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Exciton Condensation in Microcavities under Three-Dimensional Quantization Conditions

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Abstract—The dependence of the spectra of the polarized photoluminescence of excitons in microcavities under conditions of three-dimensional quantization on the optical-excitation intensity is investigated. The cascade relaxation of polaritons between quantized states of a polariton Bose condensate is observed.

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1. INTRODUCTION

Bose liquids represent an example of a new and peculiar state of matter, a quantum liquid. They possess a number of unusual properties, such as superfluidity, the formation of quantum vortices, and the macroscopic coherence of particle motion. Unfortunately, liquid helium is an almost unique example of a Bose liquid existing in nature. Thus, finding other examples of the Bose-condensed state of matter is an attractive goal. Recently, the Bose condensation of rubidium atoms was observed [1]. However, this was attained under rather extreme conditions, i.e., at ultralow temperatures in magnetic traps. The system of excitons in a semiconductor is a good candidate for observing Bose condensation. Excitons are bosons and their effective mass is low, which suggests that exciton condensation can be observed at readily obtainable temperatures. Exciton polaritons, i.e., mixed exciton–photon states in microcavities, are quasiparticles whose Bose condensation can be observed still more easily. Their mass is even lower than that of excitons, and polariton Bose condensation may, in principle, be observed even at room temperature [2]. However, there are complicating factors: first, polaritons live for a rather short time which may not be enough for them to thermalize, and second, Bose condensation in a two-dimensional system is, as a matter of fact, impossible. The first difficulty cannot be overcome, and we have to be content with nonequilibrium condensation [3]. To overcome the second difficulty, polariton condensation in cavities with three-dimensional quantum confinement is studied [4].

2. RESULTS AND DISCUSSION

Here, we study the dependence of the spectra of the polarized photoluminescence of microcavities with the three-dimensional quantum confinement of polaritons on the optical-excitation intensity. The structures under study represented mesas fabricated from microcavity structures. The microcavity consisted of a 5λ/2 GaAs layer clad between two Bragg mirrors formed by 32 $AlAs/Al_{0.15}Ga_{0.85}As$ layers. Four quantum wells were embedded in the microcavity at locations of electromagnetic-field antinodes. A set of mesas measuring from 1 to 40 μm in diameter was fabricated from these microcavities. The microcavity quality factor was as high as 20000. The microcavities featured a strong exciton–photon coupling mode, the Rabi splitting being on the order of 10 meV. The photoluminescence was excited by 100-fs-long pulses of a Ti:sapphire laser with a repetition rate of 100 MHz. The excitation photon energy was 1.62 eV, which corresponds, on the one hand, to the transparency region of Bragg mirrors and, on the other hand, to the transparency region of AlGaAs barrier layers. The photoluminescence was excited and collected using a microscope; the beam diameter was 1.5 μm. Radiation emitted from the sample within an angle of 90° was collected.

In the linear mode at low excitation intensities (i.e., below 2 mW/cm^2), the photoluminescence spectrum of the mesas represents a rather broad band spanning from 1.530 to 1.550 eV almost independently of the mesa diameter. When the excitation intensity exceeds some threshold value (i.e., upon the transition to the nonlinear mode), a single narrow peak witha full width at half-maximum smaller than 0.5 meV appears on the short-wavelength tail of this band (see Fig. 1a).

The intensity of this peak first grows very rapidly (superlinearly) with increasing excitation intensity and then stabilizes. With a further increase in the excitation intensity, other peaks appear successively in the short-wavelength region. The number and energy spacing of these peaks depend on the lateral size of the mesa. For example, only one line was observed in 40-μm-diameter mesas, while up to six lines were observed in 5-μm-diameter mesas (Fig. 1b). The intensity of these lines increases superlinearly with excitation intensity. Evidently, these narrow lines are related to the lateral quantization of exciton polaritons in the mesas. The polariton nature of the narrow photoluminescence lines is confirmed by the dependence of the degree of circular polarization of the emitted light on the magnetic field which is characteristic of excitons.

At the same time, the nature of the broad photoluminescence band looks enigmatic. Indeed, in small mesas (a few micrometers in diameter), all polariton states should be laterally quantized and manifest themselves only as a series of narrow lines independent of the excitation intensity.

We believe that the occurrence of the broad photoluminescence band in the linear mode is caused by the fact that the Q factor of the lateral modes of the microcavities is very low. Indeed, the reflection coefficient of the lateral side of a mesa is less than 30%; i.e., the photon lifetime in a mesa is about 0.05 ps, which corresponds to a decay rate of the photon mode of 20 meV. In the linear mode, the photoluminescence spectrum features a band with a width of about 20 meV and a peak located at 1.539 eV. This corresponds to emission from the bottom of the polariton band (see Fig. 2), since radiation in this mode propagates perpendicularly to the main surface of the mesa and losses are minimal.

At high excitation intensities in the nonlinear mode, where gain is sufficiently high and compensates for the losses caused by radiation output through the lateral walls of the mesa, the normal modes of the three-dimensional cavity manifest themselves.

Observation of the cavity normal modes gives no indication of whether the exciton–photon coupling in the microcavity is weak or strong. To address this issue, we examined the temperature dependence of the photoluminescence spectra.

Figure 2a shows the photoluminescence spectra at different temperatures. As the temperature increases, all spectral lines shift to lower energies. This is to be expected due to the temperature dependence of the semiconductor band gap. However, the magnitude of this shift is much smaller than the decrease in the band gap itself. Since the energy levels of purely photon modes should be temperature-independent, this result implies that the observed features have a mixed exciton–photon character; i.e., the modes have both exciton and photon components.

According to Fig. 2a, apart from the long-wavelength shift of the photoluminescence lines, an increase in the temperature brings about the appearance of new lines in the spectrum that correspond to ever higher-energy states of the lateral quantization of polaritons. The largest number of lines is observed at 60 K. This makes it possible to establish rather reliably the mutual arrangement of the photon and exciton modes in the microcavity. The dispersion curves calculated for the sample under study at a temperature of 5 K are shown in Fig. 2b. Dots show the normal modes of the mesa. One can see that the calculated mode energies and the positions of the peaks in the photoluminescence spectrum agree well with each other. As the temperature increases, the detuning between the photon and exciton modes changes, which, in turn, leads to a change in the dispersion curve. The polariton dispersion curves calculated for a temperature of 60 K are shown in Fig. 2c. One can see that the temperature shift of the normal modes is much smaller

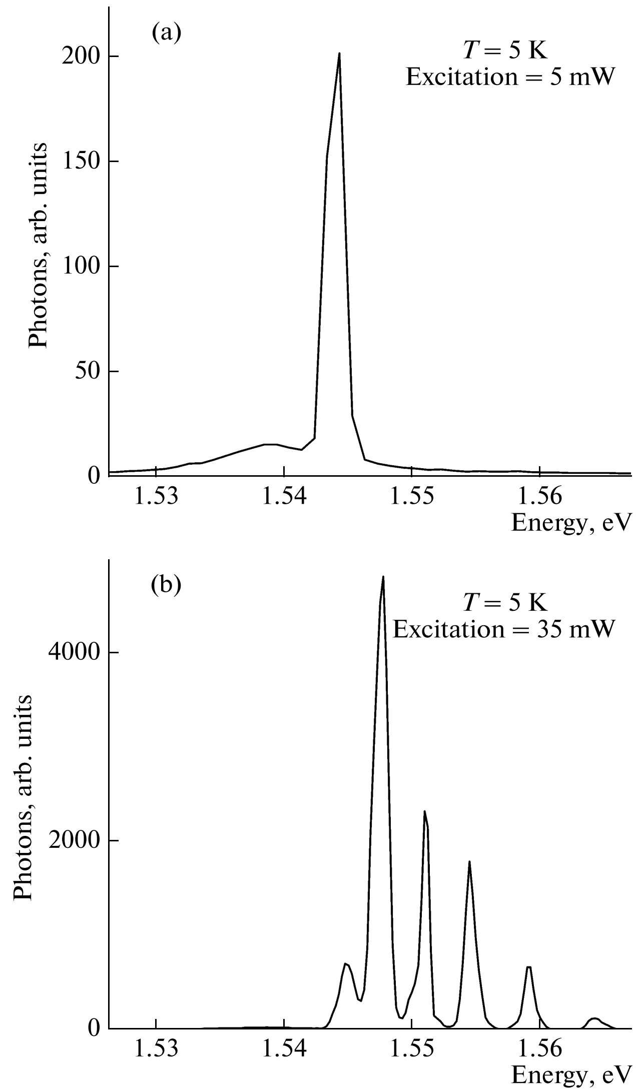

Fig. 1. Photoluminescence spectra of a 5-μm-diameter mesa under optical excitation at 1.62 eV with an intensity of (a) 5 and (b) 35 mW/cm^2.

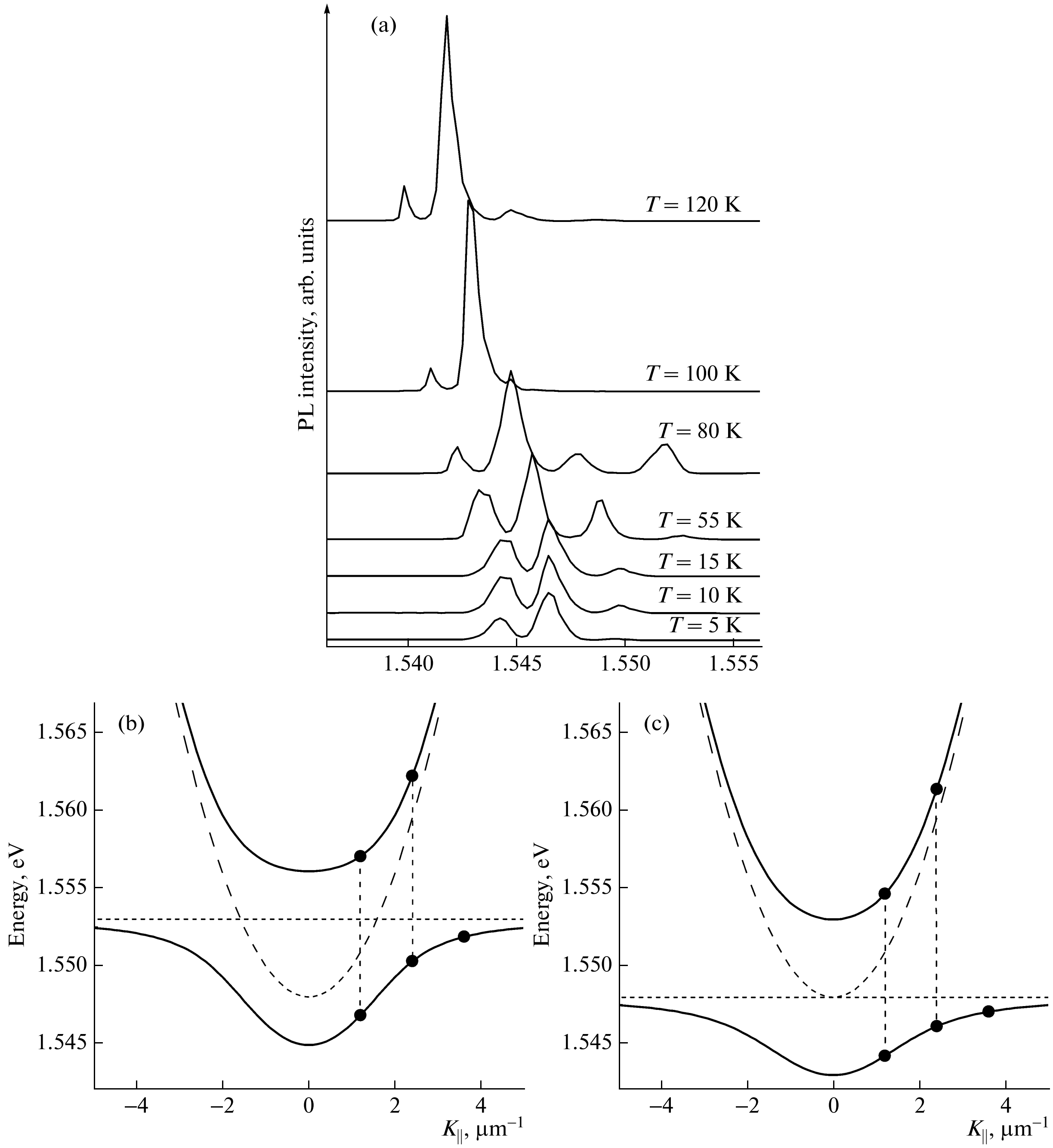

Fig. 2. (a) Temperature dependence of the photoluminescence spectra of a 5-μm-diameter mesa; the excitation intensity is 7 mW/cm^2 for all temperatures. (b) and (c) The dispersion curves of polaritons in this sample calculated for temperatures of 5 and 60 K, respectively; dots show the normal modes of the cavity.

than that of the exciton band and agrees well with the observed spectral shifts. Thus, the temperature dependence confirms that the exciton-mode energy is 5 meV higher than the photon-mode energy.

The experimentally measured decay time of the broad photoluminescence line in the linear mode is about 100 ps. Such long decay is determined by the decay time of the exciton population, since the lifetime of photons in the cavity is very short.

In the nonlinear mode, the photoluminescence decay times for all of the observed lines are on the order of a few tens of picoseconds, which is considerably shorter than the typical decay times of purely excitonic luminescence; this fact is also indicative of the mixed exciton–photon character of these states.

Investigation of the photoluminescence kinetics demonstrates that different peaks observed in the spectrum exhibit different temporal behavior. Accord-

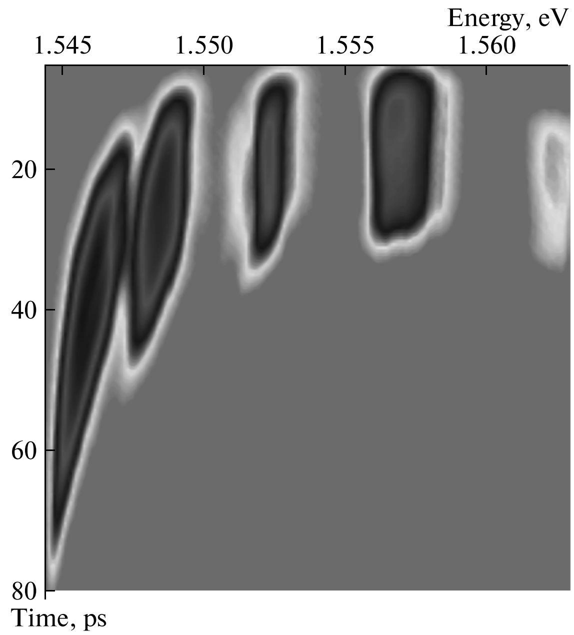

Fig. 3. Temporal dependences of the photoluminescence spectra of a 5-μm-diameter mesa.

ing to Fig. 3, these lines appear and decay in sequence one after another. This fact indicates that the cascade energy relaxation of polaritons between several quantum-confined states of a polariton Bose condensate takes place.

The longest-wavelength line both appears and disappears in the spectrum later than any other line. Furthermore, it exhibits the longest photoluminescence decay kinetics on the order of 60 ps. We attribute this line to the ground state of the polariton condensate. The shortest-wavelength spectral line, whose energy equals 1.5625 eV, has the lowest intensity and appears later than the three longer-wavelength lines. The appearance of this line may be related to the parametric scattering of polaritons. Indeed, at the spectral position symmetric to this line on the energy scale with respect to the relatively intense line at 1.557 eV there is a very weak line at 1.5515 eV, located at the long-wavelength tail of the intense line at 1.5525 eV. This weak line does not correspond to any actual quantized polariton state and originates from the parametric scattering of polaritons [4, 5] taking place upon their annihilation. In this process, one polariton occupying the level at 1.557 eV is anihilated and another polariton from the same level is excited to the level at 1.5625 eV; as a result, the emitted photon has an energy equal to 1.5515 eV. Evidently, for such scattering to occur, polaritons must interact with each other rather efficiently; i.e., they form a condensate. This process is exactly analogous to so-called shake-up processes in the recombination of trions [6]. The occurrence of such processes also supports the suggestion that the Bose condensation of exciton polaritons under conditions of their lateral quantum confinement takes place.

The behavior of the longest-wavelength line of quantized polaritons is rather remarkable. This line appears in the spectrum at times when all other lines have already disappeared and has a very long photoluminescence decay time approaching 60–80 ps. Evidently, this line should correspond to the ground state of the polariton condensate, when all polaritons have time to thermalize to the lowest-energy state.

It was found that all lines in the photoluminescence spectrum are linearly polarized. The degree of polarization varies for different lines from 20 to 60%. The direction of linear polarization also differs slightly for different lines. The degree of this spontaneous polarization is determined by the density of the polariton condensate and correlates with the intensity of the corresponding lines.

3. CONCLUSIONS

In the photoluminescence spectra of mesas fabricated from microcavity structures, peaks related to modes of the lateral quantization of polaritons have been observed. The peaks show up in the nonlinear mode upon exceeding some threshold level in the excitation intensity and appear in sequence starting from the longest-wavelength one. In the time-resolved spectra, the photoluminescence peaks corresponding to different modes disappear also one after another, starting from the shortest-wavelength peak. The longest-wavelength peak is the last one to disappear. The peak positions shift to lower energies with time; this behavior reflects the weakening of the polariton–polariton interaction with decreasing exciton concentration. Satellites related to parametric scattering were observed for some of the peaks.

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