^2} \right) \phi^\sigma(x,y) + \left[ U^c(x,y) + U^e(x,y) + U^\sigma_{\text{exch}}(x,y) \right] \phi^\sigma(x,y) = E^\sigma \phi^\sigma(x,y)
\]

(1)

where \( \sigma = \pm \frac{1}{2} \) refers to spin, \( U^c(x,y) \) and \( U^e(x,y) \) are the confinement and Hartree potentials. In the Kohn-Sham scheme, the exchange potential energy has the form:

\[
U^\sigma_{\text{exch}}(x,y) = -\frac{e^2}{\epsilon_0\epsilon \pi^{3/2}} \left[ n^\sigma(x,y) \right]^{1/2}
\]

(2)

where \( \epsilon \) is the dielectric constant of the heterostructure material and \( n^\sigma(x,y) \) is the spatial distribution for \( \sigma \)-spin electrons. We have also included the Zeeman splitting \( (g\mu_B B = 10^{-6} \text{ eV}) \) from a weak external magnetic field in order to trigger the onset of spin polarisation. This splitting turns out to be much smaller than the one caused by exchange interaction.

In order to solve Eq. (1) we applied the self-consistent iterative process with the initial total potential derived from Thomas–Fermi approximation. The electron densities are calculated at each iteration as

\[
n^\sigma(x,y) = \sum_{E_i \leq E_F} \left| \psi^\sigma_i(x,y) \right|^2.
\]

(3)
The self-consistency is reached when the energy values and the potential in successive iterations are identical within an accuracy of \(10^{-5}\) eV.

2. Results and discussion

We have shown the existence of spin-polarised states at low densities in a quantum dot and in a QPC formed by a system of coupled quantum dots. In such a situation the electron exchange energy dominates over the kinetic energy thus favouring electron spin-alignment according to the Hund’s first rule. The spin-polarisation of a single quantum dot

\[ p = n^{(\sigma=\frac{1}{2})} - n^{(\sigma=-\frac{1}{2})} \]  

(4)

is shown in Fig. 2. One can see the maximum polarisation occurs at the dot periphery where the electron density is low. In the case of a QPC, the spin-polarisation shows reach texture but again is more pronounced at the QPC edges and in the channel area, i.e. in the regions of low electron density. More thorough interpretation of the numerical data for QPCs is currently underway.

We have also found the system properties, in particular the total number of up- and down-spin electrons, strongly dependent on the parameters of the heterostructure, e.g. its donor density. This implies the operation of the potential device based on such a structure would be very sensitive to the quality of the materials used for its fabrication. On the other hand, this enables to flexibly adjust the device characteristics within a wide parameter range, thus making it feasible for various applications.

The models will also allow us to search for more esoteric states like spin-density wave and antiferromagnetic groundstates in 1D channels.

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Observation of quantum beats in photoluminescence of self-assembled quantum dots

V. Davydov†§, A. V. Fedorov§§, I. V. Ignatiev†‡, S. V. Nair†, H.-W. Ren†, M. Sugisaki†, S. Sugou† and Y. Masumoto†¶
† Single Quantum Dot Project, ERATO, JST, Tsukuba 300–2635, Japan
‡ Institute of Physics, St Petersburg State University, St Petersburg 198904, Russia
§ Vavilov State Optical Institute, St Petersburg, 190034, Russia
¶ Institute of Physics, University of Tsukuba, Tsukuba 305–8571, Japan
£ Venture Business Laboratory, University of Tsukuba, Tsukuba 305–8571, Japan

Abstract. The photoluminescence (PL) kinetics of heterostructures with InP self-assembled quantum dots were studied under quasiresonant excitation in presence of external electric field. An oscillation behavior of the PL kinetics is observed at small applied bias. Period of the oscillation is about 30 ps and decay time is 20 to 50 ps depending on spectral point. We studied the PL kinetics in magnetic and electric fields. All set of the data gives a clear evidence that the oscillations are caused by the quantum beats of the optical transitions. A possible model of the observed phenomenon is discussed.

A fine structure of the energy states of self-assembled quantum dots (QDs) is difficult to study due to the large inhomogeneous broadening. Some attempts of application of usual spectroscopy technique to a single quantum dot meet the serious problems because of weak signal and poor reproducibility. A study of fine structure in time domain by technique like pump-probe or four-wave mixing is also strongly restricted for a single layer of QD’s. Here we propose a different way to study the fine structure. We discovered a new phenomenon of quantum beats (QB’s) that are observed in the photoluminescence (PL) kinetics of QD’s in presence of external electric field. This phenomenon opens up wide possibilities to study the fine energy structure of self-assembled QD’s.

We studied a heterostructure with one layer of InP self-assembled QD’s embedded between Ga0.5In0.5P barrier layers. The areal density of the QD’s is about $10^{10}$ cm$^{-2}$. Average base diameter is about 30 nm and the height is about 5 nm. The sample was grown by gas source molecular beam epitaxy on an $n^+$ GaAs substrate. The total thickness of the undoped epitaxial layer is 500 nm. The sample was provided with a semi-transparent gold Shottky contact (thickness $\approx$20 nm) on the top surface and an ohmic contact on the back surface.

The PL of the sample was excited selectively by a mode-locked Ti:sapphire laser whose wavelength was tuned within or slightly above the PL band of the QD’s (hereafter referred to as quasiresonant excitation). The pulse duration was about 1 ps, and the pulse repetition rate was 82 MHz. To prevent the creation of more than one electron-hole pair in a QD, we used a fairly low pump power density of about 100 W/cm$^2$. The PL kinetics were studied using a 0.25 m double subtractive dispersion monochromator and a streak-camera. The time resolution of the setup is about 6 ps. The PL spectra were measured also using a continuous wave (cw) excitation, a 1 m double monochromator and a photon counting system. The sample was immersed in the superfluid helium at a temperature below 2 K.
The PL spectra of the InP QD’s measured under quasiresonant excitation at a few different negative biases applied to the sample surface are shown in Fig. 1. At zero bias, the PL spectrum has a smooth profile. When negative bias is applied, the PL is suppressed and the different resonances appear. Most prominent of them are shifted from excitation line by the transverse acoustic, longitudinal acoustic and longitudinal optic phonon energies of InP crystal. These resonances are marked in Fig. 1 by “TA”, “LA” and “LO”, respectively. The details of the behavior of the PL in electric field is discussed elsewhere [1].

The PL kinetics within the first 100 ps at the TA resonance are shown in Fig. 2. They were measured under the linearly polarized excitation and the PL detection in two linear polarizations that are parallel and orthogonal to the laser beam polarization. At zero or positive bias, the PL kinetics have a fast rising part and slow decay without any features. However at small negative bias, an oscillation type behavior of PL kinetics is clearly seen. These oscillations have exactly opposite phase in two polarizations. No oscillations are observed when nonpolarized PL is detected. Oscillations are observed for any orientation of the laser beam polarization relative to the crystallographic axes of the sample. Difference between PL kinetics measured in the parallel and orthogonal polarizations can
be well fitted by a simple expression $y = \sin(\omega t + \phi) \exp(-t/\tau)$ as is shown in Fig. 3.

The oscillations in PL kinetics are observed in the whole spectral region between excitation line and LA resonance and also at LO+TA spectral point with Stokes shift 55 meV. However the oscillations are not observed at LO resonance. Period of oscillations $T$ is about 30 ps and decay time $\tau$ is 20 to 50 ps depending on spectral point. We observed also the oscillation in PL kinetics of another sample containing slightly large InP QD’s (base diameter is about 40 nm). Period of oscillations is about 40 ps in this case.

To study the physical nature of the observed phenomenon, we measured the PL kinetics in magnetic field. We found that the magnetic field destroys the regular oscillations at the value of magnetic field of about 1.5 T. At the same time, new oscillations appear that depend strongly on magnetic field. An example of these oscillations are shown in Fig. 4. The oscillation are observable for any negative bias and any spectral points except the region between LA and LO resonances.

Presented data give a clear evidence that the observed oscillations in PL kinetics are caused by quantum beats of the coherently excited states. Period of oscillations of about
30 ps corresponds to the energy separation between excited states of about 100 μeV. Decay time $\tau$ is determined by the time of coherency $T_2$. An important and new result is that the coherency is kept after relaxation of excitation to the luminescent state by a single phonon emission.

We do not know yet the exact physical nature of the quantum states responsible for the observed phenomenon. The oscillations observed without magnetic field (see Figs. 2 and 3) could be explained as follows. Laser pulses excite coherently two optical transitions. One of them is the phonon assisted absorption when the excitation creates an electron and a hole in their ground states, and a phonon. Probability of this transition is low due to small electron-phonon interaction in InP QD’s. Another optical transition is the electronic transition between the excited electron and/or hole states. Because of quasiresonant excitation used, the excitation probably creates a hole in the excited state and an electron in the ground state. Probability of this transition should be low due to selection rules. However an external electric field may change this probability. So the probability of both transition may be equal at some bias. In this case, we may expect the quantum beats of these transitions if some splitting of the electronic and the resonant vibronic states due to their interaction takes place. In the framework of this model, the energy splitting of about 100 μeV estimated from the quantum beats reflects the interaction strength with acoustic phonons in the QD’s. The interaction with LO is presumably much stronger therefore the oscillation at LO resonance is probably much faster and is not seen due to limited time resolution of 6 ps.

Open question in this simple model is that why interaction of a discrete electronic energy level with a continuous spectrum of vibronic states creates two quasidiscrete states.

The quantum beats observed in magnetic field are caused probably by the splitting of the electron and hole ground states. A coherence is transferred from excited states to the ground states by a single phonon assisted relaxation in that QD’s where this relaxation is possible. That is why the quantum beats are not observed in the spectral points corresponding to the phonon bandgap between LA and LO phonons.

In conclusion, a new phenomenon of quantum beats in the PL of self-assembled InP QD’s is observed. This phenomenon opens up wide possibilities to study the fine energy structure of the QD’s.

References

Theoretical investigation of Auger recombination in spherical quantum dots

Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The principal mechanisms of Auger recombination (AR) of nonequilibrium carriers in spherical quantum dots (QDs) are investigated theoretically. It is shown that there exist two Auger recombination mechanisms of (i) quasithreshold and (ii) thresholdless types. These mechanisms originate from the existence of barriers but have different nature. The quasithreshold mechanism is caused by confinement of carriers within the region of a quantum dot which makes the quasimomentum conservation law approximate and enhances AR process. With increase of the dot radius this process turns to the threshold one. The thresholdless mechanism relates to the violation of the momentum conservation law at the heteroboundary and disappears with the radius tending to infinity.

1. Introduction

There are two main processes of Auger recombination (AR) in narrow gap semiconductors. The first of them corresponding to recombination of an electron and heavy hole and excitation another electron is the CHCC process. The second CHHS Auger process relates to the transition of a heavy hole to the spin-orbit split-off band. Contrary to bulk semiconductor in heterostructures the transversal momentum component doesn’t conserve thus allowing the thresholdless Auger process to appear [1, 2]. In [3, 4] it was shown that there are three types of AR processes in quantum wells (QWs): (i) threshold, (ii) quasithreshold, (iii) thresholdless; and there are two types of AR processes in quantum wires (QWRs): (i) quasithreshold and (ii) thresholdless mechanisms [5, 6]. The AR mechanism of thresholdless type in a QWR differs from those in a QW because there are two different channels (i) with transfer of a large linear momentum to the excited electron (like in planar QWs) and (ii) with transfer of an angular momentum. The later channel was predicted to be the only possible channel for quantum dots [5, 6]. The forthcoming analysis shows that there are similar mechanisms of AR in spherical semiconductor QDs.

2. Theoretical model

The Auger recombination (AR) in bulk semiconductor was investigated several decades ago [7, 8]. It was shown that Auger process calculated in the first order of perturbation theory on Coulomb interaction has a threshold nature, i.e. its coefficient exponentially depends on temperature. For the calculation of the coefficients of the two main processes of AR: CHCC and CHHS it is necessary to work in framework of the multi-band kp-theory, because complex valence band structure is necessary for the correct description of the CHHS process and the parabolicity of the spectrum in the conduction band is too rough approximations for the electron with the energy \( E \approx E_g \).
We work in 8-band Kane’s model with finite constant of spin-orbit interaction $\Delta_{s_o}$. It is convenient to take basis wave function in the conduction and valence band in form of eigenfunction of the angular momentum [9]:

$$|0\uparrow\rangle, |0\downarrow\rangle, |1, 1\uparrow\rangle, |1, 1\downarrow\rangle, |1, 0\uparrow\rangle, |1, 0\downarrow\rangle, |1, -1\uparrow\rangle, |1, -1\downarrow\rangle$$

(1)

This procedure excerts eigenfunctions having definite angular momentum and corresponding to the spherical symmetry of the QD.

3. Eigenstates of carriers in a quantum dot

Commonly, the eigenstates of carriers in a QD is deriving from the elimination the wave functions at the interface which corresponds to the infinite heights of heterobarriers [9].

We derive boundary conditions from the continuity of the transversal component of the probability flux density. These boundary conditions correspond to the finite offsets in the conduction and valence bands $V_c$ and $V_v$, respectively.

In limit $\Delta_{s_o} \to \infty$ these boundary correspond to common boundary conditions in 4-band Kane model, i.e.:

$$f<^L(r = R) = f>^L(r = R), \quad \frac{1}{2E + E^d_g} \frac{df<^L(r = R)}{dr} = \frac{1}{2E + E^m_g} \frac{df>^L(r = R)}{dr},$$

(2)

where $f<^L(r)$ and $f>^L(r)$ are solutions of the radial wave equation inside and outside the QD, respectively, and the spatial dependence of $E_g(r)$ is given by a step function $E_g(r) = E^d_g + \Theta(r - R)(E^m_g - E^d_g)$ [10, 11].

4. Matrix element of Auger recombination

The probability of AR per unit time can be calculated in terms of the first-order perturbation theory in electron-electron interaction:

$$W_{i\to f} = \frac{2\pi}{\hbar} \left| M_{fi} \right|^2 \delta(\varepsilon_f - \varepsilon_i),$$

(3)

where

$$M_{fi} = \left\langle \Psi_f(r_1, r_2, v_1, v_2) \left| \frac{e^2}{\kappa_\infty |r_1 - r_2|} \right| \Psi_i(r_1, r_2, v_1, v_2) \right\rangle$$

(4)

is the matrix element of electron-electron interaction, $r_1$ and $r_2$ are carrier co-ordinates, $v_1$ and $v_2$ are spin variables, $e$ is an electron charge, and $\kappa_\infty$ is the permittivity of the intrinsic semiconductor [3, 4].

For the spherical QD it is essential to decompose the Coulomb interaction into the spherical harmonics, using following equation:

$$\frac{1}{|r_1 - r_2|} = 4\pi \sum_{l=0}^\infty \sum_{m=-l}^l \frac{r_2}{(2l + 1)r_1^{l+1}} Y_{lm}(\theta_1, \alpha_1)Y_{lm}^*(\theta_2, \alpha_2).$$

(5)

It is easy matter to see that the matrix element of AR automatically yields conservation laws for the angular momentum and $z$-component of angular momentum (where $z$ is axis
corresponding to the second quantum number in our basis). After integration along angular coordinates matrix element takes simpler form:

\[ M_{fi} = \frac{4\pi^2}{(2l + 1)\epsilon_\infty} \int_0^\infty r_1^2 dr_1 \int_0^\infty r_2^2 dr_2 \chi_l(r_1, r_2) f_{1,4}(r_1) f_{2,3}(r_2), \]  

(6)

where \( f_{i,j} \) multiplication of radial components of corresponding wave functions and:

\[ \chi_l(r_1, r_2) = \frac{\min(r_1, r_2)^l}{\max(r_1, r_2)^{l+1}}. \]  

(7)

5. Rate of Auger recombination

To calculate the rate of AR in the first order of perturbation theory, the probabilities of transition should be averaged over all initial states of carriers with appropriate weight-occupation numbers and summed over all final states:

\[ G = \sum_{n_1, m_1, n_2, m_2, n_3, m_3, k_4} \langle M^2 \rangle f_1 f_2 (1 - f_3)(1 - f_4)\delta(E_3 + E_4 - E_1 - E_2) \]  

(8)

here \( f_1, f_2 \) are the occupancies of the initial states and \( f_3, f_4 \) are those of the final states, \( \langle M^2 \rangle \) is the sum of squared Auger matrix elements over spins of the initial and final states. In this equation is proposed that the final state of excited particle 4 lies in continuous spectrum. It is obviously true in our approximation \( V_c, V_v \ll E_g \).

6. Summary

Our analysis has shown that for the Auger processes in semiconductor structures with QDs there exist two different AR mechanisms: thresholdless and quasithreshold, similarly to the case of planar quantum wells and cylindrical quantum wires.

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References

1.3 $\mu$m photoluminescence emission from InAs/GaAs quantum dots multilayer structures

V. A. Egorov†‡, V. N. Petrov†‡, N. K. Polyakov†‡, G. E. Cirlin†‡, B. V. Volovik‡, A. E. Zhukov‡, A. F. Tsatsul’nikov‡ and V. M. Ustinov‡
† Institute for Analytical Instrumentation RAS, Rizhsky 26, 198103, St Petersburg, Russia
‡ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Optical properties of multilayer structures with quantum dots InAs on GaAs are investigated. It is shown that under optimal growth conditions the 1.3 $\mu$m emission could be achieved. Possible scenarios of quantum dots behaviour evaluation is discussed in a frame of elastic theory to explain differences in optical properties of the grown structures.

Introduction

During the last decade one of main trend in the fundamental and applied surface science is an investigation of nanostructures formation processes via exploring of the self-organisation effects during heteroepitaxial growth in lattice mismatched systems. These nanostructures — quantum dots (QDs) and quantum wires — reveals ways for design the new generation of optoelectronic devices. One of the problem in modern optoelectronic is a creation of laser diodes emitted at 1.3 $\mu$m wavelength for the applications in fibre-optic communications. To solve this problem were suggested and realised several approaches: using of InGaAsN quantum wells with low nitrogen concentration in an active region [1], placing of InAs QDs in a silicon matrix [2], and realisation of the Stranski–Krastanow growth mode for QDs formation in heteroepitaxial (In,Ga)As/GaAs system allowing to shift significantly emitting wavelength in comparison to (In,Ga)As/GaAs quantum wells.

For the last case the possible ways of reaching 1.3 $\mu$m range wavelength are:

(a) InGaAs/GaAS QDs formation by depositing sufficiently high amount ($\sim$11 monolayers (MLs) for solid solution (In,Ga)As) during molecular–beam epitaxy (MBE) via supplying of In and Ga atoms and As molecules consequently in order to increase nanoisland lateral sizes [3];

(b) placing of InAs QDs in an external (Al,In,Ga)As quantum well [4, 5] which stimulates dissociation of the solid solution and leads to increased In concentration near QD.

However, the problem to achieve 1.3 $\mu$m range for structures with InGaAs/GaAs QDs is that the total InAs amount in active region is relatively high which leads to high probability of the dislocations formation. Thus, it is actual to find new ways to create devices based on QDs emitting at 1.3 $\mu$m by minimising In amount in active region. In this paper we propose to use submonolayer migration enhanced epitaxy (SMEE) and their combination with MBE to form InAs QDs in multilayer structures allowing to observe 1.3 $\mu$m PL emission at room temperature.
Experiment

Growth experiments are carried out using EP1203 setup on singular and 7° misoriented towards [011] and [010] directions semi-insulating GaAs(100) substrates. The samples (singular and vicinal) mounted with In on the same molybdenum holder in order to minimise heater temperature field inhomogeneity and flux gradients. After desorption of an oxide layer in growth chamber at substrate temperature $T_s \approx 630 ^\circ C$ under As$_4$ flux the GaAs buffer of 3000 Å thickness is grown by conventional MBE at $T_s \approx 550 ^\circ C$ (2 $\times$ 4 surface reconstruction). Active region is placed in between 60 Å GaAs layers from both sides and consists of ten InAs QDs layers grown by SMEE or MBE+SMEE methods. For SMEE, In and As are deposited on the surface consequently, every cycle of In atoms deposition corresponds to the growth of 0.5 ML followed by 10 seconds growth interruption under As flux supplying.

Quantum dots layers are separated by 80 Å GaAs spacers. For different structures effective thickness of InAs QD layer is varied from 2.5 to 3.0 MLs. Growth temperatures $T_s \approx 480 ^\circ C$ for an active region (surface reconstruction 2 $\times$ 2 appears) and $T_s \approx 550 ^\circ C$ for the other parts of the structure are used. QDs formation is controlled in situ by RHEED technique using special registration and analysis system. Total arsenic pressure in growth chamber for all experiments are $1.5 \times 10^{-6}$ Pa. Growth rates are 0.1, 0.24 and 0.7 ML/sec for InAs, AlAs and GaAs, respectively.

For PL study, active region is confined by Al$_{0.25}$Ga$_{0.75}$As/GaAs short-period super-lattices (5 pairs, 25 Å/25 Å) from both sides. For measuring PL spectra Ar$^+$ laser ($\lambda = 514.5$ nm, excitation density $\sim 100$ W/cm$^2$) and cooled Ge photodiode as a detector are used.

Results and discussion

In Fig. 1(a) PL spectra at 77 K and 300 K for the sample which active region consisted of 10 InAs QD layers with 2.5 ML nominal thickness are shown. QDs are grown by SMEE method (5 cycles) and were separated by 80 Å GaAs spacers. Spectra are characterised by typical for quantum dots wide lines with maximuma at $\sim 1.2$ eV and $\sim 1.05$ eV for 77 K and 300 K, respectively. At room temperature only long-wave edge of PL line is in 1.3 μm region. A conclusion about relatively small nanoisland sizes from these spectra is made. This may be explained either by partial evaporation of In atoms during SMEE first cycle (which leads to effective decrease of InAs layer nominal thickness) or by effective migration of adatoms during exposing in As cycle.

To clarify the situation a serie of multilayer structures with different technological parameters is grown. Increasing InAs amount in each QD layer up to 3.0 ML leads to shift PL peak towards long-wave region. However, the shifting is accompanied with dramatically PL intensity decreasing in comparison to the previous structure. In this case only the small part of QDs with the sizes satisfying 1.3 μm emission is formed. Most of the nanoislands exceed critical volume of misfit dislocations formation and as a result — crystallographic quality of the whole structure worses.

But if the first cycle (0.5 ML of InAs) is grown by conventional MBE and the others — by SMEE (2.5 ML of InAs in total for each layer) situation changes. In Fig. 1(b) PL spectra taken at 77 K and 300 K for singular and vicinal samples are shown. At 77 K PL spectra are characterised by set of peaks in 1.0–1.2 eV range with typical shift to short-wavelength region for vicinal samples [6].

Coexistence of several peaks on these spectra causes by several reasons: presence of
Fig. 1. PL spectra taken at 77 K (solid line) and 300 K (dashed line). (a) Active region consists of 2.5 ML of InAs QDs grown by SMEE (10 QDs sheets). (b) Active region consists of 0.5 ML of InAs grown by MBE + 2.0 ML of InAs grown by SMEE (10 QDs sheets).

several groups of QD with different sizes, possible formation of QD conglomerates in upper layers as a result of lateral association of neighbour QDs [7], or contribution of excited states of QDs which is proved by changing the spectrum shape at lower excitation density. At room temperature maximum of PL peak for singular sample is at 1.3 µm, for vicinal samples — at 1.20–1.25 µm which is also correlating with PL data at 77 K. Relatively low intensity decreasing at 77 K and 300 K (about 10–20 times) reveals high quality of the grown structures.

Computer simulation data [8] of the multilayer structures with QDs formation during MBE based on elastic interaction theory show following different behaviour in vertically stacked nanoislands system depending on the nanoisland spacing:

(a) increasing of the nanoislands size from layer to layer with further association,
(b) new nanoisland birth with slight size decreasing,
(c) equilibrium state of the system with multilayered QDs for the stacking of the islands upon each other from layer to layer.

These scenarios depend critically on the spacer thickness between different layers. For the described above structures the interval of nanoisland spacing in order to get high crystallographic quality of structure should be ∼240–400 Å which is in accordance with our previous STM study [9].

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References


Photoluminescence of InAs/GaAs quantum dots under hydrostatic pressure

V. A. Gaisin†, Dinh Son Thach†, B. S. Kulinkin†, B. V. Novikov†, V. N. Petrov‡, V. M. Ustinov§ and G. E. Cirlin‡

† Institute of Physics, St. Petersburg State University, 198904 St. Petersburg, Russia
‡ Institute of Analytical Instrumentation of Russian Academy of Sciences, Rizhsky 26, 198103 St. Petersburg, Russia
§ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The baric dependencies (0–20 kbar) of luminescence spectra (77 K) of InAs quantum dots have been investigated. InAs quantum dots on vicinal substrates GaAs at misorientation angle 3[001], 7[001], 7[011] were grown. At \( P = 0 \) in the luminescence spectra a set of lines 1.235, 1.290, 1.343 eV of quantum dots of different sizes with base lengths 8–9 nm was observed. The dependence of baric coefficients on size of quantum dot was found out.

Introduction

Now the physical properties of quantum dots are researched very intensively, particularly of the quantum dots placed in crystal matrices. The molecular beam epitaxy method allows to obtain the structures with given parameters. The theory of optical transitions leading to a satisfactory correspondence to experimental properties has been developed. The theory describes a blue quantum size shift, however the structure excited states has not sorted out. The experiments with the using of high hydrostatic pressure indicated to the important role of a matrix in forming of excited states, however there remains a lot indistinct moments.

1. Experimental

The baric dependencies of luminescence spectra (77 K) of samples with InAs quantum dots, grown on vicinal substrates GaAs at misorientation angle 3[001], 7[001], 7[011] have been investigated.

The structures consist of the InAs quantum dots confined from both sides with wide-gap GaAs and \( Al_{0.25}Ga_{0.75}As/GaAs \) superlattices (5 pairs, 2 nm/2 nm each). Singular substrates GaAs [100], disoriented on 3°–7° were used. For better homogeneity of temperature field of the sample and univormity of molecular flows on substrate surface the samples were pasted using In to one holds side-side.

The luminescence spectra were excited by He–Ne laser at power 1 W/cm². The studies were conducted in the range from 0 to 20 kbar in the high-pressure camera with anvils of laicosapfire. The mixture of wood-spirit and ethyl alcohol in the ratio of 1:10 was used for transmission of the pressure to samples. Magnitude of hydrostatic pressure was determined from displacement of R1-luminescence line of ruby, placed in driving volume of camera in the immediate vicinity of the sample under study. Samples were prepared in form of parallel-plane plates of sizes 0.5 × 0.5 mm² and thickness 0.05 mm.
2. Results

At $P = 0$ in the luminescence spectra of all studied samples a set of lines at energies 1.235, 1.290, 1.343 and 1.408 eV as well as lines of free exciton of the substrate GaAs at 1.51 eV was observed. The individual components of the spectrum correspond to the emission of quantum dots of different sites with base lengths, varying within the 8–9 nm.

With increasing hydrostatic pressure the short-wave shift of all spectral peculiarities was observed. Simultaneously the increase of the energy extent of the emission spectrum with relative development of contribution of high energy components place.

At $P > 5$ kbar an additional peculiarity-WL, absent at $P = 0$, arose. The emission spectra of one samples, having the most developed structure 7[001], at different hydrostatic shifts are given in Fig. 1(a). In Fig. 1(b) two spectra at $P = 0$ and $P = 10$ kbar, superimposed at energy of free exciton in GaAs are given for comparison and illustration of the change of energy distances between the peculiarities.

![Figure 1](image)

**Fig. 1.** (a) The emission spectra of 7[001] samples at different hydrostatic pressure and $T = 77$ K. (b) The spectra at $P = 0$ and $P = 10$ kbar superimposed at energy of free exciton in GaAs. The power of He–Ne laser is 1 W/cm², $T = 77$ K.

The dependencies of energy position of the peculiarities on hydrostatic pressure and dependence of energy interval between the emission lines of quantum dots of different sizes and luminescence line of free exciton of the substrate are shown in Fig. 2(a) and Fig. 2(b). Figure 2(b) allows us to determine the magnitude of hydrostatic shift from the known meaning of baric coefficient of substance 10.2 meV/kbar (10.7 meV/kbar [4]) precisely.

3. Discussion

In literature there is a numbers of work [2–4] on investigations of the influence of hydrostatic pressure on luminescence spectra of quantum dots InAs/GaAs. Mainly the authors were interested in the range of high hydrostatic pressures $P > 40$ kbar at which the transformation of emission spectra took place in consequence of change of band structure of GaAs crystals. The observed by us dependence of hydrostatic shift magnitude on a size of pyramidal quantum dots InAs was not found out earlier. We suppose that the revealed effect
due to the influence of GaAs matrix on a spectrum of quantum dots InAs too. Indeed it is known from the theory that in parallel with the size effect the internal hydrostatic pressure as well as size effect contributes to the forming of band structure of pyramidal quantum dots [5]. It is caused by the difference between lattice constants of InAs and GaAs and depends on a length of pyramid base. It probably causes the observed in this work dependence of the baric coefficients on size of quantum dot. The difference of energy structures of pyramidal quantum dots InAs in GaAs and spherical dots InAs in polymer matrices [6] points to the possibility of such interpretation. The energies of the transitions InAs in GaAs crystals are more then those in polymer matrices by value of orders 0.3 eV owing to internal tensions.

4. Conclusion

In present paper the baric dependencies the of the luminescence spectra of InAs quantum dots, grown on vicinal substrates GaAs have been investigated. The dependencies of the baric coefficients on size of quantum dots has been found out. The possible interpretation of this effect has been given. In future for the suggestion it is proposed to carry out the direct calculation of the dependence of baric coefficient on nanocrystals size on the basic of the theory [5].

References

The spectral distribution of polaron exciton line in quantum dot

I. P. Ipatova, A. Yu. Maslov and O. V. Proshina
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The intensity of polaron exciton absorption in spherical quantum dot is calculated. The localized electrons and holes are shown to interact with long wavelength phonons with wave vectors \( q \approx 1/R \), \( R \) being the radius of the dot. The broadening of different phonon replicas are shown to be less then their separation

Compounds II–VI are good semiconductor system suitable for fabrication of blue–green lasers [1]. Since II–VI materials have high ionicity, it is natural to expect the existence of electron and hole polaron states [2].

The electron and hole polaron effect in nanostructures has been considered in [3, 4] using Kane model and Luttinger Hamiltonian approximation. It is shown that hole polaron polarization effect differ from electron polarization effect because of the degeneration of hole bands.

The paper deals with the theory of optical transition in ionic crystal were initial and final states of the interband transitions are electron and the hole polarons, respectively. The Coulomb interaction of the electron and the hole is taken into account in optical transition. It is accepted that the binding energy of polaron exciton is smaller then the size quantization energies of both the electron and the hole. It means that the polaron exciton radius \( R_{exc} \) is larger then the quantum dot radius \( R \) (strong confinement regime):

\[
\frac{R_{exc}}{R} \gg 1.
\] (1)

When the light is absorbed, there is a photon in the initial state, there are neither electrons nor holes yet. The wave function of the initial state is approximated by

\[
\Psi^{(i)} = \delta(r_e - r_h)\chi^{(i)}_{lattice}.
\] (2)

Here \( r_e, r_h \) are radius vectors of the electron and the hole, respectively, \( \chi^{(i)}_{lattice} \) is the lattice vibrations wave function. In the final state of the optical transition, there are the electron and the hole which interact one with another and with polar optical phonons. The wave function of the exciton final state in the strong confinement limit Eq. (1) is

\[
\Psi^{(f)} = \psi_e(r_e)\psi_h(r_h)\chi^{(f)}_{lattice}.
\] (3)

Since the quantum dot is considered here as a macroscopic conglomerate, the electron wave function \( \psi_e(r_e) = \psi_n(r_e)u_e(r_e) \) is the product of convolution \( \psi_n(r_e) \) by the electron modulating Bloch wave function \( u_e(r_e) \). The function \( \psi_h(r_h) = \psi_N(r_h)u_v(r_h) \) is the hole wave function where \( \psi_N(r_h) \) is the solution of Schrödinger equation with Luttinger Hamiltonian and \( u_v(r_h) \) is the hole Bloch modulating wave function. The vibrational wave functions \( \chi^{(f)}_{lattice} \) correspond to normal phonon modes with shifted equilibrium positions with respect to \( \chi^{(i)}_{lattice} \), because the different levels of the electron and the hole size quantization create different polarizations of the medium.
The rate of the absorption of a photon in the dipole approximation

\[ W_{nN}^{abs} = \frac{2\pi}{\hbar} |P_{cv}|^2 |I_{nN}|^2 \sum_f W_{if} \delta(E_{nN} - E_f - \hbar\omega). \]  

(4)

Here \( P_{cv} \) is the interband transition matrix element over Bloch modulating function, \( I_{nN} \) is the overlapping integral of the electron and the hole wave functions

\[ I_{nN} = \int d^3r \psi_n(r)\psi_N(r) \]  

(5)

and

\[ W_{if} = \left| \int dx_q \chi_{lattice}^{(i)}(x_q)\chi_{lattice}^{(f)}((x_q - s_q)) \right|^2 \]  

(6)

is the overlapping integral of lattice vibration wave functions. The polarization shift of the normal mode equilibrium positions \( s_q \) has been calculated in [4]. Both the electron and the hole polarization effects contribute to the shift

\[ s_q = \frac{e}{\omega(q)q} \left\{ \rho_N(q) - \rho_n(q) \right\} \sqrt{\frac{2\pi}{M_0 V \varepsilon}}. \]  

(7)

Here \( M_0 \) is the oscillator mass, \( V \) is the volume of the crystal, \( \varepsilon \) is the optical dielectric constant, \( \omega(q) \) is the phonon frequency with the wave vector \( q \),

\[ \rho_n(q) = \int d^3r_n e^{iqr_n} \psi_n^2(r_n), \]  

(8)

\[ \rho_N(q) = \int d^3r_h e^{iqr_h} \psi_N^2(r_h), \]  

(9)

are the electron and the hole densities.

Equation (6) can be rewritten in the form of the product of the transition rates \( w_q \) for each normal mode of lattice vibrations:

\[ W_{if} = \prod_q w_q \]  

(10)

where \( q \) numerates normal modes of the lattice vibrations and

\[ w_q = \left| \int dx_q \chi_{lattice}^{(i)}(x_q)\chi_{lattice}^{(f)}(x_q - s_q) \right|^2. \]  

(11)

Since equilibrium positions in integral (11) are shifted, there is the finite probability of transition from initial to final state with emission of arbitrary number of phonons.

The rate of the transition from the ground state of oscillator into the excited state with the emission of \( K_q \) phonons of \( q \)-th normal mode with the frequency \( \omega(q) \) is equal to [4]

\[ w_q(K_q) = \frac{\left| s_q \right|^2 M_0 \omega(q)}{2 K_q(K_q)!} K_q \exp \left[ -\frac{1}{2} \left| s_q \right|^2 \frac{M_0 \omega(q)}{\hbar} \right]. \]  

(12)
The equation (12) is the Poisson distribution of spectral lines corresponding to different number of emitted phonons with the frequency \( \omega(q) \). Each spectral line is not broadened.

The vibrational spectrum of the crystal consists of many normal modes with different \( \omega(q) \). The total set of normal modes contributes to the probability Eq. (4).

The transition rate Eq. (4) contains in the lowest dipole approximation the succession of multi-phonon transitions (peaks) each of which corresponds to the different number \( K \) of emitted phonons. The number \( K \) is obtained by the selection from the total set of normal modes only those which satisfy the condition

\[
K = \sum_q K_q. \tag{13}
\]

Each phonon replica \( K \) is broadened due to the dispersion of optical branches. The quantity \( K \) takes the values \( K = 0, 1, 2, 3, \ldots \) where \( K = 0 \) is the zero-phonon line, \( K = 1 \) is the process with the participation of the single phonon, \( K = 2 \) is one with participation of two phonons and so on.

It is necessary to find condition under which the polaron exciton optical spectrum still has the form of well seen succession of lines corresponding to multi-phonon replicas. The electron density \( \rho(q) \) is not zero inside of the quantum dot only. Therefore, electrons and holes interact with long wavelength polar optical phonons with the wave vector \( q \approx 1/R \ll \pi/a \), \( a \) being the lattice parameter. In terms of \( K \), the rate of polaron exciton absorption takes the form:

\[
W_{abs}^{nN} = \frac{2\pi}{\hbar} | P_{cv} |^2 | I_{nN} |^2 \sum_K \sum_{q_1q_2\ldots q_K} \left( \frac{4\pi e^2}{\epsilon V} \right)^K W_K(q_1, q_2, \ldots q_K) \times \delta(E_{nN} + \hbar \omega(q_1) + \hbar \omega(q_2) + \ldots + \hbar \omega(q_K) - \hbar \omega), \tag{14}
\]

where

\[
W_K(q_1 \ldots q_K) = \frac{[\rho_N(q_1) - \rho_n(q_1)] [\rho_N(q_2) - \rho_n(q_2)] \ldots [\rho_N(q_K) - \rho_n(q_K)]}{\hbar \omega(q_1) \hbar \omega(q_2) \ldots \hbar \omega(q_K) q_1^2 q_2^2 \ldots q_K^2} \times \exp \left[ -\frac{S_0}{2} \right]. \tag{15}
\]

Here \( S_0 \) is equal to

\[
S_0 = \sum_q \frac{4\pi e^2}{\epsilon V} | \rho_N(q) - \rho_n(q) |^2 \frac{1}{\hbar \omega(q) q^2} \tag{16}
\]

Notice that the summation index \( q \) in Eq. (16) means the phonon wave vector and the lable of the optical branch. When \( K = 0 \), we find from Eq. (14) the intensity of the zero-phonon line

\[
I_0 = \exp \left[ -\frac{S_0}{2} \right], \tag{17}
\]

which is not broadened. When \( K = 1 \), the intensity of the one-phonon replica \( I_1(q) \) has been calculated numerically for the model phonon spectrum \( \omega(q) = \omega_0 - \alpha q^2 \), where

\[
\alpha = \frac{\partial^2 \omega(q)}{\partial q^2} |_{q \leq 1/R}.
\]
Fig. 1. The spectral distribution of polaron exciton lines of the first phonon replica for the different values of light and heavy hole masses $\beta = m_l/m_h$.

The wave functions $\psi_n(r_e)$ and $\psi_N(r_h)$ are taken to be solutions of Schrödinger equation for the spherical quantum dot with infinite walls [5, 6]. The result of calculations is shown in Fig. 1 for different values of the ratio of the light and heavy hole masses $\beta$. The broadening of the one-phonon replica is shown to be less then the separation of peaks.

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Photoelectronic properties of InAs/GaAs nanostructures with combined quantum well and quantum dot layers grown by Metal-Organic Vapor Phase Epitaxy

I. A. Karpovich†, B. N. Zvonkov§, D. O. Filatov‡, S. B. Levichev†, N. V. Baidus§ and S. M. Nekorkin§
† University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia
‡ Research and Educational Center for Scanning Probe Microscopy, University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia
§ Physical-Technical Research Institute, University of Nizhni Novgorod, 603600 Nizhni Novgorod, Russia

Abstract. Combination of quantum well (QW) and QD layers in the GaAs/InGaaS nanostructures obtained by MOVPE result not only in well known “red shift” of the QD PL spectrum, but also to splitting of the PL spectrum and of the photosensitivity spectrum of the QW. “Red shift” of one of these spectral features relative to the other is explained by diffusion of In from the QDs to the QW at their heterointerface.

GaAs/InGaaS nanostructures with separate quantum wells (QWs) and quantum dots (QDs) have been studied well to the present time. Recently studies of the nanostructures with QD/QW combined layers have been begun [1, 2]. It is expected to observe a number of corporate effects due to interaction of the QD and QW layers: redistribution and relaxation of elastic strains, intermixing of the components, hybridization of energy spectra of QD and QW, etc. All of the above can influence on the morphology and energy spectrum both of QD and QW and, in turn, on the their electronic properties. In these structures photoluminescence and lasing in the 1.3 µm band were obtained, which is important for optoelectronics [3]. Probably it is possible to achieve the next transmitance window at 1.5 µm. Decreasing of the ground state transition energy in a QD was explained by relaxation of the elastic strain [1] or by alloy decomposition and In segregation [2].

In this work, we investigated the surface morphology, photoluminescence (PL), and capacitive photovoltage (CPV) spectra of GaAs/InGaaS nanostructures with combined QD/QW layers of various types, grown by Metal-Organic Vapor Phase Epitaxy (MOVPE).

The nanostructures were grown on semi-insulating GaAs (001), misoriented by 3° towards [110]. n-GaAs ~0.8 µm buffer layers were grown at 600°C. Then temperature was decreased down to 530°C and InAs QD layer followed by InGaaS QW layer ($x \sim 0.2$, $d \sim 5$ nm) was deposited. In other series these layers were deposited in the inverse sequence. When growing the QDs trimethyl indium and arsine were introduced into the reactor alternatively for 6 and 4 s respectively with the 4 s intervals between the cycles. Total number of the cycles was up to 10, which results in the InAs nominal thickness of 1.5 nm (~5 monolayers (ML)). The structures both with and without 15 nm GaAs cap layer for optical and morphological investigations respectively were grown. Morphology of the QD layers was studied by Atomic Force Microscopy (AFM) using TopoMetrix ‘Accurex’ TMX-2100 AFM in contact mode. PL spectra were measured at 77 K and CPV spectra — at 300 K using the technique described in [4].

According to AFM data, when isolated InAs layers are grown on the GaAs surface, the InAs clusters with 45–50 nm base and 6–10 nm height are formed. Their surface density is ~10^10 cm^-2 [5]. These parameters are close to the ones reported for the QDs grown
by molecular beam epitaxy (MBE) in [1]. For rather large fraction of In in the QDs PL peak from QD shifts to lower energies down to 0.97 eV (77 K). The corresponding CPV peak shifts down to 0.9 eV (300 K). In the nanostructures grown by MBE such shift was achieved only by deposition of an additional QW layer over the QDs [1, 2]. High PL intensity comparable to the one of QW indicates the crystal structure of the QDs to be perfect enough. Such properties of the QDs grown by AP MOCVD can be explained by assuming formation of graded composition transient region around a QD originating from diffusion of In into GaAs playing a role of the external QW.

As the ground transition energy in the QD is considerably less than the one in the QW, and as an exciton generated by photoexcitation in the QW have enough time to relax to the QD ground state before recombination so PL from QDs only can be observed at low excitation levels. In contrary, in the CPV spectrum that represents the optical absorption spectrum all over the quantized states it is possible to observe all quantum-dimensional layers [4]. However, because the surface density of QDs was rather small (< \(10^{10}\) cm\(^{-2}\)) it was sometimes difficult to resolve the QD photosensitivity above the impurity background due to the deep levels HL1 and EL2 in the substrate. Nevertheless, the InGaAs QW and the wetting layer were always well resolved. So the methods of PL and CPE appear to be complimentary allowing to resolve the objects of both types.

The morphology of combined QD/QW layers depends on consequence (order) of deposition. When depositing the QW onto the QDs (QW/QD) QDs are partly seen through the QW layer (Fig. 1(a)). Deposition of QW depressed coalescence of nanoclusters. In the QD/QW layer coalescence is stronger resulting in formation of big clusters and in decreasing the concentration of QDs (Fig. 1(b)). Tendency to the formation of groups of 2–3 clusters preceding confluence of them is seen up. It should be noted that morphology of the QD/QW layers with a cup layer differs substantially from the one of the surface dots.
Shift of the QDs’ PL peak to lower energies by 30–40 meV compared to the one in the single QD layer always takes place in the QW/QD nanostructures of both types (Fig. 2). It should be noted that such shift was much less than in the structures obtained by MBE [1, 2].

In the CPV spectrum of the QDs (Fig. 3, curve 2) the CPV edges from QDs at ∼0.98 eV and from wetting layer at ∼1.23 eV are seen. In the structures with In$_{x}$Ga$_{1-x}$As QW ($x$ ≈ 0.2, the well width ≈ 5 nm) the QW band has a sharp step near 1.2 eV (curve 1). In the QW/QD nanostructures a weak band from QD with threshold 0.94 eV is seen and its location at 300 K agrees with the PL peak position at 77 K (1.022 eV). In all the spectra of the combined layers the effects of splitting and of “red shift” of the QW band compared to the one of a single QW (QW$_{2}$ and QW$_{1}$ at ∼1.1 and 1.2 eV respectively on curve 3) were observed. The band QW$_{1}$ is likely from initial QW between the QDs. Its integration with significantly much narrower but much deeper QW from InAs wetting layer results in disappearing of the wetting layer subband and in some shift of the QW subband as a result of the QW broadening and of change of the well potential shape. However, this effect could not explain the shift as much as by 0.1 eV. Probably it is due to the parts of QW covered by QD. Shift to lower energies can be explained by diffusion of In from the QDs to the QW which can reduce the effective gap of the latter. Effect of the QW broadening in the QD/QW combined layer is also seen (curve 4) but in this case it is less expressed and the “red shift” of QW is smaller.

Similar results were observed in the combined QW/QD layer without a cup layer (Fig. 3, curves 5–7). Bigger value of “red shift” in the QW/QD than in the QD/QW can be explained by absence of In diffusion from the QW to the matrix.

Acknowledgements

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References

Semiconductor quantum dot superlattices for thermoelectric applications

A. Khitun†, A. Balandin‡, J. L. Liu† and K. L. Wang†
† Device Research Laboratory Electrical Engineering Department
University of California, Los Angeles, California 90095, USA
‡ Department of Electrical Engineering University of California,
Riverside, California 92521, USA

Abstract. We have theoretically investigated the in-plane lattice thermal conductivity of a quantum dot superlattice that consists of arrays of ordered quantum dots. Our theoretical model takes into account scattering of acoustic phonons on quantum dots made out of material with a finite acoustic mismatch with the host material. Modification of phonon dispersion in the quantum dot superlattice has also been incorporated into the model. Numerical simulation has been carried out for the quantum dot superlattice made out of multiple Si layers with Ge quantum dots. Obtained results are important for suggested applications of Si/Ge quantum dot superlattices for high-temperature thermoelectric devices.

Introduction

Quantum dot superlattices (QDS) are attractive structures that can be used in a wide range of applications [1–2]. Recently it was shown that QDS made out of suitable constituent materials might have a great potential for thermoelectric applications [3–4]. As a result, thermal properties of QDS became extremely important subject of theoretical and experimental research. Decrease of the lattice thermal conductivity due to additional phonon scattering on quantum dots and quasi-0D confinement of carriers may lead to an increase of the thermoelectric figure of merit $ZT = S^2 \varepsilon / (\kappa + \kappa_e)$ (where $S$ is the Seebeck coefficient, $\varepsilon$ is the electric conductivity, $\kappa$ is the phonon thermal conductivity, and $\kappa_e$ is the electronic thermal conductivity). Acoustic impedance mismatch between material of a quantum dot and the host material (wetting layer and spacer) leads to strong phonon scattering. At the same time, electron transport is not severely deteriorated since the band-gap offset could be small (like in Si/Ge system) and the wetting layers may act as a 2D conducting channels. In this case, QDS may represent a good example of the “phonon-blocking electron-transmitting” structure with a great promise for thermoelectric applications [5].

Significant achievements in fabrication techniques for arrays of semiconductor quantum dots made possible to control both the size and position of quantum dots [6–7]. Since the size of the quantum dots and distance between two adjacent dots are comparable with the acoustic phonon wavelength, the long-range ordering of the quantum dots may produce coherent phonon scattering that significantly enhances phonon relaxation rates and modifies phonon group velocity. The numerical calculations were carried out for a structure that consisted of multiple layers of Si with ordered Ge quantum dots separated by wetting layers and spacers.
1. Model

The expression for the lattice thermal conductivity in the relaxation-time approximation can be written as [8]

$$\kappa = \frac{1}{3} \sum_i d k v_{gi}^2(k) \tau_C S_i(k) \quad (1)$$

where \( i \) denotes particular phonon polarization branch, \( v_{gi} \) is the phonon group velocity, \( S_i(k) \) is the contribution to the specific heat from modes of the polarization branch \( i \) which the phonon wave vector interval of \( dk \). Combined relaxation time \( \tau_C \) includes all relaxation rates corresponding to the different scattering processes which do not conserve crystal momentum [9]

$$\frac{1}{\tau_C} = \sum \frac{1}{\tau} = \frac{1}{\tau_M} + \frac{1}{\tau_B} + \frac{1}{\tau_U} + \frac{1}{\tau_D}. \quad (2)$$

In Eq. (2) we included relaxation rates for three scattering processes which are dominant in Si, Ge, and SiGe. These processes are \( 1/\tau_U \) — three-phonon Umklapp processes, \( 1/\tau_M \) — point defect scattering (isotopes, impurities, etc.), \( 1/\tau_B \) — boundary scattering, and \( 1/\tau_D \) — scattering on quantum dots.

The first three terms in Eq. (2) are well known [9]. The only new term we introduced the scattering on quantum dots. To describe phonon transport in QDS, we use the continuum model approximation and an assumption that the thermal phonon wave can be represented by a sum of plane waves. A phonon wave outside the dot is a superposition of incident and scattered waves.

In order to find \( 1/\tau_D \) we integrated all scattered waves from all \( N \) dots. In some arbitrary point reflected amplitude normalized to incident one \( \Gamma \) is

$$\Gamma = \frac{|F(\theta)|^2}{r^2} \sum_{n=1}^{N} \exp(iur_n) \quad (3)$$

where \( F(\theta) \) is the scattering function

$$F(\theta) = \frac{i}{2k} \sum (2n + 1)(1 + R_n) P_n(\cos \theta) \quad (4)$$

\( R_n \) is the dot reflection coefficient, \( P_n(\cos \theta) \) are Legendre polynomials, \( u = k_0 - k \), \( k \) and \( k_0 \) are the wave vectors of the plane and scattered waves, \( r \) is the radius vector.

Due to scattering on quantum dots, the phonon dispersion and corresponding phonon group velocity in QDS will be changed. This change can be taken into account using the following relation [10]

$$k'^2 = k^2 \left( 1 + \frac{2\pi NF(0)}{Vk^2} \right)^2 - \left( \frac{2\pi NF(\pi)}{Vk^2} \right)^2 \quad (5)$$

where \( k' \) is the modified wave number in the presence of quantum dots, \( F(0) \) and \( F(\pi) \) correspond to the forward and backward scattering respectively.

Additionally, modification of the phonon group velocity may come from spatial confinement of phonon modes inside the two-dimensional spacer and wetting layers of QDS [11].
2. Results

Numerical calculations have been carried out for SiGe QDS with the spacer layer of 100 nm at temperature 300 K for different dot sizes. Lattice thermal conductivity was calculated as a function of a quantum dot volume fraction. Results are presented in Fig. 1: A, B, C curves correspond to the dot size of 1.0, 0.8, and 0.2 nm, respectively. D curve (dashed line) corresponds to the mass-different limit where each dot is just an impurity atom. Obtained results may be used for quantitative evaluation of minimum possible QDS’s lattice thermal conductivity. It was found that additional decrease (about 30% at 0.1 quantum dot volume fraction) of the Si/Ge QDS can be achieved by using ordered quantum dot array instead of QDS with randomly distributed quantum dots. The additional decrease is caused by the coherent acoustic phonons scattering. The decrease of the lattice thermal conductivity remains significant in a wide temperature range.

Fig. 1. Results of calculations of the lattice thermal conductivity of a quantum dot superlattice as a function of quantum dot volume fraction for different Dot sizes (a) (A—$a = 1.0$ nm; B—$a = 0.8$ nm; C—$a = 0.2$ nm; D—mass-different limit).

Acknowledgements

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References


Abstract. We have investigated the photoluminescence, persistent photoconductivity and hopping conductivity of InAs/GaAs quantum dot structures grown on vicinal substrates. The photoconductivity has been investigated for wavelengths $\lambda = 791 - 1120$ nm in the temperature range $T = 4.2 - 300$ K. At low temperatures we observe positive persistent photoconductivity, which is attributed to the spatial separation of photogenerated carriers. Variable range hopping conductivity (VRHC) has been observed at low temperatures. Upon reducing the temperature the resistivity shows a crossover near 3.2 K from two-dimensional Mott VRHC, $\rho(T) = \rho_0 \exp(T_0/T)^{1/3}$, to Efros–Shklovskii Coulomb gap behaviour $\rho(T) = \rho_0 \exp(T_0/T)^{1/2}$.

Introduction

In recent years, three-dimensional nanoscaled semiconductor islands have attracted significant attention due to their potential to act as quantum dot (QD) systems [1]. The lateral sizes and morphology, such as height, density and size distribution, of such islands can be adjusted within a certain range using appropriate growth conditions. Fabrication techniques, such as growth on regular patterned surfaces, have been proposed to make the distribution of quantum dot sizes more uniform [2]. While the optical properties of quantum dot structures are currently under intense investigation, less attention has been given to the transport properties. In this work, we report on the photoluminescence, persistent photoconductivity and hopping conductivity of self-assembled InAs quantum dot layers, grown on vicinal GaAs substrates.

1. Samples

The structures were grown by atmospheric pressure MOCVD at a temperature of 633°C, using trimethylindium, trimethylgallium and arsenic, on semi-insulating (001) GaAs substrates misoriented 0.14° from the (001) plane towards the [110] direction. The samples consisted of a 0.45 mm thick $i$-GaAs layer, a $\delta$-layer of Si, a spacer layer (width 18 nm), an InAs quantum dot layer, a second spacer layer (width 18 nm) and a second $\delta$-layer of Si. The Si $\delta$-layers are necessary to obtain free electrons in the QD layer. The structures were capped with a layer of GaAs (width 0.45 mm). The relevant parameters of the structures are listed in Table 1. We also prepared and investigated multi-layer InAs/GaAs quantum dot structures (see [3]).

Photoluminescence (PL) spectra were obtained at $T = 77$ K using a He–Ne laser. Transport measurements were carried out on square samples with edges along the [110]
and [110] directions. The temperature variation of the resistance was measured in the range $T = 0.6-300$ K. The magnetoresistivity $\rho(B)$ and Hall effect were measured in magnetic fields $B$ up to 10 T using a superconducting solenoid. In order to investigate the photoconductivity, the samples were illuminated using different optical filters (wavelength $783 \text{ nm} < \lambda < 799 \text{ nm}$ or $\lambda > 1120 \text{ nm}$).

2. Photoluminescence

In Fig. 1(a) the photoluminescence spectra are shown for single dot layer samples 2706, 2707, 2713, 2714. The maxima in the photoluminescence peaks are observed at 1.39–1.47 eV, while the halfwidths amount to 15–40 meV. A rough estimate of the dimensions of the QD may be extracted from the maxima of the PL spectra using the theory developed in Ref. [4]. Overlap of the electron wave functions on the nearest neighbour quantum dots should lead to a broadening of the electron (hole) energy levels in the single dot, and to 2D band formation. The QD size determined from the maxima of the PL spectra equals 5–7 nm. The QD size determined directly by atomic force microscopy amounts to 5–6 nm.

3. Persistent photoconductivity

Persistent photoconductivity (PPC) was observed in our samples. The $\rho(4.2 \text{ K})$-values, obtained before and after illumination, are listed in Table 1. In Fig. 1(b) we show $\rho(T)$ measured in dark and after illumination at $T = 4.2$ K by light passed through filter 1 ($\lambda \geq 1120 \text{ nm}$) and filter 2 ($783 \text{ nm} < \lambda < 799 \text{ nm}$) for sample 2713. In the illuminated cases, the heating rate was 5 K/min. The resistivity of all samples decreases after both types of illumination. The saturation value of the resistance after illumination through filter 2 is less than the one obtained after illumination through filter 1. For $T > 250$ K the difference between $\rho(T)$ before and after illumination was always negligible.

The Hall electron densities $n$ and the Hall mobilities $\mu$ are listed in Table 1. For all samples the Hall density and Hall mobility obtained after illumination by light with $\lambda \approx 791 \text{ nm}$ are larger than the ones obtained after illumination by light with $\lambda > 1120 \text{ nm}$.

Photo-excitation of electrons from the valence to the conduction band can not take place for incident light with energy smaller than the bandgap of GaAs. For our InAs QD layers...
Table 1. The resistivity, $\rho$, the Hall density, $n$, and the Hall mobility, $\mu$, for InAs/GaAs quantum dot structures at $T = 4.2$ K. Values are given in dark and after illumination with light with wavelengths $\lambda \approx 791$ nm or $\lambda > 1120$ nm.

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Applied wavelength (nm)</th>
<th>$\rho$ (\Omega)</th>
<th>$n$ (10$^{11}$ cm$^{-2}$)</th>
<th>$\mu$ (cm$^2$/Vs)</th>
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</thead>
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<tr>
<td>dark</td>
<td>3660</td>
<td>2.74</td>
<td>6240</td>
<td></td>
</tr>
<tr>
<td>2704</td>
<td>791</td>
<td>600</td>
<td>4.25</td>
<td>24500</td>
</tr>
<tr>
<td>$&gt;1120$</td>
<td>914</td>
<td>4.16</td>
<td>16400</td>
<td></td>
</tr>
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<td>dark</td>
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<td>1.4</td>
<td>205</td>
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</tr>
<tr>
<td>2713</td>
<td>791</td>
<td>782</td>
<td>4.5</td>
<td>1780</td>
</tr>
<tr>
<td>$&gt;1120$</td>
<td>13060</td>
<td>3.3</td>
<td>1450</td>
<td></td>
</tr>
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<td>2.03</td>
<td>1560</td>
<td></td>
</tr>
</tbody>
</table>

the energy at which the photoluminescence is maximal is about 1.39 eV (see Fig. 1(a)). We argue that in the case of illumination by light with $\lambda > 1120$ nm, PPC is attributed to the ionisation of donor Cr atoms in the GaAs substrate. The electrons excited from the Cr level move to the QDs layer and to the Si $\delta$-layer. In case of illumination by light with $\lambda \approx 791$ nm the effect of PPC can be explained by photogeneration of electron-hole pairs. The electrons flow towards the QDs layer and Si-$\delta$-layers, and the holes flow towards the substrate or recombine with the electrons trapped at the surface states [5].

In Fig. 2(a) we show the change of conductivity of sample 2713 as function of time, $\sigma(0) - \sigma(t)$, measured in dark, after illumination at $T = 4.2$ K by light with $\lambda \approx 791$ nm and 1120 nm. The relaxation of the PPC obeys a logarithmic time dependence which can be expressed as $\sigma(0) - \sigma(t) = A \ln(1 + t/\tau)$ (see [6]). This logarithmic relaxation process confirms that the persistency of the PPC is due to charge separation [6]. The parameter $\tau$

\[ W = -d(\ln \rho)/d(\ln T) \] vs $T$ for sample 1961. At $T \approx 3.2$ K the exponent changes from $p = 1/2$ to $p = 1/3$. 

Fig. 2. (a) The change of the conductivity of sample 2713 as function of time, $\sigma(0) - \sigma(t)$, measured in dark, after illumination at $T = 4.2$ K with light with $\lambda \approx 791$ nm (1) and $\lambda > 1120$ nm (2). The solid lines represent fits to the expression $\sigma(0) - \sigma(t) = A \ln(1 + t/\tau)$ with $\tau = 41$ s (1) and $\tau = 116$ s (2). The insert shows $\tau(T)$ for $\lambda > 1120$ nm. (b) $W = -d(\ln \rho)/d(\ln T)$ vs $T$ for $p = 1/2$ to $p = 1/3$. 

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Quantum Wires and Quantum Dots
decreases when the temperature is raised (see insert in Fig. 2(a)).

4. Hopping conductivity

The temperature dependence of the resistivity in the VRHC regime follows the law \( \rho(T) = \rho_0 \exp(T_0/T)^p \), where the exponent \( p \) depends on the shape of the density of states (DOS) at the Fermi energy \( E_F \). In 2D systems \( p = 1/3 \) (Mott VRHC) for a constant DOS at \( E_F \). Coulomb interactions between localized electrons produce a Coulomb gap with vanishing DOS near \( E_F \), and in this case \( p = 1/2 \) (Efros–Shklovskii (ES) VRHC). A crossover from ES to Mott regime is expected when, either the Coulomb interaction is screened, or the hopping energy exceeds the width of the Coulomb gap [7].

Figure 2(b) shows the quantity \( W = -d(\ln \rho)/d(\ln T) = p(T_0/T)^p \) plotted versus \( T \) on a log-log scale for multilayer sample 1961. The slope of the curve gives the exponent \( p \). Upon decreasing the temperature, a crossover from ES to Mott regime is observed at \( T \approx 3.2 \, K \).

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Negative differential conductivity in conducting carbon nanotubes

Ant. S. Maksimenko and G. Ya. Slepyan
Institute for Nuclear Problems, Belarus State University,
Bobruiskaya 11, 220050 Minsk, Belarus

Abstract. A theoretical model and computations of the I − V characteristics of long carbon nanotubes in a strong axial dc-fields at room temperatures have been presented. Negative differential conductivity has been predicted. It has been shown that |dI/dV| for metal carbon nanotubes in the region of the negative differential conductivity significantly exceeds corresponding values for semiconducting ones. The predicted effect makes possible the design of wave-generating nanotube-based diodes for submillimeter and infrared ranges.

Since the discovery by Iijima [1] of carbon nanotubes (CNs), a great deal of interest has been focused on these quasi-one-dimensional monomolecular structures because of their unique physical properties (mechanical, electrical, optical, etc.) and the rapid experimental progress in the controlled preparation. Processes of electron transport in strong external fields when nonlinear effects are constitutive are of a great interest for potential applications in nanoelectronics and for experimental diagnostic of CN themselves.

In this report we report a theoretical phenomenological analysis of the I − V characteristics of CNs at room temperatures, when k_B T > E_c, ΔE. In our case, the current is produced by free charge carriers — quasiparticles which are π-electrons moving in the field of the crystalline lattice. The nonlinearity of the I − V characteristic appears due to the nonlinear properties of the quasiparticle gas. We investigate the negative differential conductivity (NDC) dI/dV < 0 in CNs in a certain range of the field strength, predicted in [2].

Let us consider an undoped single-wall achiral CN (m,0) or (m,m) exposed to a homogeneous axial dc-field E_z, E_z = V/L, where V is the voltage between the CN ends. We shall apply the semi-classical approximation considering the motion of π-electrons as a classical motion of free quasi-particles with dispersion law extracted from the quantum theory. With the account to the hexagonal crystalline structure of CNs, the tight-binding approximation gives [3]:

\[ E_s(p_z) = \pm \gamma_0 \left[ 1 + 4 \cos(2\pi s) \cos \left( \frac{a}{\sqrt{3}} s \Delta p_\phi \right) + 4 \cos^2 \left( \frac{a}{\sqrt{3}} s \Delta p_\phi \right) \right]^{1/2} \] (1)

for the zigzag CNs and

\[ E_s(p_z) = \pm \gamma_0 \left[ 1 + 4 \cos(\pi s) \cos \left( \frac{a}{\sqrt{3}} p_z \right) + 4 \cos^2 \left( \frac{a}{\sqrt{3}} p_z \right) \right]^{1/2} \] (2)

for the armchair CNs. Here \( \gamma_0 \sim 3.0 \text{ eV} \) is the overlapping integral, \( a = 3b/2h, b = 1.42 \text{ Å} \) is the C-C bond length. The − and + signs correspond to the valence and conduction bands, respectively. In view of the transverse quantization of the quasi-momentum, its transverse component can take m discrete values, \( p_\phi = s \Delta p_\phi = \pi \sqrt{3} s a m \) (s = 1, ..., m) and we used \( E(s \Delta p_\phi, p_z) = E_s(p_z) \) in both equations. As different from \( p_\phi \), we assume \( p_z \)
continuously varying within the range $0 \leq p_z \leq 2\pi/a$ what corresponds to the model of infinitely long CN ($L = \infty$). This model is applicable to the case under consideration because we are restricted to temperatures and/or voltages well above the level spacing [4].

The motion of quasi-particles in an external axial electric dc-field is described by the Boltzmann kinetic equation

$$eE_z \frac{\partial f(p)}{\partial p_z} = -\frac{1}{\tau} [f(p) - F(p)],$$

where $e$ is the electron charge, $F(p)$ is the equilibrium Fermi distribution function and $\tau$ is the relaxation time. The relaxation term of the equation (3) describes the electron–phonon scattering [5, 6], electron–electron collisions etc.

Utilizing the method originally developed in the theory of quantum semiconductor superlattices we can construct an exact solution of kinetic equation (3) without assuming the electric field to be weak [2].

Then in view of Eqs. (1)–(3) one can obtain [2]

$$j_z(E_z) = -\frac{8e\gamma_0}{\sqrt{3}\hbar m} \sum_{r=1}^{\infty} \frac{r^2 \Omega \tau}{1 + (r \Omega \tau)^2} \sum_{s=1}^{m} F_{rs} E_{rs}.$$

(4)

Here $\Omega = ae E_z$ for the zigzag CNs and $\Omega = ae E_z/\sqrt{3}$ for the armchair ones is the Stark frequency. This equation states the basis for the evaluation of $I–V$ characteristics. Let us estimate the restrictions to the theoretical approach being developed. As it has been stated above, the quasi-particles motion is described classically by Boltzmann equation (3). It imposes the limitation on the external electric field strength: $|E_z| < \gamma_0/2eR$ [2]. The Coulomb electron–electron interaction has been also left out of account in our analysis. The role of this mechanism as applied to CNs was discussed in [2].

The $I–V$ characteristics obtained via numerical calculation of Eq. (4) are presented in the right panel of Fig. 1 for the metal zigzag CNs and in the left one for the armchair CNs ($m \neq 3\eta$) zigzag CNs. The figures show the linear dependence of $j_z$ on $E_z$ at weak strengths of the external field; it corresponds to the region of ohmic conductivity. As $E_z$ increases, the value $\partial j_z/\partial E_z$ growth smaller and at $E_z = E_z^{(max)}$ the current density reaches the maximum value $j_z^{(max)}$. Further increase of $E_z$ results in the decrease of $j_z$. At small $m$ the magnitudes of $E_z^{(max)}$ and $j_z^{(max)}$ for the armchair CNs turn out to be less than for zigzag CNs with similar radius by the factor approximately 2. As different from the zigzag CNs, the increase of $m$ leads to the $E_z^{(max)}$ growing up, and $I–V$ curves for zigzag and armchair CNs become practically identical at $R > 20$ nm. Thus, we predict the region with the negative differential conductivity $\partial j_z/\partial E_z < 0$, in the $I–V$ characteristics of CNs.

The external field strength $E_z^{(max)} \approx 3.2 \cdot 10^3$ V/cm for the NDC region appears to be unexpectedly weak because the nonlinearity in the structure under consideration is determined by the value of $ae E_z$: quantum superlattices with periods about $10^{-6}$ cm [8], much larger than $b$, show approximately the same fields for the NDC manifestation. It means that the nonlinearity in CNs is much higher than in superlattices. To explain this fact let us compare the mechanisms of the nonlinear conductivity in CNs and in the superlattices. The quantum superlattices are formed by alternating plain layers of different semiconducting materials, while the lattered superlattices consist of 1D chains of identical and identically coupled GaAs/AlGaAs quantum dots [8]. Both these structures are characterized by the dispersion law $E(p) = \Delta[1 - \cos(\tilde{a} p_x)]$, with $\Delta$ as the overlapping integral and $\tilde{a} = 2a/3$. 
Applying the method described above to this dispersion law, one can obtain the Esaki–Tsu relation \[ j_z(E_z) = \frac{\sigma_{zz} E_z}{(1 + U \Omega^2 I^2 \tau^2)} \] instead of Eq. (4). Here \( \sigma_{zz} = \lim_{E_z \rightarrow 0} \left( \frac{\partial j_z}{\partial E_z} \right) \) is the linear conductivity. Comparing two equations for the current density we can conclude that the specific peculiarity of the CNs is the presence of the high Stark components (summation with respect to \( r \) in Eq. (4)).

It has been shown that the electron motion in dc-field can be described as the oscillations of an ensembles of effective harmonic oscillators (Stark components) with the frequencies \( r \Omega \). The full current is a superposition of partial currents of the Stark components. Their electrical field strength corresponding to maximum current of \( r \)-th Stark component decreases with \( r \) as \( r^{-1} \) while our calculations shows that oscillator strength of these component decreases slowly. This is due to the hexagonal crystalline structure of CNs reflected in dispersion laws (1) and (2). The number the unneglectable components is 70–150 for metal and 200–300 for semiconducting CNs. As a result, the role of the high Stark components in CNs is essential and the integral nonlinearity of the CNs is much higher than in superlattices [8].

Note that the NDC provides the current instability. It can be expected that simultaneously applied dc- and ac-fields will result in the dynamic electron localization (which is the nonlinear phase of the instability) and in the 2D analogue of the self-induced transparency, like it takes a place in the semiconducting superlattices. The above mentioned effects are responsible for the absolute negative conductivity which thus is predicted to be exhibited in CNs. It must result in the appearance of the absolute negative conductivity zones and active properties of CNs providing a potentiality for the design of generative nanodiodes in microwave and infrared ranges. Such a possibility relates to both single CNs and CN ropes.

The NDC effect was also predicted in Ref. [9] for the CN-junctions generated in the result of different ways of doping the sides of the tubes or via the applying an external potential difference between them. The comparison of physical mechanisms of the NDC in [9] and [2] is actual. In [9] the electric field is localized in the narrow region of the junction while in [2] it is homogenously allocated along the CN. That is why the field strength in [9] is much higher than under the same potential. In [9] the NDC was obtained for
the junctions based on metal CNs only, while [2] also contains the prediction of the NDC in semiconducting CNs, the effect being much weaker for them. The value of $E_z^{(\text{max})}$ for armchair CNs with small radius is approximately half as great than that for metal zigzag CNs with similar radius. This result was obtained in both papers. Thus one can come to a conclusion that physical mechanisms in [9] and [2] are close. However, it is necessary to note that the $I-V$ curves in [9], as different from [2], are unsymmetric with respect to the transformation $V \rightarrow -V$.

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References

Charge separation in scrolled heterostructures

V. M. Osadchii and V. Ya. Prinz
Institute of Semiconductor Physics, 630090, Novosibirsk, Russia

Abstract. We calculate strains in the wall of semiconductor InAs/GaAs nanotube and their effects on the band edges. We show that the maxima of the electron and hole wave functions are shifted in opposite directions from the middle of the tube wall.

Recently, self-formed semiconductor nanotubes have been fabricated, and the possibility of precise control over their parameters has been shown [1, 2]. In the fabrication process, highly strained InGaAs/GaAs heterolayers are used, which, after being debonded from substrate (by selective etching of an underlying AlAs sacrificial layer), roll up in scrolls. The rolled up layers in the resultant tube were shown to perfectly stick together, thus forming a monocrystalline tube wall. It is the electrical and optical properties of these structures that are of primary interest.

Properties of a 2D electron gas on a cylindrical surface were theoretically considered in [3–5]. However, in addition to pure bending, notable strains take place in real structures, which should also be factors affecting the energy spectrum of electrons in nanotubes. In this work, within a simple model, strains in the wall of a nanotube scrolled up from a GaAs/InAs bilayer film MBE-grown on an InP substrate are calculated, as well as energy levels and wave functions of charge carriers.

The heterostructure GaAs/InAs is strained because of the mismatch of lattice parameters of two layers $f = \Delta a_i/a \left(\approx 7\%\right)$, the GaAs and InAs layers being in tension and in compression, respectively. After the bilayer film is freed from bonding with substrate, the elastic stresses are relaxed, and the film scrolls up in a tube. Provided that the layers in the tube stick together in a coherent manner, as their number increases, the outer layers of the tube becomes more and more stretched, while the tension in the inner layers also grows in value. Figure 1(a) shows the structure under consideration. Here, we assume that the tube is axially symmetrical, its elastic constants do not depend on crystallographic direction, the initial film is grown on a (100) substrate, and the axis of the tube is directed along the[010] axis. Under these conditions, only diagonal components of the strain tensor appear to be non-zero, the axial, azimuth, and radial strains being designated as $\varepsilon_z$, $\varepsilon_\theta$, and $\varepsilon_r$. Inner radius $R$ of tube is determined as a curvature radius of bilayer film [6]. We calculate the strains using the continuum elasticity theory, since this theory proved to be applicable at least to films as thick as 3 ML [7].

The axial strain $\varepsilon_z$ can be calculated as follows. It is apparent that the lattice parameter $a_z$ in the direction along the tube axis is equal to that in a free-standing planar superlattice with two alternate layers [8]:

$$a_z = \frac{a_1 G_1 h_1 + a_2 G_2 h_2}{G_1 h_1 + G_2 h_2},$$

where, for each layer, $h_i$ is the layer thickness ($i = 1, 2$), $G_i = E_i/(1 - \nu_i)$, $E_i$ is the Young modulus, $\nu_i$ is the Poisson ratio, and $a_i$ is the lattice parameter. The difference in the lattice
Fig. 1. (a) Schematic view of nanotube. Direction \((z, \theta, r)\) for the calculation of strain tensor components are shown. \(R\) is inside radius of tube. (b) Azimuth \(\varepsilon_\theta\) and axial \(\varepsilon_z\) components of strain tensor across the tube wall (4 bilayers 3 ML GaAs/3 ML InAs).

The parameter of the bulk material and strained structure determines the strain \(\varepsilon_z = (a_i - a_z)/a_z\) (Fig. 1(b), curve 1). The calculations are performed for the 4 turn tube with thicknesses of InAs and GaAs layers of 3 ML. For GaAs \(a = 0.5653\) nm, \(E = 85.5\) GPa, and \(\nu = 0.31\), while for InAs \(a = 0.6058\) nm, \(E = 51.4\) GPa, and \(\nu = 0.35\).

In calculating the azimuth strain, we assumed that the neutral surface (the boundary between the layers in tension and in compression) passes inside the tube wall. The lattice parameter \(a_\theta\) in the azimuth direction equals \(a_z\); under this condition, as our estimates showed, the energy of the elastic deformation is nearly minimal. It is apparent that, for a single-crystal tube, \(a_\theta\) varies linearly in the radial direction (Fig. 1(b), curve 2). From the known strains in two directions, the strain in the radial direction can be readily found from the Poisson relation \(\varepsilon_r = -(\nu/(1-\nu)) (\varepsilon_\theta + \varepsilon_z)\).

The strain shift band edges in a semiconductor [8]: the shift of the conduction band is \(\Delta E_c = a_c Tr(e)\) and that of the valence band is \(\Delta E_{hh,h} = a_v Tr(e) \pm b \left(\varepsilon_r - \frac{\varepsilon_\theta + \varepsilon_z}{2}\right)\) (here the upper and lower signs correspond to the band of heavy and light holes, respectively), where \(Tr(e) = \varepsilon_r + \varepsilon_\theta + \varepsilon_z\), and \(a_c\), \(a_v\), and \(b\) are the deformation potentials. In GaAs \(a_c = -7.17\) eV, \(a_v = 1.16\) eV, \(b = -1.6\) eV, and in InAs \(a_c = -5.08\) eV, \(a_v = 1\) eV, \(b = -1.6\) eV. Since the off-diagonal components of the strain tensor equal zero, the piezoelectric field equals zero too [9]. The calculated profiles of the band edges (those of the condition band and the valence band for heavy holes) across the tube wall in the non-doped structure are shown in Fig. 2(a) (solid curves). Here, the fact that the bandgaps in GaAs and in InAs are \(E_g = 1.424\) and 0.355 eV and the electron affinities are \(\chi = 4.07\) and 4.9 eV, respectively, is taken into account. As is seen, there is a modulation of the bandgap in the structure, the bandgap in each layer and the edges of both bands decreasing toward the outer surface of the tube wall. It should be noted that the near-surface bending of energy bands, in view of its being small in non-doped structures, was ignored.

From the known spatial profile of band edges, we can find the energy spectrum and wave functions of charge carriers in the structure of interest. We restrict our consideration to the ground state for electron and holes. The Schrödinger equation in the effective-mass
approximation for the radial part of the wave function of the ground state $\Psi$ is

$$-\frac{\hbar^2}{2m^*} \frac{1}{r} \frac{d}{dr} \left( r \frac{d\Psi}{dr} \right) + U\Psi = E_0\Psi. \quad (1)$$

Here $m^*$ is the effective electron (hole) mass and $U$ is the potential energy (edges of the corresponding band). The electron mass was assumed to equal $m^* = 0.067m_0$ and $0.023m_0$ in GaAs and in InAs, respectively, where $m_0$ is the free electron mass, the hole mass in both materials being equal to $0.4m_0$. The single-band effective mass approximation is known to be capable of adequately reproducing the energy levels for electron and holes in their ground states [10], whereas, for the excited states of holes, mixing of states of heavy and light holes should be necessarily taken into account. The adopted boundary conditions for the wave function were its zero value at the tube-wall surfaces and the continuity of $U\Psi$ and $1/m^* \frac{d\Psi}{dr}$ at heterointerfaces.

Since, in the Schrödinger equation, the potential energy for the structure of interest is a piecewise continuous function of the coordinate, the solution of the equation in this case cannot be expressed in terms of analytical functions and should be found numerically. In Figure 2(a), the dotted curves show the calculated energy levels of electrons and holes. Here, the electron level for the state in the $\Gamma$-minimum is shown. The effective masses in the subsidiary (L and X) minima are greater than that in the $\Gamma$-minimum, and the spatial-quantization levels here rise more slowly with decreasing dimensions of the well. However, calculation shows that the $\Gamma$-valley electron state is the ground one for the tubes with InAs layers being thicker than one monolayer, which is also the case for the free-standing GaAs/InAs superlattices [11].

Figure 2(b) shows the calculated wave functions for electrons and holes. An appreciable spatial redistribution of holes and electrons is seen to take place, which could be expected beforehand from the profiles of bend edges (Fig. 2(a)). As calculations showed, the spatial separation of charge carriers is observed in the cases where the number of rolls in the tube exceeds 2, and the degree of the charge redistribution increases with increasing number of rolls.

Now we shall dwell briefly on the magnitude of strain in tube walls (Fig. 1(b)). As is seen from the figure, the highest strain here far exceeds the strain attainable in pseudomorphic MBE-grown layers. It is possible to grow 4 ML GaAs and 4 ML InAs layers on an InP substrate, with the strain at the interface between layers being about 4%. In Fig. 1(b), the
two bilayers in the middle of the tube wall suffer a comparable strain. However, the strain in outer layers runs into 12%. At growth temperatures, such a strain would give rise to dislocations. However, in the case under study we have already formed pseudomorphic initial film, which roll up in the tube at room temperature, no dislocations being introduced here, which is confirmed by a HREM study [2] of similar tube.

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References

On depolarisation in 0D systems: Lamb-like level shift

Slava V. Rotkin
Ioffe Physico-Technical Institute, St Petersburg, Russia
Beckman Institute, UIUC, 405 N.Mathews, Urbana, IL 61801, USA

Abstract. An extremely quantized 0D system is studied for better understanding of an importance of many-body corrections to an one-electron spectrum. A new approach is proposed to describe the correction appearing as a depolarisation level shift of a (confined) electron moving within an electric field of a (confined) collective mode. For a quantum dot the correction diminishes at the size about $10^4 - 10^5$ atoms, depending also on other system parameters. For a closed-shell carbon cluster the effect does not depend on the cluster size owing to stronger quantization and the one-electron estimation does not fit anywhere. For the semiconductor quantum dot system an experimental method to check the depolarisation model is proposed.

In the paper we study an anomalous large level shift, resulted from the interaction of an electron in a 0D-system with zero-point oscillations of confined modes of the electric field. Of course, any complete many-body theory, taking into account all the Coulomb interaction, gives the correct value for the electron levels, though it is not known for present. We go to reveal an important correction treating the effect of valence electrons of a 0D object, which can be a spherical quantum dot and a closed-shell fullerene cluster, selfconsistently. Then the theory remains to be semi-classical while a nature of the effect is quantum-electrodynamical. This continues our consideration of $C_{60}$ in frame of a simple quantum mechanical model of the spherical-shell quantum well (SSQW) [1].

The energy correction depends on the system radius. This size scaling of the depolarisation is computed within an approach proposed by Migdal [2] for a calculation of Lamb shift in a hydrogen-like atom. The shift of the one-electron level is quite predictable and we will show that its amount becomes very large for the quantized system. The closed-shell fullerene depolarisation is of the order of the bare energy and independent of the cluster radius while the shift in the quantum dot decreases with the increasing number of atoms. Between two examples — a fullerene carbon nanocluster and a semiconductor quantum dot structure — the latter has not only theoretical importance. The possible experimental manifestation of the depolarisation effect is proposed basing on the spectroscopy of the quantum dot levels for different matrix materials.

1. Theory for depolarisation level shift: $C_{60}$

1. The use of the group of full rotations, SO(3), allows one to label the one-electron states and to get analytically the solution for the selfconsistent RPA response function of $C_{60}$ [1]. A peak of a collective excitation shows up in this spectrum, resulting from fast coherent oscillations of a total electron density of valence states. This surface density oscillation can be thought as a confined electrical field mode or the surface plasmon.

We have considered semiclassically the LS for an arbitrary shell object in [3], followed to Migdal [2]. The frequency of the zero-point fluctuations of the external field is much...
higher than the inverse period of the electron orbit \( \omega_p \gg \pi/\tau \). Therefore, the adiabatic approximation has to be used and one divides the fast (field) and slow (electron) variables. An electron is subjected to short fast deflections from its original orbit in the high-frequency field of the electromagnetic wave of the zero-point fluctuation. Then the energy shift is given by the second order perturbation theory as

\[
\delta E = \langle H(r + \delta) - H(r) \rangle = \left\langle \nabla H \cdot \delta + \frac{1}{2} \nabla^2 H \cdot \delta \cdot \delta + \ldots \right\rangle = \frac{1}{4} \nabla^2 H \cdot \delta^2 + o(\delta^2) \tag{1}
\]

where \( H(r) \) is the unperturbed Hamiltonian and \( H(r + \delta) \) is the Hamiltonian with account for the random electron deflection \( \delta \). The angle brackets represent the quantum mechanical average over the fast variables of the field (or, the same, over the random electron deflections). The perturbed Hamiltonian is expanded in series on the \( \delta \) and a first nonzero contribution is taken.

The expression for the mean square of the deflection, \( \delta^2 \) was deduced in Ref. [3]. Though the estimation is semiquantitative, the deflection is of the order of atomic unit, \( a_B \approx 0.53 \text{ Å} \). The \( \delta^2 \) in the SSQW does not depend on the radius, neither on the number of atoms because of the density of the valence electrons is the same.

Let suppose that one-electron model works for some cluster \( C_N \). The one-electron Hamiltonian reads as [1]:

\[
H_o = E_n + \hbar^2 / 2mR^2 \hat{L}^2, \tag{2}
\]

where \( E_n \) is the energy of a lowest level of \( n \)-th radial series; an orbital quantization energy \( \hbar^2 / mR^2 \) defines the SO(3) level spacing between eigenstates of the angular momentum operator. The SSQW level shift reads as follows:

\[
E_L = E_{L}^{(0)} \left( 1 + \kappa \hat{L}^2 / N \right), \tag{3}
\]

where \( \kappa \sim 0.36 \) is the numerical coefficient depending only on \( b \), the carbon bond length: \( \kappa = \sqrt{a_B / 2\pi^2} / 2^{2/3} \).

2. Within the closed-shell model the optical gap occurs between the levels \( |L_F \rangle \) and \( |L_F + 1 \rangle \). Within the closed-shell approximation the Fermi momentum fulfills the condition \( N = 2 \sum_{L=0}^{L_{F}} (2L + 1) = 2(L_F + 1)^2 \). The gap value does depend on the cluster size, decreasing to the zero as \( N \) going to infinity in order to approach the gapless graphite.

The depolarisation makes the gap wider. The renormalisation is universal for any closed-shell spherical cluster and amounts about 40% to the bare value:

\[
E_g = E_g^{(0)} (1 + \kappa) \approx 1.36E_g^{(0)}. \tag{4}
\]

where the parameter \( \kappa \approx 0.36 \) is the same as before.

2. **Depolarisation energy level shift in QD structure**

1. In order to evaluate the LS for the quantum dot (QD) the simplified spherical model in frame of an effective mass approximation was applied. The size scaling of the depolarisation shift is not sensitive to the model used, being dependent mainly on the corresponding density of states of the collective modes.

The simplest QD Hamiltonian is considered to have only the rotational correction which reads as:

\[
\delta H = \frac{\hat{L}^2}{2mR^2} \left( -2 \frac{\delta}{R} + \frac{3}{2} \frac{\delta^2}{R^2} + \ldots \right) \tag{5}
\]
where $R$ is about the spherical QD radius; $m$ is the electron mass which is supposed to be constant within the dot; $\hat{L}$ is the angular momentum operator.

Nearly self-evidently the bulk plasmon shift is negligible. The small factor, contained in the 3D LS, comes essentially from the expression for $\overline{\delta^2}$ which scales as $1/N$ [3], where $N$ is the number of atoms in the QD. The mean square deflection, caused by the 3D mode (which is not confined at all), decreases with $N$ too rapidly.

The square of the deflection [3] $\overline{\delta^2} = e^2 / 4m^2 \int d^3 k \ E_k^2 / \omega_k^4$ is proportional to the square of the electric field strength. The field strength can be rewritten as the zero-point oscillation frequency $E_k^2 = 2\pi \hbar \omega_k$ through the quantized field normalisation. The 3D plasmon frequency does not depend on the quantum number $k$. Hence, the mean square deflection contains the total number of states effecting on the electron level in the QD. The integral is limited above by $k_{\text{max}} \sim 1/R$. In 3D-case it brings the factor $R^{-3} \sim N^{-1}$ claimed in the beginning of the section. Then the depolarisation level shift due to 3D modes scales with $N$ as follows:

$$\Delta_{3D} = \frac{\delta E}{E_{00}} \propto N^{-5/3}. \quad (6)$$

The rude estimation of the prefactor shows that even for the small QD with $N = 100$ the shift is $10^{-6}$ of the bare energy and will not be resolved because of a number of other different factors effecting the level position.

To give a complete picture, the standard LS due to the zero-point oscillations of the free electromagnetic modes of the vacuum reads as follows: $\Delta_{\text{vac}} \propto \alpha^3 N^{-2/3}$, where $\alpha \simeq 1/137$ is the fine structure constant. Though the slope of the LS in $N$ is much slower than in Eq. (6) the prefactor is tiny because of $\alpha^3$.

2. Two possible candidates for the confined plasmon modes in the QD system are the 2D plasmon and the 0D spherical mode. The former mode can arise because of some interface possibly grown within the structure (see inset in Fig. 1). It might be a wetting layer, if it is thick enough to confine the electromagnetic field. The 2D plasmon naturally originates at the boundary between the semiconductor structure and a distinct substrate. The scaling in $N$ will have a lower exponent that reflects the different density of the confined field (plasmon) states: $\Delta_{2D} \propto N^{-7/6}$. The shift depends on the inverse size nearly linearly. However, the prefactor dominates at some moderate size of the QD and lessens the LS to $10^{-3}$ for $N = 100$. The depolarisation is still to be too small to expect experimental consequences.

3. The $\overline{\delta^2}$ considered above the less, the larger the QD size, that is not the case for the deflection due to completely localized modes like in Sec. 1. In this section the localized modes are the surface plasmons of the spherical inclusion (with the dielectric function $\epsilon_1$) in the matrix (with the different dielectric function $\epsilon_2$). $\Delta \propto N^{-2/3}$. Our estimation shows that the level correction, becoming of the order of 50%, plays the important role for the QD of 100 atoms and smaller. We collected all studied contributions to the depolarisation LS and plot them in the log–log scale versus the QD size in Fig. 1.

The depolarisation because of the localized surface QD modes is large enough to propose an experiment supporting our model. It is easy to check that $\overline{\delta^2} \sim \omega_k^{-3}$, which is nearly the frequency of the bulk plasmon in the matrix (with the weak dependence on the mode angular momentum, see [3]). Therefore, changing the optical properties of the matrix surrounding the QD, one shifts the levels. If the bare energy level lies deep in the potential well, its position is nearly independent of the well depth which changes along with the matrix parameters. The deep bare level energy depends only on the well width. Hence, the depolarisation LS is distinguishable from the standard space quantization LS.
In summary, the effect of the zero-point oscillations of the free and confined electromagnetic field on the level of the confined electron in the 0D-system, as the closed-shell carbon cluster and the spherical QD, is studied. The depolarisation due to an interaction with the zero-point fluctuations shifts up the bare one-electron state. The gap renormalisation, which follows from the angular momentum dependent LS in the fullerene cluster, is shown to be independent of the fullerene radius. The size dependence of LS in the QDs is different for 4 modes considered in the paper. Although, in general, the depolarisation decreases with the QD size, the localized surface electromagnetic mode results in the essential level shift and is to be possibly resolved experimentally for a QD made from some hundreds atoms. Another method to detect the effect could be a measurement of a deep level position of the similar QDs buried by the substrates with distinct optical characteristics.

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References

Size quantization and excited states of associated and isolated InAs quantum dots

V.G. Talalaev†, B. V. Novikov†, S. Yu. Verbin†, Dinh Son Thach†, G. Gobsch‡, R. Goldhahn‡, N. Stein‡, A. Golombek‡, J. W. Tomm§, A. Maassdorf§, G. E. Cirlin¶, V. N. Petrov¶ and V. M. Ustinov¶

† Institute of Physics, St Petersburg State University, 198904 St Petersburg, Russia
‡ Institut für Physik, Technische Universität Ilmenau, D-98684 Ilmenau, Germany
§ Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, D-12489 Berlin, Germany
¶ Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Spectra of photoluminescence (PL) including its decay time spectra and spectra of PL excitation (PLE) of heteroepitaxial structures with quantum dots (QD) InAs/GaAs have been studied. Structures had been grown by submonolayer migration-enhanced epitaxy (SMEE) mode on vicinal substrates GaAs at deposited InAs thickness close to critical (1.8 monolayer (ML)). It has been shown that PL structure under study is formed by the recombination emission of different QD families. One family consists of associated QD groups confined by vicinal terraces discretely broadened due to step bunching effect, another family — of isolated QD separated from rest QD array due to wetting layer (WL) ruptures at terrace edges. Excited exciton states of various QD groups have been detected. Their particular features are determined depending on the temperature, power and wavelength of photoexcitation.

Introduction

Additional structure of QD PL spectra is mainly formed by two mechanisms: the excited states recombination and QD size quantization of different origins. However these mechanisms have not been identified. Their evidence in PL spectra of InAs QD grown on vicinal substrates GaAs(100) has been studied here.

1. QD size quantization. Isolated QD

Two-dimensional QD arrays were grown by the SMEE mode on vicinal GaAs substrates at low temperatures (470°C) and growth rates (0.1 ML/s) from InAs layer with thickness close to critical (1.8 ML). For these QD arrays it has been established that at low excitation density (up to 50 W/cm²) the structure of low-temperature PL spectra is formed by QD groups with different sizes. Some PL components resulting from such size quantization (A₀ and A₁ bands in Fig. 1) has been observed in PL spectra of relatively high misorientation samples (5 and 7 degrees).

PLE spectra study has determined that A₀ and A₁ QD are associated with WL of InAs (Fig. 2). As the temperature grows up to 200 K the bands get narrower and shift in the long wave direction faster than InAs $E_g$. It corresponds to the model of thermal redistribution of carriers to larger QD of this group by transfer via WL.
Intensive short-wavelength band $I_1$ (Fig. 1) of PL spectrum can be attributed to the isolated QD group [1], i.e. nanoislands separated from rest array of QD due to WL ruptures at terrace edges. It has been shown that:

1. band $I_1$ is excited only by carriers from GaAs barrier (Fig. 2);
2. its red shift corresponds to the temperature dependence of InAs $E_g$;
3. as temperature rises up to 200 K the $I_1$ band broadens in contrast with A-bands [1].

Signs and values of thermal rates of A- and $I_1$-bands spectral shifts and halfwidths have been compared in the ranges 5–200 and 200–300 K. It has been concluded that thermal hopping of carriers from shallow isolated QD is not accompanied by their recapture by larger QD. It can be possible if transfer for associated QD takes place via WL and for isolated ones — via GaAs barrier.

The further study has proved that isolated QD are typical not only of vicinal surfaces where they can dominate. WL absence has also been observed at QD growth by SMEE on singular (oriented) GaAs surface. For example, the change of conventional MBE to SMEE
led to the PL band blue shift equal to 55 meV. It corresponds to transition from InAs WL to GaAs barrier.

2. Excited states of associated and isolated QD

A basically different excited state spectral detection of associated and isolated QD has been performed.

Excited states of isolated QD have been observed in PL spectra only at higher (more than 50 W/cm²) density of above-barrier excitation. Excited state J₁ of isolated QD is higher by 35 meV than ground state I₁ and at high density of excitation (more than 100 kW/cm²) J₁ dominates in PL spectra of vicinal InAs/GaAs (100) samples with 5°- and 7°-misorientation in [001] and [010] directions.

Excited states of associated QD were observed only at the below-barrier WL excitation (Fig. 3). The discrete inhomogeneity of WL thickness and the corresponding fine structure of B₀ and B₁ excited states have also been observed in this study. For example, in InAs/GaAs 7°[001] sample the state B₀ is higher by 32 meV than ground state A₀ and B₁ — by 20 meV than A₁ (Fig. 3).

It is evident that observed behavior of excited states in associated QD is the result of pyramidal QD wave function localization near the pyramid base [2], i.e. near WL. Then the high density of excitation is not necessary for the observation of B-bands in contrast to J₁ because the density of non-equilibrium carriers in two-dimensional WL is much higher than the surface density of carriers at the excitation of bulk GaAs barrier.

It had been shown earlier [1] that excited states of all QD groups are implicitly evident in the temperature dependence of PL band integral intensity as the deviation of experimental curve from Arrhenius plot. In this way we obtained values of energy gap ΔE between ground and excited states for the InAs/GaAs 7°[001] sample: 30 meV (A₀–B₀), 26 meV (A₁–B₁) and 35 meV for I₁–J₁.

PLE spectra of this sample are shown in Fig. 4. Observed excitation maxima B₀ and B₁ are shifted against A₀ and A₁ maxima bands by 30 and 23 meV. We have defined B₀
and B₁ peaks as excited states of corresponding associated QD groups.

The parity of energy gaps ΔE in PL and PLE spectra testifies that in QD PL we encounter the radiative transfer between the excited state of one carrier (electron) and the ground state of another one (hole).

Excited state of isolated QD is absent in the PLE spectra (Fig. 4) due to low excitation density. Besides, the energy gap ΔE of isolated QD at 7°[001] is equal to LO-phonon energy at InAs-GaAs interface (35 meV [3]). It may be expected that due to the localization of the excited state wave functions at the same interface this resonance leads to a very efficient relaxation of electronic excitation to ground state, i.e. to a very short lifetime of electrons in excited state. The measurement of spectral dependence of PL decay time has shown that at the energy of excited state of isolated QD the fast component of decay curve has evident minimum corresponding to 55 ps.

At the below-barrier excitation the B₀ and B₁ excited state spectrum has fine structure in contrast to the ground state under the same conditions (Fig. 3). It indicates a higher selective sensitivity of excited states to the WL arrangement. The thermal sensitivity of associated QD fine structure and temperature dependence of isolated QD PL in the 5–60 K range is discussed.

Acknowledgments

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References


Magnetooptical studies in CdSe/(Zn,Mn)Se semimagnetic nanostructures

A. A. Toropov†, S. V. Sorokin†, K. A. Kuritsyn†, S. V. Ivanov†, P. S. Kop’ev†, G. Reuscher‡, A. Waag‡, M. Wagner‡, W. M. Chen§ and B. Monemar§

† Ioffe Physico-Technical Institute, St. Petersburg, Russia
‡ Physikalisches Institut der Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany
§ University of Linköping, S-581 83 Linköping, Sweden

Recently diluted magnetic semiconductor (DMS) heterostructures have received increasing attention due to the potential optoelectronic and magnetoelectronic applications based on spin-polarized transport in semiconductors [1]. In this context, the studies of electronic spin behavior in 0D structures with a discretized electronic system are of particular interest. An excellent opportunity for studying the spin-related phenomena in DMS nanostructures is provided by recently developed molecular beam epitaxy (MBE) technique allowing growth of Zn(Cd)Se-based quantum structures with varying dimensionality, from quantum wells (QWs) to quantum disks and dots [2–5]. To produce the zero-dimensional (0D) DMS structure one can incorporate Mn ions either into the quantum objects (disks or dots) or into their surroundings. The preliminary data on magnetooptics in the quantum dot (QD) structures of this type have recently been reported by Kim et al. [6].

In this paper we report on magneto-PL studies of CdSe/(Zn,Mn)Se nanostructures, focusing on the effects of Zeeman splitting and spin polarization of strongly localized 0D excitons. Three samples were grown for this purpose by MBE pseudomorphically on (001) GaAs substrates. The structures are non-magnetic CdSe single layers of nominal thickness \( w \), embedded in the center of a 20 nm thick DMS layer of Zn\(_{0.89}\)Mn\(_{0.11}\)Se. The whole structure is surrounded by thick ZnBeSe barriers lattice-matched to GaAs. The CdSe insertion was grown using migration enhanced epitaxy (MEE) technique [2], whereas conventional MBE was used to fabricate all other layers of the samples. A, B and C samples differ only in the CdSe nominal thickness, with \( w \) being equal to 0.4, 0.8 and 1.9 monolayer (ML), respectively. According to the data obtained previously for CdSe/ZnSe (see e.g. [4, 5]), the intrinsic morphology of the insertion depends drastically on \( w \), varying from the relatively homogeneous ZnCdSe graded-gap QW (\( w < 0.6 \)–0.8 ML) with the average thickness of 4–6 ML to laterally inhomogeneous layers containing flat ZnCdSe islands with enlarged Cd content at larger \( w \). The islands arise, most probably, due to strain-driven decomposition of the alloyed layer with large Cd content. Electronically they can be regarded as quantum disks providing 3D confinement for excitons [3]. Therefore, neglecting the possible effect of Mn atoms in the barriers on the ZnCdSe islands formation, one can assume that sample A represents a 2D QW exciton. Sample B is expected to present a QW system as well, perhaps suffering larger interface disordering. Sample C is an example of the inhomogeneous ensemble of 0D excitons localized in the ZnCdSe islands of different sizes and compositions. Magneto-PL measurements were carried out in Faraday geometry in a He cryostat with a split-coil superconducting magnet at 1.8 K, using a linearly-polarized emission of a cw Ar\(^{+}\) laser at 364 nm. The PL signal was dispersed by a double-Spex monochromator and detected by a GaAs photomultiplier tube. Either
Figure 1. PL spectra of samples A, B, and C at zero magnetic field.

The $\sigma^+$ or $\sigma^-$ component of the PL was selected by a quarter-\(\lambda\) plate coupled with a linear polarizer.

Figure 1 demonstrates zero-field PL spectra of the samples A, B, and C. An increase in \(w\) causes a red shift of the PL peak attributed to the emission of localized heavy-hole excitons in a CdSe insertion (the peaks are pointed in Fig. 1 by arrows). Another effect is the peak broadening, which reflects enhancement of the layer structural disordering in good agreement with the data on similar nonmagnetic CdSe/ZnSe structures [2]. Three additional emission bands are visible for all the samples at 2.875 eV, 2.69 eV, and 2.0 eV, being attributed to the near-band-edge excitonic emission in ZnBeSe claddings, DAP recombination in the barriers, and intra-shell transitions within Mn ions, respectively. Note that only position of the peaks related to the CdSe insertions depends on \(w\). Furthermore, this peak is the only feature in the spectra, depending on a magnetic field. Particularly, an increase in a magnetic field results in (i) Zeeman shift of the PL band, (ii) rapid increase in intensity of the $\sigma^+$ component, and (iii) large enhancement of the integral PL intensity. The latter observation was also reported by Kim et al. [6], who observed a field-induced increase in the integral intensity of exitonic emission from CdSe/(Zn,Mn)Se nanostructures, accompanied by quenching of the PL related to Mn inter-shell transitions. Owing to this observation, the authors of [6] explained this effect in terms of an efficient transfer of excitation between the excitons and the Mn ions, which is suppressed by a magnetic field. In our samples, we do not observe any field-induced changes of the Mn-related PL and, hence, the observed enhancement of the excitonic PL intensity implies some other mechanism of non-radiative recombination, quenched by a magnetic field. In our samples, we do not observe any field-induced changes of the Mn-related PL and, hence, the observed enhancement of the excitonic PL intensity implies some other mechanism of non-radiative recombination, quenched by a magnetic field. As it is seen from Fig. 2, the absolute value of the PL intensity decreases with \(w\) for any field in the range studied. The \(w\)-dependent behavior results probably from Mn ions clustering [7], which can be especially enhanced in the vicinity of the laterally inhomogeneous CdSe insertions. In this case, the centers of nonradiative recombination can be formed due to the locally enhanced concentration of Mn ions, with the cluster density and distribution depending on \(w\).

Figure 3 illustrates the Zeeman shifts of the PL peak maximum for all three samples. For samples A and B the dependencies are alike and generally consistent with the giant Zeeman splitting effect observed in DMS QWs (see e.g. [8]). For sample C the dependence is almost twice weaker and the curve shape is more complicated. It is well known that the magnitude of Zeeman splitting in the structures containing both DMS and nonmagnetic
layers depends on many factors. Particularly, the Zeeman splitting of a given state reflects its weighted probability distribution over the two media, which can be a function of the magnetic-field-dependent band offsets in the heterostructure. Another important factor is the structure of an interface between the DMS and non-DMS materials, because manganese ions near to the interface possess the larger magnetism relative to those in bulk material [7]. In regard to the QW structures, an increase in $w$ is expected to decrease Zeeman splitting due to simultaneous action of both the factors. Really, the larger content of Cd results in the deeper wells for confined carriers and, hence, in smaller penetration of the carrier wave function into the DMS barriers. Simultaneously, the probability to find an electron near the interface is smaller for a deeper QW, which should decrease the interface contribution.

However, very small difference between the curves in Fig. 3 for the QW samples (A and B) indicates negligible effect of $w$ on the relative probability to find electron within the regions with higher effective concentration of Mn ions. Most probably, this results from the effect of Zn(Cd)-Mn interdiffusion during the structure growth, which causes unintentional incorporation of Mn into the thin layer of otherwise non-DMS CdZnSe alloy. To contribute to understanding of the different behavior of the sample C, we plot in Figs. 4(a) and (b) (samples A and C, respectively) the spectral contours of the PL intensity and circular polarization rate for two values of a magnetic field. For the sample A, the PL line shape weakly depends on a magnetic field. The respective polarization-rate contour follows the PL line when the latter is shifted by a magnetic field. The shape of the contour depends on the field rather weakly as well. The difference concerns mainly the region of two visible LO-phonon replica of the PL band. In contrast to that, the PL line of the sample C changes its shape noticeably with a magnetic field increase. Furthermore, the respective polarization-rate contour does not follow the PL line, but rather changes its shape, expanding to the red side with a field increase. These observations for the sample C can be explained, considering inhomogeneous nature of the PL line involving responses from 0D excitons localized in different cites. However, they hardly can be attributed only to the energy dispersion among the sites, because no large field-induced changes of the contour shape are expected in this case. The analyses of the inhomogeneous contours at different fields indicate that the observed behavior is rather related to the dispersion of the excitons magnetic properties, particularly of their Zeeman splittings, which assumes inhomogeneous distribution of Mn ions in the vicinity of the CdSe-based islands.

In conclusion, the performed magneto-PL measurements exhibit formation of 0D excitons in CdSe/(Zn,Mn)Se nanostructures. The excitonic states forming the inhomogeneously broadened emission band differ both in their energy (due to fluctuations in the sizes and compositions among the localization sites) and in magnetic properties (most probably,
due to the effect of Mn ions clustering in the nearby regions). The structure potential for spin-related optoelectronic applications is demonstrated, owing to almost complete circular polarization of the emitted light within the wide spectral range (∼100 meV) at moderate magnetic fields.

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References

Spontaneous spin polarization in a quantum wire

V. V'yurkov and A. Vetrov
Institute of Physics and Technology, RAS
Nakhimovsky prosp. 34, Moscow, 117218, Russia
e-mail: vyurkov@mail.imvs.ru

Firstly the miraculous “0.7 structure” was seen in the experiments with quantum wires (QWRs) in 1996 [1]. There was registered the pronounced additional step of a quantum wire conductance quantization at the level 0.7 of a conductance quantum \( G_0 = \frac{2e^2}{\hbar} \). A bit later an apparent deviation from a conductance quantum \( G_0 = \frac{2e^2}{\hbar} \) was also observed in the most perfect for today long QWRs fabricated by cleaved overgrowth of GaAs/AlGaAs quantum well heterostructures [2]. Even a decrease up to 50% was registered for 20 \( \mu \) wire. Worth mentioning that the effect was present for multiple quantization steps visible in the experiment. Recently a novel observation of the 0.7 structure was made in quantum wires manufactured by split-gate technology [3]. The authors saw again an additional step of quantization at the level \( 0.7 \cdot \frac{2e^2}{\hbar} \). The fact that quite different structures revealed the same effect pointed out to its fundamental origin.

For a while there was no adequate explanation consistent with all available experimental data. Attempts to apply the spin and spinless Luttinger liquid theories seemed the most appropriate to unravel the problem. Indeed, these theories gave the corrections to QWR conductance caused by Coulomb interaction between electrons [4]:

\[
G = G_0 \left( 1 + \frac{V(0)}{\pi v_F} \right)^{-1/2}
\]

where \( V(0) \) represents the Fourier transform \( V(q) \) of the real space interaction potential between electrons for the transfer momentum \( q \) equal to zero, \( v_F \) is the Fermi velocity. However, these corrections have monotonic dependence on the Fermi energy that contradicts with abrupt transition to common integer steps of conductance quantum with rising a gate voltage observed in the experiment [3] and flat plateaus observed in [2].

When a disorder was involved [5] this also gave rise to an obvious decrease of the conductance but dependent on the electron density in the wire. The calculations fulfilled for realistic QWR wall roughness also revealed a strong dependence of scattering rate on the Fermi energy and subband number [6]. Thus such a scattering can not be at all a feasible reason of conductance deviation because it do not accord with observed flat plateaus (within 5%) and conductance steps of equal height [2].

Moreover, the latest experiments discovered an obvious connection of the “0.7 structure” with spin polarization of electrons in a QWR. They saw a smooth transition of the “0.7 structure” for zero magnetic field to the “0.5 structure” when a magnetic field was going up [3]. This experimental evidence crucially sustains the hypothesis of spontaneous spin polarization of electrons in a QWR was beforehand put forward in [7].

Here we argue that the strong deviation from conductance quantum is caused just by a spontaneous spin polarization due to exchange interaction between electrons in a QWR. As for Coulomb interaction, it can be put into consideration in an audible self-consistent way. However, a realistic Coulomb potential in a QWR should be taken into account [8]. Surely, the electrostatic potential induced by internal electrons in a QWR can even blockade

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the wire conductance. But we adhere to the experimental conditions when the wire was quite penetrable for electrons and electrostatic potential can not influence on the linear response to infinitesimal bias applied to the wire.

We assume that exchange energy is small compared with kinetic energy so that pair interaction between electrons is efficient approximation. Suppose that two electrons move across QWR in the same direction (left or right moving fields) with sufficiently small longitudinal momentum difference $\hbar \Delta k$ so that

$$h \Delta k < h/\lambda$$

where $\lambda$ is an effective screening length ($\lambda < L$) and $L$ is a wire length. These electrons possess exchange energy almost as great as Coulomb energy, i.e.

$$\left(\frac{e^2}{\kappa L}\right) \ln\left(\frac{\lambda}{d}\right).$$

Here $d$ is a QWR diameter. In our calculations the conventional Coulomb potential $V(x) = 1/\kappa x$ was cut off for distances $x$ smaller than $d$. For greater momentum mismatch exchange integrals involve fast oscillating functions and tend to zero. A sign of exchange energy depends on the spin configuration. If electrons have an antisymmetric spin configuration (total spin equals unity) then their space wave function is symmetric and the sign of exchange energy is positive, i.e. the same as that of Coulomb energy. Otherwise, when a total spin equals zero, the exchange energy is negative and reduces total energy of electron system.

It was found out that due to exchange interaction the ground state ($T = 0$) corresponding to the minimum of the total energy (including kinetic one) can be that of predominant symmetrical spin configuration for electrons near the Fermi level, i.e. spin polarized. The condition of the cross-over from conventional unpolarized state to polarized one is as follows

$$\frac{\pi a_B k_F}{4 \ln(\lambda/d)} < 1$$

where $k_F$ is the Fermi electron wave vector and $a_B = h^2/\kappa me^2$ is a Bohr radius. It should be noted that much higher Fermi energies obey the above condition than that obeying the condition of 1D Wigner crystallization derived in [12]. Although these conditions differ only by coefficients and arguments in logarithmic function this difference is essential. Once the condition (4) is met the polarized phase arises in the energy interval under the Fermi level

$$\delta \epsilon = \left(\frac{e^2}{\kappa \lambda}\right) \ln(\lambda/d).$$

The magnitude of $\delta \epsilon$ equals the exchange energy per one Fermi electron. Worth mentioning that it does not depend upon Fermi energy of electrons in any subband of a QWR. This is in a good qualitative agreement with the experimental evidence for conductance corrections to be insensitive to the Fermi energy [2].

In a polarized phase the density of states of electrons adjacent to the Fermi level much decreases although at the same moment energy corrections (consequently the corrections of Fermi velocity $v_F$) are rather small. We obtained the relative decrease of the electron density of states in the energy interval $\delta \epsilon$ under the Fermi level as follows:

$$\frac{1}{\pi a_B k_F} \ln(\lambda/d).$$

According to inequality (4) the decrease of density of states cannot be less than $1/4$ in spin polarized phase. The “diluted” density of states results in corresponding decrease of
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a conductance. However, as the parameter (6) is not small a non-perturbation approach should be developed to get a precise number.

Our estimations show that the condition of a cross-over (4) to polarized phase is valid even for top Fermi energies attained in the experiment [2] (unlike to that in [3]). We accepted for evaluations a screening length $\lambda$ equal to a several timed distance from the QWR to the nearest gate electrode and the wire diameter $d$ consistent with subband spacing (20 meV) pointed out there. Then we gained $\delta e$ exceeding $kT$ ($T$ about 1 K). When the temperature is rising the polarized phase is smeared and the conductance quantum restates. This explains the abnormal temperature dependence of the QWR conductance seen in the experiment. When the bias $V$ exceeds the value of $\delta e/e$ “undiluted” electrons, i.e. outside the polarized layer, are involved in the conductance and thus a conductance quantum restates too.

To be consistent with over-all experimental data [2] a wire length should be introduced in the theory. The experiment revealed a quite weak dependence of the conductance deviation on the wire length, at least, sub-linear one. The conductance deviation was only doubled while the wire length varied from 1 $\mu$m to 20 $\mu$m. Two possibilities look plausible. The first one is that in the experimental structure the wire diameter diminishes as wire lengths. An indirect allusion to this very dependence was that the less negative gate voltage pinched off a longer wire. The second possibility is an interaction of a wire with leads which partially ruins polarized phase in the pre-contact region. Although the presence of the leads was already modeled in [9–11] this consideration looks quite deficient yet.

In conclusion, an existence of predominant symmetrical spin configuration (spin polarized phase) and “diluted” density of states under the Fermi level in the quantum wire is considered. The reduction of quantum wire conductance is in agreement with recent experimental data.

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References

Spectroscopy of inhomogeneous strain in silicon-based quantum dots

A. Zaslavsky†, Jun Liu‡, B. R. Perkins† and L. B. Freund†
† Division of Engineering, Brown University, Providence, RI 02912, USA
‡ Dept. of Physics, Brown University, Providence, RI 02912, USA

Abstract. Resonant tunneling is employed to probe the inhomogeneous strain in silicon-based quantum dots. When submicron structures are etched from a $p$-Si/SiGe/Si double-barrier heterostructure, the resonant $I(V)$ peaks shift and develop a fine structure consistent with pronounced strain relaxation in the SiGe quantum well. We calculate the strain dependence on dot size by finite element techniques and convert the strain to an effective lateral confining potential. In sufficiently small dots, we find that the inhomogeneous strain confines carriers not only to the central core, as in GaAs-based dots, but also to a ring-like region at the perimeter. We probe the resulting density of states by magnetotunneling $I(V,B)$ measurements.

Introduction

When a semiconductor nanostructure, like a quantum wire or a quantum dot, is fabricated from strained epitaxially grown material, the originally homogeneous strain is replaced by geometry-specific strain gradients. The symmetry-based analytic treatment [1] for handling the biaxial strain in lattice-mismatched materials no longer suffices, particularly for quantum dots which lack translational symmetry altogether. Instead one turns to finite-element calculations of the strain field based on linear elastic models, but their applicability to structures whose size $D$ might be down to tens of lattice constants is not self-evident.

Thus, any experimental technique that is sensitive to inhomogeneous strains provides a valuable test bed for the validity of finite-element techniques for strained nanostructures—in our work, the experimental probe will be the resonant tunneling current-voltage $I(V)$ measurements.

Further, inhomogeneous strain in semiconductor nanostructures is taking on additional technological relevance, as advances in strained layer epitaxy and ongoing device miniaturization promise the arrival of deep submicron bandgap-engineered devices, such as strained Si/SiGe HBTs for high-frequency analog and digital applications. Particularly interesting is the use of strain-driven self-assembly of quantum dots in semiconductor lasers [2] to enhance gain and shift the lasing wavelength. In such devices, strain relaxation is the key to the size and morphology of the quantum dots, as well as the optical transition energies.

Finally, it is important to note that inhomogeneous strains in quantum nanostructures can contribute to carrier localization in unpredictable ways. Over the past decade, quantum dots have been extensively investigated as systems containing a few spatially confined charge carriers [3–5]. However, most of these experiments probed dots made from lattice-matched GaAs/AlGaAs heterostructures, in which the carriers are confined to the central region of the dot by a roughly parabolic lateral potential arising from the gate potential or the pinning of the Fermi level at the surface. In our strained Si/SiGe quantum dots, the inhomogeneous-strain-induced lateral confinement potential is nonmonotonic, leading to an effective potential minimum near the perimeter of the dot. For sufficiently small dots,
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Fig. 1. (a) SEM micrograph of a representative double-barrier nanostructure with lateral dimension $D \sim 0.15 \mu m$. (b) Self-consistent potential profile of the Si/SiGe double-barrier active region at $V = 150 \mu V$. (c) Resonant tunneling $I(V)$ of a large $D = 2 \mu m$ device at 1.7 K, with the HH peak magnified $\times 35$ for clarity.

this potential confines the ground state to a one-dimensional (1D) ring-like region near the perimeter — a new type of structure expected to have interesting magnetic properties.

1. Tunneling and strain in SiGe double-barrier microstructures

Our devices begin with $p$-$Si/Si_{1-x}Ge_x$/Si double-barrier heterostructures described in detail in previous publications [6, 7, 8]. They are grown on $p$-$Si$ substrates, with an undoped active region consisting of Si barriers confining a Si$_{1-x}$Ge$_x$ QW that is 35 Å wide with Ge content $x = 0.25$ or 0.2 (corresponding to a lattice mismatch of $\sim 1$ and $\sim 0.8\%$ respectively). Outside the barriers are $p$-$Si_{1-x}Ge_x$ emitter and collector regions that serve as reservoirs for tunneling holes.

When a bias $V$ is applied between the emitter and collector, the holes in the emitter tunnel via the quantized 2D hole subbands, subject to the usual energy $E$ and transverse momentum $k_\perp$ conservation rules [9]. In large devices, the strain in the SiGe well can be taken as biaxial and homogeneous, so the energies of the 2D subbands can be reliably calculated numerically [6]. Figure 1 shows an SEM photograph of a device, together with a self-consistent potential distribution in the active region under bias, and the $I(V)$ characteristic of a large $D = 2 \mu m$ Si/Si$_{0.75}$Ge$_{0.25}$ device at $T = 1.7 \text{ K}$. The peaks correspond to tunneling through the two confined 2D subbands, labeled HH and LH for the heavy-hole and light-hole branches of the dispersion. The agreement with the predicted peak positions is excellent [6, 7], so the peak positions reflect the energies of the quantized states in the SiGe well. In particular, the energy separation $\Delta E$ between the HH and LH subbands arises in part from the strain-induced splitting, which lifts the HH-LH degeneracy in the SiGe valence band [1, 10]. Thus, any significant change in the strain should be reflected in $\Delta E$. 
Fig. 2. $I(V)$ characteristics with nominal lateral diameters of $D = 1.0 - 0.25 \mu m$ at 1.7K. Current scale corresponds to the smallest device, other curves are rescaled and the HH peaks magnified for clarity. Vertical lines mark the HH and LH peak positions in large devices.

2. Average strain relaxation in SiGe double-barrier microstructures

When a narrow mesa is etched through the active region of our devices, the biaxially compressed SiGe strained layers can relax by lateral displacement at the sidewall. We first observed this effect experimentally some years ago [7] and it has been confirmed by others [11]. Figure 2 shows the effect in a series of identically fabricated Si/Si$_{0.75}$Ge$_{0.25}$ devices with $D$ ranging from 1 down to 0.25 $\mu$m. The $I(V)$ curves exhibit a consistent shift of HH and LH peaks towards each other as $D$ decreases, even as the $I(V)$ lineshape remains largely unaffected. The effect is large, with changes in $\Delta E$ indicating a significant amount of strain relaxation.

We compared the strain relaxation inferred from the data in Fig. 2 to finite-element simulations based on a linear elastic model, in which the cylindrical structure was allowed to relax to a minimum energy configuration [12]. The average values of strain relaxation predicted by these calculations agree quite well with our experimental data, with the dominant radial strain component $\epsilon_{rr}$ relaxing to $\sim 0.7$ of the full lattice-mismatch strain when $D$ falls to 0.3 $\mu$m. Interestingly, the simulated strain relaxation in the SiGe layers is non-monotonic in the radial direction $r_\perp$ and significant strain gradients exist throughout the SiGe well in sufficiently small structures, $D \leq 0.25 \mu m$. The inhomogeneous strain leads to additional lateral quantization in the confined 2D subbands, which we first observed in [8].

3. Inhomogeneous strain in SiGe quantum dots

The calculated radial strain $\epsilon_{rr}$ in the SiGe QW and the corresponding strain-induced lateral potential for HH states are shown in Fig. 3 as a function of $r_\perp$ for $D = 0.1 - 0.2 \mu m$ devices with a Si$_{0.8}$Ge$_{0.2}$ well. The inset shows the calculation geometry and lateral displacement of the strained SiGe layers. First consider the radial strain $\epsilon_{rr}(r_\perp)$ curves at the top of Fig. 3. For $D = 0.2 \mu m$, $\epsilon_{rr}$ decreases gradually with $r_\perp$ except for a region of increasing strain near the perimeter that extends $\sim 100 \AA$ and reaches $\epsilon_{rr} \sim 0.6$.

This strained ring-like region exists for all $D$ [8], [12], but for devices much larger than
Fig. 3. The top curves show the calculated radial strain component $\varepsilon_{rr}(r_\perp)$ for $D = 0.1-0.2 \mu m$ devices at the mid-plane of the Si$_{0.8}$Ge$_{0.2}$ well (full strain is $\varepsilon_{rr} = 1$). Inset shows the magnified displacement of the finite-element mesh near the sidewall. The bottom curves are the corresponding in-plane confining potentials for the HH states as function of $r_\perp$. Dashed lines mark the confined ring subbands in a $D \sim 0.15 \mu m$ dot.

$D = 0.2 \mu m$ it can be taken as a perturbation to the inner core. On the other hand, in small devices, $D \leq 0.15 \mu m$, the ring at the perimeter becomes the most highly strained region in the device. The corresponding confining potential for HH states is shown by the bottom set of curves in Fig. 3: in the smallest dots, the strain would confine holes to a 1D ring at the perimeter.

Figure 4 shows the HH $I(V)$ peak of a $D \sim 0.15 \mu m$ device fabricated from the Si$_{0.8}$Ge$_{0.2}$ double-barrier material, together with a reference lineshape from a large device. The $I(V)$ lineshape exhibits very strong fine structure, corresponding to strong lateral quantization due to inhomogeneous strain [8]. A full-blown calculation of the expected density of states in such a quantum dot in the presence of the inhomogeneous strain is complicated by the anisotropy and nonparabolicity of the in-plane effective mass in the quantum well, but taking the in-plane HH effective mass $m^* \sim 0.25$ we obtain several radially quantized subbands in the perimeter ring separated by a few meV, see Fig. 3. Since quantized ring-like subbands overlap in energy with the states in the relaxed central core, they would be expected to contribute structure on top of the relatively smooth overall HH peak lineshape. The energy separation of the ring subbands extracted from the structure in the $I(V)$ agrees reasonably with the calculations of Fig. 3.

4. Magnetotunneling spectroscopy of strained SiGe quantum dots

The additional ring-like confinement of hole states in sufficiently small inhomogeneously strained SiGe quantum dots provides a new and interesting system for magnetic field effects. Figure 5(a) shows the evolution of the HH $I(V)$ peak fine structure in magnetic fields $B = 0-10 T (B \parallel I)$, while Fig. 5(b) indicates the evolution of the $I(V)$ peaks converted to an energy scale. In the absence of inhomogeneous strain, the $B$ field would compress the 2D subband density of states into Landau levels. Given a constant in-plane effective mass, as in $n$-GaAs/AlGaAs double-barrier structures, the Landau level separation would
Fig. 4. HH resonant peak $I(V)$ and $dI/dV$ characteristics of a $D \sim 0.15 \mu m$ Si/Si$_{0.8}$Ge$_{0.2}$ dot. The dashed line shows the smooth HH $I(V)$ lineshape in a large device.

Fig. 5. (a) Evolution of HH $I(V, B)$ lineshape of the $D \sim 0.15 \mu m$ dot with $B$; (b) $B$-field-induced shifts and splittings in the density of states peaks in the dot.

be linear in $B$ and the $I(V, B)$ lineshape would show equally spaced features falling on the usual Landau fan diagram [13]. Given the complex, nonparabolic dispersion of holes in SiGe quantum wells, the Landau level spectrum is quite complicated even in a uniformly strained well [14] and $B$-induced structure has only been seen experimentally at fairly large $B$ [6]. The data in Fig. 5 shows rather complex behavior, with some of the peaks shifting towards lower energy with $B$, others appearing to repel each other, and some even splitting around $B \sim 5 T$. A theoretical analysis of ring-like hole states in a B field remains to be performed.

5. Conclusions

We have investigated the effects of size-induced strain relaxation in strained SiGe quantum dots. Our data indicate that large strain relaxation and nonmonotonic strain gradients appear in deep submicron structures, in agreement with finite element simulations. We also see evidence for confinement of carriers to ring-like regions at the perimeter due to inhom-
geneous strain. Our measurements prove resonant tunneling to be a viable spectroscopic probe for strain effects in individual nanostructures.

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References

Effect of alloying on growth of GeSi self-assembled islands

N. V. Vostokov†, S. A. Gusev†, Yu. N. Drozdov‡, Z. F. Krasil’nik†,
D. N. Lobanov†, L. D. Moldavskaya†, A. V. Novikov†, V. V. Postnikov†,
M. Miura‡, N. Usami‡ and Y. Shiraki‡
† Institute for Physics of Microstructures RAS,
603600, GSP-105 Nizhny Novgorod, Russia
‡ Research Center for Advanced Science and Technology (RCAST),
The University of Tokyo, Japan

Abstract. The effect of alloying on growth and parameters of GeSi self-assembled islands has been investigated at different growth temperatures by the atomic force microscopy and X-ray diffraction. The correlation between the sizes and composition of islands has been obtained. The Ostwald ripening and alloying significantly broaden the islands sizes distribution at growth temperature 750°C.

Introduction

Heterostructures of consisting GeSi strain-induced self-assembled islands provide a promising material to realise Si-based optoelectronic devices operating at 1.2–2 µm. This range is particularly important for telecommunication industry. High crystal quality of structures is critical to the success of such applications. In general, rather high growth temperatures (500–800°C) are used for fabrication of GeSi structures with good crystal quality. The diffusion and alloying process largely affect growth and parameters of structures at these temperatures. In case of islands growth the non-uniform elastic strain fields arise which result in additional flow of atoms [1].

In this paper, we present the results of investigation of self-assembled GeSi islands growth on Si (001) at various temperatures. The effect of alloying on the size and shape of GeSi self-assembled islands is investigated by X-ray diffraction, atomic force and electron microscopy.

1. Results and discussion

The samples under investigation were grown by molecular beam epitaxy from a solid source (SSMBE) (BALZERS UMS 500P) and a gas source (GSMBE) (Daido-Hoxan VCE S2020) on Si (001) substrates at growth temperatures \( T_g = 600°C, 700°C \) and 750°C. Further details of the growth process are presented elsewhere [2, 3]. After substrate cleaning and growth of Si buffer layer a Ge layer with the equivalent thickness from 3 monolayers (ML) to 11 ML was deposited. Sample morphology was determined by \textit{ex situ} atomic force microscopy (AFM) (NT-MDT “Solver-P4” and Digital Instruments Nanoscope IIIa). The electron microscopy measurements were performed on “JEOL EM-200EX”. The X-ray diffraction measurements were performed at room temperature using a “DRON-4” double crystal X-ray diffractometer.

The parameters (sizes, surface density, composition and elastic strain) of GeSi islands grown from a gas source at 700°C coincided with the parameters of the islands, grown from a solid source and investigated earlier [3, 4]. The structures with a narrow size distribution of dome-islands (dispersions of lateral sizes and height less than 10%) were obtained at
Fig. 1. The dependence of the island lateral size on its height obtained from AFM images. The dashed line separates the regions of different island shape.

this growth temperature by both deposition techniques. Average composition and elastic strain of dome-islands were defined from X-ray diffraction investigation using a strained layer approximation. In the framework of this model the content of Si in dome-islands is about 50% regardless of a deposition technique. Such a high value of Si content is related to strain-driven surface and volume diffusion of Si from the region of maximum elastic strain near the islands bases [4, 5].

A decrease of growth temperature to 600°C inhibits the surface and volume diffusion of Si and Ge. As a result of reduction of Ge adatom surface diffusion, the surface density of islands at $T_g = 600\degree C$ increases four times in comparison with that $T_g = 700\degree C$. Besides, the critical sizes of islands with pyramid-like shape (“pyramid”-islands), at which they transform to a dome shape, decrease about 2 times (Fig. 1). One reason of this change of size is reduction of Si content in islands with a decrease of $T_g$. Modification of island composition with a change of growth temperature was confirmed by X-ray analysis. Figure 2 displays the $\Theta/2\Theta$ scans of samples grown at different temperature near the symmetric (004) Si reflection. It is seen that the small peak from the islands shifts from the Si substrate peak to the region of diffraction from a pure Ge layer. The Ge concentration in islands obtained by analysis of the symmetric (004) and asymmetric (224) X-ray reflections changes from 50% at $T_g = 700\degree C$ to 75% at $T_g = 600\degree C$. The increase of Ge content results in an increased elastic strain in islands and, according to the model suggested by Tersoff et al[6], a decreased critical pyramid-islands volume.

The increase of islands surface density is another reason for changes in the pyramid and dome islands size. The high surface density of the islands gives rise to elastic interactions between them and can significantly reduce the equilibrium transition volume [7].

The effect of alloying on growth of GeSi islands is most pronounced at $T_g = 750\degree C$. Besides the increase of pyramid and dome size (Fig. 1), the high diffusion rate of Si into islands at this growth temperature initiates a reverse transformation of islands from dome to pyramid shape during deposition. As a result, the islands of an intermediate shape (with pyramid-like base and dome-like apex) were seen on AFM image. Similar changes of the islands shape were observed during a post-growth annealing of structures grown at $T_g < 700\degree C$ and connected with a decrease of elastic strain in dome islands with an increase of Si content in the islands during annealing [4, 8].

The Ostwald ripening mechanism also affects islands growth at $T_g = 750\degree C$. During Ostwald ripening some islands build up at the expense of other islands that shrink and
dissipate completely. This results in the spread of the islands sizes distribution. The surface of samples grown at $T_g = 750^\circ$C keeps traces of the dissociated islands as circumference-like grooves. Here the situation is also similar to annealing of structures grown at $T_g < 700^\circ$C [4].

In summary, we investigated the effect of alloying on growth and parameters of GeSi self-assembled islands. The change of maximum pyramid-islands sizes with an increase of growth temperature is related to enhancement of alloying in GeSi islands. The Ostwald ripening and alloying broaden the islands sizes distribution at $T_g = 750^\circ$C.

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Raman scattering from Ge nanocrystals on Si substrates: problems and solutions

A. V. Kolobov†‡§, Y. Maeda¶, H. Oyanagi‡, K. Tanaka† and Z. Cernosek∥
† Joint Research Center for Atom Technology – National Institute for Advanced Interdisciplinary Research, 1-1-4 Higashi, Tsukuba, Ibaraki 305, Japan
‡ Electrotechnical Laboratory, 1-1-4 Umezono, Tsukuba, Ibaraki 305, Japan
§ Ioffe Physico-Technical Institute, St Petersburg, Russia
¶ Osaka Prefecture University, 1-1 Gakuencho, Sakai, Osaka 599-8531, Japan
∥ University of Pardubice, Pardubice, Czech Republic

1. Introduction

Recent years have witnessed growing interest in Ge and Si nanostructures. A number of groups have reported the formation of Ge nanocrystals embedded in SiO₂ by different methods such as annealing of Ge-containing oxides [1, 2], ion implantation [3], UV-assisted oxidation [4] and intense photoluminescence around 2 eV has been observed [2, 4].

The technique most often used to characterize the structure of the nanocrystals is Raman scattering. However, as some of us have argued earlier [5], in many cases the experimentally observed peak attributed to Ge is in fact coming from the Si substrate (See Fig. 1). The three upper curves represent Raman spectra of Ge nanocrystals formed in SiO₂ by different methods [1, 3, 4]. The lower curve is a Raman spectrum of a silicon wafer [6]. The striking line shape similarity of the “Ge nanocrystal” samples with that of the Si wafer and one-to-one correspondence of features located at 229, 300 and 435 cm⁻¹ clearly demonstrate that the reported spectra are strongly dominated by the two-phonon acoustic Si peak. It is thus very important that the Ge signal be separated from the Si peak and below we present results of different approaches to achieve his goal.

Fig. 1. Raman spectra from Ge nanocrystals samples prepared through different techniques (curves 1–3, respectively after Refs) and that from a Si wafer (4).
Fig. 2. Raman spectra of as-made samples containing 60.2 mol.% Ge deposited onto Si (1) and quartz glass (3). The dotted line (2) is a Raman spectrum of the Si wafer. At the bottom, HY-polarized spectrum (4) is shown. The excitation light is a 633 nm line from a He–Ne laser. The spectra obtained under the non-polarized and polarized conditions are normalized.

2. Experimental details

The samples were prepared by co-deposition of Si and Ge oxides onto Si(001) substrates by magnetron sputtering. The Ge concentration varied from 25 to 60 mol.%. After deposition, the samples were annealed for 1 h at 800 °C in an argon atmosphere which produced nanocrystals with a characteristic size of 6–8 nm. More details can be found elsewhere [2].

Raman spectra were taken at room temperature in the back-scattering geometry. A YAG:Nd³⁺ laser (λ = 1064 nm), a He–Ne laser (λ = 633 nm) and an Ar-ion laser (λ = 488 nm) were used as excitation light source. The temperature dependence was studied using the He–Ne excitation in the temperature range from ∼77 K to 300 K. In the case of polarized measurements, a thin film polarizer was placed at the entrance slit of the monochromator.

The X-ray absorption measurements were performed at BL13B station at the Photon Factory using a 27-pole wiggler magnet inserted in a 2.5 GeV storage ring. More details can be found elsewhere [7].

3. Experimental results

Figure 2 shows Raman spectra for an as-made sample (60.2 mol. % Ge) together with the signal obtained from the back side of the same Si substrate under the same conditions. The excitation source is a He–Ne laser. The origin of features located at 229, 300, and 435 cm⁻¹ is obvious: they come from the substrate. The only contribution from Ge is a broad feature located below 300 cm⁻¹. This is further evidenced by the spectrum of an identical sample deposited onto a quartz glass substrate.

One possible way to decrease the contribution from the substrate is to make use of resonant Raman scattering. We found that indeed if an Ar-ion laser is used (the dielectric constant, ε₂, of Ge reaches maximum at around 2.3 eV) the signal from the substrate is
considerably suppressed. Use of 1064 nm light did not result in improvement of the Ge/Si peak ratio.

Since the Si peak located at 300 cm$^{-1}$ is a two-phonon peak while that of Ge is a single phonon peak, they should have different temperature dependencies. Our measurements show that while the intensity of the Si peak does decrease at lower temperatures it still remains significant.

Finally, we used various scattering configurations. It is known [6, 8] that the acoustic two-phonon peak disappears in the configuration when the polarisation planes of the incident and scattered beams are parallel to each other. Curve 4 in Fig. 1 shows the polarised Raman spectrum. The spectrum does not contain the contribution from the Si substrate: the features at 229, 300 and 435 cm$^{-1}$ disappear and the remaining broad peak is very similar to the one detected in the sample on a quartz substrate (curve 3).

4. Discussion

The above results allow us to conclude that the most efficient way to suppress the relative intensity of the Si two-phonon signal is to use an Ar-ion laser as the excitation source and/or utilize an appropriate scattering configuration.

Based on the peak position and line shape we conclude that the Ge peak observed in as-made samples is due to amorphous Ge present in the matrix. Upon annealing, Ge nanocrystals are formed. The formation of Ge crystals was checked by Raman spectroscopy. The line shape of the observed peak is asymmetric as should be for nanocrystals. The formation of nanocrystals was also verified by HR-TEM.

In order to confirm these conclusions we have also performed EXAFS (extended X-ray absorption fine structure) measurements. We found no peaks above the first Ge–Ge neighbour which confirms our conclusion that Ge is mainly present in the amorphous phase. After annealing the Ge–Ge peak gets stronger and peaks corresponding to higher shells emerge which is another evidence of crystallisation. Details of this work will be published elsewhere [9].

In samples with lower Ge concentration we were not able to observe any Ge-related features in the Raman spectra although our TEM and EXAFS studies indicated the formation of Ge nanocrystals.

At present we do not completely understand why there are no Ge-related peaks in the Raman spectra containing 25 and 40 mol.% Ge. A possible explanation is that in the case of lower Ge concentration the formed nanocrystals are mush smaller (our TEM data) and the line broadening due to the size distribution can be more significant. Further studies are needed and are currently underway.

5. Conclusion

We have demonstrated that in many cases the experimentally observed peak at $\sim 300$ cm$^{-1}$ is in fact originating from the Si substrate. We have found that the most promising ways to tackle this problem are (1) use of resonant Raman and/or (2) use of polarized Raman scattering. The former is more efficient to excite the Ge nanoclusters but also results in stronger photoluminescence which can make the measurement more difficult.

Our results also demonstrate that Raman scattering can be used as an efficient probe only if Ge concentration exceeds a certain value.
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References

The effect of deuterium on the optical properties of free standing porous silicon layers

A. I. Belogorokhov†, L. I. Belogorokhova, Y. Masumoto§‡, T. Matsumoto§ and E. A. Zhukov
M. V. Lomonosov Moscow State University, 119899 Moscow, Russia
† Institute of Rare metals, 117571 Moscow, Russia
§ Single Quantum Dot Project, ERATO, JST, Tsukuba Research Consortium, Tsukuba, Ibaraki 300-2635, Japan
‡ University of Tsukuba, Tsukuba, Ibaraki 305-8571, 305-2635, Japan

Abstract. Free-standing porous silicon films with high deuterium concentration have been fabricated. Optical properties of the freshly produced samples have been studied using infrared spectroscopy and photoluminescence. A detailed analysis of photoluminescence and infrared absorption spectra was performed. Changes in the optical and photoluminescence properties, modification of the surface states of porous silicon layers during their exposure in the ambient air have been studied. The precise value of the vibration frequencies of the silicon-deuterium modes were obtained. The estimated value of refractive index of the porous silicon samples with deuterium is a factor of 1.6 higher than that of the samples with hydrogen.

Introduction

While there is strong evidence that light emission originates from Si crystallites, the difference of only 0.3 eV between the fundamental band gap $E_g$ of Si and the red photoluminescence (PL) peak position, as well as the effect of chemical treatment on the PL spectra, indicates that the radiation process should be more complex than the simple confinement picture, and suggests, that the PL properties, the porous silicon (PS) microstructure and the surface chemical composition of the porous material should be correlated. The steady interest to these problems is justified from the point of view of their application of PS to electronic and optoelectronic devices.

Some researchers attempt to solve these problems by the variation of the current density, treatment time or electrolyte composition or by using electrolytes such as DF:C$_2$D$_5$OD [1] and HCl:HF:C$_2$H$_5$OH solutions [2] were made. They were able to verify that samples prepared by means of these electrolytes exhibit considerably better PL properties than samples obtained by means of the standard procedure.

Ipatova and co-worker [3] demonstrated that the quantity of deuterium (D) atoms in a sample of silicon must be greater than that of hydrogen atoms. Naturally that this result evoked great interest of the researchers which investigate problems of surface passivation of PS and its degradation.

Methods of the sample preparation and experimental techniques

Hydrogen terminated porous Si (H–PS, N1), and deuterium terminated porous Si (D–PS, N2) were formed by electrochemical anodizations. The H–PS was fabricated in the dark to avoid the oxidation using HF-ethanol solution (HF:H$_2$O:C$_2$H$_5$OH = 1:1:2) by applying
positive bias to a p-type 3–5 Ω cm Si substrate with a current density in the range of 15 A/cm² for 45 min. The D–PS were also fabricated in the same condition except for the use of DF-ethanol-D₆ solution (DF:D₂O:C₂D₅OD = 1:1:2) for 65 min. The free-standing porous Si layers were prepared by applying the electropolishing technique after the fabrication of the porous layer — we abruptly increased the current density up to 700 mA/cm² to remove the porous layer from the Si substrate.

The PL spectra were measured at sample temperature 300 K by using monochromator DFS-24. The PL was excited by a beam from Ar⁺-ion laser (ℏω = 2.540 and 2.409 eV, P = 5 mW in a spot of 3 mm² square). Absorbance and reflectance measurements were performed at room temperature in the wave number range from 20 to 5000 cm⁻¹ using a Bruker IFS 113v Fourier transform infrared (FTIR) spectrometer. Instrumental resolution was 0.5 cm⁻¹. The spectrophotometer Hitachi 330 was used for registration of the absorption spectra. All spectra of the PS samples were measured two times: (1) as-grown; (2) after 6 months after they were stored in atmospheric ambient.

**Experimental results and discussions**

**FTIR spectra**

FTIR spectra of the samples N1 and N2 are shown in Fig. 1. The spectrum of the sample N2 shows a strong absorption in the range 1500–1570 cm⁻¹. On the contrary, this band is absent in a spectrum of the sample N1. The feature in this part of spectrum we can explain by the absorption of Si–D bonds in PS sample with deuterium. The fine structure of that absorption peak is demonstrated on the insertion of Fig. 1(a). It is important to note the next two factors. First, the structures of absorption bands SiHₙ and SiDₙ are similar. Second, isotopic shift of Si–D peak is 1.375. It coincides with the theoretical value of 1.376 [3]. In additionally, other well resolved another features have been observed in absorption spectra of the sample N1 and N2. The transmission spectra of the sample N1 and N2 in the range
Fig. 2. (a) Optical-transmission and PL spectra of the as-grown sample N1 (dotted curve) and sample N2 (solid curve). (b) FTIR vibrational spectra of the samples after their degradation.

from 400 to 1300 cm\(^{-1}\) are demonstrated in Fig. 1(b).

Comparison of the hydrogen concentration which was estimated in the sample N1 and deuterium concentration in the sample N2 gives the ratio of \(N_{D_2}/N_{H_2} = 1.06\). This ratio doesn’t reach the value 5–6 obtained from [3]. However, the calculation obtained by Ipatova et al. [3] is valid for the equilibrium condition. Furthermore, the calculation which were carried out in [3] are related to the bulk semiconductors, but the diffusion of hydrogen and deuterium atoms on PS layer surface, especially during the process of nanocrystall surface formation, can be different from that in the case of the bulk monocrystall Si.

The transmission and reflection spectra of the sample N1 and N2 in the far-infrared (FIR) wavelength region (25–500 cm\(^{-1}\)) have the features connected with the light interference in a thin film of PS. It is possible to estimate the refractive index (\(n\)) of samples from these spectra knowing the thickness of the layer (\(d_{N1} = 50.0\, \mu\text{m}\) and \(d_{N1} = 30.2\, \mu\text{m}\)). We obtained \(n = 1.52\) and 2.44 and from the transmission spectra \(n = 1.60\) and 2.41 for the samples N1 and N2 correspondingly.

Photoluminescence and light absorption in the visible spectral range

The absorption edge (\(E_g\)) of 1.81 and 1.83 eV for the sample N1 and N2, correspondingly was determined by the absorption measurements. Additionally, the photoluminescence spectra of these sample are shown in Fig. 2(a). The usual asymmetric form of the PL spectra with maximum at 1.769 eV (N1) and at 1.764 eV (N2) have been observed on the as-grown samples N1 and N2. The PL intensity of the fresh sample with hydrogen was similar to the sample with deuterium.

Degradation of the samples

The transmission spectra in the spectral region as in Fig. 1(b) of the sample N1 and N2 obtained after 6 months of exposure in the ambient air are shown in the Fig. 2(b). Careful analysis reveals essential peculiarities in the behaviour of the spectra. It is clearly seen the changes occurs at frequencies corresponding to the Si\(H_n\) and Si\(D_n\) stretching modes. From the change of the absorption ratio of the stretching modes (Si–D versus Si–H) before
and after 6 months, we can show the effect of the gradual substitution from deuterium termination to hydrogen one as a function of aging time by comparing the results of Fig. 1(a) and Fig. 2(b). Also intensity of the absorption on the C = O (1720 cm\(^{-1}\)) bonds is increased. The spectral position of the PL peak of the sample with deuterium has not been changed. Very small change have been observed only in a high energy part of the spectrum. At the same time the PL peak of the sample N1 (with hydrogen) is shifted to lower energy (\(\delta = 35\) meV). The dominant mechanism of radiative recombination in samples with hydrogen at the first moment after preparation is via excitons. The red shift upon oxidation is related to recombination involving a trapped electron or exciton [4].

It is known that the deuterium termination can significantly reduce the rate of the light-induced oxidation compared to hydrogen termination under the same oxidation condition [5].

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References
Resonant acceptors states in Ge/Ge$_{1-x}$Si$_x$ MQW heterostructures

V. Ya. Aleshkin†, I. V. Erofeeva†, V. I. Gavrilenko†, D. V. Kozlov† and O. A. Kuznetsov§

† Institute for Physics of Microstructures of Russian Academy of Sciences, GSP-105, Nizhny Novgorod, 603600, Russia
§ Physico-Technical Research Institute of Nizhny Novgorod State University, Russia

Abstract. New nonvariational theoretical approach allowing to calculate both the localized and the continuum states of charge carriers in QW heterostructures in the presence of the Coulomb potential has been developed. The method has been used to calculated the energy spectra of shallow acceptors in strained Ge/GeSi MQW heterostructures. Optical transitions from the acceptor ground states to the resonant states have been revealed in the measured far IR photoconductivity spectra of the heterostructures.

1. Introduction

In recent years there has been considerable interest to the resonant impurity states in semiconductors (see for example [1]). This interest was stimulated by the observation of stimulated emission of far infrared (FIR) radiation observed in uniaxially compressed p-Ge at high electric fields [1]. In uniaxially stressed p-Ge the resonant states arise due to the splitting of light and heavy hole subbands at high enough stress value when the impurity levels pertain to the upper split-off subband turn out in the energy continuum of the lower subband. The similar situation can be easily realized in quantum well (QW) heterostructures where the resonant acceptor states arise due to QW confinement. The paper is devoted to the theoretical and experimental study of the resonant acceptor states in strained Ge/GeSi(111) multiple-quantum-well (MQW) heterostructures.

2. Theoretical formalism

The new nonvariational approach has been developed for the calculation of the acceptors energy spectra. The spectra of shallow acceptors in Ge/GeSi QW heterostructures were calculated in the framework of the envelope function approximation. The acceptor Hamiltonian used was $4 \times 4$ matrix operator included Luttinger Hamiltonian, the deformation term, the QW confinement potential due to the valence band discontinuity and the Coulomb potential. Similar to the work [2] the axial approximation was used, i.e. the Hamiltonian was considered to be invariant by rotation about the $z||[111]$ axis (the axis perpendicular to the growth plane of the structure). In this case the $z$-component of the total angular momentum $J_z$ is a good quantum number and the acceptor states are two-fold degenerate by the sign of the component of the angular momentum [2]. The acceptor envelope function was expanded in the basis of free hole envelope functions in the QW $v_n^r(k, z)e^{ikr||}$, which are eigenstates of the Hamiltonian not containing the Coulomb potential:

$$F^r(r) = \sum_n \int d\mathbf{k} C_n(k) v_n^r(k, z)e^{ikr||}$$  \hspace{1cm} (1)
where \( n \) is the number of subband, \( \mathbf{r} \) is the radius-vector, \( \mathbf{r}_\parallel = (r_\parallel, \theta) \) is the in-plane coordinate and \( \mathbf{k} = (k, \alpha) \) is the hole wave vector. In the axial approximation the expansion coefficients take the following form:

\[
C_n(k) = C_n(k)e^{im\alpha}
\]

(2)

where \( m \) is the projection of the total angular momentum on the z-axis. By substituting the expansion (1) into the Schrödinger equation for the envelope function the integral equations for the above expansion coefficients were obtained. The integrals were approximated by the discrete sums over the hole momentum \( k \), the step difference being chosen much less than the inverse Bohr radius. The sums were interrupted at \( k \) much greater than the inverse Bohr radius. Therefore the problem was reduced to the diagonalization of the finite symmetric matrix (typically \( 500 \times 500 \)). Using this method the energies and wave functions of states, which correspond to the values of total angular momentum projection \( J_z = \pm \frac{3}{2}, J_z = \pm \frac{1}{2} \) and \( J_z = \pm \frac{5}{2} \) were calculated. Dipole optical transition from the acceptor ground state \( (J_z = \pm \frac{3}{2}) \) are allowed into localized excited states and the states of the continuum corresponding to \( J_z = \pm \frac{1}{2} \) and \( J_z = \pm \frac{5}{2} \).

The acceptor wave functions have the complicated structure. Expansion (1) include terms referred to different hole subbands in the QW. If the expansion of a wave function mainly consists of free hole envelope functions of the certain subband, we’ll ascribe the state corresponding to the function to this subband. The acceptor ground state pertains to the 1st subband \( hh_1 \) and has a negative energy (with respect to the bottom of \( hh_1 \) subband). The energy of a state pertaining to an upper subband can be either negative (localized state) or positive (resonant state). The localized state which pertains to an upper subband becomes resonant if the width of the quantum well is reduced. The part of the wave functions of resonant state consisting of the hole envelope functions of the subband to which this state is pertained to looks like the wave function of the similar localized state corresponding to 1st hole subband.

3. Results and discussion

Measured FIR photoconductivity spectra of undoped Ge/GeSi MQW heterostructure are shown in Fig. 1 by solid lines. The residual shallow acceptors concentration is estimated to be about \( 10^{14} \) cm\(^{-3}\). In the long wavelength range (Fig. 1(a)) the photoconductivity results from the optical transitions from the acceptor ground state to the continuum and to the excited localized states (and subsequent thermoionization). The binding energies of shallow impurities in QW heterostructures are known to be dependents on the impurity position in the well. This is clearly seen in Fig. 2 for the acceptor ground state in the investigated heterostructure. The dashed line in Fig. 1(a) shows the photoconductivity spectrum calculated at the supposition that acceptors are uniformly distributed over QW (details of the procedure are described in [3]). One can see that in this spectrum the photoconductivity maximum corresponds to the binding energy of on-center acceptors at about 7.4 meV. This results from the fact that the impurities situated near QW center (about 50% of QW width) have nearly the same binding energy (Fig. 2, cf. [3]). However the measured spectrum has quite a different form. There is a strong photoconductivity band in the spectrum in between 2.5 and 5 meV (Fig. 1(a)). The short wavelength edge of this band just corresponds to the binding energy of the acceptors situated at the heterointerface (on-edge acceptor, see arrows 1b in Fig. 1, 2). So it is naturally to assume that in addition to the uniformly distributed over QW width acceptors there are some centers concentrated near the heterointerface. A heterointerface is known to be a source of point defects and the observed residual acceptors
Fig. 1. Long (a) and short (b) wavelength parts of the measured (solid line) and the calculated photoconductivity spectra of Ge/Ge_{0.88}Si_{0.12} heterostructure #306 ($d_{QW} = 200 \, \text{Å}, N_{QW} = 162$, elastic deformation $\epsilon = 2.4 \times 10^{-3}$). Dashed and dotted lines correspond to two types of acceptor distribution over QW: uniform and uniform plus $\delta$-layers at the heterointerfaces respectively. The sheet concentration at the heterointerfaces was taken half of that for the uniformly distributed acceptors per well. Arrows indicate binding energies for on-center (a) and on-edge (b) acceptors.

are likely to be connected with vacancies rather than with the chemical impurities. The dotted line in Fig. 1(a) represents the calculated photoconductivity spectra taking into account both the uniformly distributed and localized near heterointerfaces acceptors. It is clearly seen that the above model of the acceptor distribution describes fairly well both the photoconductivity peak at 7 meV and the short wavelength edge of the main photoconductivity band in between 2.5–5 meV. The long wavelength part of this band (that was observed in this particular sample only, cf. [4]) can be attributed to the photoionization either of the acceptors localized in barriers near heterointerfaces or of shallow $A^+$ centers in QWs [4].

Fig. 2. Energy diagram of hole states in Ge/Ge_{0.88}Si_{0.12} QW heterostructure #306. Solid and dashed lines correspond to free holesubbands (heavy holes: hh1–hh4, light holes: lh1) and acceptor levels (ground state and two “excited” resonant states). The arrows indicate the optical transitions introduced in the caption to Fig. 1.

The represented above approach allowed to calculate not only the localized acceptor states but all the states of the continuum including resonant states as well. The dotted curve in Fig. 1(b) shows the calculated spectrum of the photoconductivity resulted from the optical transitions from the acceptor ground state to the states of the continuum. To fit the spectral positions of the main features designated by arrows 3a and 3b in Fig. 1(b) to the experimentally observed ones we have to reduce the well width at the calculation down
to 185 Å (if compared with 200 Å obtained from X-ray analysis). Arrows 2a,b and 3a,b in Fig. 1(b), 2 indicate the optical transitions from the acceptor ground state \( J_z = \pm \frac{3}{2} \) to the “excited” resonant states \( (J_z = \pm \frac{1}{2} \) and \( J_z = \pm \frac{5}{2} \) pertained with the upper hole subbands hh2 (arrows 2) and hh3 (arrows 3) respectively for the on-center (a) and on-edge (b) acceptors. There are no doubts that the doublet 3a-3b really presents in the measured spectra in Fig. 1(b). Moreover there are noticeable spectral features in between 9–12 meV in the measured spectra that can be attributed to the doublet 2a-2b. To the authors knowledge this is the first observation of the resonant acceptor states in QW heterostructures.

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References

Transition from a single- to double-quantum-well magnetotransport in
the p-GeSi/Ge/p-GeSi heterosystem

M. V. Yakunin†, G. A. Alshanskii†, Yu. G. Arapov†, O. A. Kuznetsov‡ and
V. N. Neverov†
† Institute of Metal Physics RAS, Ekaterinburg, GSP-170, 620219, Russia
‡ Scientific-Research Institute at Nizhnii Novgorod State University, Russia
e-mail: yakunin@imp.uran.ru

Abstract. As revealed from the high field low temperature magnetotransport measurements, the
joint quasi two dimensional hole gas with the density \( p_s = \left(3 - 5\right) \times 10^{15} \text{ m}^{-2} \) exists in a p-
GeSi/Ge/p-GeSi quantum well up to the Ge layer widths \( d_w \lessgtr 25 \text{ nm} \), but it separates into two
sublayers located at the well’s sidewalls at \( p_s \approx 5 \times 10^{15} \text{ m}^{-2} \) and \( d_w \gtrsim 35 \text{ nm} \). The sublayers
have approximately equal resistances suggesting similarity of the normal and inverted interfaces
in this heterosystem. The recently discovered quantized Hall insulator phase was detected here
only in the intermediate range of sublayer separations, yielding evidence that the intersublayer
correlations are important to stabilize this state.

A system of two closely spaced two-dimensional (2D) layers — a double quantum well
(DQW) — creates an intriguing new physics since the distance between the layers can be
made comparable or less than an average distance between free carriers within a single
layer. Under these conditions the interlayer correlation effects may be stronger than the
intralayer ones. A picture of Landau levels in a DQW system differs considerably from
that in a single 2D layer reflecting the following essentials.

1) If the two layers are close enough, the tunneling between them results in a gap \( \Delta_{\text{SAS}} \)
between the generalized DQW symmetric and antisymmetric levels; each of these levels
genrating its own fan of Landau levels.
2) The combination of intra- and interlayer correlations modifies the gap; this effect de-
dpends on magnetic field. The tunneling and correlations, acting in concert, can either enhace or destroy the gap. Consequently, the Landau levels of a single 2D layer would
be split in the DQW system into doublets each presenting a combination of symmetric and
antisymmetric levels. In high enough magnetic fields, such that the orbital and Zeeman
splittings exceed \( \Delta_{\text{SAS}} \), and in sufficiently perfect samples able to satisfy the condition that
the level width is smaller than the gap, the first integer quantum Hall (QH) state would
relate to the gap between the lowest split-off levels, i.e. to the resultant gap in a DQW, the
same being for all the odd numbered QH states. Thus the odd QH states can serve as a tool
for exploring the gap in a DQW system. The situation is unique in that the gaps between
magnetic split-off levels do not necessarily increase with field. Just this case was observed
in [1] where the odd numbered QH states, although quite distinct in the intermediate field
range, collapsed in the high fields. On the other hand, the interlayer correlations may cre-
ate a gap even when the single electron gap \( \Delta_{\text{SAS}} \) is absent (the case of infinitely high but
narrow barrier), although this gap is easily destroyed by the increasing temperature or by
the in-plain magnetic field [2].
The other feature of the DQW is the ability to stabilize the hardly achievable or even forbidden states of 2D electron gas (2DEG) due to additional degree of freedom stemming from the possibility for the carrier to locate in each of the coupled wells. So the $\nu = 1/2$ fractional QH effect state, a forbidden state in a single 2DEG, was observed in a DQW [3]. Also a transition into an insulating phase, probably into a Wigner crystal, was achieved in a DQW under conditions for which it has never been observed in a single 2DEG [4].

Deliberately introducing a barrier into the well is not the only way to create a DQW system. A similar physics can be realized in a sufficiently wide and intensively populated single well [5]. The idea is when electrons are introduced in a wide QW, the electrostatic repulsion between the electrons forces them into a stable configuration in which two 2DEG’s are formed at the wall wells. A major advantage of this system over a conventional DQW is the minimization of alloy scattering since the barrier between the two 2DEG’s is of the same material, it’s not a heterointerface. Also, both $\Delta_{\text{SAS}}$ and inter-sublayer distance can be changed with varying the carrier density in the well.

So far almost all of the researches in the DQW were performed in the electron systems. The hole system offers some new properties in the DQW, particularly the large value of the heavy hole mass allows to achieve easily the configuration with Coulomb coupled hole gases without tunneling between them.

We performed low temperature high magnetic field ($B \leq 30$ T) measurements of the resistivity and Hall voltage in a series of multilayered p-Ge$_{1-x}$Si$_x$/Ge heterostructures differed by the Ge layer width (which is the QW for holes) in the range $d_w = 12-40$ nm, and by the hole densities per single Ge layer $p_h = (1-6) \times 10^{15}$ m$^{-2}$. The structures were grown by the vapor deposition and selectively doped with boron in the central parts of the Ge$_{1-x}$Si$_x$ barriers. The barriers were sufficiently wide to avoid the inter-Ge-layer tunneling. The low temperature hole mobilities were in the range $1-1.4$ m$^2$/Vs.

Data for four samples are presented in the Fig. 1. The sample parameters are depicted in Fig. 2, additionally for the sample 1124b3: $p_h = 2.8 \times 10^{15}$ m$^{-2}$, $d_w = 20$ nm. For the
samples 1006 and 1124, with the narrowest Ge layers (12.5 and 20 nm), a usual QH picture was obtained: the plateaus in $\rho_{xy}(B)$ at integer filling factors $i \geq 1$ (per single Ge layer) and concomitant minima in $\rho_{xx}(B)$. A similar picture was obtained for the samples with the widest Ge layers and highest hole densities: $d_w = 38$ nm and $p_s = 5 \times 10^{15}$ m$^{-2}$ (sample 476 in Fig. 1; we have examined a group of 4 similar samples and obtained similar results), but with an essential difference: the QH state for $i = 1$ was absent here. Considering that the spin splitting in the valence band of 2D Ge is as high as nearly a half of the orbital one [6] and that the $i = 1$ QH state is well resolved in the narrow well samples, the missing of the $i = 1$ QH state in the wide wells means unambiguously that conductivity in each Ge layer is via two parallel 2D sublayers here. That the first plateau on the high field side is close to the fundamental value $\rho_{xy} \approx h/2e^2$ means that the two sublayers in a Ge layer have approximately equal resistances $\rho_{xy} \approx h/e^2$. This result is surprising on the background of similar researches in a wide GaAs/Ga$_{1-x}$Al$_x$As QW [7] where up to 40 times difference in mobility was obtained for electron sublayers located at the normal and inverted interfaces. Bad mobility near the inverted interface was attributed to the scattering on Si dopands floated up from the lower Ga$_{1-x}$Al$_x$As barrier during the growth. Our
results on the QH effect indicate that it is possible to grow a Ge$_{1-x}$Si$_x$/Ge system so that the effects of the boron dopant floating up are negligibly weak and both the normal and inverted interfaces of the Ge layer are similar. The multilayered structure of our samples may help the high symmetry of the wells [8]. Although some deviations of the $\rho_{xy}(B)$ plateau from the $i = 2$ horizontal and its finite slope may be attributed to some inequality of sublayers. Also approximately 20% positive magnetoresistance in the weakest fields has been observed only in this group of samples indicating the participation of two kinds of carriers with slightly different mobilities and densities.

The most unusual results were obtained on a group of three samples within the intermediate range of sublayer separations: $d_w = 34-41$ nm, comparable with the previously described sample group, but with lower hole densities $p_h = (1-2) \times 10^{15}$ m$^{-2}$. Each of these samples exhibited bistable behavior. In one of their two metastable states a weak $i = 1$ QH peculiarity was present (see sample 451b4 in Fig. 1) concomitant with the weak $\rho_{xy}(B)$ minimum, while an extremely wide plateau was revealed in the second metastable state at $\rho_{xy}(B)$ close to the value 13 kΩ, which corresponds to $i = 2$. We interpret the latter as a manifestation of a double layer quantized Hall insulator state stabilized by the interlayer correlations [9].

The potential profile of the QWs together with the energy levels and wave functions were calculated self consistently from a system of Schrödinger and Poisson equations — see Fig. 2. In general, these results confirm our explanations. For the sample 1006 all the levels are higher than the bottom bending amplitude, and the ground wave function is uniformly distributed within the well. On the contrary, in the samples 475/476 the two lowest levels $E_0$ and $E_1$ coincide and their wave functions are confined within triangular wells close to the well walls. Only the third level $E_2$ corresponds to the state extended through the whole layer width, but the Fermi energy is lower than this level.

In the intermediate case, for the sample 451, the first two levels $E_0$ and $E_1$, as well as the Fermi level, are lower but very close to the bottom bending amplitude. Even for the level $E_0$ the wave function $\Psi_0$ is far from zero in the center of the well. The differences from the samples 475/476 are due to much lower hole density in the well. All these features indicate the possibility for existence of two coupled hole sublayers in the Ge layer. Considering the big hole mass, even a lowest barrier suppresses tunneling that is reflected in the small splitting between $E_0$ and $E_1$ levels. But a small depth of a $\Psi_0$ minimum in the center of the well indicates that the effective distance between the sublayers might be small enough for the interlayer correlations were substantial.

Acknowledgements

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References

Room temperature single electron devices by STM/AFM nano-oxidation process

Kazuhiko Matsumoto
Electrotechnical Laboratory, MITI, 1-1-4, Umezono, Tsukuba, 305-8568, Japan
e-mail: kmatumot@etl.go.jp

Abstract. A single electron transistor (SET) and a single electron memory were fabricated using the improved pulse-mode AFM nano-oxidation process. A single electron transistor which works as an electrometer for detecting the potential of the memory node of the single electron memory showed the clear Coulomb oscillation characteristics with the periods of 2.1 V at room temperature. A single electron memory showed the hysteresis loop by the return trip of the memory bias when starting from 0 V to 10 V and again coming back to 0 V.

Introduction

A various types of single electron transistors and memories were, so far, studied. Almost all single electron devices so far studied adopted the self-organized nano-structure because of the difficulty of the artificial fabrication process of the nano-structure. The some examples of the self-organized nano-structure for the single electron devices are the polycrystalline silicon film [1], the poly-crystal silicon dot [2], and the squeezed delta-doped GaAlAs/GaAs layer [3] for the multi-tunnel junctions and/or memory node. They showed the clear single electron device characteristics such as the digitized threshold voltage shifts [1] or hysteresys loop [3] by the injected few number of electrons. Those devices, however, include the problem of the poor reproducibility of the device characteristics because of the application of the spontaneously formed nano-structures. On the other hand, in the present paper, we show the new device fabrication process [4–6], which could artificially fabricate the few tens of nano-meter order small structure and room temperature operated single electron transistor and single electron memory were realized.

1. AFM nano-oxidation process

The new fabrication process used the atomic force microscopy (AFM) cantilever as an ultra small cathode, and pulse bias was applied between cantilever and the 2.5 nm thick titanium (Ti) thin film that was on the atomically flat $\alpha - Al_2O_3$ substrate as shown in Fig. 1. AFM nano-oxidation process so far we proposed used DC bias between the cantilever and the titanium metal, and the surface of the Ti metal was anodized to form the oxidized titanium line (TiO_x) which works as a tunnel junction for the single electron devices [4–6]. During this DC-mode AFM nano-oxidation process, however, the positive hydrogen ions H^+ formed the space charge at the front end of the TiO_x/Ti interface, which prevents the further oxidation. This causes the difficulty for the deeper oxidation. In order to solve the problem, the $\pm 1.5 \sim 2$ V pulse bias was applied instead of the DC bias between the conductive AFM cantilever and the Ti metal [7]. The negative pulse anodized the surface of the metal and the positive pulse neutralized the positive space charge from the front end of TiO_x. Therefore, using the pulse bias for the oxidation, the deeper oxidation becomes possible than the DC bias at the same applied bias. In another words, a narrower and deeper
oxidized Ti line could be formed using the lower applied pulse bias. Figure 2 shows the AFM image of the TiO$_x$ line folded six times on the atomically flat Ti/$\alpha$-Al$_2$O$_3$ substrate fabricated by the present new AFM nano-oxidation process. The line width is 20 nm, and line height 1.2 nm. Using this new process, the single electron transistor and the single electron memory were fabricated.

2. Single electron transistor

Figure 3 shows the schematic structure and AFM image of the fabricated one island side gate single electron transistor (SET) on the atomically flat $\alpha$-Al$_2$O$_3$ substrate. Using the pulse mode AFM nano-oxidation process, two tunnel junctions for SET were formed by TiO$_x$ lines. The size of the tunnel junction is 19 nm (width) $\times$ 26 nm (length) $\times$ 2 nm (thickness) and the island size is 8 nm $\times$ 26 nm.
3. Single electron memory

Figure 6 shows the equivalent circuit of the single electron memory which consists of the multi-tunnel junction and the normal memory capacitance. The memory node of the single electron memory where small number of electrons are stored is connected by the single electron transistor (SET) through the normal capacitance $C_g$. Using the equivalent circuit of the single electron memory shown in Fig. 6, the principle of the operation of the single electron memory is explained as follows. By increasing the memory bias $V_{MEM}$, the potential of the memory node $V_t$ also increases in proportion to the ratio of the memory capacitance $C_{gt}$ and the multi-tunnel junction capacitance $C_{it}$, i.e.,

$$V_t = V_{MEM}C_{gt}/(C_{it} + C_{gt}).$$

When the potential of the memory node $V_t$ becomes larger than the Coulomb gap bias of the multi-tunnel junction $V_0$, one electron can pass through the multi-tunnel junction to reach the memory node, which lowers the potential of the memory node as $e/(C_{it} + C_{gt})$. 

The width of the gate insulator for the side gate SET was increased up to 964 nm to completely prevent the gate leakage current at room temperature. (In the previous work, the width of the gate insulator was 300 nm).

Figure 4 shows the drain current-gate bias characteristics of the SET at room temperature using the drain bias as a parameter. The drain current of the SET oscillates with the period of 2.1 V at room temperature in the gate bias range of 0 V to 10 V at the drain bias of $V_D = 0.25$ V and 0.3 V. Five peaks are clearly seen for each drain bias condition. The current modulation rate is from 20% to 30%. Even at the different drain bias, the drain current shows the oscillation peaks at the same gate bias points. The gate capacitance estimated from the periods of the Coulomb oscillation is $8 \times 10^{-20}$ F.

Figure 5 shows the drain current-gate bias characteristics when the gate bias was changed from $-10$ V to 10 V at the drain bias of $V_D = 0.25$ V. The peaks of the Coulomb oscillation appear periodically and symmetrically against the plus and minus region in the gate bias. Thus, the fabricated SET shows the clear gate action even at room temperature.
and the second electron can not pass through the multi-tunnel junction. By increasing the memory bias further, the potential of the memory node \( V_t \) again reaches to the Coulomb gap bias, \( V_0 \), then the second electron can pass through the multi-tunnel junction to reach the memory node. Thus the one by one electron transfer becomes possible through the multi-tunnel junction to the memory node. When the \( n \) electrons are stored at the memory node, the potential of the memory node which is lowered by \( n \) electrons is described as follows:

\[
V_t = \frac{(V_{MEM}C_{gt} - ne)}{(C_{tt} + C_{gt})},
\]

where \( e \) is the electron charge. When the memory bias, \( V_{MEM} \) comes back to the lower bias, the potential of the memory node \( V_t \) goes into the negative region because of the stored \( n \) electrons. When \( V_t \) becomes smaller than the negative Coulomb gap bias of the multi-tunnel junction \( -V_0 \), the electrons are ejected one by one to the ground. Therefore, the potential of the memory node \( V_t \) shows the hysteresis loop. The relation between the memory node potential \( V_t \) and the memory bias \( V_{MEM} \) is shown in Fig. 7. Thus, few number of electrons are memorized at the memory node for few hundred seconds and this device works as a single electron memory. The change of the potential at the memory node by the injection and the ejection of electrons through the multi-tunnel junction is detected by the change of the drain current of the SET, which works as an electrometer. The change of the drain current of the electrometer is considered to be linearly proportional to the potential change of the memory node.

Figure 8 shows the schematic structure of the fabricated single electron memory which sits on the atomically flat \( \alpha-Al_2O_3 \) substrate. Figure 9 is the AFM image of the multi-tunnel junction area and two tunnel SET junctions of the fabricated single electron memory. The device consists of the multi-tunnel junction and normal capacitance for the memory and two tunnel junctions for the single electron transistor. There are four electrodes, i.e. the memory bias electrode for the memory, the source, drain and gate electrodes for the single electron transistor. The multi tunnel junction consists of five or seven tunnel junctions which are 15-25 nm in width, 15-25 nm in length, 2 nm in thickness, and the spacing between them is 10-15 nm. The normal memory capacitance is 341 nm in width and 15-25 nm in length, and 2 nm in thickness. The area between the multi-tunnel junction and the memory capacitance is designated as a memory node where few number of electrons are stored. The distance between the memory node and the island of the single electron transistor is 486 nm which determined the sensitivity of the SET. The size of the two tunnel
Fig. 8. Schematic structure of single electron memory on atomically flat $\alpha - \text{Al}_2\text{O}_3$ substrate made by pulse-mode AFM nano-oxidation process.

junctions for SET is $15 \sim 25$ nm in width, $15 \sim 25$ nm in length, 2 nm in thickness, and the spacing between them is $10 \sim 15$ nm, i.e., the island size is $10 \sim 15$ nm $\times$ $15 \sim 25$ nm.

For the measurement of the single electron memory, the condition of the electrometer was set as the drain bias of $V_D = 3$ V, and the gate bias of $V_G = 1.5$ V to detect the potential change of the memory node. Figure 10 shows the drain current of the electrometer, i.e., the single electron transistor versus the memory bias characteristics at room temperature. When the memory bias $V_{MEM}$ of the single electron memory increases from 0 V to 10 V, the drain current of the electrometer increases at the beginning and begins to oscillate. The drain current oscillation is attributed to the two effects, i.e., one is owing to the injection of the individual electrons through the multi-tunnel junction to the memory node which lowers the potential of the memory node, and the other is owing to the direct coupling of the memory bias to the island of the single electron transistor which causes the Coulomb oscillation. When the memory bias comes back from 10 V to 0 V, the drain current follows the different trace owing to the stored electrons at the memory node and shows the clear hysteresis loop even at room temperature. The measured hysteresis loop is quite similar to that shown in Fig. 2(b). Owing to the large noise level in Fig. 10, however, it is difficult to distinguish the oscillation peaks owing to the electron injection to the memory node from noise peaks.

The time transient of the drain current at room temperature when the memory bias was suddenly cut off from 10 V to 0 V showed the step-like transient trace indicating the one by one electron ejection from the memory node to the ground. The retention time is found to be 600 seconds.

The number of electrons stored in the memory node when the memory bias of 10 V was applied could be calculated from Eq. (2) and

$$n = (C_{gt} V_{MEM} - (C_{gt} + C_{gt}) V_t) / e,$$

where $V_{MEM} \gg V_t$ when $V_{MEM} = 10$ V. Therefore, $n$ is roughly calculated as

$$n = C_{gt} V_{MEM} / e.$$
Fig. 9. Plain AFM image of the fabricated single electron memory on atomically flat substrate. Multi-tunnel junctions for memory and SET junctions for electrometer are seen.

\[ C_{gt} = 4 \times 10^{-19} \text{ F.} \] Using this value, the number of electrons stored in the memory node is calculated to be about \( n = 25 \).

Using the improved pulse-mode AFM nano-oxidation process, the single electron memory was fabricated on the atomically flat \( \alpha - \text{Al}_2\text{O}_3 \) substrate. The single electron memory stored about 25 electrons at the memory node with the applied memory bias of 10 V, and showed the hysteresis loop even at room temperature. The retention time of the single electron memory is 600 seconds.

4. Conclusions

Using the improved pulse-mode AFM nano-oxidation process, the single electron transistor and single electron memory were fabricated on the atomically flat \( \alpha - \text{Al}_2\text{O}_3 \) substrate. The single electron transistor shows the Coulomb oscillation with the periods of 2.1 V at room temperature. The single electron memory shows the hysteresis loop even at room temperature.

References

Charge transfer phenomena in carbon nanotube heterodevices

A. A. Odintsov
Nuclear Physics Institute, Moscow State University, Moscow 119899 GSP, Russia
and Delft University of Technology, 2628 CJ Delft, The Netherlands

Abstract. We describe the transfer of electric charge in junctions between a metal and carbon nanotube as well as between metallic and semiconducting carbon nanotubes. The long range Coulomb interaction drastically modifies the charge transfer phenomena in one-dimensional nanotube systems compared to conventional semiconductor heterostructures. Being brought into a contact with a metal, conducting nanotube accumulates electric charge whose density decays slowly with the distance from the junction. The length of the Schottky barrier in nanotube heterojunctions varies from the distances of the order of the nanotube radius (nanometers) to the distances of the order of the nanotube length (microns) depending on a doping strength. The Schottky barrier height shows pronounced asymmetry under the forward and reverse bias. This results in rectifying behavior of heterojunctions, in agreement with recent experimental observations by Z. Yao et al. and M. Fuhrer et al.

Introduction

Physical properties of single-wall carbon nanotubes (SWNTs) are determined by their geometry [1]. Depending on the wrapping vector, SWNTs can be either metallic or semiconducting with the energy gap in sub-electronvolt range [2, 3]. Strong Coulomb interaction in 1D SWNTs results in a variety of correlation phenomena. Away from half-filling the correlations are well described by the Luttinger liquid-like model. In particular, the non-Fermi liquid ground state of the system results in a power-law suppression of the density of electronic states near the Fermi level. This was observed in single-[4] and, presumably, multi-wall [5] nanotubes, as well as in junctions between metallic SWNTs [6].

Of particular interest are carbon nanotube devices. The simplest can be fabricated by contacting two nanotubes with different conducting properties. Electron transport in such nanotube heterojunctions has been investigated in two recent experiments. Yao et al. have treated heterojunctions at the kinks in SWNTs [6] whereas McEuen et al. have explored contacts of crossed nanotubes [7]. Both groups have reported asymmetry in the $I-V$ characteristics of heterojunctions (rectifying behavior). This might be surprising, since one expects no charge transfer in contacts between two isolated SWNTs made of the same material.

In this work we study charge transfer phenomena in nanotube heterojunctions taking true long-range Coulomb interaction into account. We concentrate on junctions between a metal and a nanotube and between metallic and semiconducting SWNTs. Their equilibrium and non-equilibrium characteristics are analysed by solving the Poisson equation self-consistently.

1. Method

Consider metallic or semiconducting SWNT with the axis $z$. We assume that conducting $p_z$ electrons in SWNT are confined to the surface of a cylinder of radius $R$. SWNT is
surrounded by coaxial cylindrical metallic gate of radius $R_s \gg R$. The Poisson equation relates the potential $\phi(z)$ at the surface of SWNT to 1D charge density $\rho(z)$ in SWNT and the potential $\Phi(z)$ of the gate,

$$\phi_q = \phi_q^{(p)} + \phi_q^{(g)} = U_q \rho_q + M_q \Phi_q,$$

with

$$U_q = \frac{2}{\kappa} \left\{ I_0(qR) K_0(qR) - I_0^2(qR) \frac{K_0(qR_s)}{I_0(qR_s)} \right\},$$

$$M_q = \frac{I_0(qR)}{I_0(qR_s)}.$$  \hspace{1cm} (1)

Here $\kappa$ is the dielectric constant of the media and $I_0$, $K_0$ are the modified Bessel functions. The kernel (2) describes the long-range Coulomb interaction, $U(x) = 1/\kappa |z|$, for $R \ll |z|$. The interaction is screened at large distances $|z| \gg R_s$, so that $U_q(q = 0) = 1/C = (2/\kappa) \ln(R_s/R)$, $C$ being the capacitance of SWNT per unit length.

In equilibrium, the charge density is related to the energy $\bar{E}_0(z) = E_0(z) - E_F$ of the gapless point (charge neutrality level) of graphite $E_0$ counted from the Fermi level $E_F$. At zero temperature we obtain [8],

$$\rho(z) = e \int_0 dE \nu(E),$$  \hspace{1cm} (4)

with the density of electronic states $\nu$ and $e > 0$. Equation (4) is valid if $\bar{E}_0(z)$ varies slowly on the scale of the Fermi wavelength.

We restrict our consideration to low energies $|\bar{E}_0(z)| < \Delta^{(1)}$ and neglect the effect of higher 1D subbands ($\Delta^{(1)}/(h v_F/R) = 1/2$ for metallic/semiconducting SWNT). The densities of states in metallic and semiconducting SWNTs are given by,

$$\nu_M = \frac{4}{\pi h v_F}, \quad \nu_S = \frac{4}{\pi h v_F} \frac{|E| \Theta(|E| - \Delta)}{\sqrt{E^2 - \Delta^2}},$$

$$\Delta = h v_F/3R$$ being the half energy gap in semiconducting SWNT, and $v_F \simeq 8.1 \times 10^5$ m/s being the Fermi velocity.

In turn, the charge neutrality level is related to the electrostatic potential (1),

$$\bar{E}_0(z) + e \phi(z) = \text{const}.$$  \hspace{1cm} (6)

In what follows we will solve Eqs. (1), (4), (6) self-consistently for two situations.

2. Metal-nanotube contacts

We consider first metallic SWNT ($z > 0$) contacting the $xy$-plane of a metallic electrode ($z < 0$) at $z = 0$ (Fig. 1(a)). The potential of the $xy$-plane is chosen to be zero, and the potential of the gate is $V_g$. Assuming the metal electrode to be a perfect conductor we neglect the band bending in it. The charge neutrality point in SWNT at $z \to +0$ is shifted from the Fermi level by an amount $\bar{E}_0(+0) = \Delta W$ equal to the difference $\Delta W = W_M - W_{NT}$ of the work functions of the electrode and SWNT, so that const = $\Delta W$ in Eq. (6).
Fig. 1. Two systems under consideration: SWNT contacted to a metal (a) and heterojunction between metallic (M) and semiconducting (SC) nanotubes (b). The potential $V_g$ is applied to the gate electrode.

In order to fulfill the boundary condition $\varphi(r, z = 0) = 0$ for the potential in $xy$-plane we solve Eqs. (1), (4), (6) with antisymmetric sources. The solution for the charge density has the form,

$$\rho_q = -\frac{2i q}{q^2 + \alpha^2} \frac{(\Delta W/e - M_q V_g)}{U_q + 1/(e^2 v_M)}$$

with $\alpha \to 0$.

To evaluate the solution in the intermediate distance range $R \ll z \ll R_s$ we use the asymptotic $U_q = -(2/\kappa) \ln(qR)$. The deviation of the charge neutrality point from the Fermi level is given by,

$$\bar{E}_0(z) = U_{\nu} \ell_{\omega} W - e V_g \frac{1}{g \ln(R_s/R)}$$

where $c = (1/\pi) \int dx |I_0(x)|^{-1} \simeq 1.33$ and the Coulomb interaction $g = 2e^2 v_M/\kappa$ is supposed to be strong, $g \gg 1$. One can infer the interaction parameter $g \simeq 5$ from the experimental data [4, 6].

Using the asymptotics $U_q = (2/\kappa) \ln(R_s/R)$, and $M_q = 1$ for $|q| \ll R_s^{-1}$ we obtain the result at large distances $z \gg R_s$,

$$\bar{E}_0 = \frac{\Delta W - e V_g}{1 + g \ln(R_s/R)}.$$  

In this regime the electric field of the electrode at $z = 0$ is well screened by the gate and the charge density in SWNT can be effectively controlled by the gate voltage.

3. All-nanotube heterojunctions

We will consider the heterojunction joining metallic ($z < 0$) and semiconducting ($z > 0$) SWNTs at the angle $\pi$ (Fig. 1b) and perform its modeling using the formalism of Section 1. Since experimental values [6, 7] of the heterojunction conductance are typically small, $G/(e^2/h) \sim 10^{-3} - 10^{-2}$, we will assume low transparency $T \ll 1$ of the barrier at the interface between the nanotubes. In this case the electrons in SWNTs are described by the equilibrium Fermi distribution, even if the voltage $V$ is applied to the junction. Assuming that the electrodes are connected to SWNTs at large distances $d \gg R_s$ from the junction we obtain $\varphi^{(g)}(z) = V_g$. This allows us to rewrite Eq. (6) as follows,

$$\bar{E}_0(z) + e \varphi^{(g)}(z) = \mu \pm e V/2.$$  

$$\bar{E}_0(z) + e \varphi^{(g)}(z) = \mu \pm e V/2,$$
where $\mu \pm eV/2$ are the electro-chemical potentials for holes in metallic and semiconducting SWNTs and $\mu = \Delta W - eV_g$. Note that the problem becomes non-linear due to the presence of semiconducting SWNT.

Figure 2(a) shows the result for the SB height $u$ defined as the minimum energy of electron or hole excitation required to transfer the elementary charge across the junction in the absence of tunneling through the SB. The SB height shows pronounced asymmetry as a function of the bias voltage. At small electrochemical potential the SB height at forward (reverse) bias is determined by the energy of the valence (conduction) band in the "bulk" of semiconducting SWNT, $x \to \infty$, with respect to the Fermi level, $u^{(b)}_{f(r)} = \Delta \mp \tilde{E}_0(\infty)$, with $\tilde{E}_0(\infty) = \mu + eV/2$. This corresponds to straight portions of contour lines in the lower part of Fig. 2 (a). In particular, the positive $V_+$ and negative $V_-$ threshold voltages at which the SB vanishes ($u^{(b)}_{f(r)} = 0$) are given by $eV^{(b)}_{\pm} = \pm 2\Delta - 2\mu$.

Straight portions of contour lines (Fig. 2(a)) are interrupted by cusps. At forward bias the cusps occur along the line $eV = 2\mu$ (Fig. 2(a)) where the charge density in metallic SWNT and the band bending change sign. Above the point of a cusp, the height of a SB at forward (reverse) bias corresponds to the energy of the valence band at the interface of SWNTs, $x = 0$, counted from the Fermi level of semiconducting (metallic) nanotube, $u^{(f)}_{\pm} = \Delta - \tilde{E}_0(\pm0)$ . The threshold voltages $V^{(f)}_{\pm}$, correspond to suppression of a SB at the interface, $u^{(f)}_{\pm} = 0$. Note that at $\mu \sim \Delta/2$ the positive threshold $eV^{(f)}_+ \simeq \Delta$ is relatively insensitive to the electro-chemical potential, Fig. 2(a). This can be used for a rough estimate of the gap from experimental data.

The asymmetry of the $I - V$ characteristics and threshold voltages has been discovered
in recent experiments [6, 7]. According to the data of Ref. [6], both the thresholds $V_+$, $V_-$ shift upwards with the gate voltage. Moreover, the positive threshold shifts less than the negative one. Such behavior is consistent with our model in the regime of moderate doping, $0.5 < \mu / \Delta < 1.8$ (Fig. 2). However, the blockade region of $3-4$ V detected in the experiment is somewhat wider than the theoretical estimate, $V_+ - V_- < 6.5 \Delta \simeq 2$ eV. The extra voltage drop could be due to potential disorder in semiconducting SWNT [9] and/or an additional SB at the interface between semiconducting SWNT and metallic electrode.

We now check the model against the experimental data of Ref. [7]. The measured width of the blockade region, $0.5-0.7$ V, agrees with the theoretical estimate. The gap in semiconducting SWNT, $\Delta \simeq eV_+$, evaluates at $\Delta = 0.19, 0.29$ eV for the two devices studied [7]. These values are in the expected range $\Delta \sim 0.25-0.35$ eV [2, 3]. A smooth onset of the current over the range $\sim 0.1-0.3$ eV around threshold voltages is naturally associated with quantum tunneling through a “leaky” SB (thermal energies are much smaller, $k_B T \simeq 5$ meV). Finally, the step-like feature of the current under reverse bias almost certainly corresponds to the reconstruction of the band profile due to the Fermi level entering the conduction band of semiconducting SWNT.

4. Conclusions

To conclude, we have studied the electronic properties of carbon nanotube heterojunctions and provided explanation for the main features of recent experimental data [6, 7]. Due to the long-range Coulomb interaction, the charge transfer phenomena in one-dimensional nanotube systems differs drastically from those in conventional semiconductor heterostructures. This creates new challenges in the design of novel electronic devices.

References

[8] We assume that $\rho$ is averaged over few atomic distances. Our approach does not describe phenomena at atomic lengthscale, for instance, the Friedel oscillations.
Semimagnetic resonant tunneling diodes for electron spin manipulation

Th. Gruber, M. Keim, R. Fiederling, G. Reuscher, A. Waag, W. Ossau, G. Schmidt and L. Molenkamp
Physikalisches Institut, EP III, Universität Würzburg, 97074 Würzburg, Germany

Abstract. Recently, efficient electrical injection of spin polarized electrons into GaAs has been accomplished by using a semimagnetic II–VI single layer, namely BeMnZnSe, as a spin aligner [1]. In such spin aligner materials, the orientation of the electron spins can only be controlled by varying the magnetic field. Here, we introduce an approach aiming at manipulating spins by an external voltage. For that we developed BeTe/ZnMnSe/BeTe semimagnetic resonant tunneling diodes as a spin injector into nonmagnetic III–V material. By changing the resonance condition from a spin-up to spin-down Zeeman level, it should be possible to actually switch from one spin orientation to the other. An AlGaAs/GaAs/AlGaAs p-i-n Light Emitting Diode was used to detect the efficiency of the spin injection, because the circular polarization of the electroluminescence is a direct measure of the spin polarization of the injected electron current. Although no spin-splitting of the bound electron states in the ZnMnSe quantum well could be observed in current-voltage-measurements, electroluminescence from the LED revealed a high circular polarization of up to 70%, indicating a strongly spin-polarized electron current. The use of a semimagnetic RTD as a spin-switch device would represent a further important step towards spin manipulation in semiconductors.

Introduction

Until today, common electronic devices are operated with the charge of the electrons only. But electrons also carry spin, and since several years theory predicts immense advantages, if one could employ this attribute in electronic devices. In spite of great efforts dedicated to the development of such spin-coherent electronics, very basic problems seem to oppose a real breakthrough in this field. One major task is to find an efficient spin injector. Much work focused on utilizing ferromagnetic metallic contacts, but it proved extremely difficult to show spin injection into a semiconductor material. Only very small effects of spin-polarized injection from a NiFe contact have been reported until today [2, 3] (0.9% and 0.2% resistance change, respectively). Therefore more attention is recently being paid to another set of materials, i.e. diluted magnetic semiconductors, and very promising results have already been reported. Recently, we have been able to demonstrate a very efficient spin injection into non-magnetic GaAs via a single layer of the semimagnetic II–VI material BeMnZnSe [1]. A spin polarization of close to 100% in the BeMnZnSe together with a high quality II–VI/III–V interface and a small conduction band offset in that system are the prerequisites for efficient spin injection. In fact it has been shown theoretically that a spin polarization of less than 100%, as is the case for ferromagnetic metal contacts, will result in a very small degree of spin injection into a non-magnetic semiconductor [4]. A major limitation of this simple spin aligner approach, however, is that the spin orientation of the injected electron current can only be changed by reversing the magnetic field, which is a slow and dissipative process.

Here we report on the development of BeTe-ZnMnSe-BeTe resonant tunneling diodes, aiming at the fabrication of a voltage operated spin-switch device. The incorporation of
the magnetic Mn ions (with their magnetic moment arising from the half-filled d-shell) into ZnSe presents a semimagnetic system with a well known magnetic behaviour. Most significantly, the sp-d exchange interaction between the Mn ion spins and carrier spins leads to a giant effective $g$-factor of up to 100, and hence a large Zeeman-splitting in external magnetic fields [5]. Using ZnMnSe in the quantum well of a double barrier RTD and applying an external magnetic field therefore results in a splitting of every subband into two spin-split bands, each corresponding to one spin component (Fig. 1(a)). Theoretical considerations using the Transfer-Matrix-Method predict a spin-dependent transmission coefficient, leading to a spin polarized tunneling current [6]. Consequently, the orientation of the spin-polarization should be tunable by the voltage applied across the RTD.

**Experimental details**

The AlGaAs-GaAs p-i-n LED structures have been grown in a separate III–V MBE reactor, using Be and Si as dopants. The BeTe/Zn(Mn)Se Resonant Tunneling Diodes were then grown directly onto the LED in a neighbored II–VI reactor, being transferred through a UHV transfer system to avoid any surface contamination. Figure 1(b) shows the general setup of such a structure. Flux calibration was done by the RHEED oscillation technique and the lasting and very pronounced RHEED oscillations were an indication for the high quality of the growth and a very low density of extended defects at the II–VI/III–V interface, as has been shown earlier for the BeTe growth start on GaAs [7].

First we have investigated electron transport in the BeTe/Zn(Mn)Se RTDs by means of electrical methods and standard dc measurements were done to record their I/V characteristics. The heteropair ZnSe and BeTe is characterized by a type II band alignment with extremely high band-offsets of 2.3 eV in the conduction band and 0.9 eV in the valence band, which enabled us to study up to four clearly separated resonances with high peak to valley ratios (PVR) and additional features such as LO-phonon assisted tunneling [8]. A giant electro-optical anisotropy of the spatially indirect transition from the ZnSe conduction band into the BeTe barriers has been interpreted in terms of quantum-confined Pockels-Effect, providing further proof for the high structural quality of the samples, in particular of the BeTe/ZnSe interfaces [9]. The next step was to use combined electro-optical methods to learn more about the LED-RTDs, by analyzing the LED electroluminescence in Faraday geometry with a conventional CCD camera.
Results and discussion

The resonance peaks in the I/V characteristics of the semimagnetic LED-RTDs showed no direct indication of Zeeman splitting in external magnetic fields, which is consistent with the results of Keim et al.[10]. This could be due to the existence of a lateral fluctuation of the band offset because of “ZnTe” and “BeSe” terminated interfaces, leading to a broadening of the bound electron states in the quantum well. Another reason could be a dominating non-coherent, sequential tunneling current, leading to an efficient thermalization of carriers into the lower Zeeman level. In contrast to I/V, the polarization behaviour of the LED under electrical injection through the RTD shows a large effect of the semimagnetic quantum well: We found a very high degree of circular polarization of the emitted light, which is a direct measure of the spin-polarization of the injected electron current, since the recombination of the electrons with the split-off hh band in the non-magnetic GaAs qw follows the relevant selection rule $\Delta m_j = \pm 1$. In fact no hh transition has been observed in the samples. Figure 2(a) shows the magnetic field dependence of the circular polarization for a RTD with 10% Mn in the qw and the corresponding non-magnetic reference structure at 1.6 K and an applied voltage of 1.9 V. While the magnetic sample reveals a polarization of up to 70%, the non-magnetic reference emits almost unpolarized light, and its small polarization is due to the intrinsic $g$ factor of the GaAs qw. The increase and consequent saturation of the polarization of the magnetic RTD with increasing magnetic field can be theoretically accounted for by considering the typical behaviour of the Zeeman splitting in diluted magnetic semiconductors. This is an important indication that the subbands in the RTD really split in magnetic fields, although this could not be observed in pure electrical measurements. We also examined the behaviour of the polarization degree with respect to the applied voltage, especially around the first resonance in the I/V characteristic of the RTD. Such a result is shown in Fig. 2(b) for a fixed magnetic field of 5.5 T. Around the first resonance, the spin polarization is sharply decreasing from 80% down to 60%. Presently its unclear whether the decrease in polarization with increasing bias voltage is due to the expected resonance effects, or just due to a heating of the Mn system at higher currents, leading to a smaller spin polarization. Even though no clear effect of the 2 Zeeman resonances could be detected at the present stage, semimagnetic BeTe/ZnMnSe RTD obviously

![Figure 2](image_url)

**Fig. 2.** (a) Circular polarization vs magnetic field and (b) circular polarization and current vs voltage.
presents a very efficient spin injector into non-magnetic GaAs. Further work has to be done in order to optimize the resonant tunneling structures.

References


Resonant tunneling diodes based on stacked self-assembled Ge/Si islands

O. G. Schmidt†, U. Denker‡, O. Kienzle§, F. Ernst§, R. J. Haug‡ and K. Eberl†
† Max-Planck-Institut für Festkörperforschung,
Heisenbergstraße 1, 70569 Stuttgart, Germany
‡ Institut für Festkörperphysik, Universität Hannover,
Appelstrasse 2, 30167 Hannover, Germany
§ Max-Planck-Institut für Metallforschung,
Seestraße 92, 70174 Stuttgart, Germany

Abstract. We present a new concept and first results of resonant tunneling diodes based on self-assembled Ge/Si islands. The proposed structure consists of closely stacked and vertically aligned Ge islands which create vertical channels with energetically deep thermalization layers and high Si double barriers for holes in the valence band. The current–voltage (I–V) curve of such a layer sequence shows two resonances which we attribute to the heavy-heavy hole and heavy-light hole (lh) transition. The lh resonance is pronounced and negative differential resistance is conserved up to over 45 K. Magnetic field dependence of the resonances suggest that the tunneling current through the structure is of 2-dimensional character.

Introduction

Self-assembled Ge/Si islands have been of broad interest during the last years and much effort has been put into their structural [1–5], vibrational [6] and electronic investigation [7–9]. Despite the large effort to understand their fundamental properties, devices incorporating Ge/Si islands as active material are only scarcely found. RTDs with SiGe/Si quantum wells are commonly found in the literature [10, 11]. However, their performance is rather poor, yielding negative differential resistance only up to moderate temperatures around 150 K. Valence band (VB) offsets of typical RTDs are about 200 meV (Ge concentrations of 20%) and thermal quenching plays a major role at higher temperatures. In this contribution we propose a new concept for resonant tunneling diodes (RTDs) which makes use of self-assembled Ge islands. The purpose of the concept is two-fold: First, the large valence band offsets of Ge islands are used to create deep Ge rich vertical channels with high Si double barriers. Second, vertically and self-aligned Ge/Si island stacks offer the possibility of perfect side passivation.

1. Results and discussion

Figure 1(a) shows a cross-section transmission electron microscopy (TEM) image of a typical active layer arrangement in quantum well RTDs. Two thick Si$_{1-x}$Ge$_x$ quantum wells at the bottom and the top serve as thermalization layers for holes. The double barriers are created by a thin Si$_{1-x}$Ge$_x$ quantum well sandwiched between two Si barriers. The schematic band edge alignment for the VB is given next to the TEM image.

A new concept is presented in Fig. 1(b). It is based on the vertical self-alignment of closely stacked Ge/Si islands, as has been investigated in detail in the literature [4, 5]. In this case the thermalization layers are created by very closely stacked Ge islands yielding small minibands due to strong electronic carrier...
coupling. The two Si barriers are formed by leaving a slightly thicker spacer layer between the 5-th and 6-th as well as the 6-th and 7-th island layer. The schematic band edge alignment (without bias and doping) yields much larger valence band offsets than for the quantum well case. A slight modification of this concept can be found in Fig. 1(c), where the first two island layers are substituted by a thick SiGe quantum well.

2. Samples and experimental setup

We concentrate our investigation on one sample which was grown by solid source molecular beam epitaxy. The exact growth procedure follows the concept of Fig. 1(c): On a p$^+$ Si(001) substrate a 100 nm Si:B (1 × 10$^{19}$) layer is grown followed by a 12 nm thick Si$_{0.82}$Ge$_{0.18}$ quantum well. After that a stack of 4 bilayers of 6 ML Ge and Si layers is deposited, where the Si spacer layer between the 1-st and 2-nd island layer is 9 nm and all other Si spacers are only 5 nm thick. The structure is finished with a 150 nm thick Si:B (1 × 10$^{19}$) cap. The growth temperature for the Ge islands was 600 $^\circ$C, resulting in low density and large dome-like Ge islands with diameters of about 80 nm [7]. A schematic of the layer sequence is given as an inset in Fig. 2. The sample is then processed into mesas (≈300 nm wide) using optical lithography and wet-chemical etching. For such diameters we expect less than 5 island stacks in the mesa. Electrical measurements are carried out in a 3He evaporation cryostat equipped with a 12 T magnet.
3. Electrical characterisation and discussion

The I–V curve of the RTD is presented in Fig. 2 (solid curve) and shows two resonances, one at 0.49 V, which exhibits negative differential resistance (NDR) and a second shoulder at about 0.34 V. The two features can be explained by resonant tunnelling through the hh and lh subbands of the middle Ge island. In fact, the I–V characteristic looks very similar to what has been observed for conventional RTDs using Si/SiGe quantum wells [10]. The similarity between our island and the QW RTDs suggests a 2D density of states in the Ge islands. This is a reasonable assumption since the lateral extension of these islands is much larger than the de-Broglie wavelength of charge carriers. Figure 2 also shows (upper dotted curve) that NDR is conserved up to 45 K. Information about the dimensionality of the tunneling structure can be obtained by magnetic field dependences of the resonant current peaks. The I–V curve for a magnetic field \( B = 12 \, \text{T} \) parallel to the current is also given in Fig. 2. It is striking that only a minor voltage shift of about 4 mV is observed for the resonance at 0.49 V. This is characteristic for 2D structures and in clear contrast to a 0D quantum dot, where we would expect a pronounced shift of the resonance peak with increasing magnetic field.

Although our first results look very promising we want to address some problems faced during the fabrication of such RTDs. Recently, we showed that the degree of material intermixing in upper layers is more pronounced [9]. Additionally, penetrating strain fields and different island sizes are likely to modify the electronic band structure in the island stacks and in the sandwiched Si barriers. All these effects make it very difficult to produce vertically homogeneous islands and hence to evaluate the exact band edge alignment. On the other hand, it is well known from QW RTDs that a careful tuning of the band edge alignment is essential to obtain good device performance. Hence, the fundamental electronic properties of Ge/Si island stacks need to be investigated further to improve the promising RTD results presented in this paper.
4. Conclusion

In conclusion, we have introduced a new concept to fabricate RTDs based on stacked Ge/Si islands. The concept exploits the effect of vertical self-alignment and creates vertical Ge channels with larger valence band offsets than in conventional Si/SiGe quantum well RTDs. A first sample shows pronounced resonance peaks in the I–V curve, which are attributed to the hh and lh resonant tunneling currents. The lh resonance is distinct and exhibits negative differential resistance for temperatures larger than 45 K. Magnetic field dependent measurements suggest a 2D density of states in the Ge islands. Problems in the fabrication of homogeneous islands are addressed.

References


Decrease of MODFET channel conductivity with increasing sheet electron concentration

J. Požela, K. Požela and V. Jucienė
Semiconductor Physics Institute, A. Goàtavio 11, 2600 Vilnius, Lithuania
e-mail: pozela@uj.pfi.lt

In modulation-doped field-effect transistor (MODFET) structures, spatial separation of carriers from their parent donors increases electron mobility and enables a modulation doping level with donors, and, consequently, electron concentration in a MODFET channel to be enhanced. Both these factors enhance transconductance and operation speed of MODFET’s. There are a lot of attempts to improve MODFET parameters by increasing the modulation doping level with donors.

As is known, AlGaAs/GaAs/AlGaAs and AlGaAs/InGaAs/GaAs MODFET’s with the cutoff frequency as high as 400 GHz are created. But the further improvement of high-speed MODFET parameters is restricted because of a decrease of electron mobility with increasing a doping level of the structure.

In the paper, the factors responsible for limitation of MODFET channel conductivity enhancement with increasing sheet electron concentration are considered.

Using the dielectric continuum approximation [1] the calculations of scattering rates of confined electrons by confined polar optical (PO) phonons depending on sheet electron concentration are performed.

A strange effect is observed: the heterolayer conductivity decreases with increasing the electron concentration in the layer. The decrease of mobility exceeds the increase of sheet electron concentration when $n_s > 5 \times 10^{15} \text{ m}^{-2}$.

Taking into account the electron degeneration, the scattering rate of an electron from the initial state in subband $i$ with the energy $E$ to final states in subband $f$ with the energy $E \pm \hbar \omega_v$ is written as

$$W_{if}(E) = \sum_v W_{ifv} \frac{1 - f(E - \hbar \omega_v)}{1 - f(E)} + W_{ifv} \frac{1 - f(E + \hbar \omega_v)}{1 - f(E)}$$

(1)

where $f(E)$ is the Fermi–Dirac distribution function, the superscripts $e$ and $a$ correspond to the phonon emission and absorption, respectively. The inverse electron life time $\tau_i$ in the state $E$ of subband $i$ limited by optical phonon scattering can be determined as

$$\frac{1}{\tau_i(E)} = \sum_f W_{if}(E).$$

(2)

For estimation of the electron mobility limited by PO phonon scattering we involve the life time $\tau_i(E)$ as momentum relaxation time. Then the mobility in subband $i$ is determined as

$$\mu_i = \frac{2}{m} \left( \frac{1}{\tau_i(E)} \right)^{-1}$$

(3)
where the brackets $\langle \rangle$ mean the average value:

$$
\langle A \rangle = \frac{\int A f(E) dE}{\int f(E) dE}.
$$

The average electron mobility in the QW is

$$
\mu = \sum_i \mu_i \frac{n_{si}}{n_s},
$$

(4)

where

$$
n_{si} = D \int_{E_{si}}^{\infty} f(E) dE
$$

(5)

is the concentration of electrons in subband with the bottom energy $E_{si}$, $D = m/\pi \hbar^2$ and

$$
n_s = \sum_i n_{si}.
$$

In Fig. 1 the calculated electron mobility as a function of sheet electron concentration $n_s$ in the Al$_{0.25}$Ga$_{0.75}$As/GaAs/Al$_{0.25}$Ga$_{0.75}$As QW is presented.

One can see that, taking into account only electron-PO phonon scattering, calculated mobility decrease at 100 K exceeds the sheet electron concentration increase in the range of $n_s = (6-10) \times 10^{15} \text{ m}^{-2}$. As a result, the negative change of the channel conductivity (represented in Fig. 1 as the mobility multiplied by the electron concentration: $\mu n_s$) takes place.

It allows us to expect that the great electron-PO phonon scattering increase is the main factor responsible for the great decrease of the mobility and conductivity observed experimentally at high sheet electron concentrations in AlGaAs/GaAs/AlGaAs QW’s.

In the Al$_{0.25}$Ga$_{0.75}$As/GaAs/Al$_{0.25}$Ga$_{0.75}$As QW the alternate increase and decrease of the calculated channel conductivity $\mu n_s$ with increasing $n_s$ are observed. The channel QW...
conductivity of MODFET can be increased by increasing the doping level. The conductivity when \( n_s = 2.5 \times 10^{16} \text{ m}^{-2} \) exceeds the conductivity at \( n_s = 6 \times 10^{15} \text{ m}^{-2} \) (see Fig. 1).

Each cycle of the alternate decrease-increase conductivity change with increasing \( n_s \) corresponds to the change of the Fermi level position \( E_F \) with respect to the QW subband energy level \( E_s \). In the Al\(_{0.25}\)Ga\(_{0.75}\)As/GaAs/Al\(_{0.25}\)Ga\(_{0.75}\)As QW at 100 K, the Fermi level crosses two subband energy levels when the sheet electron concentration changes from \( n_s = 10^{15} \text{ m}^{-2} \) to \( n_s = 10^{17} \text{ m}^{-2} \). Correspondingly, two conductivity increase-decrease cycles are observed (see Fig. 1).

The insertion of a thin AlAs barrier into the GaAs QW center changes the electron subband energies. This admits a possibility for increasing the doping level and the maximal channel conductivity. This is shown in Fig. 1 where the calculated mobility \( \mu \) and channel conductivity \( \mu n_s \) for Al\(_{0.25}\)Ga\(_{0.75}\)As/GaAs/Al\(_{0.25}\)Ga\(_{0.75}\)As QW with an inserted thin AlAs barrier as functions of doping level are represented.

The increase of maximal doping limits determinates the possibilities of enhancement of high-speed parameters for Al\(_{0.25}\)Ga\(_{0.75}\)As/GaAs/Al\(_{0.25}\)Ga\(_{0.75}\)As MODFET’s.

References

Multiple-barrier resonant tunneling structures for application in a microwave generator stabilized by microstrip resonator

P. N. Lebedev Physical Institute, Leninsky pr., 53, 117924 Moscow, Russia

Abstract. One of the main goals of contemporary electronics is the expansion of device capability to high frequencies up to the terahertz range. In this paper the results of comparison investigation of vertical transport and high frequency oscillatory properties of double-barrier and triple-barrier resonant tunneling structures (DBRTS and TBRTS) combined with a microstrip resonator stabilizing circuit are presented. The I–V characteristic of measured TBRTS shows higher values of a peak-to-valley current ratio than the values measured for DBRTS in agreement with theoretical predictions. Microwave oscillations from semiconductor quantum well resonant-tunneling structures, stabilized by use of the microstrip resonator, are observed for the first time. The microstrip system, compatible with MBE methods, provides appropriate circuit conditions for realization of high frequency oscillations by use of planar active structures and looks rather encouraging for millimeter and submillimeter wavelength applications.

Double barrier and triple barrier resonant tunneling structures as microwave oscillators

The recent theoretical and experimental investigations of negative differential conductivity (NDC) effects in double barrier resonant tunneling structures (DBRTS) prove the extremely fast frequency response of charge transport (less than about 100 fs) [1, 2]. Systems, which are expected to be more efficient, are the asymmetric double- and triple-quantum-well structures [3–5]. A variation of the structure parameters in such systems allows one to control independently the current peak-to-valley ratio and peak current values, which is impossible in the case of DBRTSs. The numerical simulation data confirm [3–5] the higher efficiency of a triple-barrier resonant structure (TBRTS) compared with a double-barrier one. The peak current value \( J_p \) for the TBRTS may be significantly higher than for a DBRTS with the same current peak-to-valley ratio \( J_p/J_v \). This is due to sharp resonant properties of the inner interwell region and high electron transparency of the outer barriers, that is rather promising to achieve required power level and discernible oscillations at upper frequencies.

Resonant tunneling diode samples and microstrip resonator circuitry

The implementation of a resonant tunneling GaAs/AlAs quantum-well generator stabilized by use of the microstrip resonator is presented. Resonance-tunneling structures with one and two quantum wells (QW) were fabricated in the same MBE technique conditions. The structures were grown by MBE on semi-insulating GaAs(001) substrates in “Tsna-18” MBE system. Silicon was used as a donor impurity. The substrate temperature during growth was 630 and 690°C for GaAs and AlAs respectively. GaAs growth rate 0.58 µm/h, and the ratio of As-to-Ga equivalent beam pressures 15:1. Substrate temperature was changed
during growth interruption under arsenic stream. The duration of interruption has made 30 seconds. The quality of the heteroepitaxial film was carried out in situ by RHEED technique. As-stabilized \((2 \times 4)\) — reconstruction was observed.

On an \(n^+\)-GaAs buffer layer by a thickness 1 \(\mu\)m doped to \(2 \times 10^{18}\) cm\(^{-3}\), which acts as the lower contact layer and barrier for a diffusion of impurity from substrate, structure of barriers and quantum wells claded by spacer layers from undoped GaAs was grown. The undoped spacer layers inserted between the heavily doped electrodes and tunneling barriers are used to prevent the incorporation of segregated impurities into the active part of the structure during epitaxial growth, and to improve its frequency response [3, 6–9]. The thicknesses of lower and upper undoped layers have made 10 and 5 nm respectively. The upper contact layer consists of a 50 nm n-GaAs layer doped to \(2 \times 10^{17}\) cm\(^{-3}\) and 500 nm \(n^+\)-GaAs doped to \(2 \times 10^{18}\) cm\(^{-3}\). AlAs by a 3 nm thickness was used as barriers of resonance-tunneling structures. GaAs is a quantum well material. In case of DBRTS the width of QW has made 4 nm, and in case of TBRTS — 5 and 4 nm for the lower and upper quantum well respectively.

After MBE grow to the highly doped semiconductor layers 0.35 \(\mu\)m Au-Ni – (Au + 12 percent Ge) ohmic contacts were created. Resonant tunneling diode was formed as the mesa-structures with 16×16 \(\mu\)m\(^2\) area of upper contact. The other contact was formed sideways from the mesa-structure on lower \(n^+\)-GaAs layer. The system of coplanar contacts used provides the extremely low RC time delay in the negative-differential conduction (NDC) region of the current-voltage (I–V) curve due to a decrease in the capacitance and the series resistance of a device.

The asymmetrical quantum well resonant tunneling structures are investigated in combination with the microstrip resonator configuration providing a best circuit conditions for realization of high frequency (up to THz band) oscillations by use of planar active structures. In preparation for high-frequency experiments, the fabricated chips containing DBRTS mesas were mounted in a microstrip resonator. The resonator was designed as a quarter-wave tee coupled microstrip line with one end short-circuited [3].

**Comparison static vertical transport characteristics**

The measured current-voltage (I–V) characteristics of the representative DBRTS are shown in Fig. 1. The data reveal asymmetry in values of \(J_p/J_v\), which are equal to 4.1 and 2.4 at \(T = 77\) K for negative and positive bias applied to the top contact of mesa, respectively.

![Fig. 1. Measured current-voltage curve of the double-barrier resonant tunneling structure.](image-url)
The corresponding peak current densities are around $2.3 \times 10^2$ A/cm$^2$ and $3.2 \times 10^2$ A/cm$^2$ at 77 K and resonant bias values are $-292$ mV and $+435$ mV. The observed asymmetry is caused by different electrical properties of lower and upper cladding layers (mainly of spacer layers) of the structure and is characteristic of the regimes of MBE process. These results were used to estimate the real profiles of impurity distribution in the cladding layers.

![Graph](image)

**Fig. 2.** Measured current-voltage curve of the triple-barrier resonant tunneling structure.

The so obtained impurity distribution profiles in cladding layers were exploited for numerical simulations of the measured I–V characteristics of TBRTS, shown in Fig. 2. The modeling dependence agrees with the experimental data rather well and allows the interpretation of the observed features in the I–V characteristic of this asymmetric double-well structure. The I–V characteristic of measured TBRTS shows higher values of $J_p/J_i$ ratio around 5 (77 K) than the values around 4 (77 K) measured for DBRTS grown under the similar conditions in agreement with theoretical predictions. Obtained results are encouraging for numerical simulation and development of real efficient multi-barrier resonant tunneling systems with predictable resonant properties.

**Resonant tunneling microwave oscillations in DBRTS and TBRTS**

Microwave oscillations have been found from GaAs/AlAs TBRTS stabilized by a microstrip resonator for the first time. The microwave oscillations in the spacer-cladded TBRTS were observed in the microstrip resonator at frequencies around 1 GHz with negative or positive bias applied in any point of the NDC region (Fig. 2). The output power was about $10^{-5}$–$10^{-6}$ W. The stable monochromatic microwave oscillations were obtained at 77 K. The comparative analysis of the TBRTS and DBRTS statical and dynamical characteristics manifests the use of multiple quantum well-barrier resonant tunneling structures in narrow-band as well as in broad-band pulsed mm and sub-mm applications.

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**References**


Does the quasibound-state lifetime restrict the high-frequency operation of resonant-tunneling diodes?

M. Feiginov  
Institute of Radioengineering and Electronics RAS,  
11 Mokhovaya St., Moscow 103907, Russia  
e-mail: misha@mail.cplire.ru

Abstract. We have shown that, firstly, the response time ($\tau_{\text{resp}}$) of the resonant-tunneling diode (RTD) can be much smaller as well as much larger than the quasibound-state lifetime in the quantum well ($\tau_{\text{dwell}}$); secondly, the real part of the RTD conductance can be negative at the frequencies higher than the reciprocal $\tau_{\text{dwell}}$. The Coulomb interaction of the electrons in the quantum well with emitter and collector is responsible for the effects. A simple analytical expression for the impedance of the RTD has been derived and it is in fairly good agreement with experimental data.

1. Introduction

The resonant-tunneling diodes (RTD) on the basis of the double-barrier heterostructures are extensively studied for a long time. It was demonstrated that they work up to the frequency of several THz. The complicated numerical approaches to simulation of the dynamic response of RTD have been developed recently. They allow one to deal more or less accurately with the problem. Nevertheless, up to now there was not a simple analytical and physically clear approach, that would allow one to describe quantitatively the high frequency behavior of RTD. The problem has been solved in the present work. Also, it is widely believed that the RTD response time ($\tau_{\text{resp}}$) cannot be less than the quasibound-state lifetime ($\tau_{\text{dwell}}$) in the quantum well (QW). We show that it is not true.

All the results of the present work are obtained in the sequential tunneling approximation and the problems were treated self-consistently. The details of derivation could be found in [1]. Here we present just the new and main results, discussion and comparison with experiment.

2. The response time and conductance of RTD

The following equations have been derived for the linear response of RTD:

\[
\frac{\partial}{\partial t} + \frac{1}{\tau_{\text{resp}}} \delta N_{2D}(t) \propto \delta E_{fc}(t),
\]

\[
\frac{1}{\tau_{\text{resp}}} = \frac{1}{\tau_c} + \frac{1}{\tau_e(U_w)} + \frac{e^2 \rho_{2D}}{C} \left\{ \frac{1}{\tau_e(U_w)} - \left[ E_{fe} - E_{fw} \right] \frac{\partial}{\partial U_w} \left( \frac{1}{\tau_e(U_w)} \right) \right\},
\]

here $\tau_c$ and $\tau_e$ are the electron dwell times in QW due to the tunneling to collector and emitter, respectively; $e$ is the electron charge, $\rho_{2D}$ is the 2D density of states in QW, $C = \epsilon (L + d)/4\pi$ is the capacitance of QW per unit area, the effective emitter-well distance ($d$) is more than the emitter-barrier thickness by the Thomas–Fermi screening length and the half width of QW; $L$ is the similar well-collector distance, that includes...
Fig. 1. On the left are the plots of $G' \equiv \text{Re}(G)$ for different bias voltages (the upper 3 are for biases in PDC region and the lower one in NDC region) and on the right are the corresponding plots of $G'' - \omega C_{ec}$, where $G'' \equiv \text{Im}(G)$. The experimental points are taken from [2]. $C_{ec}$ was calculated in [2] by averaging $G''$ between 3.9 and 4.0 GHz. Continuous lines in the figures are $G'(\omega)$ and $G''(\omega)$ calculated with the help of (4). The necessary parameters were extracted as follows. The value of $G_{nonres}$ was extracted from the plots with bias 1.69 V: $G$ is independent on frequency in the case, i.e. $G'_{RT} \approx 0$. $G''_{RT}$ is known for all biases from the experimental points, $G'$ at large frequencies gives the ratio of $\tau_d/(L+d)$ and $\tau_{resp}$. As it follows from (4), $G'(\omega) = (G'(0) + G'(\infty))/2$ at $\omega = 1/\tau_{resp}$. It should be noted that the negative sign of $G'' - \omega C_{ec}$ at low frequencies and biases 1.4 V and 1.65 V corresponds to $C < C_{ec}$; and positive sign at 1.72 V corresponds to $C > C_{ec}$. 
also the thickness of the depletion region; $N_{2D}$ is the electron concentration in the QW, by $\delta$ we denote small variations, $E_{f e}$ is the collector Fermi level, emitter is supposed to be grounded ($\delta E_{f e} = 0$). $\tau_{\text{resp}}$ has the sense of the tunnel relaxation time of the fluctuation of $N_{2D}$. The first and the second terms in (2) describe the relaxation due to the electron tunneling to collector and emitter, respectively, and they give the electron dwell time in QW: $1/\tau_{\text{dwell}} \equiv 1/\tau_{c} + 1/\tau_{e}(U_{w})$. As the Fermi level in QW ($E_{f w}$) changes, the energy of the bottom of the 2D subband in the QW ($U_{w}$) also shifts due to the Coulomb interaction of the electrons in the QW with emitter and collector. As a result, firstly, an additional contribution in emitter-well current appears due to the change of the number of the free states in QW available for tunneling (third term). Secondly, the current changes owing to the variation of $\tau_{e}(U_{w})$ with the shift of the bottoms of the 2D subband in QW, that is described by the fourth term in (2). Variation of $\tau_{e}(U_{w})$ is significant, $\tau_{e} \to \infty$ when $U_{w}$ becomes lower then the bottom of the conduction band in the emitter; the variation of $\tau_{c}$ is supposed to be negligibly small. The factor before the figure brackets in (2) equals to $\delta U_{w}/\delta E_{f w}$ and its typical value is $5-10$. Due to the factor the third term in (2) is always much larger than the second one. The last term in (2) can increase as well as diminish $\tau_{\text{resp}}$. If the Coulomb effects are omitted ($C \to \infty$), then $\tau_{\text{resp}} = \tau_{\text{dwell}}$. The Coulomb interaction significantly changes $\tau_{\text{resp}}$.

Next the following equation has been derived, it relates $\tau_{\text{resp}}$ to the static differential conductance due to the resonant current ($G_{\text{RT}}^{0}$):

$$G_{\text{RT}}^{0} = \left[ 1 - \frac{\tau_{\text{resp}}}{\tau_{\text{dwell}}} \right] \frac{C_{\text{wc}}}{\tau_{c}}.$$  

where $C_{\text{wc}} = \epsilon/4\pi L$ is the well-collector capacitance. Eq. (3) gives possibility to get $\tau_{\text{resp}}$ in the static measurement of the I–V curve.

An expression for the linear conductance ($G$) of RTD has been derived also:

$$G(\omega) \equiv \frac{e\delta J_{\text{RTD}}}{\delta E_{f e}} = i\omega C_{\text{ec}} + G_{\text{RT}}^{0} \frac{1 + i\omega \tau_{c} d/(L + d)}{1 + i\omega \tau_{\text{resp}}} + G_{\text{nonres}}.$$  

$C_{\text{ec}} = \epsilon/4\pi (L + d)$ is the emitter-collector capacitance, $J_{\text{RTD}}$ is the RTD current, $G_{\text{nonres}}$ is the conductance due to nonresonant component of current, e.g., thermionic emission. The contact resistance should be connected in series with $G$ (4), if it is significant one.

3. Discussion

It follows from (3) that $\tau_{\text{resp}}$ is always less than $\tau_{\text{dwell}}$ in the positive differential conductance (PDC) region of the I–V curve. In the case of RTD with 3D emitter just the first three terms are left in (2) ($\tau_{e}(U_{w}) \approx \text{const}$ in the PDC region) and $\tau_{\text{dwell}}/\tau_{\text{resp}} \approx 5-10$ due to the third one, if $\tau_{e} \ll \tau_{c}$. $\tau_{\text{resp}}$ is always more than $\tau_{\text{dwell}}$ in the negative differential conductance (NDC) region, as it follows from (3), and $\tau_{\text{resp}}$ grows up with NDC ($\tau_{\text{resp}} \to \infty$ when $G_{\text{RT}}^{0} \to -\infty$).

The comparison of the frequency dependence of the RTD conductance (4) and the experimental data from [2] is shown in Fig. 1. The theory and experiment are in excellent agreement with each other, as it follows from the figure.

From (4) follows that RTD conductance can be approximated by RC-circuit in the low frequency limit, when $\omega \tau_{c} d/(L + d) \ll 1$ and $\omega \tau_{\text{resp}} \ll 1$:

$$G(\omega) \approx i\omega \tilde{C} + G_{\text{RT}}^{0} + G_{\text{nonres}}, \quad \tilde{C} = C_{\text{ec}} + G_{\text{RT}}^{0} \left[ \frac{d}{L + d} - \tau_{\text{resp}} \right].$$  

(5)
The analysis of (5) and (3) shows that $\tilde{C} > 0$, although it can be essentially less than $C_{ec}$. The low frequency C–V characteristic has specific features in the PDC and NDC regions. In the PDC region $\tilde{C}$ can have an increase (if $\tau_c d/(L + d) > \tau_{resp}$) or dip (if $\tau_c d/(L + d) < \tau_{resp}$). Both cases are real and they are observed in plenty of experiments, e.g., the increase was observed in [3] and dip in [2] (see Fig. 1 and subscript there), the experimental data are in very good quantitative agreement with (5). As a rule, $\tilde{C}$ has peak in the NDC region and it is also in good quantitative agreement with (5).

RTD conductance can be approximated by a different RC-circuit in the high frequency region ($\omega \tau_{resp} > 1$ and $\omega \tau_{dwell} \ll 1$):

$$G(\omega) \approx i\omega C_{cc} + G_{RT}^0 \frac{d}{L + d} \tau_{c} \omega + G_{nonres}.$$  \hfill (6)

$G' \equiv \text{Re}(G) \approx -dC_{wc}/(L + d)\tau_{dwell}$, if $G_{nonres}$ is sufficiently small and $G_{RT}^0 \gg C_{wc}/\tau_c$. In the case, $G'$ does not depend on static differential conductance and is substantiated by $\tau_{dwell}$ and the geometry of RTD only. At low frequencies the variation of bias ($\delta E_{fc}$) leads to a significant variation of $N_{2D}$ and $J_{RTD}$ changes in result. At the high frequencies the situation is different. $\delta N_{2D}$ is very small and its phase is shifted by $\pi/2$ with respect to $\delta E_{fc}$ (see Eq. (1)), but it leads to variation of the electric field distribution in RTD and, as a consequence, to variation of the charges in emitter and collector. The variation of the charges, in its turn, leads to variation of $J_{RTD}$ with the phase shift of $\pi/2$. The negative $G'$ appears precisely due to the two phase shifts. So, $G'$ can be finite and negative when $\omega \tau_{resp} > 1$ and it should be possible to use RTD as generator at such frequencies.

RTD conductance (4) has formally the form coinciding with RLC-circuit [4] in the intermediate frequency range ($\omega \tau_{resp} \lesssim 1$ and $\omega \tau_c d/(L + d) \ll 1$):

$$G(\omega) \approx i\omega C_{cc} + \frac{G_{RT}^0}{1 + i\omega \tau_{resp}} + G_{nonres}.$$  \hfill (7)

The “inductance” describes the delay of current with respect to bias [4] and the delay is $\tau_{resp}$ (“inductance” $l = \tau_{resp}/G_{RT}^0$) rather than $\tau_{dwell}$ ($l [4] = \tau_{dwell}/G_{RT}^0$).

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References

Soft breakdown in the bistable MOS tunnel structures

I. V. Grekhov, A. F. Shulekin, S. E. Tyaginov and M. I. Vexler
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. The S-shape reverse-bias characteristics of MOS tunnel structure on n-Si (d < 3 nm, N_d ≈ 10^{16} cm^{-3}) and their post-soft-breakdown transformation are analyzed. Turn-on voltages and holding voltages are found theoretically and measured. The change of these parameters with oxide damage is suggested to reflect a complicated interplay between the injection efficiency, energy distribution of injected electrons and conductivity of an inversion layer.

Introduction

MOS tunnel structures with 1–3 nm oxide thickness are now being intensively studied worldwide as components of future sub-0.1 µm-channel MOSFETs [1] and of several other interesting devices, e.g. tunnel emitter transistors [2]. A central point of these studies is the reliability limitation by a soft breakdown (SB) which is defined as an appearance of non-extending local breakdown spot [3]. Along with values of a transported charge-to-breakdown, commonly used as the measure of reliability [3], it is, of course, very important to obtain information about the transformations of structure characteristics in general. Special attention deserves the case of reverse bias, as corresponding to a conventional operational mode of most MOS tunnel devices.

1. Reverse current–voltage characteristics

The reversely biased MOS tunnel structures on n-Si were shown to exhibit bistable behavior [2, 4] (Fig. 1). That the nature of this effect is Auger ionization is known long ago [4], but neither the parameters of an S-shape segment, nor their change after the SB of the oxide have been studied in detail earlier. The purpose of this work is twofold: first, we aim to determine the parameters (turn-on voltage V_sw, holding voltage V_h and current), and secondly to clarify how these are changed after the soft breakdown in the oxide. A basic equation for a MOS structure under inversion is the balance equation:

\[ \int j_h dS + \int j_{diff} dS = J_{ext} + \int j_e (M - 1) dS \]  

for minority carriers, where \( j_h \) and \( j_e \) are densities of hole and electron tunnel currents, \( J_{ext} = (j_{ext} S) \) is eventual external hole supply, \( j_{diff} \) is density of hole diffusion current and \( M \) is carrier multiplication factor, \( S \) is for device area.

2. Model of electron energy relaxation

The process of hot electron thermalization in silicon can be divided into two stages (Fig. 2). The first one takes place just beyond the inversion layer and the second one proceeds in the space charge region (like in p-n-junction). Taking both these stages in mind, the multiplication factor can be written as:

\[ M = (1 + P)(1 + \gamma) \]
Fig. 1. Typical transformation of the reverse current–voltage characteristics (averaged current vs applied bias) of a MOS tunnel diode after the SB.

Fig. 2. Band diagram of an n-Si MOS tunnel structure under reverse bias.

\[ P = \frac{1}{j_e} \int_0^{+\infty} \frac{dR}{dE} P_{\text{Auger}} dE \]

\[ \gamma = a_0 q N_d w^2 \frac{1}{b_0 \epsilon_0 \epsilon_s} \exp \left( -\frac{b_0 \epsilon_0 \epsilon_s}{q N_d w} \right), \quad a_0, b_0 = \text{const} \]  

(3)

\( P \) denotes the effective quantum yield of Auger generation and \( \gamma \) of impact ionization (Fig. 3). In earlier studies, impact ionization was ignored. Integration over \( dE \) is to regard the energy distribution of injected electrons. The factor \( \gamma \) depends on the depletion layer width \( w \) (if \( V \gg U \) it is a function of just \( V \)).

Fig. 3. Interrelation between the insulator voltage \( U \), applied bias \( V \) and inversion layer potential \( \phi \). Relative contribution of impact/Auger ionization \( \gamma/(\gamma + P) \), and of inversion layer/depletion layer charge \( N_s/(N_s + N_{\text{depl}}) \).

3. OFF state

For an Al/SiO$_2$/n-Si structure, it is only the electron component that provides a total tunnel current in an OFF state (see Fig. 1 for notation) at moderate \( V \). In our thickness range, the inversion layer does not exist in an OFF state. For this reason, the large-area device
4. ON state

For a fresh and homogeneous structure, the parameters of the holding point (Fig. 4) may be found disregarding the contribution of depletion layer charge to the insulator voltage $U$. Further, in the ON state (Fig. 1), there is no impact ionization ($P > 0, \gamma = 0$). Therefore, we have for the voltage $U$ at holding point:

$$P(E_{\text{eff}}) = \beta^{-1}$$

$$E_{\text{eff}} = qU + q\sqrt{\frac{72}{11}} \left[ \frac{\hbar I}{\varepsilon_s \sqrt{q m_h}} \right]^{2/3} \left( \frac{U}{d} \right)^{2/3}$$

where $\beta$ is for the ratio of electron-to-hole tunneling currents. The second term in (5) regards the additional energy gain by monoenergetic [4] electrons while passing through inversion layer. In the ON state, higher from the holding point (Fig. 1), the insulator voltage and the diffusion current $j_{\text{diff}}$ increase. The ON state of a MOS tunnel structure is very sensitive to the oxide degradation (Fig. 4). The matter is that the inversion layer tends to become equipotential along the whole structure, even if the oxide is locally destroyed. The leakage in one place strongly influences neighboring area. Oxide breakdown may cause the local changes of $\beta$ and also the (local) changes in the transport mechanism which lead to the alterations of interrelation $E(U)$.

5. Turn-on voltage

The value of a turn-on voltage (Fig. 5) is predominately determined by the impact ionization (see Fig. 3). In fact, the major contribution toward the insulator voltage is done by holes, while the extension of a depletion area was shown to play a minor role. Due to hole generation by impact ionization, the insulator voltage grows with increasing $V$ in the OFF state. Except for the very weak inversion, injection efficiency $\beta$ only insignificantly depends on $U$. Turn-on voltage is sensitive to the oxide degradation through the change of $\beta$. 

Fig. 4. Parameters of holding point of MOS tunnel structures for the case of low $N_d$. Holding voltages $V_h$ are by $\sim 0.7$ V lower than $\phi_{\text{hold}}$. 

operates as a conjunction of multiple quasi independent MOS structures. Defect area, if any, turns to be isolated from the rest part of structure. As a result, only minor changes of the current–voltage curve after the SB can be seen (Fig. 1). This is in contrast to the accumulation case when the electrical conductivity along the Si/SiO$_2$ interface is large.
Fig. 5. Turn-on voltage of MOS tunnel structures. Its value after the oxide soft breakdown is limited by the breakdown of space charge area.

and $P$. In fact, calculations show that both $P$ and $\gamma$ give a substantial contribution toward the multiplication factor $M$ (Fig. 3). With decreasing $d$, $\beta$ decreases and $V_{sw}$ therefore increases.

6. Conclusion

The transformations of reverse current–voltage characteristics of n-Si MOS tunnel structures resulting from a soft breakdown of the oxide layer have been considered. After the soft breakdown, both the turn-on voltage and the holding voltage have been shown to increase. Three factors are responsible for this: (a) decrease of MOS emitter injection efficiency with a reduction of the oxide thickness; (b) decrease of electron energy and quantum yield of Auger ionization inside the breakdown spot(s); (c) strong current crowding in the thinnest device sections.

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References

Phase coherent electron transport in open quantum dots and quantum dot arrays

J. P. Bird†, R. Akis†, D. K. Ferry†, M. El Hassan†, A. Shailos†, C. Prasad†, L.-H. Lin‡, N. Aoki‡, K. Nakao‡, Y. Ochiai‡, K. Ishibashi§ and Y. Aoyagi§

† Department of Electrical Engineering & Center for Solid State Electronics Research, Arizona State University, Tempe, AZ 85287-5706, USA
‡ Department of Materials Technology, Chiba University, 1-33 Yayoi-cho, Inage-ku, Chiba 236-8522, Japan
§ The Institute for Physical and Chemical Research (RIKEN), 2-1 Hirosawa, Wako-shi, Saitama 351-0198, Japan

Abstract. Recent studies of coherent electron transport in open quantum dots and quantum dot arrays are reviewed. Our interest focuses on the connection between the quantum and semi-classical descriptions of transport in these structures, which provide ideal systems for the experimental study of quantum chaos.

1. Introduction

An important issue in the study of quantum chaos concerns the connection between the discrete level spectrum of a strongly confined quantum system and the periodic-orbit structure of its corresponding classical counterpart. Semiconductor quantum dots [1, 2], consisting of a sub-micron-sized cavity coupled to quantum point contact leads, are ideally suited for the study of this problem. The discrete density of states in these dots remains well resolved in experiment, even with the point contacts configured to support a number of propagating modes [3]. At temperatures well below a degree Kelvin, the magneto-resistance of these devices exhibits highly regular and reproducible fluctuations, which may be interpreted as arising from a spectroscopy of the discrete density of states of the dot [3–5]. Equivalently, the oscillations may be associated with the interference of a small number of semi-classical orbits within the dot. These transport features therefore provide a connection between the density of states of the dot and its periodic orbits. In this invited contribution, we review our recent studies of coherent electron transport in open quantum dots. We discuss how, by modifying the details of the coupling between the dot and its quantum mechanical leads, it is possible to select different wavefunction states, giving rise to directly measurable transport results. We also briefly discuss our more recent studies of open dot arrays [6], which reveal evidence for collective transport signatures when the coupling between the dots is sufficiently strong.

2. Experimental details

Both the fabrication and basic characterization of the devices we study are described in detail elsewhere [5]. Split-gate quantum dots and quantum dot arrays were formed on the surface of GaAs/AlGaAs heterojunction wafers using electron-beam lithography and metal-deposition techniques. At 4.2 K, the carrier density of the samples was found to vary from $3.8 - 5.5 \times 10^{11} \text{cm}^{-2}$, while the electron mobility was as high as $1,000,000 \text{cm}^2/\text{V}s$.  

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3. Transport studies of single dots

In Figure 1, we show typical results of measurements of the conductance of single dots as a function of magnetic field and gate voltage. At the low temperatures considered here, electron phase coherence is maintained over long distances [7, 8] and the discrete level spectrum of the dot remains well resolved, in spite of the coupling that exists between the dot and the quantum point contact leads [3]. The conductance fluctuations observed in Fig. 1 then result as the magnetic field or the gate voltage is used to sweep the discrete states of this spectrum past the Fermi level [3–5].

An important feature of the conductance fluctuations shown in Fig. 1 is their highly regular nature. This property is confirmed by a Fourier analysis of the fluctuations, which reveals the presence of a small number of dominant frequencies at discretely separated values [3, 5]. This characteristic suggests that transport through these dots is dominated by a small number of orbits. Furthermore, the experimental finding that the frequency
content of the fluctuations does not depend sensitively on gate voltage points to the highly stable nature of these orbits [5]. Temperature dependent studies show that the fluctuations disappear on warming to above a degree Kelvin, indicating they are built up in a highly recursive process in which electrons undergo multiple traversals of the same basic orbits while maintaining their phase coherence [5]. In support of this notion, it has been found possible to reproduce the dominant experimental frequencies in numerical simulations of classical cavities in which electrons are assumed to be injected into the dot in a highly-directed, or collimated, beam [9]. The collimation is generated by the quantum point contact leads, which are configured to support only a small number of one-dimensional modes. Electrons entering these leads from the external reservoirs may therefore only do so by matching their transverse momentum component to one of the discretely quantized values within the contact itself.

While the discussion above attributes the conductance fluctuations to an interference effect involving a small-number of semi-classical orbits, they may also be understood to arise from quantum-mechanical mode-matching considerations between the central dot and the one-dimensional leads [3, 4]. According to this interpretation, the fluctuations provide a “spectroscopy” of the discrete level spectrum of the open dot. The crucial point here is that the use of quantum point contacts to couple the dot to the external reservoirs does not obscure its discrete energy spectrum. Rather, it results in a small set of dot states being preferentially excited in transport. The connection between the eigenstates of a closed dot and the conductance of its open counterpart can be seen in Fig. 2 (for further details on the numerical simulations we refer the interested reader to [5, 9]). Note how the energy levels of the isolated dot shift almost linearly over the entire range of gate voltage shown. A similar variation is also apparent in the conductance contour of the open dot; the linear striations that run through this grayscale plot clearly follow the motion of certain closed-dot states, indicating that these states play a strong role in mediating transport through the open dot. Crucially, however, we note that not all states of the isolated dot give rise to a
marked modulation of the conductance of its open counterpart. Instead, the conductance provides a filtered probe of the density of states of the closed dot. As we discuss in greater detail in [4], similar considerations have also been found to apply to the results of magneto-conductance measurements. In these, the magnetic field sweeps the density of states of the open dot past the Fermi level and the resulting oscillations in the conductance also provide a spectroscopic probe of the open dot [4].

An important conclusion of these studies is that the transport properties of open dots are critically influenced by the details of their coupling to the external environment. (This conclusion is further suggested by studies of the phase-breaking time in these structures [8].) An interesting possibility is that it should therefore be possible to modify electron transport in the dots, simply by changing the orientation of their point contact leads. In order to investigate this possibility, we have fabricated dots whose leads are positioned at different points on their perimeter (Fig. 3) [3]. The conductance fluctuations in these dots show different frequency content, which we attribute to the role of the leads in coupling to different dot states [3]. Numerical studies show that the wavefunction in the dots is strongly scarred by the remnants of a small number of semi-classical orbits. Consistent with the discussion above, the scarring is thought to result from the interference of orbits that are preferentially excited through their coupling to the leads. As the gate voltage is varied, the scars are found to recur periodically with frequencies that correspond very closely to the dominant components of the conductance fluctuations in experiment. The simulations also show that the different fluctuation frequencies measured in the different dots is correlated to the recurrence of different kinds of scars in these geometries [3]. That is, it would appear that, simply by changing the nature of the coupling between the dot and its external environment, it is possible to excite different wavefunction states, giving rise to directly measurable transport results.

4. Transport studies of quantum dot arrays

In coherently-coupled arrays of dots, the transport behavior should be somewhat more complicated than that discussed above, since the details of electron interference in any one dot will now be determined by the nature of the collective coupling that exists to the other dots in the array. Motivated by this consideration, we have studied the nature
of coherent electron transport in linear quantum-dot arrays and in Fig. 4 we show the results of measurements of one such array. While in many regards the behavior shown here appears reminiscent of that found in studies of single dots, a Fourier analysis of the magneto-conductance fluctuations shown here reveals that, as the coupling between the dots is increased by opening the quantum point contact leads, a marked increase in the high-frequency content of the fluctuations occurs. We believe that this increase may signal an evolution from single-dot, atomic-like, transport to molecular-like behavior in the arrays. Further studies are currently underway to investigate this possibility, however.

5. Conclusions

Our recent studies of coherent electron transport in open quantum dots and quantum dot arrays have been briefly reviewed. These devices are particularly suited to experimental studies of quantum chaos, since they reveal a picture in which electron transport is dominated by the excitation of a small number of stable orbits. These orbits are selected by means of coupling conditions between the dot and its quantum mechanical leads. At low temperatures, where phase coherence is maintained over long distances, electrons that undergo multiple traversals of these orbits give rise to strong wavefunction scarring and associated transport results.

References


Current instability and shot noise in nanometric semiconductor heterostructures

V. Ya. Aleshkin†, L. Reggiani‡ and A. Reklaitis∗
† Institute for Physics of Microstructures, Nizhny Novgorod GSP-105, 603600, Russia
‡ Dipartimento di Ingegneria dell’ Innovazione and Istituto Nazionale di Fisica della Materia, Università di Lecce, Via Arnesano s/n, 73100 Lecce, Italy
∗ Semiconductor Physics Institute, Gostauto 11, 2600 Vilnius, Lithuania

Abstract. We investigate electron transport and shot noise in single barrier GaAs/AlGaAs heterostructure of nanometric sizes. The coupling between space charge and the dependence of the transmission coefficient on energy is found to provide the positive feedback which enhances shot noise and ultimately leads to a current instability of $S$-type. Theoretical results are in qualitative agreement with existing experiments and confirm recent Monte Carlo simulations evidencing shot-noise enhancement in GaAs/AlGaAs heterostructures.

Introduction

The aim of this work is to provide a theoretical basis for the understanding of the current-voltage (I–V) and shot-noise as measured by the Fano factor $\gamma = S_I/(2qI)$ (with $S_I$ the noise power and $q$ the unit charge) in single barrier semiconductor structures characterized by ultra-short distances between the emitter and the barrier. The theory is able to explain both sub and super Poissonian shot noise behaviors in terms of the interplay among tunneling, space charge, and ballistic transport. In particular we investigate the positive feedback between tunneling and space charge due to the dependence of the transmission coefficient on the energy of ballistic moving electrons. When this feedback is negligible, long range Coulomb correlations between current pulses dominate and shot noise is suppressed. When this feedback is strong enough, shot noise is enhanced and ultimately we observe the onset of an $S$-type I–V characteristic.

1. Results and discussion

The physical system we analyze is a single barrier structure, as depicted in Fig. 1. By taking a one-dimensional $x$-space and a three dimensional momentum space, it is assumed the presence of an applied voltage high enough so that carrier injection from the collector is negligible. Electron transport is then described by the following analytical model [1]. The distribution function of particles injected from the emitter is taken of Maxwell type with concentration and temperature independent of applied voltage. In the region between the emitter and the barrier carriers are subdivided into two groups: one group contains ballistic particles, which do not perform any scattering, and the other contains thermalised particles which performed at least one scattering. Tunneling is finally described by a quasi classical triangular barrier.

The theory concerns with a model material of static dielectric constant $\kappa = 12.9$ and parabolic conduction band with effective mass $m = 0.067m_0$, $m_0$ being the free electron
mass. These values correspond to GaAs, being this a material appropriate for an experimental validation of the results. The theory is based on five independent external parameters, respectively, injected carrier concentration \( n_b \), barrier energy \( U \), fraction of ballistic carriers \( \beta \), temperature \( T \), barrier-collector length \( L \). We note that in the present model the distance between the emitter and the barrier is not considered explicitly but it influences the current through the value of \( \beta \).

Accordingly, \( \beta \to 1 \), is associated with ballistic particles and corresponds to ultra-short emitter-barrier distances. By contrast, \( \beta \to 0 \), is associated with scattered particles, and corresponds to long distances. A limited series of a complete set of results is presented in Fig. 2 where transport and noise are investigated at different values of \( \beta \) (see Fig. 2(a)) and \( T \) (see Fig. 2(b)). The I–V characteristics are reported in the left scale of Fig. 2.

As a general trend, the current increases monotonically at increasing voltages to finally saturates at sufficiently high voltages. In the increasing region, the current exhibits a strong super-Ohmic behavior due to tunneling processes. In the saturation region the value of the current equals that of the current injected from the emitter, \( I_0 \). The reason for such a value of current saturation is the absence of any current flow to the emitter coming from both: (i) ballistic particles reflected by the barrier, and (ii) thermalised particle. Indeed, because of the high voltages ballistic particles pass over the barrier, and thermalized particles remain confined in the potential well just before the barrier. The value of the voltage corresponding to the onset for current saturation rises with the increase of \( n_b \), \( U \), \( \beta \) and \( L \), and with the decrease of \( T \). In the case of \( \beta \) and \( T \) in Fig. 2. To understand these behaviors we note that the current starts saturating always when the voltage drop between the emitter and the barrier, \( u \), is a little bit greater than \( U \). In the current saturation region, the ratio of the ballistic current to the total current is independent of \( n_b \) and it is determined only by \( \beta \). From Fig. 2 it is clear that the increase of \( \beta \) or the decrease of \( T \) leads to the increase of the maximum value of the differential conductance \( dI/dV \) towards I–V characteristics of S-type.

The dependence of the Fano factor on voltage is reported on the right scale of Fig. 2. One can see that \( \gamma \) exhibits minima and maxima. In all cases, at the lowest and highest voltages (above current saturation) \( \gamma = 1 \) indicating full shot-noise. In the intermediate region of voltages for the considered values of injection concentration \( n_b = 5 \times 10^{17} \text{ cm}^{-3} \), \( \gamma \) exhibits both a maximum, corresponding to shot noise enhancement, followed by a minimum corresponding to shot noise suppression. The maximum value of \( \gamma \) exhibits a dramatic increase with increasing \( \beta \) and/or decreasing \( T \) while the minimum value remains close to 0.5. Deviations from full shot noise are interpreted in terms of the voltage dependence.
of the collector lifetime $\tau_c$ associated with the rate equation for the fluctuation of the total number of carrier inside the sample controlled by the collector $\delta N_c$. When $\tau_c$ is positive, any $\delta N_c$ is damped and only shot-noise suppression due to long range Coulomb interaction is possible. When $\tau_c$ is negative, a positive feed-back between tunneling and space charge amplifies any $\delta N_c$ and shot-noise enhancement becomes possible. When this positive feedback is rather weak (i.e. $|\tau_c|$ is very long) the maximum of the Fano factor is absent, as seen in Fig. 2(a) by the curve for $\beta = 0.2$. The general trends are that the increase of $\beta$ and the decrease of $T$ rises the strength of the positive feedback and in turn enhanced shot noise becomes more pronounced as reported in Fig. 2. This positive feedback is ultimately responsible for a decrease of the electrical stability of the structure. Accordingly, when its strength is sufficiently strong (i.e. $|\tau_c|$ is very short), the I-V characteristics become of $S$-type as seen by the curve corresponding to $T = 150$ K in Fig. 2(b). It is well known, that a region where the differential conductance is negative is unstable under a constant applied voltage. Therefore, instead of being damped, fluctuations grow being only limited by boundary conditions. In the unstable region, the Fano factor loses of physical meaning. However, at voltages near to the instability the Fano factor increases dramatically and tends to infinity. We remark that shot noise is for these structures a very sensitive indicator of the fact that the system is moving towards an instability region, this situation resembling that of phase transitions.

**Fig. 2.** Current normalized to the saturation value $I_0$ and Fano factors vs applied voltage for the barrier structure in Fig. 1 with $U = 0.2$ eV for different $\beta$ (a) and for different $T$ (b).
2. Conclusions

A theoretical analysis of electron transport and shot noise in ultra-short single barrier structures is presented. Results are interpreted in terms of a positive feedback between Coulomb interactions and the dependence of the barrier transparency on energy for ballistic particles which, in turn, weakens significantly the damping of carrier number fluctuations. This positive feedback is responsible for shot noise enhancement in triangular barrier structure and if its strength is high enough we observe the onset of an $S$-type I–V instability. By contrast, when the strength of the feedback is weak shot noise suppression is observed. The general trends of the model developed here is in qualitative agreement with existing experiments [2] and numerical simulations performed with Monte Carlo techniques [3].

Acknowledgements

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References


Hopping transport equation for electrons in superlattices with vertical disorder

I. P. Zvyagin, M. A. Ormont and K. E. Borisov
Faculty of Physics, Moscow State University, 119899 Moscow, Russia

Abstract. We develop a theory of vertical hopping transport in doped superlattices with intentional vertical disorder introduced by controlled random variations of well widths. For structures with sufficiently large disorder, the vertical conductance (in the direction of the growth axis) is limited by phonon-assisted hopping between the wells. It is shown that due to quasi-equilibrium situation within the wells, the master rate equation for transitions between the electronic states of the structure can be reduced to a truncated rate equation for inter-well transitions only. At low bias, the solution of this rate equation is shown to be equivalent to finding total resistance of a quasi-one-dimensional network of resistances expressed in terms of integral transition rates between the wells. This network is generally different from the Miller–Abrahams network and contains multisite resistors.

Introduction

Superlattices with intentional disorder (SLIDs) in which vertical disorder (in the direction of the growth axis) was introduced by random controlled variations of well widths in the process of structure deposition were first discussed in [1] and were experimentally realized in [2]; vertical transport in such structures was studied both by optical methods, in particular, by stationary and picosecond luminescence spectroscopy (e.g., see [2]), and by direct measurements of the vertical conductance [3]. Optical experiments clearly showed that with increasing vertical disorder, localization of electronic states in the direction of the structure growth increases. It was argued that for small overlap of the wave functions of neighboring wells even for superlattices without intentionally introduced disorder, vertical transport can be due to phonon-assisted inter-well transitions (hopping) [4, 5]. Introduction of intentional disorder substantially enhances localization of electronic states; for short-period superlattices GaAs/AlAs with random fluctuations of well and barrier widths (from 1 to 3 monolayers), the onset of localization was studied in [6]. At low energies, the decay length of the wave function can become smaller than the monolayer width so that structures considered seem to be similar to a one-dimensional chain of localized states. For structures with sufficiently large disorder studied in [3], the miniband width was estimated to be smaller than the width of the level distribution; therefore one expects that the states are strongly localized in the vertical direction. Thus it appears that vertical transport in SLIDs is similar to that in one-dimensional systems of localized states (sites), and SLIDs are very promising model systems to study the effects of disorder and Coulomb interactions on electronic states and transport. Indeed, phonon-assisted hopping for usual one-dimensional and quasi-one-dimensional of sites was extensively studied by using the quantum transport equation techniques and percolation theory (e.g., see [7, 8]); therefore, comparison of experimental data for SLIDs with localization and transport theories might appear to be a useful test of current theoretical concepts.

However, it recently became clear that there are essential differences between SLIDs and standard systems of point sites. In particular, it appears that a finite extent of the “cores” of
localized states in the vertical direction is important, giving rise to virtual-tunneling-assisted hopping [9] - [11]. Moreover, sites cannot be assigned to individual quantum states; one must rather associate sites with quantum wells, which are macroscopically populated. In this paper we discuss an approach that takes account of the specific natures of SLIDs, derive a truncated rate equation for macroscopic sites and establish the equivalence of the problem to that of a generalized resistance network.

1. Transport equation

As usual, in the envelope function approximation the wave functions of electronic states in SLIDs can be written in the form
\[ \psi_{\lambda k} = A u_{\lambda}(z) \exp(i k_\parallel \rho), \]
where \( A \) is a normalizing factor, \( z \) is the coordinate in the SLID growth direction, \( k_\parallel \) and \( \rho \) are the in-plane position and momentum vectors, \( u_{\lambda}(z) \) is the wave function for an eigenstate \( \lambda \) with energy \( \epsilon_\lambda \) corresponding to the solution of the one-dimensional problem with the potential \( V(z) = \sum_n V_n(z) \) describing the modulation of the conduction band edge, and \( V_n(z) \) is the potential of the \( n \)th well. The energies of the states \( \{\lambda k\} \) are \( E_{\lambda k} = \epsilon_\lambda + \frac{\hbar^2}{2m} k_\parallel^2 \).

For the structures considered, typical well widths are such that for a single-well problem, upper size quantization levels in the wells lie much higher than the lowest levels. Therefore, in what follows we neglect contributions from all dimensional subbands except the lowest one. For strong disorder, the states \( \{\lambda k\} \) are strongly localized in the \( z \)-direction and the \( u_{\lambda} \) are close to the corresponding “atomic-like” wave functions with small admixture of wave functions of neighboring wells.

The quantum transport equation describing transition between the states \( \{\lambda k\} \) has a form
\[ \frac{df_{\lambda k}}{dt} = -\sum_{\lambda' k', k} W_{\lambda' k', \lambda k} f_{\lambda' k'} \left( 1 - f_{\lambda' k'} \right) - W_{\lambda k, \lambda' k} f_{\lambda k} \left( 1 - f_{\lambda' k} \right), \]
where \( f_{\lambda k} \) is an average occupation number of the state \( \{\lambda k\} \) and \( W_{\lambda' k', \lambda k} \) is the probability of transitions from the state \( \{\lambda k\} \) to \( \{\lambda' k'\} \). Equation (1) contains both intra- and interwell transitions. If we neglect interwell transitions, we arrive at a system of standard independent Boltzmann transport equations; each of these describes transport and relaxation in the corresponding well. Since we are not interested in transport along the well planes, we do not write out explicitly intrawell diffusion and drift terms.

We show that Eq. (1) can be reduced to a truncated rate equation that contains only interwell transition rates. To this end, we sum both sides of Eq. (1) over \( k_\parallel \):
\[ \frac{dv_\lambda}{dt} = -S^{-1} \sum_{\lambda, k, k', \lambda' \neq \lambda} \left[ W_{\lambda' k', \lambda k} f_{\lambda' k'}^{(qe)} \left( 1 - f_{\lambda' k'}^{(qe)} \right) - W_{\lambda k, \lambda' k} f_{\lambda k}^{(qe)} \left( 1 - f_{\lambda' k}^{(qe)} \right) \right], \]
where \( v_\lambda = S^{-1} \sum_{k_\parallel} f_{\lambda k}^{(qe)} \) are nonequilibrium aerial concentrations, \( f_{\lambda k}^{(qe)} \) are the local quasi-equilibrium (quasi-Fermi) distributions characterized by the quasi-Fermi levels \( \mu_\lambda \) and \( S \) is the structure area. In Eq. (2) the intrawell transitions canceled out. Moreover, since for the structures considered the interwell transition rates are much greater than those of interwell transitions, a “local” equilibrium is established in each of the wells, and the functions \( f_{\lambda k} \) in the interwell transition rates can be replaced by \( f_{\lambda k}^{(qe)} \). Thus Eq. (2) is considerably simpler than the master equation (2), since it contains no intrawell transition rates.
2. Linear theory and resistance network

Now let an electric field be applied in the direction of the z-axis; for states strongly localized in the z-direction, its effect can be described by the corresponding energy shifts $\epsilon_\lambda \rightarrow \epsilon_\lambda + U_\lambda$, where $U_\lambda$ is the potential energy at the localization region of the state $\lambda$. An applied field generally produces variations of carrier concentrations in the wells related to the shifts of local quasi-Fermi levels $\delta \mu_\lambda$. Just as in the standard hopping theory [8], we can linearize Eq. (2) taking account of the fact that transition probabilities $W_{\lambda k_\parallel, \lambda k'_\parallel}$ are energy-dependent. Then we arrive at the linearized equation

$$\frac{dv_\lambda}{dt} = \frac{1}{SkT} \sum_{\lambda' \neq \lambda} \Gamma_{\lambda, \lambda'}(U_\lambda + \delta \mu_\lambda - U_{\lambda'} - \delta \mu_{\lambda'}), \quad (3)$$

where the “integral” transition rates are

$$\Gamma_{\lambda, \lambda'} = \sum_{k_\parallel, k'_\parallel} W_{\lambda k_\parallel, \lambda' k'_\parallel} f(0)_{\lambda k_\parallel} \left( 1 - f(0)_{\lambda' k'_\parallel} \right), \quad (4)$$

the transition probabilities are taken with unshifted energies and $f(0)_{\lambda k_\parallel}$ are the equilibrium (Fermi) distribution.

Equation (4) is similar to the standard linearized hopping rate equation [8] with some important differences. First, the sites here are not single-electron states but rather multi-electron: they can be related to “macroscopic” quantum wells that contain many electrons. It follows that correlation effects such as Hubbard repulsion and Coulomb gap effects are expected to be much weaker than in standard hopping problems. Second, the transition rates $\Gamma_{\lambda, \lambda'}$ are integral rates obtained by summing over the initial and final states of the wells with different $k_\parallel$ and $k'_\parallel$.

It follows from Eq. (2) that just as in the standard hopping theory [7, 8, 12], the low-field problem can be reduced to an equivalent resistance network; in our case the resistances $R_{\lambda \lambda'}$ between the states $\lambda$ and $\lambda'$ are expressed in terms of the integral rates by $R_{\lambda \lambda'}^{-1} = (e/ST)\Gamma_{\lambda, \lambda'}$.

3. Discussion

It should be noted that for weak overlap of wave functions of neighboring wells and strong disorder, instead of the eigenstates $\{|\lambda k_\parallel\}$, we can use a basis of “atomic-like” wave functions $\phi_n(z)$ localized at the corresponding wells $n$; in this case one can associate the sites of the resistance network with the wells. It was shown in [9, 10] that for sites with finite-size cores, hybridization effects can be important even for small overlap. In this case the interwell transition probabilities depend on parameters (in particular, on energies) of intermediate virtual states, and network resistances are no more reduced to two-well resistances $R_{nn'}$ but are “multisite”. Moreover, hybridized (cluster) states corresponding to different energies can include different sites and, on the other hand, the same site can belong to different cluster states, i.e., different multisite resistances can “overlap”.

In some cases (e.g., for nearest-neighbor hopping or for systems with sufficiently large disorder) the complicated structure of the network is not crucial for the calculation of the total network resistance. In fact, if the total resistance is determined by the resistance of the critical region, percolation arguments hold, and the network resistance can be evaluated in a straightforward way [9, 10].
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References

High magnetic field dependence of the edge and bulk state electron transport in single-crystalline tungsten nanostructures

G. M. Mikhailov†, A. V. Chernykh†, J. C. Maan‡, J. G. S. Lok‡, A. K. Geim‡ and D. Esteve§

† Institute of Microelectronics and High Purity Materials RAS, 142432 Chernogolovka, Moscow District, Russia
‡ High field magnet lab, Nijmegen University, Nijmegen, the Netherlands
§ SPEC-CEA Saclay, France

Abstract. Electron conductivity of single crystalline nanostructures both of bridge- and cross-type has been investigated at 4.2 K. It shows strong dependence in magnetic field and also exhibits anisotropy against magnetic field direction. It was found new reentrance effect in ballistic properties of multi-terminal cross-type nanostructure, where ballistic properties are suppressed in moderate magnetic field and restored again at higher field.

Introduction

In magnetic field, one should consider both skipping orbits (edge states) and spiral orbits (bulk states) electron transport in solid state nanostructures. Electron interactions with surface scatters depend also on magnetic field direction against the current and the film surface. Developing of fabrication of single-crystalline planar nanostructures with large bulk mean free path, that capable of ballistic electron transport, open an opportunity to investigate new geometric effects in magnetoresistance. Here we present the results of investigation of single-crystalline tungsten nanostructures in high magnetic field, which is parallel to the sample surface (in-plane B-field), but may be along or perpendicular to the current. Multi-terminal and geometrical effects are also investigated.

1. Experimental results and consideration

Single-crystalline (001) tungsten planar nanostructures were fabricated on r-plane sapphire. Film thickness was of \( d = 170 \) nm, 290 and 340 nm, the width of the bridge was of \( W = 7d \) and the length of \( L = 2W \). Cross-type nanostructure arms were of \( 7d \) width. Magnetoresistance of the samples has been measured at 4.2 K by four terminal method, applying conventional AC lock-in system.

1.1. Magnetoresistance of the bridge

As found, magnetoresistance shows giant effect at B-field to be perpendicular to the current, resistance increasing mostly 10 times (Fig. 1., 1p-3p). It goes through the inflection point near \( d/r_L = 0.3–0.5 \) from quasi-linear to linear dependence and has a tendency to quadratic dependence at \( d/r_L \) coming to 2. For B-field being parallel to the current it goes through the maximum at near \( d/r_L = 0.3–0.5 \) and then decreases more than 5 times at \( d/r_L = 2 \) (Fig. 1, 1a-3a). When B-field is perpendicular to the current angular dependence of resistance is weak in all B-field range. When it is along the current it may show strong effect at B-declination within few degrees (Fig. 2). However, angular dependence at \( d \) smaller than 0.5\( r_L \) is weak, but may come stronger at higher magnetic field. Sharp anisotropy of the bridge resistance at high magnetic field is presented in the inserting in
Fig. 1. Experimental data for bridge-type structures. $B$-field is perpendicular (1p-3p) or parallel to the current (1a-3a), thickness $d = 340$ nm (1), 290 (2) and 170 (3), respectively, $r_L$ is cyclotron radios.

Fig. 2. Magnetoresistance data for the bridge with $d = 170$ nm. 1—$B$-field is along the current, 2—$\Delta \alpha = 1.43^\circ$, 3—$\Delta \alpha = 2.25^\circ$, where $\alpha$ is angle between the surface normal and the $B$-field direction. Rotation axis of the sample is perpendicular to the surface normal and the current. In the inserting an angular dependence of $R(B)/R(0)$ for the bridge with $d = 290$ nm (squares, $B = 16.5$ T) and $d = 170$ nm (solid line, $B = 20$ T) is shown.
Fig. 3. Bend resistance $(U_{4,3}/I_{1,2})$ of the cross versus the $B$-field, $d = 170$ nm, $\Delta \alpha = 0^\circ$ (1), 0.82 (2), 1.23 (3), 1.85 (4), 2.25 (5). For comparison, the data for the $B$-field being along the other cross arms are presented, $\Delta \alpha = 0^\circ$ (6), 2.25 (7). In the inserting (bottom), angular dependence of $R_{\text{bend}}(\alpha)/R_{\text{bend}}(0)$ is shown for $B = 2$ (a), 5 (b), 10 (c) and 20 T (d). Inserting in the top is the cross-scheme with leads numbers.

Fig. 2. Declination of the $B$-field from the direction of the current causes transformation of $R(B)$ (1-3 in Fig. 2) from 3a to 3p type as in Fig. 1. It may be explained by an interplay between skipping orbits (edge states) and spiral orbits (bulk states) transport, controlled by magnetic field, as well as by spiral orbit scattering on the interfaces. More pronounce effect of the $B$-field is found for spiral orbits at high magnetic field, where magnetic focusing may suppress electron surface scattering. Estimation of electron bulk mean free path gives tens of microns for the investigated bridge-type structures.

1.2. Magnetoresistance of the cross

Geometrical and multi-terminal effects have been investigated for the cross-type nanostructures. At low magnetic field ($d/r_L$ lower than 0.3) the bend resistance is negative, as a consequence of ballistic properties of the cross-type nanostructure, for all angles of $B$-declinations. At the $B$-field come stronger the bend resistance increases with its absolute value approaching zero. For the $d/r_L$ more than 0.3–0.5 it may cross zero or goes down at higher field depending on declination angles (Fig. 3). Ballistic properties of the cross are restored at high field, the bend resistance coming to be negative again. At low magnetic field, angular dependence is weak (see also inserting in Fig. 3). Due to symmetry of the cross magnetoresistance shows mostly the same dependence upon the $B$-field at its direction being both along one pair of the arms and the other ones (compare 1 and 6 or 5 and 7 in Fig. 3).
Conclusion

Cross-over from low field to higher field range changes the skipping orbit (edge state) transport to the spiral orbit (bulk state) one. As a consequence, it leads to giant effect in magnetoresistance in bridge-type nanostructures both upon magnetic field and its angle declination in the case when B-field is parallel to the current. New reentrance effect in ballistic properties of multi-terminal cross-type nanostructures has been found, where ballistic properties of the structure are depressed at moderate B-field and restored again at its higher magnitude.

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Fractional charge in transport through a 1D correlated insulator of finite length

V. V. Ponomarenko
Dept. of Physics and Astronomy SUNY at Stony Brook, NY 11794, USA
Ioffe Physico-Technical Institute, St Petersburg, Russia

Abstract. Transport between two Fermi liquid reservoirs through a one channel wire of length $L$ is examined when the 1D electron system has an energy gap $2M > T_L \equiv v_c/L$ induced by the interaction in its charge mode ($v_c$: charge velocity in the wire). In the spinless case transformation of reservoir electrons into solitons of fractional charge entails a crossover from a Fermi liquid regime of integer charge transfer below the crossover energy $T_x \ll T_L$ to the insulating one of the fractional charge transfer in the current vs voltage, conductance vs temperature, and the shot noise. Similar behavior is predicted for a spin Mott insulator of filling factor $v = n/(2m')$ ($n, m'$: positive integer).

Introduction

Fractional charge (FC) is one of the central issues in the physics of strongly correlated electronic systems. However its direct observation in transport through these systems involves transfer of FC between the Fermi liquid (FL) source and drain reservoirs. Since the charge carriers of the reservoirs are electron-like quasiparticles one can doubt to which extent this transfer is possible even between the ideal infinite reservoirs. Recently a few remarkable experiments in transport through 1D [1] and 2D [2] systems have been attempted to measure the FC of elementary excitations. In the 2D systems of incompressible Fractional Quantum Hall liquid (FQHL) the quantum transport is dominated by the edge mode, which has been described [3] as a chiral Tomonaga–Luttinger liquid (TLL). Another realization of TLL is a metallic phase of 1D interacting electrons in the quantum wire. Therefore it had been expected that the same model describes both transports despite the different origin of their effective interaction. The above experiments however have revealed a principal difference. It has been attributed [4] to mechanisms of mutual transformation of the fractionally charged excitations into electrons at the interfaces between the strongly correlated system and the (FL) reservoirs.

1D transport model

In this work we examine transport through the wire of length $L$ when its 1D electrons are in a Mott insulating phase characterized by the gap $2M \gg v_c/L \equiv T_L$ in the energy spectrum ($v_c$: charge velocity in the wire). This situation has been approached in experiment by Kouwenhoven et al. [5] who built a periodic potential into a 1 channel wire of spinless electrons. Varying the electron density with a gate voltage they observed suppression of the conductance at integer and some rational fillings. Recently Tarucha et al. has succeeded [6] in introducing a shorter period potential. This allows Umklapp scattering and, hence, insulating behavior due to correlations at some rational fillings $v = n/m$. In the spinless case this is a 1D charge density wave insulator (CDW). It is described by a sin-Gordon model whose carriers are (anti)solitons of FC $q = 1/m \leq e \equiv 1$ and mass $M$. For the finite length
insulator, however, the transport is described by an inhomogeneous sinh-Gordon model and involves mutual transformations between reservoir electrons and CDW (anti)solitons. The latter see the reservoirs as a slowly decaying interaction along the boundaries whose strength is proportional to $q^2$, meanwhile the electrons see the CDW condensate as a quantization of their phase at the boundaries, whose values $2\pi q \times \text{integer}$ relate to the CDW vacua. A general solution to this model is unknown. Below we concentrate on the low energy limit of $T \ll T_L < M$ ($T$: temperature) when the carriers in the wire are rare.

Solution and results

By applying an instant on technique the model reduces [7] through a Duality Transform to a point scatterer in a TLL whose solution is known. Behavior of the low temperature conductance is ruled by a scaling dimension of the tunneling operator $\bar{g} = q^2$. The latter is always relevant $\bar{g} < 1$ for the correlated insulator opposed to the band insulator where it is marginal. With lowering temperature/voltage the mutual transformations between reservoir electrons and CDW (anti)solitons eventually produce a crossover at some energy $T_x = \text{cst} \sqrt{T_L M} \exp[-\varepsilon/(1 - \bar{g})] \ll T_L$. $\varepsilon = \sqrt{M^2 - (q\mu)^2}/T_L$, ($\mu$: chemical potential) in the transport from an insulating regime of FC transfer to a FL regime of integer charge current. This conclusion is confirmed by analysis of the shot noise, which reveals that above the crossover the charge of carriers is $q$. It takes the same values as FC in FQHL theory. Meanwhile below $T_x$ it recovers an integer electron value because the interaction induced by the interface couples tunneling (anti)solitons in groups. The correlated insulator conductance recovers its free electron value in the absence of impurities below the crossover at the temperature $T_x$. The current versus voltage has its maximum at $V \approx T_x$ and a negative differential conductance above. Therefore the FC transfer is indicated in the transport experiments by the non-monotonic conductance vs temperature and current vs voltage below $T_L$. The inverse of $T_x$ estimates the time during which the FC exists in the reservoirs. This conclusion complies with consideration of capacitance spectroscopy for the system where one of our reservoirs is changed into a metallic dot of charging energy $E_c$ following [8]. In neglect of the one-electron level spacing of the dot smeared by a small temperature it may be shown that the fractional quantization of the charge emerges if $E_c/T_x > 1$.

This description of the CDW condensate is also applicable to the charge mode of electrons with spin at rational fillings of even denominator $\nu = 1/(2m') \times \text{integer}$. The above results just need redefinitions: $q = 1/m'$, $\bar{g} = q^2/2$. In particular, the charge $q$ of the Mott–Hubbard insulator $\nu = 1/2$ is now 1. This marginality shows up in saturation of the current as the voltage exceeds $T_x$ [9].

References

Electron transport in a mesoscopic wire: the charging and exchange interaction effects

V. A. Sablikov and S. V. Polyakov
Institute of Radio Engineering and Electronics, RAS,
Moscow District, Fryazino, 141120, Russia

Abstract. We have shown that the difference of the chemical potentials appears between a semiconductor quantum wire and electron reservoirs. It can be comparable with the Fermi energy of electrons. Due to the chemical potential difference the electron density is redistributed between the wire and reservoirs and the wire acquires the charge. We have analyzed the effect of this charge and the exchange interaction of electrons on the potential shape of the wire and the conductance. Under far from equilibrium condition we predict the possibility for several stable states to appear at a given external voltage.

Introduction

Recent experiments and theoretical investigations of coherent transport in semiconductor quantum wires (QW) have clearly shown that observed transport properties are significantly determined by the exchange interaction of electrons [1], and by the presence of electron reservoirs, to which the QW is coupled [3, 2]. Previously [4] we have shown that the strong effect of reservoirs on electron transport appears as a result of the electron density redistribution between the QW and the reservoirs and the charging of the QW. In the present paper we investigate the charging effect on the electron transport taking into account the exchange interaction of electrons with the use of the model [4, 5] based on the Hartree-Fock approximation.

The electron transport and the distribution of the electron density and the potential are investigated considering a QW and reservoirs as a unified system. We show that the chemical potentials of uncoupled QW and reservoirs are essentially different. When a QW and reservoirs are brought into the contact, the electron density is redistributed between the QW and the reservoirs. As a result the QW acquires the net charge and contact potential difference appears which affects electron transport. The acquired charge changes the linear conductance of a QW with non-adiabatic transition to reservoirs. However the most interesting consequence of a charge acquired in the QW is the instability that arises under a high enough applied voltage. The development of instability results in the appearance of multi-stable states, i.e. in the existence of several stable states at a given voltage.

The exchange interaction gives rise to the increase in the Friedel oscillation potential near the non-adiabatic contacts. However the Friedel oscillations do not anywhere significantly suppress the conductance because the QW becomes inhomogeneous as a result of its charging. The essential effect due to exchange interaction is the lowering of the effective potential profile in the QW and correspondingly the increase in the electron density.
1. Contact potential difference

The decoupled QW and the electron reservoirs have their own chemical potentials which are generally not equal each other. The chemical potential is determined by the Fermi energy of non-interacting electrons and the self-energy $\Sigma(k_F)$ which contains the contributions arising from the exchange and correlation interaction as well as from the electron interaction with the positive background charge. We find the chemical potential difference $\Delta \mu$ between a QW and a reservoir. The results are illustrated in Fig. 1 for different Wigner–Seitz parameters $r_s$. It is seen that $\Delta \mu$ can be positive and negative, its magnitude being as high as several effective Rydbergs.

![Fig. 1. Dependence of the chemical potential difference $\Delta \mu$ between the decoupled QW and the 2D electron reservoir on the wire radius $a$ for various density parameters: (a) $r_s < 1$, (b) $r_s > 1$. $R_s$ is effective Rydberg, $a_B$ is the effective Bohr radius. The 2D layer thickness was put equal to the QW diameter, the density parameter of the 2D electrons $R_s$ was equal to 0.5.](image)

If the wire is now coupled to the 2D reservoir, the electron density is redistributed. The QW acquires the net charge and contact potential difference appears. In contrast to the similar phenomenon in the classical case of 3D conductors, there are two features: (i) the contact electric field is not screened over finite length but spreads over the length determined by the size of conductors; (ii) the spatial distribution of the electron density is governed by quantum mechanics.

The self-consistent calculation of the electron density distribution, electric potential and electric current were done using the model of our previous works [4, 5]

2. Linear conductance

The conductance is investigated in the case of non-adiabatic contacts between a QW and reservoirs. The linear DC conductance has been studied as a function of the chemical potential $\mu_0$ in the system. The conductance oscillates with $\mu_0$ because of the resonances of the electron waves over the QW length, $L$. The acquired charge (or the contact potential difference) leads to the change in the kinetic energy of electrons and in such a way affects the oscillation period.

The exchange interaction makes the conductance oscillations more frequent. The reason for this effect is that the exchange interaction results in an effective lowering of the potential energy of electrons and consequently in the increase of their kinetic energy. The
Fig. 2. (a) Average exchange ($E_{\text{ex}}$) and Hartree ($E_{\text{H}}$) energies of electrons incident on the QW with the energy $\varepsilon$. (b) The electron density spectrum averaged over the QW. Thick line (1) is obtained taking into account the exchange interaction. Thin line (2) is the Hartree approximation. The parameters used in the calculation are: $a = 5 \cdot 10^{-7}$ cm, $L/a = 30$, $U_0 = 20$ meV, $\mu_0 - U_0 = 4$ meV.

The exchange interaction effect on the effective potential is illustrated in Fig. 2. Here the exchange interaction energy and the Hartree energy are compared. The exchange interaction is seen to diminish potential energy stronger than the Hartree energy increases it. Also shown is the spectral distribution of the electron density in the QW with and without the exchange interaction. Owing to the exchange interaction the electrons with energy below the transverse quantization energy $U_0$ can pass through the QW.

3. Non-linear transport and multi-stability

A significant redistribution of the electron density between the QW and the reservoirs occurs under far from equilibrium conditions arising when the applied voltage exceeds the Fermi energy. Electrons are injected from the left reservoir (cathode) while the electrons entering the QW from the positive reservoir (anode) are scattered back inside the QW. As

Fig. 3. Dependence of the average kinetic energy of electrons on the applied voltage. The calculations are for the following parameters: $a = 5 \cdot 10^{-7}$ cm, $L/a = 20$, $U_0 = 10$ meV, $\mu_0 - U_0 = 3$ meV.
a consequence the electron density decreases in the QW (roughly speaking to one-half of the equilibrium density) though the positive background charge is unchanged. Because the positive charge is dominant, a potential well appears in the QW, with the potential shape being distorted by the external potential.

When the applied voltage is high enough in comparison with the Fermi energy, the electron density and potential distribution becomes instable under the fixed voltage condition: there are several stable potential shapes and the system can switch between them. These potential shapes have the same potential difference between the QW ends, but differ each other by the depth and the shape of the potential well. A parameter that characterizes uniquely the nonequilibrium state is the mean kinetic energy of electrons in the QW. This parameter is shown in Fig. 3 as a function of the applied voltage $V_a$. The kinetic energy (and hence the potential energy as well) is seen to be multivalude at a given voltage.

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References

Coherent transients in semiconductor nanostructures as the base of optical logic operating

I. Ya. Gerlovin, V. V. Ovsyankin, B. V. Stroganov and V. S. Zapasskii
S. I. Vavilov State Optical Institute, St Petersburg, Russia

Abstract. The idea of the design is proposed for the photonic quantum gates (PG) with their input/output properties based on nonlinear optical dynamics of 2D arrays of two-level systems (TLS) in the resonance field of coherent optical pulses. As a particular example of the 2D medium with functions of the PG, we consider the GaAs/AlGaAs-based 2D excitonic structures. By methods of femtosecond chronopolarimetry of delayed photoreflection, we have studied the energy and phase relaxation rates and the biexciton binding energy in these structures and have shown that the GaAs/AlGaAs MQW can serve as a good basis for building the PG.

Fundamental limitations for speed of electronic computers stimulates physicists to searching for alternate ways of implementation of computational operations. As the most promising trend is considered replacement of electrical methods of information processing by optical ones, characterized by extremely high bandwidth and possibilities of global parallelism of calculations [1]. At the same time, a serious problem in development of optical computer is related to the absence of constructive ideas about the ways of implementation of the optical logical operations that could be competitive, in terms of their speed, smallness of energy consumption, and possibilities to be miniaturized, with contemporary elements of micro- and nanoelectronics [2].

In this communication, based on analysis of optical dynamics of the epitaxial semiconductor heterostructures, we propose a specific method of the photonic logical element, capable of overpassing, in its main parameters, not only the present-day but also anticipated potentialities of electronic circuits. The photonic logical element is referred to a structure with two stable states capable of passing from one state to the other under the action of combination of the input (controlling) light pulses. The output light signal carries information about final state of the logical element, with the shape of the input and output signals being standardized. The latter requirement is related to necessity to perform several successive computational operations, with the output signal of the preceding element being the input for the subsequent one.

The operation of the proposed logical element is based on properties of resonance ensemble of two-level systems, which can be inverted by a short light pulse (by the so-called pi-pulse [3]). As can be found by use of the Maxwell–Bloch equations [4], for the case of exact resonance and with no allowance made for relaxation the inverted ensemble is capable of emitting the secondary light pulse virtually identical to the exciting one. If to ascribe unity to the inverting light pulse and zero to the pulse of essentially smaller amplitude, such an ensemble can be used to build a logical element of optical computer. This conclusion is confirmed by the results of numerical calculations of the response of ensemble of two-level systems to the pulse with the small amplitude as shown in Fig. 1. It follows herefrom that weak pulses of standard shape can be used to read out the state of the ensemble, and the result of the read-out will have the form of a logical light pulse.
Thus, the analysis of dynamics of ensemble of two-level systems in the field of coherent light pulses confirms fundamental opportunity to use the ensemble for implementation of all-optical logical operations. We have to estimate, however, to what extent the approximations made in analysis are justified for real physical systems.

The first serious assumption, was about exact resonance between the frequency of the light oscillations and eigenfrequency of each two-level system of the ensemble. This means, in fact, that the spread of eigenfrequencies of the two-level systems (i.e., the inhomogeneous broadening) should be essentially smaller than the Rabi frequency. For quasi 2D-structures the Rabi frequency is proportional to the product of the dipole moment of two-level systems on the concentration of systems. So, to meet the above condition, one has to use two-level systems with allowed electric-dipole transitions and with greatest concentration. This is the reason why the free excitons in quantum-confined semiconductor structures are considered as the most promising objects. The dipole moment of optical transitions in such structures is the greatest, while the concentration is equal to the number of sites of the crystalline lattice in the layer [5].

The second important assumption — ignoring relaxation processes — can be considered justified if the switching time of the cell determined by the light pulse width is much smaller than relaxation times of the ensemble. From this point of view, the high energy of the exciton-phonon coupling in quasi-2D heterostructures is also a merit.

In addition, a fundamental question arises about applicability of two-level approximation to the excitons, since the exciton energy structure, in a perfect crystal, represents a set of equidistant levels. In real crystals, this is not the case due to exciton–exciton interaction, and if the energy of this interaction essentially exceeds the energy of excitations with photonic field, the two-level approximation can be considered justified.

To make clear, to what extent parameters of real heterostructures can satisfy all the above conditions, we have performed experimental study of nonlinear optical dynamics of HH-excitons in a sample of standard quality grown by the MBE technique.

The samples under study were 20-period superlattices GaAs/Al_{0.3}Ga_{0.7}As with 7-nm thick GaAs and AlGaAs layers grown on the GaAs (100) plane without growth interruption on heteroboundaries. We studied dynamics of the pulsed differential photoreflection in the pump-probe configuration. In this scheme, the sample is successively subjected to action of two short optical pulses (pump and probe) with a time delay of \( t_d \), and the probe beam amplitude is measured as a function of the time delay.

The pump and probe beams were formed by splitting the output pulses of a tunable Ti:sapphire laser. The pulse width was 70 ps. The frequency of the laser beam was tuned in resonance with that of the HH-exciton transition of the structure under study. The probe
beam was transmitted through an optical delay line, and then both beams were converged on the sample by means of a thin lens ($f = 20$ cm).

Dependence of the signal amplitude on the time delay $t_d$ is shown in Fig. 2. The main signal is associated with two-photon coherent scattering by the population inversion of the HH-excitons, and its shape reflects population dynamics of the excitonic states. Exponential approximation of trailing edge of the pulse yields the relaxation time $\tau = 6 \cdot 10^{-12}$ s. The detailed analysis shows that the measured time is predominantly related to radiative recombination of the excitons. The above value well agrees with theoretical estimates of the radiative lifetime of free excitons in quasi-2D GaAs/AlGaAs structures [5].

In the experiments with time-integrated signal detection, the presence of the dephasing processes should be accompanied by a decrease of the detected amplitude of the two-photon coherent scattering. As has been shown experimentally, the signal from the sample under study remained virtually the same up to 25 K and varied linearly with the pump power density. This means that the phase relaxation rates related to the exciton–photon and exciton–exciton scattering were essentially lower than the radiative decay rate of the excitons.

In Fig. 2, along with the main signal, one can see, at the initial stage of the pulse, several periods of oscillations with the frequency close to $10^{12}$ Hz. As is known [6], the source of such oscillations in the coherent signal are the beats between two transitions with close frequencies. At the same time, analysis of the luminescence and absorption spectra of the structure under study has shown no splitting near the energy 4 meV, corresponding to the observed modulation frequency. Therefore, the only source of the oscillating signal can be quantum beats between the biexciton and two single-exciton states [7]. This means that the inter-exciton coupling energy equals 4 meV and exceeds essentially the exciton–photon interaction energy 0.1 meV determined from the measured radiative decay rate.

Thus, the standard GaAs/AlGaAs heterostructures, in terms of their relaxation parameters, correspond fairly well to the requirements mentioned above. The main obstacle that does no at allow one to implement, with their help, photonic logical operations is too large inhomogeneous broadening 5 meV. However, according to the literature data [8], contemporary level of the MBE technology allows one to decrease the inhomogeneous broadening in structures of this type at least to 200–300 $\mu$V, i.e., down to the value comparable with the radiative broadening. Such high-quality structures are, in principle, already suitable for implementation of a planar matrix of photonic logical elements operating in parallel. The
transverse size each element, determined by the light-wavelength, will lie in the range of 1 \( \mu \), i.e., about \( 10^9 \) elements can be arranged inside the area 1 mm\(^2\). The clock frequency, for these elements, is limited only by the exciton relaxation rate and can approach \( 10^{11} \) Hz, with the switching energy lying around \( 10^{-16} \) J. In terms of the relation between these parameters, the photonic logical elements can exceed, by several orders of magnitude, the performance of electronic systems. It is important also that the switching energy, in such elements, is not converted into heat, as in all electronic circuits, but rather remains in the system in the form of a logical signal.

References

Continuous quantum measurement of a qubit state

Alexander N. Korotkov
Department of Physics and Astronomy, State University of New York, Stony Brook, NY 11794-3800, USA
and Nuclear Physics Institute, Moscow State University, Moscow 119899, Russia

Abstract. We consider a two-level quantum system (qubit) which is continuously measured by a detector. The conventional formalism, which implies the ensemble averaging, describes the gradual decoherence of the qubit state due to measurement. However, in each particular realization of the measurement process we can have the opposite effect: gradual purification of the qubit density matrix. This can be described by the recently developed Bayesian formalism suitable for individual quantum systems. The purification effect may be verified experimentally using present-day technology and can be useful for quantum computing. In particular, the decoherence of a single qubit can be suppressed using continuous measurement and the feedback loop.

The significant progress in experimental techniques during recent years as well as the active research on quantum computing have motivated renewed interest in the problems of quantum measurement, including the long-standing “philosophical” questions. In contrast to the usual case of averaging over a large ensemble of similar quantum systems, it is becoming possible to study experimentally the evolution of an individual quantum system. In this paper we consider the continuous measurement of a qubit (two-level system) state by a “weakly responding” detector which can be treated as a classical device [1]. As examples, we will discuss the following solid-state realizations of such a measurement. First, the location of a single electron in a double-quantum-dot can be measured by a nearby quantum point contact (QPC) in such a way that the QPC barrier height and, hence, the current through the detector are sensitive to the measured electron position [2, 3]. The second possible setup is a single Cooper pair box, the charge state of which is measured by a capacitively coupled single-electron transistor (SET) [4]. Finally, the flux state of a SQUID can be continuously measured by another inductively coupled SQUID [5].

The conventional approach describes the measurement process by the following equations for the qubit density matrix $\rho_{ij}$ in the basis of “localized” states:

$$
\dot{\rho}_{11} = -\dot{\rho}_{22} = -2 \frac{H}{\hbar} \text{Im} \rho_{12},
$$

$$
\dot{\rho}_{12} = i \frac{\epsilon}{\hbar} \rho_{12} + i \frac{H}{\hbar} (\rho_{11} - \rho_{22}) - \Gamma \rho_{12},
$$

where $H$ is the mixing (tunneling) and $\epsilon$ is the energy asymmetry of the qubit states, while $\Gamma$ is the dephasing due to measurement. In the case of the QPC as a detector $\Gamma = (\Delta I)^2 / 4S_I$ [2, 3, 6] where $\Delta I = I_2 - I_1$ is the difference between the average detector currents corresponding to two localized states of the qubit (we assume $|\Delta I| \ll (I_1 + I_2)/2$) and $S_I$ is the low frequency spectral density of the detector shot noise.
Notice that Eqs. (1)–(2) do not depend on the detector output that is a consequence of averaging over the ensemble of systems. The situation is completely different in the case of an individual quantum system since the system evolution should become dependent ("conditioned") on the particular detector output. The theory of conditioned (selective) evolution of a pure wavefunction was developed relatively long ago, mainly for the purposes of quantum optics (see, e.g. Ref. [7] and references therein). However, for solid state structures the problem of continuous quantum measurement with an account of the measurement result has been addressed only recently [1], with the main emphasis on mixed quantum states and detector nonideality.

In the Bayesian formalism developed in Ref. [1] the evolution of the qubit density matrix $\rho$ is described by the equations

$$
\dot{\rho}_{11} = -\dot{\rho}_{22} = -2 \frac{H}{\hbar} \text{Im} \rho_{12} - \frac{2\Delta I}{S_I} \rho_{11} \rho_{22} [I(t) - I_0],
$$

$$
\dot{\rho}_{12} = i \frac{\epsilon}{\hbar} \rho_{12} + i \frac{H}{\hbar} (\rho_{11} - \rho_{22}) + \frac{\Delta I}{S_I} (\rho_{11} - \rho_{22}) [I(t) - I_0] \rho_{12} - \gamma \rho_{12},
$$

where $I(t)$ is the particular detector output, $I_0 \equiv (I_1 + I_2)/2$, and $\gamma$ is the extra dephasing due to the "pure environment",

$$\gamma = \Gamma - (\Delta I)^2/4S_I.$$  

There is no extra dephasing, $\gamma = 0$, for the measurement by a QPC, which thus represents an ideal detector, while the SET in a typical operation point is a significantly nonideal detector, $\gamma \approx \Gamma$ [4]. The SQUID can be an ideal detector only if its total sensitivity is quantum-limited [8] ($\hbar/2$ in energy units).

Equations (3)–(4) allow us to calculate the evolution of the system density matrix for a given detector output $I(t)$. In order to analyze the behavior of $I(t)$, these equations should be supplemented by the formula

$$I(t) - I_0 = \frac{\Delta I}{2} (\rho_{22} - \rho_{11}) + \xi(t),$$

where the zero-correlated ("white") random process $\xi(t)$ has zero average and the same spectral density as the detector current, $S_\xi = S_I$. (We use the Stratonovich formalism for stochastic differential equations.) Notice that even though the Bayesian formalism is valid only for "weakly responding" detectors, $|\Delta I| \ll I_0$, the dimensionless coupling $\hbar \Gamma / H$ is arbitrary, so the formalism can be used in the Quantum Zeno regime as well as in the case of weakly perturbed quantum (Rabi) oscillations.

Figure 1 shows the result of the Monte-Carlo simulation of the continuous measurement by a slightly nonideal detector, $\gamma = 0.1 \Gamma^*$, in the case when the evolution starts from the maximally mixed state, $\rho_{11} = \rho_{22} = 0.5$, $\rho_{12} = 0$. One can see that $\rho_{12}$ gradually appears during the measurement, eventually leading to well-pronounced quantum oscillations. In the case $\gamma = 0$ the density matrix becomes pure after a sufficiently long time. This gradual purification can be interpreted as being due to the gradual acquisition of information about the system. The detector nonideality, $\gamma \neq 0$, causes decoherence and competes with the purification due to measurement.

In contrast to QPC, the SET as a detector directly affects the qubit energy asymmetry $\epsilon$ because of the fluctuating potential $\phi(t)$ of SET’s central island. Since there is typically a correlation between fluctuations of $I(t)$ and $\phi(t)$ [5], Eqs. (3)–(4) can be further improved.
taking into account the information about the likely fluctuations $\epsilon(t)$ caused by the SET. It is natural to add into Eq. (4) the term $i\rho_{12}K[I(t) - (\rho_{11}I_1 + \rho_{22}I_2)] = i\rho_{12}K\xi(t)$, where $K = (d\epsilon/d\phi)S_{\phi I}/S_I\hbar$ and $S_{\phi I}$ is the mutual low-frequency spectral density. One can easily check that the addition of this term corresponds to the partial recovery of coherence, so that the dephasing rate $\gamma$ should be replaced with $\gamma = \gamma - K^2S_I/4$.

To observe the density matrix purification experimentally, it is necessary to record the detector output with sufficiently wide bandwidth, $\Delta f \gg \Gamma$ (possibly, $\Delta f \sim 10^9$ Hz), and plug it into Eqs. (3)–(4). Calculations will show the development of quantum oscillations with precisely known phase. Stopping the evolution by rapidly raising the qubit barrier ($H \rightarrow 0$) when $\rho_{11} \simeq 1$ and checking that the system is really localized in the first state, it is possible to verify the presented results.

Density matrix purification can be used in quantum computing to suppress the gradual qubit decoherence due to interaction with the environment (to keep a qubit “fresh”). The idea is to use the feedback loop which controls the qubit parameter $H$ (control of $\epsilon$ is also possible) in order to decrease the difference between the desired phase of quantum oscillations and the fluctuating phase continuously monitored by a detector supplemented by the “calculator” which computes Eqs. (3)–(4). The preliminary Monte-Carlo results show very significant suppression of decoherence in the case when the detector coupling is stronger than the coupling with extra environment.

Several other predictions related to experiments with single quantum systems can also be made using the Bayesian formalism. In particular, we can show that the quantum oscillations of the qubit state can never be seen directly by the continuous detector (while they can be computed using the noisy detector output and Eqs. (3)–(4)). Quantitatively, the spectral peak of the detector output $I(t)$ at the frequency $\Omega = (4H^2 + \epsilon^2)^{1/2}/\hbar$ of the oscillations cannot exceed $4S_I$ [10]. This is still twice as high as the classically possible limit, that is explained in the Bayesian approach by the correlation between the detector noise and the qubit evolution.
Acknowledgements

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References

Coherent charge qubits based on GaAs quantum dots with a built-in barrier

L. Fedichkin, M. Yanchenko and K. A. Valiev
Institute of Physics and Technology, RAS, 117218, Moscow, Nakhimovskiy pr., 34

Abstract. We investigated using as basic elements of the quantum computer — quantum bits (qubits) semiconductor quantum dots containing one electron and consisting each of two tunnel-connected parts, as shown in Fig. 1. The numerical solution of a Schrödinger equation with the account of Coulomb field of adjacent electrons shows, that in such structures the realization of a full set of basic logic operations which are necessary for fulfillment of quantum computations is possible. Decoherence rates due to spontaneous emission of phonons and acoustic phonons (both piezoelectric and deformational) are evaluated. Durations of one- and two-qubit operations versus qubit geometry are obtained.

1. Introduction

Quantum computing attracts much attention recently because it allows solving some classical problems by using quantum algorithms for exponentially smaller (depending upon length of input data) number of steps compared with the best classical algorithms. A. Ekert et al. [1] proposed using as qubit basis states (“0” and “1”) the two states of spatial quantization of one-electron semiconductor quantum dot. Unfortunately they found soon [2] that their proposal could not be a real qubit because of low coherence of such system. For realization of bi-qubit operations the authors of [1, 2] have offered to use electrical dipole interaction. Potential in one of these quantum points thus was guessed by asymmetric. Distance between dot states was 10–100 meV. The variants of such structures despite of their intrinsic decoherence were studied further by other groups of researchers [3, 4].

In this work we propose to use as qubit quantum dot with a symmetric lateral view, as shown in Fig. 1.

![Fig. 1. Sketch of quantum bit (left) and sketch of CNOT gate (right).](image-url)
2. **Qubit structure**

To avoid high decoherence rates we propose to use special constriction potential for each dot. The presence of two minimums of potential, disjointed by thick barrier is essential. It is supposed that there is only one electron in each qubit. Presence of the second electron is excluded by Coulombic interelectronic repulsion. We conduct hereinafter evaluations for gallium arsenide quantum dots. At distance between minimums $r = 10$ nm (see Fig. 1) the Coulomb energy will be about $e^2/kr = 11$ meV, that allows to eliminate spontaneous charging of a point by the second electron. To obtain clock rates and other performance parameters of qubit offered we numerically solved a two-dimensional Schrödinger equation for an electron in a GaAs quantum dot with potential $V$

$$V = \frac{m\omega^2}{2} \left( x^2 + y^2 \right) + V_B \exp \left( -\frac{x^2}{W^2} \right)$$

(1)

where $m = 0.067 m_e$, $W = nl$, $\omega = \hbar/ml$, $V_B = 1.510^{-19}$ J, $l = 20$ nm, $n$ varies within the range 0.095–0.35.

For logical “0” and “1” it is convenient to take not states with particular energy ($\Psi_1$ and $\Psi_2$), but their Hadamard transform

$$|0\rangle = \frac{\Psi_1 + \Psi_2}{\sqrt{2}} \quad |1\rangle = \frac{\Psi_1 - \Psi_2}{\sqrt{2}}$$

It allows to record input data and read out results by single-electronics procedures by using read-out gates, as shown in Fig. 1. The central control gate serves for lowering of a potential barrier to perform quantum unitary transformations.

3. **Basic unitary transformations**

Let the qubit be in state $\Psi_0$

$$\Psi_0 = c_0 |0\rangle + c_1 |1\rangle$$

When a barrier is high, two states evolve with one frequency $\omega_1$, as two lower states are practically merged.

$$\Psi(t) = (c_0 |0\rangle + c_1 |1\rangle) \exp (i\omega_1 t)$$

The lowering of a barrier leads to an inequality of frequencies ($\Delta \omega = \omega_2 - \omega_1 > 0$) and to periodic gyration of a vector of a qubit state in basis $\{|0\rangle, |1\rangle\}$.

$$\Psi(t) = \frac{1}{\sqrt{2}} \left[c_0 \cos (\Delta \omega t/2) + c_1 \sin (\Delta \omega t/2)\right] \exp \left[\frac{i(\omega_1 + \omega_2)t}{2}\right]$$

Having given an impulse of a positive voltage of particular duration $\tau_{\text{NOT}}$, equal to $\pi/\Delta \omega$, we (throwing away an incidental phase factor) unitary transform a qubit into state

$$\text{NOT}(\Psi_0) = c_1 |0\rangle + c_0 |1\rangle$$

So, with the help of the given procedure it is possible to exchange amplitudes at 0 and 1, that is to realize the unitary operation NOT. Changing a pulse length it is possible to realize qubit rotation by any required angle.

For build-up of the universal quantum computer it is necessary also to know how to realize one nontrivial two-qubit operation (not decomposable in a sequence of one-qubit
ones). We consider realization of the operation CNOT. Depending on a state of control qubit, target qubit should stay the same or be reversed after CNOT transform (i.e. we should apply to target qubit operation of identity or NOT). For realization of a two-qubit CNOT operation between adjacent qubits we use a Coulomb interaction. Let’s arrange two qubits as shown in Fig. 1. It is supposed that there is completely opaque barrier between qubits. Height of a barrier dividing right (target) qubit, depends on a state of left-hand (control) qubit. Having slightly opened the barrier in target qubit with the help of corresponding central gate it is possible to achieve realization above operation, i.e. the operation of identity, when control qubit is in a state 0 and operation NOT, when control qubit is in a state 1, that is operation CNOT. It is achieved by varying pulse duration so that if control qubit is “0”, then the action of an impulse is equivalent to sequential application even number of operations NOT to target qubit, that is operation of identity. If control qubit is “1”, then the action of an impulse is equivalent to sequential application odd number of operations NOT to second qubit, that is operation NOT.

4. Qubit modelling

While modelling, the qubit-qubit interaction was calculated directly from a Coulomb’s law (in all nodes of a two-dimensional grid the field influencing target electron due to a partial charge of control qubit electron from all other nodes of a grid was evaluated). Exchange effects were neglected. The dependences of durations of operation NOT, depending on geometrical parameters are shown in Fig. 2. While estimating a coherence of proposed structure, we have considered a low-temperature limit ($T \rightarrow 0$). It is justified, as the modern cryogenic engineering allows to realize operations of structures at temperatures down to several millikelvins, that is sufficiently lower than the distances between the basic and first excited levels in qubits. The case of high temperatures is not favourable for qubits because of inevitable prompt losses of a coherence and impossibility of correct operation of the quantum computer. However, in solid-state structures even at absolute zero of temperature the processes of losses of a coherence owing to a spontaneous emission of quantums or acoustic phonons and transition of an electron from excited on a ground
level are possible. These processes also will limit a degree of a coherence in our structure. We considered spontaneous radiation of quanta and acoustic phonons (both due to deformational and piezoelectric electron–phonon interaction), spontaneous radiation of a ultrasonic deformation phonon, spontaneous radiation of a ultrasonic polarization phonon. The dominant mechanism of losses of a coherence occurred to be the emission of polarization acoustic phonons. However, our calculations show that even this process has small probability at of clock tick relevant to clock rates for wide structures (wider 15 nm).

So, new quantum bit is offered, on the basis of electrons in symmetric semiconductor quantum points controllable with the help of voltages on electrodes. The frequencies of switching of the quantum register lay in convenient for electronic control range and reach 1 GHz. The offered quantum register is controllable and scalable.

References

Nuclear magnetic resonance spectrum of $^{31}$P donors in silicon quantum computer

A. A. Larionov, L. E. Fedichkin, A. A. Kokin and K. A. Valiev
Institute of Physics and Technology, RAS
34, Nakhimovsky pr., Moscow, 117218, Russia

Abstract. The influence of an electric field created by a gate potential of the silicon quantum computer on the hyperfine interaction constant (HIC) is obtained. The errors due to technological inaccuracy of location of donor atoms under a gate are evaluated. The energy spectra of electron-nuclear spin system of two interacting donor atoms with various values of HIC are calculated. The presence of two pairs of anticrossing levels in the ground electronic state is shown. Parameters of the structure at which errors rate can be greatly minimized are found.

Introduction

One of the most perspective ideas on experimental realization of the quantum computer is the possibility of creating artificial multispin systems. Such an approach, not yet realized, was offered by Kane [1] and was studied further in [2–4]. The special feature of this variant consists in the realization of the individual addressing to separate spins-qubits. For this purpose it is supposed to use a silicon structure of the MOS type, where donor atoms of stable isotope of phosphorus $^{31}$P replacing silicon atoms in the knots of a crystalline lattice are introduced into the thin layer of the spinfree silicon $^{28}$Si isotope. Such a donor has nuclear spin with $I = 1/2$ and states which are characterized by a high value of an effective Bohr radius. In the considered approach it is necessary to use rather low temperatures so that the electrons of donor atoms occupied only the lower spin state in the magnetic field, that is $T \ll 2\mu_B B/k$, where $\mu_B = 9.27 \times 10^{-24}$ J/T is Bohr magneton, $B$ is induction of the external constant magnetic field, $k = 1.38 \cdot 10^{-23}$ J/K is Boltsman constant. At fields $B > 2$ T it corresponds to temperatures $T < 0.1$ K, which are much lower, than the temperature of freezing out the donors electronic state. Therefore, the donors will remain in the neutral ground orbital S-state.

Each donor atom with its nuclear spin in a semiconductor structure is supposed to be arranged regularly with an adequate accuracy under “its” own control metal gate (gate A), separated from the surface of silicon by a thin dielectric. The gates A form a linear chain of any length with the period $l$. The variation of the electrical potential of gates J, located between gates A, allows to control a degree of overlapping of electrons’ wave functions located on the adjacent donors a and b and the constant of their exchange interaction $J$ as well as the constant of scalar interaction of their nuclear spins $I_{ab}$.

This is achieved by redistribution of electronic density between adjacent donor atoms. It is supposed that with the help of electrical field created by gates A it is possible to change distribution of an electronic density near the nucleus in the main state by adjusting, respectively, individual resonance frequency of a nuclear spin of each donor atom, which is determined by hyperfine interaction between the atom and electronic spin. It allows to realize quantum operations by selective influence of resonance radio-frequent impulses on nuclear spins of certain donors.
Thus, for the development of a variant of the semiconductor quantum computer proposed in [1], it is important to have a detailed information about properties of a donor atom energy spectrum hyperfine structure.

1. Dependence of the hyperfine interaction constant on the electrical field

The spin Hamiltonian of hyperfine interaction is:

\[ \hat{H}_{HS} = A \left( \hat{I} \cdot \hat{S} \right), \]  

(1)

where \( \hat{I} \) and \( \hat{S} \) are nuclear and electron spins respectively,

\[ A(V) = \frac{8\pi}{3} |\Psi_0(0,V)|^2 2\mu_B g_N \mu_N \frac{\mu_0}{4\pi}, \]

(2)

\( \mu_N = 5.05 \cdot 10^{-27} \text{ J/T} \) is nuclear magneton, \( \mu_0 = 4\pi \cdot 10^{-1} \text{ T^2 cm^3/J} \), \( g_N = 2.26 \) is Lande’s factor of \( ^{31}\text{P} \), \( \mu_B = 9.27 \cdot 10^{-24} \text{ J/T} \) is Bohr’s magneton, \( \Psi_0(0,V) \) is the wave function of the electron near nucleus. Taking into account the existing electric field the Schrödinger equation for this wave function should be supplemented by a perturbation operator \( \hat{U} = e\phi(\rho',z') \) \( (e = 1.6 \cdot 10^{-19} \text{ C}, \rho'^2 = x'^2 + y'^2) \) [2], where the potential \( \phi \) is induced by the gate which has the form of round disk of radius \( a = l_A/2 \). By factorizing this potential and taking as undisturbed wave functions hydrogen-like functions, we calculate matrix elements of transitions necessary for calculation of error corrections for the wave function within the framework of the theory of perturbations. Limiting ourselves by the second order of the perturbations theory and conducting necessary number substitutions, we shall receive the following expression for a relative error correction of the donor atom hyperfine interaction constant (with \( c = 2a = 10 \text{ nm}, V \) is expressed in volts):

\[ \frac{\Delta A(V)}{A} = 0.55V - 0.09V^2. \]

(3)

Let us consider the case, when gates \( A \) have the form of endlessly long strips with the breadth of \( 2a = l_A \) located at distance \( D \) from the conducting substrate, from which the potential of a gate is accounted. Conducting similar operations, we shall receive for a relative error correction of the donor atom HIC the following expression (with \( c = 2a = 10 \text{ nm}, D/a = 100 \)):

\[ \frac{\Delta A(V)}{A} = -0.063V^2. \]

(4)

The voltage in (4) is expressed, similar to the above, in volts.

2. Influence of the technological location spread of donor atoms on the HIC. Voltage error on a gate

Technological inaccuracies of a donor atom location under a gate in respect of the position of \( x = 0, z = c \) shall be designated by \( \delta x \) and \( \delta z \). We restrict ourselves here to the consideration of only the case of a strip gate. Factorizing the electric field strength and its gradient along axes \( x \) and \( z \) to degrees \( \delta x \) and \( \delta z \), we receive a resultant expression for a relative error of the HIC:

\[ \frac{\delta A}{A} = \delta z \left( 0.063V^2 \frac{2c}{a^2 + c^2} + (\delta x)^2 \right) + (\delta x)^2 \left( \frac{0.063V^2}{2c^2 - a^2} - 0.085V \frac{2c^4 - a^4}{2c^2 (a^2 + c^2)^2} \right) \]

(5)
Fig. 1. Levels of energy of spins for a system of two atoms $^{31}\text{P}$ in a magnetic field ($A_a/J = 0.3; A_b/J = 0.4$).

The second item in (5) can be turned to zero with different values of $a$, $c$ and $V$. That is why by setting a required relative error, it is possible to determine parameters of structure $a$ and $c$ and voltage $V$ at which the required accuracy will be realized. Permissible deviation of the donor atom along axis $z$ connected with the technological inaccuracy of location in 2–3 nm corresponds to a 1% preset relative error of hyperfine interaction. Voltage error at a gate allowed for a correctly controlled quantum computing process can be determined from resultant expressions for changes of the hyperfine interaction constant. The bandwidth of resonance impulse frequencies should be, at least, one order less than the resonance frequency of nuclear spins which is equal to hundreds of kHz. Taking the value of the error for the hyperfine structure constant corresponding to the bandwidth of the resonance impulse $\delta(U_{\Delta \lambda}) \sim 10^4 \text{Hz}$, we shall find the value of the error for the voltage at the gate being equal to $10^{-3} - 10^{-4} \text{V}$. Let us note, that in case of round disks gates because of the linear $V$ summand in expression (4), there is a chosen voltage at which the allowable voltage error greatly increases.

3. Energy spectrum of an electronic-nuclear spin system of two interacting donor atoms

Let us consider two donor atoms being at a distance of $l$ from each other. This distance should be such that constant $J$ of the effective exchange interaction of electrons caused by partial overlapping of their wave functions, had the magnitude sufficient for organizing double qubit operations. The spin Hamiltonian of such a system looks as follows:

$$\hat{H} = 2\mu_B \vec{B} \left( \hat{S}_a + \hat{S}_b \right) + J \left( \hat{S}_a \hat{S}_b \right) - g_N \mu_N \vec{B} \left( \hat{I}_a + \hat{I}_b \right) + A_a \left( \hat{I}_a \hat{S}_a \right) + A_b \left( \hat{I}_b \hat{S}_b \right)$$

(6)

where, $\hat{S}_a$, $\hat{I}_a$, $\hat{S}_b$, $\hat{I}_b$ are spin operators of both an electron and a nucleus for the first and second atoms respectively, $\vec{B}$ is magnetic field (it is directed along the $z$ axis), $A_a$ and $A_b$ are constants of hyperfine interaction depending on the potentials on gates, $\mu_B$ is Bohr magneton, $\mu_N$ is nuclear magneton, $g_N$ is Lande’s factor for $^{31}\text{P}$. The eigen values of a Hamiltonian was accurately calculated. With the help of this matrix the energy levels were numerically established.
4. The conclusion

The proposed paper has a detailed analysis of the influence of the electric field created by gate A on the hyperfine interaction constant for two types of gates: in the form of a disk and in the form of a strip. A weaker dependence of the hyperfine interaction constant on the depth of the donors position under a gate as well as decreasing of its value as the distance between the gate and the substrate was increasing were noted. The conducted calculations with the two types of gates make it possible to conclude that there is a significant dependence of the hyperfine interaction constant on the form of a gate.

The obtained expressions enable to determine permissible values of technological inaccuracies $\delta x$ and $\delta z$ with the set value of the $A$ constant error and working values of the $V$ gate potential.

The paper contains calculation of the full energy spectrum of an electronic-nucleus spin system of two interacting donor atoms for different values of HIC. The presence of two pairs of anticrossing levels in the main electronic condition corresponding to the projection of full electronic and nuclear spins-1 (dotted line) and 0 (dashed line) is shown. For organizing computing and measuring process both pairs anticrossing levels can be used.

References

Abstract. Recent advances in quantum cascade (QC) lasers include widely tunable single-mode distributed feedback (DFB) lasers with high optical power at $\lambda \approx 4.6 \mu$m, and single-mode DFB lasers at $\lambda \approx 16 \mu$m based on new surface plasmon waveguides with dual-metal gratings. Single-mode and tunable QC-DFB lasers have successfully been used in many collaborations for various trace-gas sensing applications. Examples are the detection of stratospheric methane and nitrous oxide, the sub-ppb detection of ammonia by cavity ring down spectroscopy, methane concentration and isotopic composition measurements, and the detection of complex molecules in open air by direct absorption spectroscopy. The versatility of band structure engineering enabled the design and realization of QC-lasers, which operate under both, positive and negative polarity displaying distinct characteristics in each polarity if an asymmetric structure is employed.

Introduction

In the mid-infrared wavelength range only few lasers are established as high power and continuously tunable, narrow linewidth light sources for high-resolution gas sensing applications. The recent development of quantum cascade (QC) and single-mode QC distributed feedback (DFB) lasers [1, 2], however, offers a new and promising alternative. The possibility of achieving amplification from electronic transitions within one band of a semiconductor heterostructure was first postulated by R. F. Kazarinov and R.A. Suris [3]. Semiconductor QC-lasers are consequently based on such electronic transitions between quantized conduction band states of a multiple quantum well structure. They are designed through band-structure engineering and grown by molecular beam epitaxy (MBE).

1. Quantum cascade distributed feedback lasers

Here, we first present QC-DFB lasers designed and fabricated for various trace gas sensing applications. Figure 1 shows an overview of the continuous single-mode tuning ranges of selected lasers operated in pulsed mode. Incorporation of a strong Bragg-grating into the waveguide of the QC-DFB laser leads to very large tuning ranges of approximately 100 and 150 nm for 5 $\mu$m and 10 $\mu$m wavelength lasers, respectively. In the following we will discuss as two examples the shortest (4.6 $\mu$m) [4] and longest (16.2 $\mu$m) [5] QC-DFB lasers in more detail.

While the early work on QC-DFB lasers was motivated by feasibility proofs, our later work on QC-DFB lasers is strongly guided by applications. Trace-gases such as CO or CO$_2$ and their isotopes are of great environmental and medical importance; however, the short wavelengths of their fundamental rotational-vibrational transitions between 4 and 5 $\mu$m wavelength are largely out of reach of conventional QC-lasers. Here, we present short-wavelength single-mode and tunable QC-DFB lasers using strained InGaAs/AlInAs...
Fig. 1. Top: The area under the curve shows the transmission of the atmosphere at sea level. Bottom: Single-mode tuning ranges of selected QC-DFB lasers operated in pulsed mode.

material in the active region [6]. Strained layers increase the band-offset between the quantum wells and barriers (In$_{0.62}$Ga$_{0.38}$As wells and Al$_{0.4}$In$_{0.6}$As barriers result in a band-offset of $\sim 0.725$ eV), thus allowing good quantum confinement also for the upper laser state and a concomitant good device performance. The lasers emit in the wavelength range of 4.6–4.7 $\mu$m, where they overlap with one major branch of the CO rotational-vibrational absorption spectrum. These new devices have unprecedented pulsed peak output power levels of $\sim 100$ mW at room temperature and $\sim 150$ mW in continuous wave at 80 K.

The design of the active region is of the so-called ‘three well vertical’ type with InGaAs and AlInAs lattice-matched to InP. This operating principle is for the $\lambda = 4.6$ $\mu$m laser in more detail discussed in reference [4] and references therein. These lasers contained a waveguide core comprised of $N = 26$ active region stages alternated with electron injectors. A high number of stages is essential for high optical output power and slope efficiency. The waveguide core further contained low doped InGaAs layers below and above the active regions/injectors stack. The lower waveguide cladding is formed by the low-doped InP-substrate. An inner low-doped AlInAs layer and an outer highly doped AlInAs layer, designed for plasmon-enhanced confinement [7], form the upper waveguide cladding. The first-order Bragg-grating of appropriate period ($\Lambda = 730–750$ nm) is transferred into this last layer using optical contact lithography and wet chemical etching to a depth that essentially removes the plasmon confining layer in the grating grooves. The grating is subsequently covered with top contact metallization of 30 nm Ti / 300 nm Au, resulting in a complex coupling scheme of the DFB laser. The coupling coefficient amounts to $|\kappa| \sim 16 \text{ cm}^{-1}$, and is dominated by the modulation of the effective refractive index. Reliable single-mode output is achieved from 90 to 300 K. The tuning with heat sink temperature is well approximated by a quadratic function and covers $\sim 65$ nm with the tuning coefficient increasing linearly from 0.2 nm/K to 0.33 nm/K. In single-mode operation we achieve a side mode suppression ratio of $\sim 30$ dB independent of the current level. Peak power levels of 100 mW and a slope efficiency of 180 mW/A at room temperature and $\sim 400$ mW/A at low temperatures are achieved.

QC-DFB lasers as the ones described above rely on dielectric waveguides and a Bragg-
grating that was etched into one or more layers of that dielectric waveguide. This approach is very successful for QC-DFB lasers with wavelengths \( \lambda \sim 11 \mu m \). It becomes, however, impractical for very long (\( \lambda \sim 17 \mu m \)) wavelength lasers as thick waveguides and very deep gratings would be needed. As a solution to this problem another type of optical confinement—at the interface between two different homogeneous materials—can be used. These ‘surface plasmon’ modes, characterized by an exponentially decaying intensity in the two directions normal to the interface, exist provided the dielectric constants of the two materials have real parts of opposite sign. Negative dielectric constants are encountered e.g. in metals. For wavelengths approaching the far-infrared (\( \lambda > 15 \mu m \)), the penetration depth into the metal is greatly reduced, yielding low optical losses and a concomitant large transverse optical-mode confinement factor.

Here we present a high performance surface plasmon laser operating at \( (\lambda \sim 16 \mu m \). In order to extend the concept of DFB to such surface plasmon lasers, a two-metal grating deposited on top of the semiconductor was used to produce a strong index contrast due to the large spatial modulation of the skin depth.

Our device employs QC active material with a variable period superlattice active region of the type described in reference [8]. Samples comprising 35 to 40 superlattice/injector stages were grown by MBE. The growth of the active material was preceded by a \( \sim 1 \mu m \) thick InGaAs layer and followed by thin, highly doped contact layers. The wafers were processed into deep etched ridges. A very wide portion of the top of the ridges was left open for the deposition of the metallic surface-plasmon carrying layer. Deposition of 10 nm of titanium before the final gold layer already results in a relative change of the refractive index of \( \sim 1.8 \times 10^{-3} \), compared to pure gold, and of the losses of \( \sim 1.5 \times 10^{-2} \). To spatially modulate this variation along the ridge, we fabricated a first-order Bragg grating of an alternate sequence of Ti/Au and pure Au stripes along the surface plasmon propagation. For a grating period \( \lambda = 2.05 \mu m \), we compute a complex coupling coefficient for the DFB mode \(|\kappa| = 6.9 \text{ cm}^{-1}\) which is dominated by the real part. Figure 1 also shows the single mode tuning range of this laser. The change of the modal refractive index with temperature can again be used as an effective way to tune the laser frequency; a linear tuning coefficient is found at the highest temperatures with a value of 1 nm/K.

### 2. Trace gas sensing application using QC-DFB lasers

Single-mode and tunable QC-DFB lasers at various wavelengths from 5.2 to 8.6 \( \mu m \) are presently used in collaborations with expert spectroscopists for various trace gas sensing applications, several of which are referenced below. To achieve a narrow linewidth, the lasers are operated in continuous wave. The tuning of the single-mode output is accomplished by varying the current through the device and by Joule’s heating. C. Webster and coworkers [9] at the Jet Propulsion Laboratory, conducted measurements of the concentration of \( \text{CH}_4 \) and \( \text{N}_2\text{O} \) in Earth’s atmosphere from ground level to the stratosphere using a 7.95 \( \mu m \) QC-laser on board a high-altitude air-plane. B. Paldus and coworkers [10] at Informed Diagnostics have also demonstrated sub-ppbv (\( \text{NH}_3 \)) sensitivity measurements using cavity ring down spectroscopy. A. A. Kosterev et al. [11] at Rice University, TX, reported on measurements of the concentration of \( ^{12}\text{CH}_4 \), its natural isotopes \( ^{13}\text{CH}_4 \) and \( ^{12}\text{CH}_3\text{D} \), \( \text{H}_2\text{O} \), \( \text{N}_2\text{O} \), and \( \text{C}_2\text{H}_5\text{OH} \) diluted in standard air using a direct absorption technique around 7.95 \( \mu m \) wavelength. R. Williams et al. [12] at PNNL in collaboration with M. Taubman and J. Hall at JILA, CO, measured the intrinsic linewidth of several of our QC-DFB lasers around 8 \( \mu m \) wavelength as \( \sim 1 \text{ MHz} \). The laser could furthermore be electronically stabilized to a linewidth \( < 20 \text{ kHz} \).
3. Bi-directional quantum cascade laser

Band-structure engineering of unipolar structures is a very powerful tool for the design and fabrication of devices with particular characteristics. We recently demonstrated a bi-directional QC-laser with emission wavelength dependent on bias-polarity, $\lambda^- \approx 6.5 \, \mu m$ and $\lambda^+ \approx 6.35 \, \mu m$ [13]. This is a new concept for the generation of two wavelengths from a single laser device. In fact, a single device appears as being made of two different laser materials according to the two bias-polarities. Few constraints are posed upon the two wavelengths, which can even be arbitrarily close. We demonstrated this by another bi-directional laser with $\lambda^- \approx 6.75 \, \mu m$. The wavelengths are excited separately in time, which simplifies multiple-wavelength detection schemes, such as differential LIDAR (light detection and ranging).

Figure 2, left, shows the principle of operation of the bi-directional QC-laser. The devices used a so-called ‘diagonal’ laser transition [14], where the upper laser state is the ground state of the injector region and the lower laser state is residing in a single ‘active region’ quantum well, bridging successive injectors together. To obtain an asymmetric bi-directional laser, the injector is designed asymmetric by locally modifying quantum well and barrier thickness, while for a symmetric laser the injector region is designed entirely symmetric. The lasers displayed a good threshold current density of $\sim 3 \, kAcm^{-2}$, and peak powers of $\sim 300 \, mW$ at cryogenic temperatures. Maximum pulsed operating temperature at present is $\sim 150 \, K$. As expected, for the symmetric structure, the emission wavelength was independent of bias polarity, $\lambda^- \approx \lambda^+ \approx 6.75 \, \mu m$. Figure 2, right, shows the emission spectra of a device from the asymmetric, bi-directional QC-laser. The laser emits at the different wavelengths $\lambda^- \approx 6.5 \, \mu m$ and $\lambda^+ \approx 6.35 \, \mu m$, and displays the Fabry–Perot spectra typical for ridge waveguide structures. The lasers produce a peak output power of $\geq 100 \, mW$ at cryogenic temperatures independent of the bias polarity.

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References


Long wavelength quantum dot lasers on GaAs substrates

V. M. Ustinov†, A. E. Zhukov†, A. R. Kovsh†, S. S. Mikhrin†, N. A. Maleev†,
B. V. Volovik†, Yu. G. Musikhin†, Yu. M. Shernyakov†, E. Yu. Kondrat’eva†,
M. V. Maximov†, A. F. Tsatsul’nikov†, N. N. Ledentsov†, Zh. I. Alferov†,
J. A. Lott‡ and D. Bimberg§
† Ioffe Physico-Technical Institute, St Petersburg, Russia
‡ Air Force Institute of Technology, Wright-Patterson Air Force Base, OH, USA
§ Technische Universität Berlin, D-10623 Berlin, Germany

Abstract. We study 1.3 µm diode lasers based on self-organized InAs quantum dots grown by MBE on GaAs substrates. Overgrowing the InAs quantum dot array with thin InGaAs layer allows us to achieve 1.3 µm emission and keep sufficiently high surface density of quantum dots. Using transmission electron microscopy we show that the main reason for the long wavelength PL shift in InAs/InGaAs quantum dots is non-uniform distribution of In in InGaAs leading to the increase in effective volume of a quantum dot. Long stripe lasers showed low threshold current density (<100 A/cm²), high differential efficiency (>50%), and low internal loss (~1−2 cm⁻¹). Maximum output power for wide stripe lasers was as high as 2.7 W and for single-mode devices 110 mW. The lasing wavelength for VCSELs was 1.28 µm. The threshold current for the device with the 12 µm apperture was 1.8 mA. The output power of 220 mW at drive current of 2.4 mA was observed under pulsed mode.

Introduction

Diode lasers based on InGaAsP/InP heterostructures emitting at 1.3 and 1.55 µm are currently widely used in fiber optical communication systems. These systems are mainly used for long-distance data transmission whereas their application in local networks is limited due to the lack of inexpensive light emitters. Two factors mainly lead to the high cost of the InGaAsP/InP lasers. They are: strong temperature dependence of threshold current density due to small conduction band discontinuity and weak variation of refractive index in InGaAsP system making the synthesis of the VCSEL structure in a single growth run hardly possible. The fabrication of the 1.3 µm light emitters on GaAs substrates will allow to overcome those problems and also to avoid the use of expensive InP substrates of inferior quality as compared to that of GaAs.

The increase in emission wavelength from InGaAs/GaAs quantum wells is limited to 1.2 µm due to the limitations of pseudomorphic growth. InGaAsN/GaAs quantum wells [1] and InAs/GaAs quantum dots [2] are currently considered as the most promising candidates for increasing the emission wavelength from GaAs based heterostructures. In the present work we show that the emission wavelength of the InAs/GaAs quantum dot heterostructures can be increased to 1.3 µm and low-threshold diode lasers with high output power can be realized on their basis.

1. Formation of the InAs quantum dots emitting at 1.3 µm

InAs/GaAs quantum dot heterostructures were synthesized by molecular beam epitaxy (MBE) in situ at the initial stages of strained layer heteroepitaxy via Stranski–Krastanow
growth mode [2]. We have shown earlier that the maximum emission wavelength which can be observed from the InAs/GaAs quantum dot ensemble upon increasing the effective deposition thickness of InAs is 1.24 \( \mu m \) at 300 K [3]. Alternative deposition of In, Ga, and As fluxes leads to the formation of larger InAs quantum islands as compared to the usual quantum dots obtained by conventional MBE. This result allowed the authors of [4] to realize 1.3 \( \mu m \) lasing from InAs quantum dots on GaAs substrate. However, in this case the surface density of quantum dots is low (\( \sim 1.3 \times 10^{10} \) cm\(^{-2}\)), and the problem of insufficient gain forces the authors to use extremely long laser diodes or highly reflective coatings which leads to low differential efficiency. In the present work we use the overgrowth of the InAs quantum dot ensemble by a thin In\(_{x}\)Ga\(_{1-x}\)As layer [5]. TEM image shows that in this case the surface density of quantum dots is about 5 \( \times 10^{10} \) cm\(^{-2}\), Fig. 1(a). Both the increase in the effective thickness of deposited InAs (QInAs) and the increase in the InAs mole fraction \( x \) in the In\(_{x}\)Ga\(_{1-x}\)As ternary leads to gradual increase in the emission wavelength which can achieve 1.3 \( \mu m \) at certain values of QInAs and \( x \) [6], Fig. 2(a). Figure 2(b) shows characteristic photoluminescence (PL) spectra of InAs/In\(_{x}\)Ga\(_{1-x}\)As quantum dots. This figure also shows that we observe the red shift of the PL line even when we overgrow InAs quantum dots with the In\(_{x}\)Al\(_{y}\)Ga\(_{1-x-y}\)As quaternary whose band-gap is approximately equal to that of GaAs. This unexpected result was explained after studying the cross-section TEM images of the structures, Fig. 1(b). This image shows the characteristic

Fig. 1. Plan-view (a) and cross-section (b) TEM images of single plane of InAs quantum dots covered by 5-nm thick In\(_{0.15}\)Ga\(_{0.85}\)As QW.

Fig. 2. (a) Dependence of the PL peak position (at room temperature) on the effective thickness of InAs QD plane covered by InGaAs QW of various InAs mole fraction; (b) PL spectra of 2.5-ML InAs QDs covered by various materials.
increase in dark contrast just above the InAs islands which is due to local nonuniformities in the InAs distribution in the InGaAs ternary. As the result, the effective volume of the InAs islands is increased which leads to the long wavelength shift of the PL line. The characteristic PL spectrum (Fig. 2(b)) consists of two features, the main peak is due to the ground state emission and the second (shorter wavelength peak) is due to the excited state quantum dot emission. We should note that the InAs/InGaAs quantum dot structures are characterized by high indium content which could potentially lead to strain relaxation with the formation of misfit dislocations. Therefore, we have minimized the overall In content in the structure keeping 1.3 µm emission. We have found that in this case the structures demonstrate maximum PL intensity indicating the lowest defect concentration [6].

The problem of gain saturation characteristic for quantum dot lasers [7] leads to the use of multi-plane quantum dot structures in the active area of quantum dot lasers. We have found that in the case of 1.3 µm InAs quantum dots the use of thick (>20 nm) GaAs spacers between quantum dot planes allows us to avoid misfit dislocations and to keep high luminescence intensity.

2. InAs/InGaAs 1.3 µm quantum dot lasers

The structures for diode lasers studied in the present work were grown by MBE and the quantum dot active region was placed into GRINSCH AlGaAs/GaAs structure with graded waveguide. Si or Be doped 1.2 µm Al0.8Ga0.2As layers were used as emitters. The thickness of the waveguide layer was 0.4 µm. InAs/In0.15Ga0.85As quantum dots consisted of 2 ML of InAs and 5 nm of In0.15Ga0.85As. The GaAs spacer thickness between quantum dot planes was 30 nm. Laser characteristics were studied as a function of the number of quantum dot sheets. All structures demonstrated luminescence in the 1.25–1.29 µm range. Broad area (100 and 200 µm wide) stripe lasers with various cavity lengths were studied under pulsed mode. CW characteristics were measured after mounting the diode p-side down on a copper heat sink. Narrow (7 µm) stripes were formed to study the single mode operation.

2.1. Threshold characteristics

It is well known that, to have ground state lasing, the following expression should be valid:

\[ \alpha_i + \frac{1}{2L} \ln \left( \frac{1}{R_1 R_2} \right) \leq g_{\text{sat}} \]  

(1)

here \( \alpha_i \) is internal loss, \( R_{1,2} \) are mirror reflective indexes, \( L \) is cavity length, \( g_{\text{sat}} \) is saturated gain on the ground state. Important problem for quantum dot lasers is relatively low saturated gain due to a finite surface density of quantum dots and their size dispersion [7]. As the result, in our experiments we failed to realize ground state lasing for single-sheet quantum dot structure. Ground-state lasing at 1.25 µm was observed for three-sheet quantum dot laser. The threshold current density as low as 65 A/cm² was measured on the four-cleaved sample. Characteristic temperature \( T_0 \) was 150 K up to 310 K and then decreased to 50 K presumably due to the population of excited states. Stripe lasers demonstrated differential efficiency of 55% and low internal loss (1.5 cm⁻¹). Internal quantum efficiency was estimated as 70%, (Fig. 3). Figure 3 also shows lasing wavelength as a function of the cavity length. In the 2–1.2 mm range ground state lasing is observed (\( \lambda \sim 1.25 \) µm) and \( J_{th} \) increases only slightly. However, at \( L \sim 1 \) mm lasing jumps from the ground to excited state (\( \lambda \sim 1.17 \) µm) and threshold current density is abruptly increased. We attribute this effect to the gain saturation at the ground state. In this case mirror losses are increased with the decrease in the cavity length and finally the overall
losses become more than the saturated gain at the ground state. The excited state can accumulate more carriers than the ground state due to the degeneration and, therefore, is characterized by higher $g_{\text{sat}}$ and transparency current density. As the result, lasing proceeds via excited state and the threshold current density is increased. Based on the experimental data given above we estimated the value of saturated gain per the quantum dot layer as $4 \, \text{cm}^{-1}$. From this result it follows that, to achieve ground state lasing from the short (400–500 µm) diodes, one has to use multi-plane quantum dot structures in the active region of a diode laser. In the present work we observed 1.29 µm ground state lasing from the 10-sheet InAs/InGaAs quantum dot laser.

2.2. Power characteristics

High differential quantum efficiency and low internal loss allowed us to demonstrate room temperature CW operation of a quantum dot laser for the structure containing three planes of quantum dots. Broad area laser diodes with the stripe width 100–200 µm showed high output power 1.9–2.7 W (output power density 19–13.5 mW/µm) at 17°C and was limited by thermal roll-over, Fig. 4. Maximum conversion efficiency was 25%. The lasing wavelength at maximum output power was 1.28 µm due to the overheating of the active region.

2.3. Single-mode operation

Broad area lasers demonstrated above give multiple mode radiation. However, fiber optical communication systems require single-mode devices. In the present work we studied single-mode devices by fabricating 7-µm wide stripes from the 3-QD-plane structure. Figure 5 shows output power vs pumping current for the 2-mm cavity laser. Threshold current was 60 mA and differential efficiency 37%. The shapes of the far-field patterns for the output power less than 110 mW are close to gaussians which is indicative of a spatially single-mode operation. Further increasing the pumping current leads to the formation of the mixed mode accompanied by the decrease in the output power.
3. VCSELs

In the present work the VCSEL structure was fabricated by placing the 1.3 μm InAs/InGaAs quantum dot layers into a λ GaAs cavity which was inside the optical cavity formed by AlO/GaAs Bragg mirrors [8]. The AlO layers and the current aperture were formed by selective wet oxidation. Current injection is provided via intra-cavity contacts. The structure showed room temperature lasing at 1.28 μm. The threshold current density for the 12 μm aperture device was \( \sim 1.5 \) kA/cm\(^2\) \((I_{th} = 1.8\) mA\)). The maximum output power under pulsed mode was 220 μW with the external differential of 46%, Fig. 6. To the best of our knowledge this is the longest wavelength current injection VCSEL on a GaAs substrate reported to date.

4. Conclusion

Quantum dot diode lasers emitting in the 1.3 μm range grown by MBE on GaAs substrates have been realized. They showed low threshold current density, high differential efficiency, high output power and single-mode operation. Some characteristics approached to that of the InP based 1.3 μm lasers or even exceed them. In combination with the first data on
the 1.28 \( \mu m \) GaAs based VCSEL these results are very promising for applications in fiber optical communication systems.

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References

New prospects for terabit-scale integration

Konstantin K. Likharev
State University of New York, Stony Brook, NY 11794-3800, U.S.A.

Abstract. I will review several recent new ideas in the field of nanoelectronics that may eventually revolutionize the semiconductor memory and data storage technologies.

Crested barriers

The electronics industry predicts [1] that the current progress in scaling down silicon MOSFETs will lead eventually to gigabit-scale electronic circuits, most importantly, dynamic random-access memories (DRAM) with density of the order of 5 Gbits/cm² and integration scale up to 64 Gbits per die. Further evolution in this direction is, however, very much in doubt: though silicon field-effect transistors may apparently be scaled down to sub-10-nm channel lengths [2, 3], DRAM storage capacitors are non-scalable since their capacitance has to be large enough to charge the output lines all the way down to sense amplifiers. On the other hand, floating-gate cells of nonvolatile memories [4] are scalable, but their write/erase times are so slow (of the order of 1 microsecond) that they cannot serve as a basis for mainstream, bit-addressable memories.

The situation may be changed by the recently proposed [5, 6] “crested” tunnel barriers, with an electrostatic potential peaking in the middle. In these barriers, applied voltage increases the barrier transparency much more quickly than in usual uniform (say, SiO₂) barriers. Calculations have shown that crested barriers may combine a sufficiently long retention time (above 10 years) with fast write/erase time (below 10 nanoseconds). This performance may be reached at low electric fields (below 10 MV/cm), promising high barrier endurance under electric stress. In addition, low write/erase fields limit disturb effects and as a result allow the simplest floating gate memory architectures to be used, increasing circuit density.

NOVORAM

This radical improvement may be used, first of all, for the development of fast nonvolatile random-access memories (NOVORAM) [5]. The NOVORAM cell structure is similar to that of the usual floating-gate memories, except that it uses Fowler-Nordheim tunneling through a crested barrier for both write and erase operations. Estimates show that NOVORAM may be denser than DRAM even at the current (few-100-nm) patterning technology level, adding the convenience of non-volatility.

Moreover, the incorporation of nanoscale MOSFETs with ballistic electron transfer along undoped channels [2, 3] may allow NOVORAM cells to be scaled down to a minimum feature size about 10 nm, corresponding to a memory density of about 100 Gbits/cm², and integration scale up to 16 Tbits per die [7, 6].
Single-electron memories

The NOVORAM scaling limit mentioned above is determined by MOSFET scaling [2, 3] and may be overcome by replacing field-effect transistors with single-electron transistors (SET). The fundamental effect of background charge randomness which hounds single-electron devices [7] may be circumvented in memories using either the so-called “SET/FET hybrids” with dynamic SET read-out in background-charge-insensitive mode [8, 9], or purely single-electron memories equipped with “nanofuses” [10] to exclude cells with inappropriate background charge values [11].

Estimates show that single-electron memories may be scaled down to at least 3 nm minimum feature size, enabling density beyond 1 Tbit/cm² and integration scale of at least 64 Tbits. If equipped with crested tunnel barriers, they may apparently sustain bit-addressable applications.

ESTOR

Crested barriers may also be used in the “ESTOR”, a system for electrostatic data storage [12, 5]. In this system, a read/write head (tip) is flown over a substrate (disk) on which the crested barrier separates a conducting ground layer from a layer of nanometer-size metallic grains, not necessarily all of the same size or shape. The binary unity is coded by the few-electron charging of a small group of grains. Write/erase operation is achieved by the application of a sufficiently high voltage to the circuit on the tip. The recorded data may be read out by the activation of the single-electron transistor which is located on the tip. The SET output signal is further amplified by a closely located, FET-based sense amplifier and then sent out. Recent experiments by a Bell Labs group [13] may be considered as the first step toward the implementation of such readout.

Preliminary estimates show that electrostatic recording may provide data storage density about 1 Tbits/in², i.e., about two orders of magnitude higher than the presently demonstrated level, and an order of magnitude higher than the apparent fundamental limit, of magnetic recording density, if the read/write head can be flown at a comparable height (of the order of 15 nm) above the substrate surface. In contrast to earlier approaches to electrostatic data recording, the use of crested tunnel barriers may make possible a write/read speed up to 1 Gbps per channel, which seems adequate even for this unparalleled bit density.

Conclusion

It seems that the implementation of NOVORAM may revolutionize fast semiconductor memories, while ESTOR may play a similar role in the field of digital data storage, in the very near future. (Single-electron memories require a-few-nm patterning of VLSI scale, and shall take much more time to implement.) Moreover, if combined with the ultrafast superconductor-based RSFQ logic [14, 15], they may allow to develop affordable desktop-size teraflops-scale computers [16] within the next decade.

In my talk at the meeting I will describe these exciting opportunities in detail.

Acknowledgments

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[10] A. Korotkov, New device ideas for terabit-scale digital technologies, Invited talk E14.03 at the APS March meeting (Minneapolis, MN, March 21, 2000), and to be published.

[11] Notice that in logic circuits such exclusion is problematic due to much higher number of interconnects, affecting feasibility of single-electron logic circuits [7].


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