Defense Technical Information Center Compilation Part Notice

ADP013189

TITLE: Nonlinear Effects in Dense Two-Dimensional Exciton-Polariton System

This paper is part of the following report:

TITLE: Nanostructures: Physics and Technology International Symposium [9th], St. Petersburg, Russia, June 18-22, 2001 Proceedings

To order the complete compilation report, use: ADA408025

9th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 18–22, 2001

MPC.01i

Nonlinear effects in dense two-dimensional exciton–polariton system

V. D. Kulakovskii[1], A. I. Tartakovskii[1], D. N. Krizhanovskii[1],
N. A. Gippius[2] and M. S. Skolnick[3]
[1] Institute of Solid State Physics, Chernogolovka, 142432, Russia
[2] Institute of General Physics, Moscow, Russia
[3] University of Sheffield, Sheffield, UK

Abstract. Angle resolved emission spectra of MCs with quantum wells embedded in the active layer have been investigated under conditions of resonant excitation into the lower polariton (LP) branch. The conditions have been found at which macroscopic filling at the LP branch bottom is reached in the strong coupling regime. Under these conditions strong nonlinear effects in the intensity and the degree of polarisation of polariton emission have been observed and investigated. The experimental dispersion of renormalized cavity mode and its strong narrowing have been explained using an interacting polariton model predicting that the coupling between the LP mode $E_{\mathrm{LP}}(k)$ and the mode $E^*(k) = 2\hbar\omega - E_{\mathrm{LP}}(k + 2k_{\mathrm{excitation}})$ is qualitatively different from that of exciton and photon.

Semiconductor microcavities (MCs) with planar Bragg mirrors change substantially the properties of excitons in quantum wells located at the antinodes of electric field [1–3]. In high quality MCs where the interaction between photon (C) and exciton (X) modes in a quantum well exceeds broadening of the modes, these interacting photons and excitons are considered as MC exciton polaritons revealing a number of peculiar features. The exciton polaritons in bulk semiconductors are stable three-dimensional quasi-particles, their energy tends to zero (at low polariton branch (LPB)) as the wave vector k decreases. In planar MC the polaritons are quasi-two-dimensional and their annihilation does not require conservation of the momentum in the direction perpendicular to the MC plane. As a result, (i) the life time of MC polaritons is finite and is of the order of picoseconds in MCs with a finesse about several thousands; (ii) their energy is finite at $k = 0$. In such a system a strong two-dimensional (2D) confinement of light makes accessible very high densities of the photonic field which may result in a different type of nonlinear effects [4, 5]. Another intriguing property is connected to the boson nature of the mixed exciton-photon states in MCs, 2D polaritons, characterized by an extremely small ($< 10^{-4}m_0$, m_0 being the free electron mass) in-plane effective mass. The low density of polariton states makes possible the high filling at the LP band bottom to be achieved at relatively low densities when the influence of the fermion nature of electron and hole in exciton compound is yet negligible. In this case, the bosonic nature of polaritons can favor stimulated scattering [5, 6] and allow Bose condensation. Experimental observation of these, presumably, nonlinear effects in the strong coupling regime is usually prevented by a slow relaxation of photoexcited polaritons into the LP band bottom [7, 8]. Only recently the stimulated character of polariton relaxation has been demonstrated using resonant excitation into the lower or upper polariton branch [9, 10].

In this work we discuss the properties of dense polariton system generated with the use of resonant excitations by circularly and elliptically polarised light. We succeeded (i) in generation of polaritons at the LPB bottom with a high degree of polarisation that

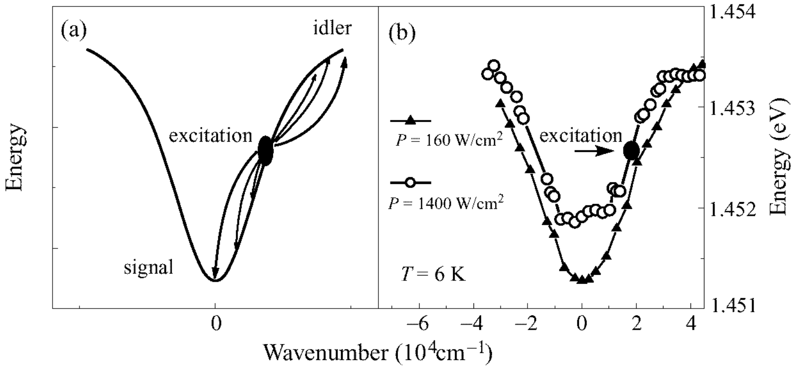

Fig. 1. (a) Polariton dispersion, measured at low and high excitation densities at $\Phi = 16°$. (b) Schematic diagram of two-photon scattering.

under particular conditions exceeds markedly that of the exciting light and (ii) in observing nonlinear effects both in the emission intensity and in the circular polarisation degree.

A GaAs/AlAs MC containing 6 InGaAs quantum wells in the active layer (Rabi splitting, Ω of 6–7 meV) have been studied under the conditions of resonance between the photon and the exciton modes, when polaritons at $k = 0$ have half-exciton-half-photon character. We used a tunable Ti-Sapphire laser to excite luminescence in the MC. A sample was mounted in a cryostat at temperature $T = 1.8-20$ K.

Figure 1 displays the dispersion dependence for polaritons, that was measured at low excitation density. The excitation with the energy lower than the energy E_X of free excitons near the LPB bottom close to the inflection point of the dispersion curve, is the most promising for achieving high density of polaritons without filling of exciton states with large k. However experimental realisation of the idea is problematic because the time of acoustic phonon-mediated scattering into polaritons with $k < 10^4$ cm^{-1} is comparable to the life time of $k = 0$ polaritons. For this reason the k-distribution of polaritons is not in thermodynamic equilibrium. This situation is illustrated in Fig. 2, where LP emission spectra recorded with high angular resolution ($< 1°$) at $T = 2$ K and low density of resonant excitation at $\Phi = 16°$ ($k \sim 2 \cdot 10^4$ cm^{-1}) are displayed. It is seen that the LP emission intensity and, hence, their population decreases as k reduces, while thermodynamically equilibrium system should have demonstrated a sharp increase of LP concentration at the LPB bottom at so low temperature (2 K).

Thus, the phonon mechanism does not provide any effective relaxation of the energy of photoexcited polaritons. The problem can be solved in the case when the exciting light scatters directly into polariton states at the LPB bottom. Following the dispersion law (Fig. 1), that is possible at resonant excitation near the inflection point ($k_{\text{ex}} \sim 1.8 \cdot 10^4$ cm^{-1}, or $\Phi \sim 16°$), when a direct two-photon scattering into polariton states at $k = 0$ and $2k_{\text{ex}}$ can occur with the conservation of the energy and the momentum.

Figure 2 displays experimental spectra of polariton emission recorded for two circular polarisations at $\Phi = 0°$ and $T = 2$ Š under the conditions of circularly (σ^+) polarised excitation at $\Phi \sim 16°$. In the spectrum there is a single line corresponding to the LP emission at $k = 0$. The fine structure of the line is related to the light interference in the sample (the sample thickness is 0.5 mm) and is not considered below. For the σ^+ polarisation the emission intensity at low density of excitation (the LP$^+$ peak) is slightly stronger than that for the σ^- polarisation (LP$^-$ peak), which implies that the spin relaxation time is larger than the lifetime. With increasing P the LP$^+$ peak at $k = 0$ shifts slightly

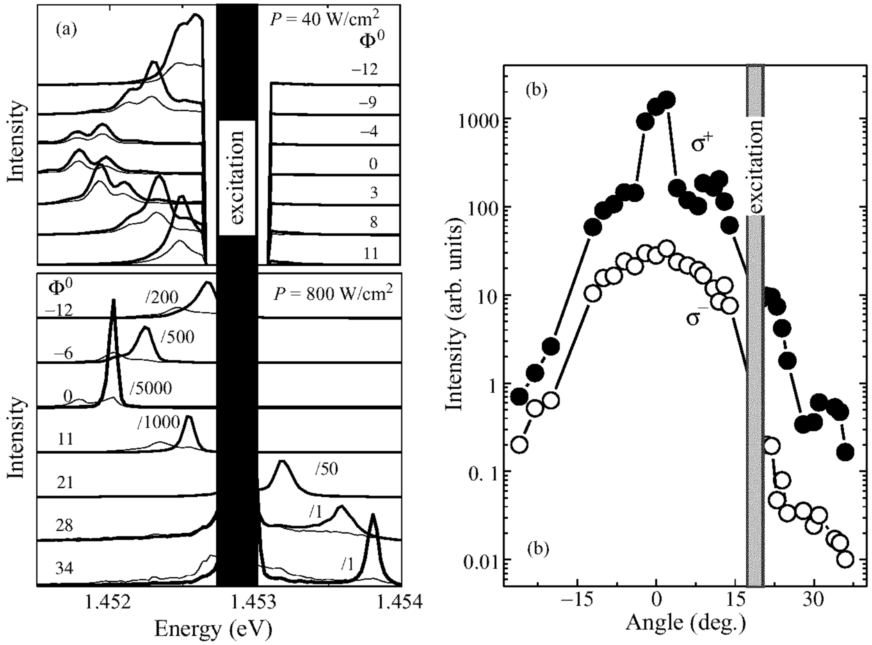

Fig. 2. (a) Polarized σ^+ (thick lines) and σ^- (thin lines) emission spectra of MC recorded for various detection angles under σ^+ polarized excitation into the LPB at angle $\Phi = 16^\circ$ at low and high excitation densities. (b) Angle dependence of the intensity of the LP$^+$ and LP$^-$ lines.

to higher energies, increases superlinearly, and becomes narrow. The LP$^-$ peak, on the contrary, increases linearly with P and its energy does not notably shift. As a result, at high P, the degree of circular polarisation of the LP line reaches 95%. Note that even at the highest P the shift of the LP$^+$ peak is much smaller than the Rabi splitting, which suggests that nonlinear emission effects are related to mixed exciton-photon system.

The dependences of the LP line intensity I_{LP} and polarisation degree ρ_{LP} on the density of excitation at $k = 0$ are plotted in Figs.3(a) and 3(b). There are four regions of the density, revealing qualitatively different behaviour. In the first region ($P < 150$ W/cm^2) $I_{\mathrm{LP}}+$ and $I_{\mathrm{LP}}-$ are nearly linear in P whereas $\rho_{\mathrm{LP}}(P)$ changes rather weakly (between 30 and 40%). In this range the light excites mainly localised excitons, and the quantum yield of LP emission does not exceed 0.1%.

In the second range ($P = 150$–200 W/cm^2) the dependence $I_{\mathrm{LP}} + (P)$ is replaced by the squared one, while $I_{\mathrm{LP}} - (P)$ remains close to linear and hence ρ_{LP} starts increasing. In this region the most effective mechanism of filling of polariton states near $k = 0$ is direct two-photon scattering: $2\hbar\omega(k_{\mathrm{ex}}) = E(k = 0) + E(k = 2k_{\mathrm{ex}})$, which increases the quantum yield of the polariton emission at $k = 0$. A strong narrow line arising in the MC emission spectrum at the detection angles $\Phi = 30$–34° exactly corresponding to $k = 2k_{\mathrm{ex}}$ is a direct proof of the process. Figure 1 shows as well that this line has the energy $\hbar\omega = 2\hbar\omega(k_{\mathrm{ex}}) - E_{\mathrm{LP}}(k = 0)$ and the same polarisation as the LP line at $k = 0$.

As the excitation density rises above 450 W/cm^2 the squared dependence of $I_{\mathrm{LP}}+(k = 0)$ first changes to steep function close to the exponential one, and then saturates at $P >$ 650 W/cm^2. Since the increase in $I_{\mathrm{LP}}-$ continues to be sublinear, the degree of circular polarisation of the LP line at $k = 0$ and $k = 2k_{\mathrm{ex}}$ increases highly and approaches unity. The quantum yield of the emission at the LP$^+(k = 0)$ mode at $P = 1000$ W/cm^2 approaches

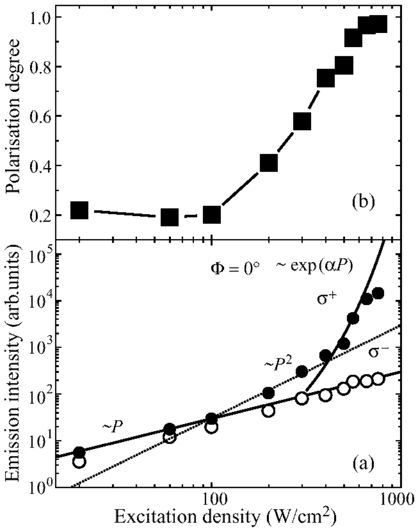

Fig. 3. (a) Dependence of σ^+ and σ^- emission intensity at $\Phi = 0°$ on the density of σ^+ excitation into the LPB at $\Phi = 16°$. (b) Degree of polarisation of polariton emission at $\Phi = 0°$.

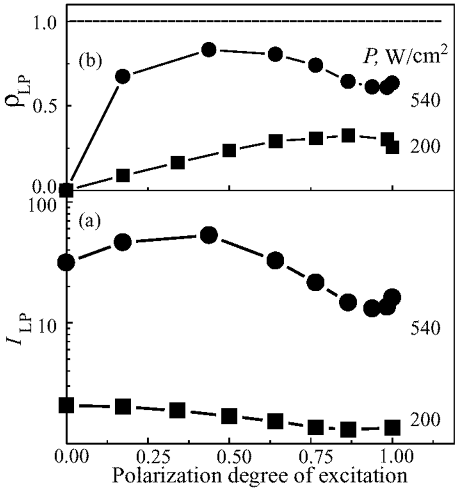

Fig. 4. Dependencies of degree of circular polarisation $\rho_{\rm LP}$ (b) and intensity of the LP line $I_{\rm LP} = I_{\rm LP+} + I_{\rm LP-}$ (a) on the degree of circular polarisation of the excitation at $P = 200$ and 540 W/cm^2.

10%.

The change from the squared dependence of $I_{\rm LP} + (P)$ to the near-exponential suggests the initiation of the stimulated two-photon scattering, and is related to the bosonic nature of polaritons. An additional argument in favour of the stimulated origin of the process is nearly 100% polarisation of the LP line at $k = 0$. The stimulated process develops at increased LP filling factors $\nu_{\rm LP}(k = 0)$. The exponential growth of $I_{\rm LP} + (k = 0)$ is caused not only by increased efficiency of the direct two-photon scattering shown in Fig. 1 but also by stimulated scattering of photoexcited localised excitons and polaritons with $k > 0$. Scattering of localised excitons occurs with emission of acoustic phonons and becomes effective at $\nu_{\rm LP}(E) > \nu_{\rm ph}(E)$, where $\nu_{\rm ph}(E)$ is the filling factor of the phonon mode at the energy E counted from the LPB bottom.

As seen from Fig. 1(b), under the excitation into the inflection point $k_{\rm inf}$ there is no way to obtain effective two-photon scattering into the LP states outside the range $0 < k < 2k_{\rm inf}$. However inside this range the scattering is possible for a wide range of k due to a small difference between $E_{\rm LP}(k)$ and $E^*(k) = 2\hbar\omega(k_{\rm inf}) - E_{\rm LP}(k)$. As a result, one can expect the LP emission under high excitation densities to depend not only on the energy but also on the k direction. Thereby we have measured LP emission spectra in a wide range of angles from −35 to +35°. Experimental results are shown in Fig. 2(a) and (b). In the range of $k < 0$ the emission intensity decreases while the PL peaks width becomes broad with increasing $|k|$. In contrast, at positive k the LP line is very strong and relatively narrow in the whole range of $k < 2k_{\rm inf}$, especially at $k = 0$, $2k_{\rm inf}$, and near $k_{\rm inf}$.

So far we have discussed the behaviour of polaritons excited by circularly polarised light and found them to result in a well pronounced polariton-polariton scattering despite the absence of any absorption resonance near the two photon energy. Note, however, that $2\hbar\omega_{\rm ex}$ is close (lower by about 1 meV) to the energy of the ground biexciton state in the

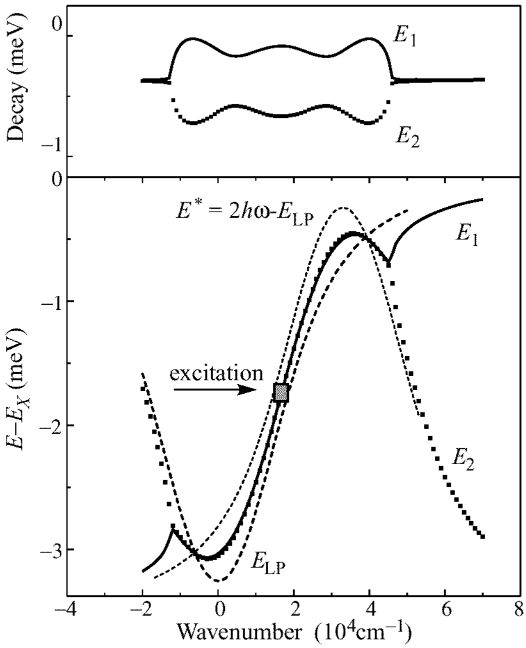

Fig. 5. Calculated dependences of the real (a) and imaginary parts (b) for the polariton modes in the highly excited MC. Dash lines show the dispersion of the modes $E_{\rm LP}$ and $E^*(k) = 2\hbar\omega - E_{\rm LP}(2k_{\rm ex} - k)$ in the weakly excited MC. The parameters used in the calculations are $\Omega = 6.3$ meV, $\Delta_{XC} = 0.09$ meV and the decay of exciton and cavity modes was taken as 0.3 meV, Renormalization of polariton energy due to exciton-exciton interaction is neglected.

quantum well. The biexciton state being a spin singlet is expected to result in an additional resonant contribution in the two-photon scattering only for not completely polarised light, the contribution being increased with decreasing degree of the circular polarisation of exciting light $\rho_{\rm ex}$ from 1 to 0. Thus, the two-photon scattering is expected to increase markedly with decreasing $\rho_{\rm ex}$.

Figure 5 displays the dependences of $\rho_{\rm LP}$ and total intensity of the LP line $I_{\rm LP} = I_{\rm LP} + +I_{\rm LP}-$ on $\rho_{\rm ex}$ at two excitation densities. As seen, $I_{\rm LP}$ increases when the circularly polarised light is replaced by the linearly polarised one. At low excitation density (i.e. under the conditions of spontaneous two-photon scattering) the degree of circular polarisation of the LP emission at $k = 0$ $\rho_{\rm LP}(0)$ monotonically decreases with depolarization of the exciting light. In contrast, at high P the magnitude of $\rho_{\rm LP}$ first grows considerably up to $\rho_{\rm ex} \sim 0.6$ where it even exceeds the degree of polarisation of exciting light. Only at $\rho_{\rm ex} < 0.4$ the value $\rho_{\rm LP}$ decreases rapidly down to zero. Such a behaviour of $I_{\rm LP}$ and $\rho_{\rm LP}$ is an additional strong evidence in favour of (i) that the two- photon scattering process is highly enhanced due to the resonance of the two-photon energy to the biexciton one and (ii) that the process acquires the stimulated character at high P.

The experimental results can be partly explained using an interacting polariton model. The coupling between photon and exciton which give rise to the polaritons results in the repulsion of cavity and exciton modes whereas their imaginary parts become averaging. In MCs highly excited into the LP branch, the wave mixing results in the appearance of an additional branch $E^*(k)$ with the dispersion $E^*(k) = 2\hbar\omega - E_{\rm LP}(2k_{\rm ex} - k)$. Calculations have shown that the coupling between the waves $E_{\mathrm{LP}}(k)$ and $E^*(k)$ is qualitatively different from that of exciton and photon. In case of the polariton formation the cavity mode transforms to exciton and vice versa, in case of four wave mixing the coupling occurs between pairs of the modes $E_{\mathrm{LP}}(k)$, $E_{\mathrm{LP}}(2k_{\mathrm{ex}}-k)$, and the two quanta of the pump. When the pump is strong enough the LP modes start to grow up. In the frequency domain it manifests in the attracting of the levels $E_{\mathrm{LP}}(k)$ and $E^*(k)$ whereas their imaginary parts repel with one of the level exhibiting gain (when the imaginary part of the mode energy becomes positive) and the other mode becoming overdamped. Figure 5 displays the calculated dependence of the real and imaginary parts for the "gain" mode in the MC highly excited under $\Phi = 14^\circ$. The figure shows that the calculated mode dispersion $ReE(k)$ is very similar to the experimental one. In a qualitative agreement with the experiment the calculations show as well the strong line narrowing in the whole range $0 < k < 2k_{\mathrm{ex}}$. However the more detailed comparison shows that the calculations predict the appearance of the gain first at the points of the crossing of $E^*(k)$ and $E_{\mathrm{LP}}(k)$ whereas the experiment shows that this occurs just at $k \sim 0$. It seems that exciton localisation effects as well as relaxation processes have to be taken into account in order to fit the experimental k-dependence of decay of the renormalized polariton mode. Thus, investigating angle resolved emission spectra of MCs with quantum wells embedded in the active layer we have found the conditions at which macroscopic filling is reached at the LPB bottom. Under these conditions we observed and studied strong nonlinear effects in the intensity and the degree of polarisation of polariton emission, and demonstrated that these nonlinearities occur in the strong coupling regime. The experimental dispersion of renormalized cavity mode and its strong narrowing have been explained using an interacting polariton model predicting that the coupling between the modes $E_{\mathrm{LP}}(k)$ and $E^*(k)$ is qualitatively different from that of exciton and photon.

Acknowledgements

We wish to thank M. Bayer, L. V. Keldysh, V. B. Timofeev and S. G. Tikhodeev for fruitful discussions. The work was supported by the RFBR and the Program 'Nanostructures'.

References

[1] C. Weisbush *et al.*, *Phys. Rev. Lett.* **69**, 3314 (1992).
[2] M. S. Skolnick, *et al.*, *Semicond. Sci. Technol.* **13**, 645 (1998).
[3] G. Khitrova, *et al.*, *Rev. Mod. Phys.* **71**, 1591 (1999).
[4] D. M. Whittaker *et al.*, *Phys. Rev. Lett.* **77**, 4792 (1996);
V. Savona *et al.*, *Phys. Rev. Lett.* **78**, 4470 (1997);
R. Houdre *et al.*, *Phys. Rev. Lett.* **73**, 2043 (1994).
[5] A. Imamoglu *et al.*, *Phys. Rev. A* **53**, 4250 (1996).
[6] S. Pau, *et al.*, *Phys. Rev. A* **54**, 1789 (1996).
[7] F. Tassone *et al.*, *Phys. Rev. B* **56**, 7554 (1997).
[8] A. I. Tartakovskii, *et al.*, *Phys.Rev. B* **61**, R2283 (2000).
[9] A. I. Tartakovskii, *et al.*, *Phys. Rev. B* **60**, R11293 (1999).
[10] P. N. Savvidis *et al.*, *Phys. Rev. Lett.* **84**, 1547 (2000);