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Spectroscopy of the high energy quantum confined excitonic states in the thick GaAs quantum wells

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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 coinsides 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 demonsrate 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 $Al_{0.3}Ga_{0.7}As$ barrier layers. The QW in QDP1779 with the thickness 150 nm was embedded between $In_{0.51}Ga_{0.49}P$ 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, respectivelly) 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

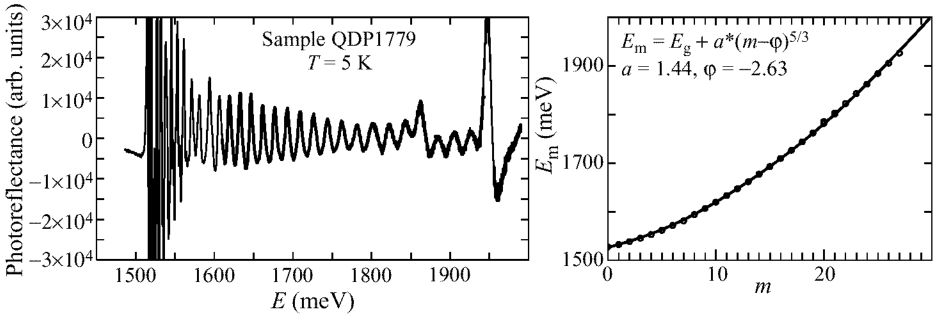

Fig. 1. PR spectrum of the sample QDP1779 with QW 150nm (left panel) and the energy dependence of the oscillation maxima (right panel). Open circles represent experimental data, solid line is the fit by the "power law 5/3".

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.

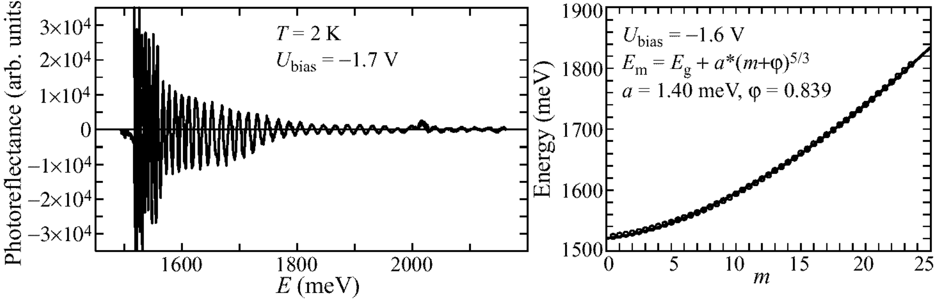

Fig. 2. ER spectrum of the sample QDP1779 measured at bias −1.7 V applied to the sample surface. The other is as in Fig. 1.

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 $E_m - E_g \sim m^{2/3}$ (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.

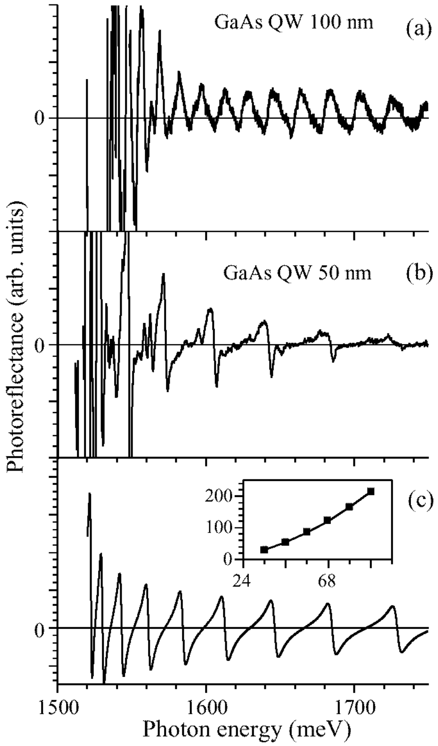

Fig. 3. PR spectra of the samples e188 (a) and e187 (b) and the theoretical simulation of the PR spectrum (c) as described in the text. The energy positions of right zeros in the spectrum (b) and their fit is shown in inset.

We theoretically studied this possibility for the case when electric field changes lineary 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 aproximation 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 thinest QW have a complicate shape.

In order to describe the experimental data, we developed a simple model. The dielectric function of quantum well $\varepsilon = \varepsilon_1 + i\varepsilon_2$ has been calculated with regard to the width of optical transitions, where

$$\varepsilon_1 = \varepsilon_\infty + \frac{2\mu\,|d_{cv}|^2}{\hbar^2 L}\sum_n \ln\left(\frac{\tilde{E}^2}{\Delta_n^2+\gamma_n^2}\right), \tag{1}$$

$$\varepsilon_2 = \frac{4\mu\,|d_{cv}|^2}{\hbar^2 L}\sum_n \left[\frac{\pi}{2} + \arctan\left(\frac{\Delta_n}{\gamma_n}\right)\right], \tag{2}$$

ε_∞ is the background dielectric constant, μ 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 γ_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 γ_n and the change in the reflectivity (ΔR) is determined by the imaginary part (ε_2) of dielectric constant; then

$$\Delta R \sim \frac{d\varepsilon_2}{d\gamma} = -\frac{4\mu\,|d_{cv}|^2}{\hbar^2 L}\sum_n \frac{\Delta_n}{\Delta_n^2+\gamma_n^2}. \tag{3}$$

It is easily to see that this function oscillates with respect to $\hbar\omega$ (Fig. 3(c)). The right zeros of ΔR 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 Δ_n and the change in the reflectivity (ΔR) is determined by the real part (ε_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 $\hbar\omega_{0,n} - E_g = 3.6n^{1.966}$ [meV] for GaAs quantum well ($L = 50$ 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.

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