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3. Quantum dots and wells, mesoscopic networks

Nonlinear effects in dense two-dimensional exciton polariton system

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Abstract. The interaction between quantum well excitons and cavity photons in semiconductor microcavities in the strong coupling regime results in mixed 2D exciton-photon states, called exciton polaritons. The behavior of a dense polariton system is of particular interest due to the fact that these particles have integer spin and, hence, obey Bose-Einstein statistics. Drastic nonlinearities have been observed both in the low polariton (LP) emission intensity and polarization in the case of the resonant excitation into the LP branch under condition that $2\hbar\omega(k_{ex}) = \hbar\omega(k=0) + \hbar\omega(2k_{ex})$. The experiments have shown a very strong final state stimulation of a two polariton scattering due to the bosonic nature of the polaritons. The filling of k = 0 LP state significantly exceeding 1 has been realized under continuous excitation at T = 1.8 K. The dependence of the effect on the polarization of photoexcited light and temperature is discussed.

Semiconductor microcavities (MC) 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 and exciton 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, whose 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 lifetime of MC polaritons is finite and is of the order of picoseconds in cavities with a finesse about several thousands; (ii) their energy is finite at k = 0. The MC polaritons have low effective masses $\sim 10^{-5}m_0$, and large sizes of the order of a micron [4, 5].

Due to their unique features the quasi-two-dimensional MC polaritons are a very intriguing object of investigations.

V D Kulakovskii, A I Tartakovskii, D N Krizhanovskii Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow Region, Russian Federation M S Skolnick University of Sheffield, Sheffield, UK Polaritons are Bose particles like excitons but their effective mass is 3-4 orders less than that of excitons. For this reason, one can obtain high filling of polariton states even at low total density when the influence of internal fermion structure of excitons is small. Then the MC polaritons should show peculiarities typical for bosons such as stimulated scattering and condensation in the *k*-space (Bose condensation) [6].

One of manifestations of stimulated scattering is a superlinear increase in the emission of polaritons at high density of excitation. This effect was observed in [7] where GaAs/AlAs MC were studied under conditions of strong excitation by light with the photon energy $\hbar\omega$ exceeding the GaAs/AlAs band gap E_{g} . However, this effect was detected at rather high densities when Coulomb interaction was strongly screened and exciton-photon interaction was suppressed. Recently, the superlinear regime of polariton luminescence near the bleaching threshold of the exciton system has been observed in II-VI semiconductor MC where the critical density is substantially higher than that in GaAs [8]. Notice that excitation of polaritons with $k \approx 0$ by photons with $\hbar \omega > E_{g}$ is not effective, because the time required to relax into the polariton states at the LPB bottom is comparable to the lifetime of polaritons [9].

In this work high densities of polaritons in the LPB at 2-4 meV above its minimum were generated under conditions of resonant excitations by circularly polarized light. Since the relaxation time t_s of spin polarization of polaritons exceeds their lifetime t_1 [10], we succeeded in creation of polaritons with a high degree of polarization and in observing nonlinear effects both in the emission intensity and in the circular polarization degree.

We have studied GaAs/AlAs MC containing six InGaAs quantum wells in the active layer, 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 MC. A sample was mounted in a cryostat at temperature 1.8 - 20 K. The experiments were made on the structure with Rabi splitting of 6–7 meV and detuning $|\Delta|$ in the energies of exciton (X) and photon (C) modes less than 1 meV. Figure 1a shows the dispersion dependence for polaritons, which was measured at low excitation density.

As seen in Figure 1, the excitation with the energy lower than the energy E_x of free excitons near the LPB bottom close to inflection point of the dispersion curve, is the most promising for achieving high density of polaritons [11]. In this case there is reason to hope that exciton states with large k will be not filled. However, experimental realization of the idea is problematic. Light with energy $\hbar\omega < E_x$ excites mainly the localized excitons [12], whose scattering time into polaritons with $k < 10^4$ cm⁻¹ exceeds greatly the lifetime of



Figure 1. Polariton dispersion, measured at low excitation density (a). Schematic diagram of two-photon scattering (b) (see text).

polaritons at k = 0 (which is several ps) [13]. For this reason the *k*-distribution of polaritons is far from thermodynamic equilibrium, and the polariton population at the bottom is not high, as a rule [14]. This situation is depicted in Fig. 2, where polariton emission spectra recorded with high angular resolution (< 1°) at T = 2 K and low density of resonant excitation at 16° ($k \approx 2 \times 10^4$ cm⁻¹) are plotted. It is seen that the emission intensity of polaritons and, hence, their population slightly decrease as k reduces, while thermodynamically equilibrium system should demonstrate a sharp increase of polariton concentration at the LPB bottom at so low temperature (2 K).

Thus, the phonon mechanism does not provide an effective energy relaxation of photoexcited polaritons and excitons. The problem can be solved in the case when the exciting light scatters into polariton states at the LPB bottom bypassing the localized exciton states. Following the dispersion law (Fig. 1a), this case takes place at resonant excitation near the inflection point ($k_{ex} \approx 1.8 \times 10^4$ cm⁻¹, or $\Phi \approx 16^\circ$), when a direct two-photon scattering into polariton states at $k = k_{ex} - \Delta k$ and $k = k_{ex} + \Delta k$ is allowed with the conservation of the energy and momentum:

$$2\hbar\omega(k_{\rm ex}) = E_{\rm LP}(k_{\rm ex} - \Delta k) + E_{\rm LP}(k_{\rm ex} + \Delta k) \,.$$

The region of allowed transitions is easily found by comparison of the curves with $E = E_{LP}(2k_{ex} - k)$ (polariton) and $E = 2\hbar\omega(k_{ex}) - E_{LP}(k)$ (the energy after emission of polar-



Figure 2. Emission spectra of MC recorded for various detection angles at low density of excitation into the LPB at angle $\Phi = 16^{\circ}$. The narrow line at 1.4523 eV corresponds to the scattered laser emission.

iton from the two-photon state $E = 2\hbar\omega(k_{ex})$ and $k = 2k_{ex})$). These curves are plotted in Fig. 1b by the solid and dashed lines, respectively. The crossing points of the curves indicate the allowed transitions [15]. It is seen that for the excitation near the inflection point of the curve $E_{LP}(k)$ the allowed transitions are in a wide range from $k = 2k_{ex}$ to k = 0.

Figure 3a plots experimental spectra of polariton emission, recorded for two values of polarization at $\Phi = 0^{\circ}$ and T = 2 K under the conditions of circularly (σ^+) polarized excitation at 16°. In the luminescence spectrum there is a single line corresponding to the polariton 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. It is seen that for the σ^+ polarization the emission intensity at low density of excitation (the LP⁺ peak) is slightly stronger than that for the σ^- polarization (LP⁻ peak), which implies that $t_s > t_1$. As the density of excitation increases, the LP⁺ peak shifts to higher energies approximately by 0.3 meV. At P > 200 W cm⁻², it reveals a strong superlinear increase and becomes more narrow. The LPpeak, on the contrary, increases linearly as density of excitations rises and its energy does not notably shift. As a result, at high density of excitation, the degree of circular polarization of the LP line reaches 95%. Note that even at the highest density of excitation the shift of the LP⁺ peak is much smaller than the Rabi splitting, which suggests that nonlinear emission effects are observed in the strong coupling regime, i.e., they are related to mixed exciton-photon modes rather than to pure photon ones.

The detailed dependences of the emission intensity of the LP line I_{LP} and its polarization ρ_{LP} on the density of excitation are plotted in Figs. 3b, c. As is seen, there are four regions of the density, revealing qualitatively different behavior. In the first region when P < 150 W cm⁻², the dependences of I_{LP^+} and I_{LP^-} on P are close to the linear ones. The dependence $\rho_{LP}(P)$ changes weakly, the degree of polarization varies within the limits 30–40%. In this region the excitation of localized excitons mostly occurs, the scattering into the polariton states at the LPB bottom is



Figure 3. (a) Polarized σ^+ (thick lines) and σ^- (thin lines) emission spectra of MC at $\Phi = 0^\circ$ recorded with angular resolution 0.5° under σ^+ polarized excitation into the LPB at $\Phi = 16^\circ$. (b) Dependence of σ^+ emission intensity (squares) and σ^- (circles) at $\Phi = 0^\circ$ on the density of σ^+ excitation into the LPB at $\Phi = 16^\circ$. (c) Degree of polarization of polariton emission at $\Phi = 0^\circ$.

rather weak, and the quantum yield does not exceed 0.1%. At $P = 150-200 \text{ W cm}^{-2}$ this dependence $I_{\text{LP}^+}(P)$ is replaced by the squared one, while $I_{\text{LP}^-}(P)$ remains close to linear, and the degree of polarization ρ_{LP} starts increasing. In this region the most effective mechanism of filling of polariton states is direct two-photon scattering:

$$2\hbar\omega(k_{\rm ex}) = E(0) + E(2k_{\rm ex})\,,$$

which increases the yield of the polariton emission at k = 0. A narrow line arising in the emission spectrum of MC above the excited photon energy is a direct proof of the process. This line (Fig. 4) has the energy $\hbar \omega \approx 2\hbar \omega (k_{\rm ex}) - \hbar \omega (k = 0)$ and the same polarization as the LP line at k = 0, it is observed at the detection angles $\Phi = 30-34^{\circ}$ exactly corresponding to $k = 2k_{\rm ex}$.

As the excitation density rises above 450 W cm⁻² the squared dependence of $I_{LP^+}(k=0)$ first changes to steep function close to the exponential one, and then saturates at P > 650 W cm⁻². Since the increase in I_{LP^-} is slightly



Figure 4. Spectra of σ^+ polarized polariton emission at 2 K and various detection angles, observed at high density of σ^+ excitation into the LPB at $\Phi = 16^{\circ}$. Thick lines indicate spectra corresponding to the emission of polaritons at k > 0, which satisfy the two-photon process condition.

sublinear, the degree of circular polarization of the LP line at k = 0 and $k = 2k_{ex}$ is close to unity. The quantum yield of the emission at the k = 0 mode and P = 1000 W cm⁻² approaches 10%.

The change-over from the squared dependence of $I_{LP^+}(P)$ to the near exponential one suggests the initiation of the stimulated two-photon scattering, and is related to the bosonic nature of polaritons. An additional argument in favor of the stimulated origin of the process is nearly 100% polarization of the LP line at k = 0. The stimulated process runs more intensively at increased filling factors, $v_{LP}(k=0)$, of polariton states. The exponential growth of $I_{LP^+}(k=0)$ is caused not only by increased efficiency of the direct twophoton scattering but also by stimulated scattering of photoexcited localized excitons and polaritons at k > 0. Scattering of localized excitons occurs with emission of acoustic phonons and becomes effective at $v_{LP}(E) > v_{ph}(E)$, where $v_{ph}(E)$ is the filling factor of the phonon mode and the energy E is counted from the LPB bottom. At T = 2-5 K v_{ph} decreases rapidly as E rises and inequality $v_x(E) > v_{ph}(E)$ is fulfilled even at $E \approx 0.5$ meV. The exponential growth of the luminescence with $\hbar \omega = E_{LP}(k=0)$ saturates at the values of the quantum yield of about 10%. This value is rather high as compared to known types of optical parametric transducers.

As is seen in Fig. 1b, under the excitation near the inflection point of the LP dispersion E(k) there is no way to obtain effective two-photon scattering into polariton states with wave vectors outside the range $0 < k < k_{ex}$. However, inside this region it is possible for a wide range of k due to a small difference between $E_{LP}(k)$ and

$$E^*(k) = 2\hbar\omega(k_{\rm ex}) - E_{\rm LP}(2k_{\rm ex} - k) \,.$$

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There is experimental evidence in favor of this suggestion. Figure 4 presents spectra of polariton emission recorded at high angular resolution and high density of resonant excitation. In the range k < 0, the signal is seen to decrease as |k| rises. But the behavior changes at $0 < k < k_{ex}$, when intense two-photon scattering is accompanied by a sharp narrowing of the line observed at k = 0, $2k_{ex}$, and near $k = k_{ex}$.

So far we have investigated the behavior of polaritons excited by circularly polarized light. Note, however, that the energy $2\hbar\omega(k_{ex})$ is close (lower by 1-2 meV) to the ground energy of the biexciton state in the quantum well. The state is spin singlet and optically active for two-photon scattering of linearly polarized light. Under excitation by elliptically polarized light its optical activity decreases gradually down to zero as the degree ρ_{ex} of circular polarization of exciting light varies from 0 to 1. Thus, the two-photon scattering is expected to be resonant and to increase sharply at linearly polarized light.

Figure 5 presents the dependences ρ_{LP} and $I_{LP} =$ $I_{LP^+} + I_{LP^-}$ on ρ_{ex} at two different densities of excitations. As is seen, I_{LP} increases when the circularly polarized light is replaced by the linearly polarized one. The effects become more pronounced as the density of excitation increases. At P = 540 W cm⁻² $I_{\rm LP}$ increases by several times, growing mainly at $\rho_{\rm ex} < 0.6$. Under the conditions of spontaneous two-photon scattering, the degree of circular polarization of the LP line should disappear monotonically with decreasing $\rho_{\rm ex}$. Conversely, at high excitation density of excitations $\rho_{\rm LP}$ grows considerably as ρ_{ex} decreases and at $\rho_{ex} \sim 0.6$ even exceeds the polarization degree of exciting light. Only at $\rho_{\rm ex} < 0.4$ the value of $\rho_{\rm LP}$ decreases rapidly down to zero. The behavior of the circular polarization of the LP line is an additional evidence that the two-photon scattering process is excited by elliptically polarized light and that the process is stimulated.

Thus, studying the angle-resolved emission spectra of MC with quantum wells in a wide range of densities of the



Figure 5. Dependencies of degree of circular polarization ρ_{LP} (a) and intensity of the LP line $I_{LP} = I_{LP^+} + I_{LP^-}$ (b) on the degree of circular polarization of the excitation at P = 200 and 540 W cm⁻².

excitation we have found the conditions at which polaritons with high filling factors are excited at the LPB bottom. Under these conditions we observed and studied strong nonlinear effects in intensity and polarization degree of polariton emission, and demonstrated that these nonlinearities occur in the strong coupling regime.

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- 15. The scheme is only qualitative. Renormalization of polariton states, caused by polariton-polariton interactions should be taken into account for precise calculations

Spectroscopy of electron – electron scattering in a 2DEG

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<u>Abstract</u>. Experimentally electron-beam injection and detection via quantum point-contacts is used to investigate the scattering of a non-equilibrium electron distribution in a twodimensional electron gas (2DEG) of a GaAs/(Ga,Al)As heterostructure. The energy dependence of electron–electron scattering processes has been studied in a weak magnetic field by investigating the detector signal. Assuming electron beams with a narrow opening angle a magnetic field *B* perpendicular to the 2DEG plane causes only electrons which are scattered in a point *O* at an angle α to reach the detector. Thus, it is possible to measure directly the energy dependence of the angular electron distribution after scattering. The experimental data give a clear evidence for the importance of small angle scattering processes in two-dimensional systems, as predicted theoretically.

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