Laser emission from semiconductor microcavities: The role of cavity polaritons

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We present an experimental study on the role of cavity polaritons in laser emissions from a GaAs quantum-well microcavity. We show that cavity polaritons play no role in the laser emission process when the cavity is nearly resonant with the excitons. The laser emissions emerge from the bare cavity mode instead of from a cavity-polariton branch and the threshold density is much higher than the saturation density at which cavity polaritons vanish. We also show that the presence of emission doublets near the lasing threshold, which was previously taken as an evidence for laser emission from cavity polaritons, is primarily the result of spatial and/or temporal variations of exciton densities within the excitation volume. [S1050-2947(97)05010-5]

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Nonequilibrium condensation of exciton polaritons (coupled exciton-photon modes) was first discussed for bulk crystals with dipole-allowed interband optical transitions [1]. In such systems, the presence of a relaxation bottleneck near the turning point of the lower polariton dispersion leads to accumulation of polaritons. Stimulated transitions of polaritons into the bottleneck region become important when the occupation number of polaritons in this region exceeds 1. This stimulated transition process is very similar to stimulated emission of photons in a laser and could similarly lead to non-equilibrium condensation of polaritons in the bottleneck region.

Coupled exciton-photon modes are qualitatively modified near $k=0$ in a semiconductor microcavity due to the quantization of photon wave vectors and are referred to as cavity-polaritons [2–3]. Nonequilibrium condensation of cavity polaritons can in principle occur at $k=0$ instead of at the bottleneck region. In the limit excitons couple strongly to the cavity mode, the mass of cavity polaritons near $k=0$ can become much smaller than the mass of bare excitons. For GaAs quantum-well (QW) microcavity structures, the polariton mass corresponds to a thermal de Broglie wavelength of 7 μm at 4 K, far greater than the exciton Bohr radius (of order 0.01 μm). It was argued that because of this extremely large thermal de Broglie wavelength, nonequilibrium condensation of cavity polaritons could occur at a density far below the exciton Mott density and therefore could be realized experimentally [4,5]. Optical emissions from such a condensate are shown theoretically to be approximately in a coherent state, providing a mechanism for generating coherent laserlike emissions [5].

Evidence of nonequilibrium condensation of cavity-polaritons has been reported recently in a GaAs QW microcavity [4]. A doublet was observed in emission spectra with one of the emission resonance exhibiting laserlike threshold behaviors. It was argued that this doublet is due to emissions from two branches of cavity polaritons and the laserlike resonance is due to emissions from a nonequilibrium condensate of cavity polaritons. The physical origin of the emission doublet, especially, the role of cavity polaritons in the lasing process has been a subject of considerable debate.

In this paper we present an experimental study on the role of cavity polaritons in laser emission from a GaAs QW microcavity. We show that laser emissions emerge from the bare cavity mode instead of from a cavity-polariton branch and that cavity polaritons vanish at densities far below the lasing threshold. Physically, the lasing process is due to stimulated emission of photons rather than condensation of cavity polaritons. Measurements using a pinhole aperture to probe a small region within the excitation volume also reveal that emission doublets observed near the lasing threshold are the result of spatial and/or temporal variations of exciton densities within the excitation volume and cannot be taken as an evidence of persistence of cavity polaritons at the lasing threshold. In addition, exciton localization due to interface fluctuations is suggested as a major obstacle for achieving the elusive nonequilibrium condensation of cavity polaritons.

The GaAs QW microcavity used in our study has four 13 nm GaAs QW's placed at the center of a wavelength-long cavity and uses 16 (22) pairs of $Al_{0.11}Ga_{0.89}As$/AlAs Bragg reflectors as the top (bottom) mirror. The cavity length is tapered such that the cavity resonance varies slightly across the sample while the energy of excitons remains nearly constant. All measurements were performed at 10 K unless otherwise noted.

Figure 1(a) shows reflection spectra of the sample when the cavity is tuned slightly above the heavy-hole exciton absorption line center. At low excitation limit, the reflection spectrum is characterized by two well-resolved cavity-polariton resonances (the minimum normal mode splitting of the sample is 2.6 nm). At high excitation limit, the normal mode splitting collapses and cavity-polaritons disappear [6–8]. In this limit, reflectivity spectra are characterized by the bare cavity resonance as shown by the dashed curve in Fig. 1(a). The collapse of the normal splitting was shown to be due to ionization of excitons in an earlier study [8]. With a further increase in excitation levels the bare cavity resonance moves toward lower wavelength due to mode pulling of the cavity resonance (not shown).

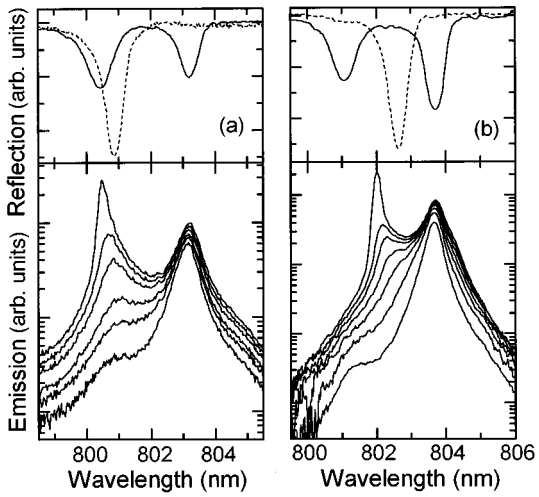

FIG. 1. Reflection and emission spectra with pulsed excitation at two different exciton-cavity detunings. The solid and dashed reflection spectra are obtained at low and high excitation limits, respectively. Emission spectra are obtained at input intensities of 80, 120, 150, 230, 290, 360 W/cm^2 for (a) and of 40, 80, 120, 160, 200, 240, 280 W/cm^2 for (b). The top emission spectrum is obtained at the threshold pumping intensity.

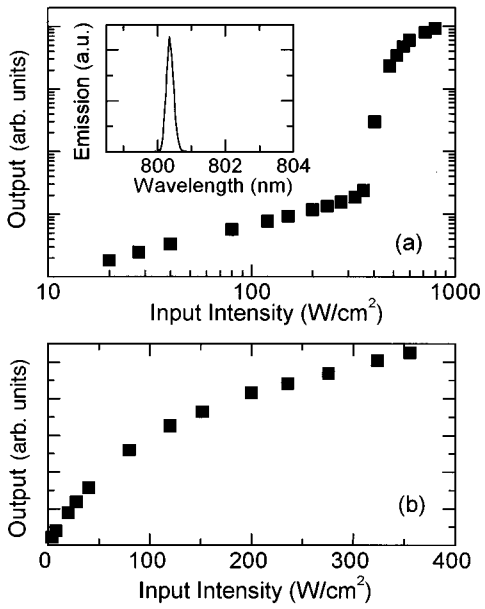

FIG. 2. (a) Total output intensity as a function of the input intensity. (b) Output intensity from the lower energy emission resonance as a function of the input intensity. The exciton-cavity detuning is the same as in Fig. 1(a). The inset shows an emission spectrum above the lasing threshold.

Corresponding emission spectra at various excitation levels are shown in Fig. 1(a). For the emission measurement, the sample is excited off resonantly at a reflection minimum near 755 nm with output from a mode-locked Ti:Sapphire laser. A doublet is observed in emission spectra. At lowest input intensities, the doublet corresponds to the two cavity-polariton resonances in the reflection spectrum and is due to emissions from the two branches of cavity polaritons. Emissions from upper (higher energy) cavity polaritons are much weaker because of the very low temperature.

With increasing excitation levels, the total emission intensity (spectrally integrated) increases rapidly while the intensity of the lower energy resonance in the doublet saturates. Figure 2(a) shows the threshold behavior of the total emission intensity as a function of the input intensity. Figure 2(b) shows saturation of the intensity of the lower energy resonance as a function of the input intensity. An emission spectrum above the lasing threshold is also shown as an inset in Fig. 2(a) and is completely dominated by the higher energy resonance. Behaviors qualitatively similar to those shown in Fig. 2 are also observed at other exciton-cavity detunings as long as the cavity is resonant or nearly resonant with the excitons.

Emission spectra shown in Fig. 1(a) along with the threshold behavior shown in Fig. 2(a) might lead to an assignment that the doublet in Fig. 1(a) is due to emissions from two cavity-polariton branches at all input intensities with the upper polariton going above the threshold with increasing input intensities. This assignment would imply that emissions from the lower (lower energy) polariton branch should saturate at an input intensity near the lasing threshold. Figure 2(b), however, shows that the output from the lower energy resonance saturates at an input intensity near 50 W/cm^2 much lower than the threshold intensity of 360 W/cm^2.

Problems associated with the above assignment become more evident when we examine emission spectra obtained with the cavity tuned slightly below the exciton resonance [see Fig. 1(b)]. At lowest input intensities, the emission spectra feature a doublet that corresponds to the two cavity-polariton resonances in the reflection spectrum. With increasing input intensities, however, the emission resonance from the upper cavity polariton disappears and a new resonance emerges from between the two cavity-polariton resonances. The energy position of the new resonance agrees with that of the bare cavity resonance and moves toward higher energy with increasing input intensities. We emphasize that the same behavior is also observed when we tune the cavity resonance above the exciton resonance. The approximate agreement in energy position between the upper cavity polariton and the higher energy emission resonance near the lasing threshold shown in Fig. 1(a) is coincidental since in this case the bare cavity resonance is close to the upper cavity polariton.

The main issue in understanding the above experimental result is whether near the lasing threshold cavity polaritons still remain a valid description for optical excitations in the microcavity. The observation of optical emissions from the bare cavity resonance far below the lasing threshold clearly indicates that the threshold density n_{th} is much greater than the saturation exciton density n_s at which normal mode splitting collapses and cavity polaritons vanish [9]. This is also supported by the saturation of the lower energy emission resonance at densities much smaller than n_{th} [see Fig. 2(b)] since optical emissions from the lower cavity polaritons are expected to saturate at n_s. We therefore conclude that cavity polaritons play no roles in the lasing process and that the laser emission is due to stimulated optical transitions rather than condensation of polaritons.

The above model satisfactorily explains the behavior of laser emission and the saturation of optical emission from

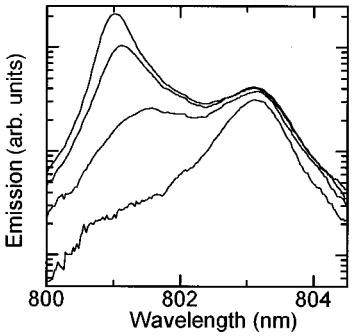

FIG. 3. Emission spectra collected from the center of the excitation volume. Input intensities used are 80, 160, 240, 280 W/cm^2. Other experimental conditions are similar to that of Fig. 1(a).

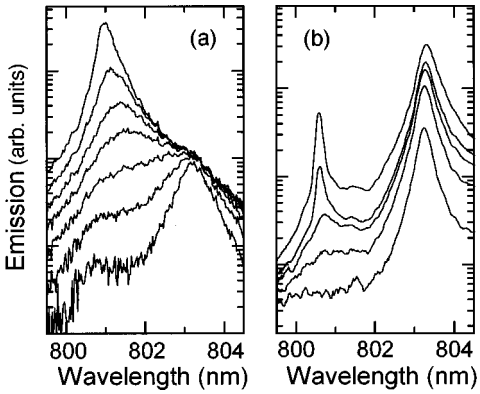

FIG. 4. Emission spectra with cw exciton and collected from the center of the excitation volume (a) and near the edge of the excitation volume (b). Input intensities used are $0.05I_0$, $0.1I_0$, $0.15I_0$, $0.2I_0$, $0.3I_0$, $0.45I_0$, $0.65I_0$ for (a) and $0.15I_0$, $0.5I_0$, $0.85I_0$, I_0, $1.5I_0$ for (b) where I_0 is the threshold pumping intensity. Other experimental conditions are similar to that of Fig. 1(a).

lower cavity polaritons but does not account for the persistence of emission doublets at or near the lasing threshold. As shown in Fig. 1, the energy position of the lower energy emission resonance follows that of the lower cavity polariton and remains nearly independent of the input intensities. The lower energy emission resonance is therefore associated with the lower cavity polaritons at all input intensities. In contrast, the higher energy emission resonance is due to emissions from the upper cavity polaritons only at very low input intensities and switches to the bare cavity resonance approaching the lasing threshold, as shown earlier. This raises the question that if $n_{\rm th} > n_s$, why strong optical emissions from the lower cavity-polariton can still be observed at the threshold pumping intensity. In fact, the presence of a doublet near the lasing threshold was taken to be a crucial evidence for the role of cavity polaritons in the lasing process.

Properties of optical excitations in a microcavity depend critically on the density of excitons when the exciton density is near n_s. The persistence of lower cavity-polariton emissions near the lasing threshold reflects important effects of spatial and/or temporal variations of exciton densities within the excitation volume. In a typical optical measurement and at a given pumping intensity, the exciton (or carrier) density varies greatly from the center to the edge. There are always outer regions where the density of excitons falls below n_s even when the density of excitons exceeds n_s at the center. Optical emissions from these outer regions are characterized by emissions from two branches of cavity polaritons. As a result, emission spectra can feature simultaneously contributions from the bare cavity resonance as well as the cavity polaritons. Note that emissions from the upper-cavity polaritons are more than one order of magnitude smaller than that of the lower cavity polaritons and can be overwhelmed by emissions emerging from the bare cavity resonance near the threshold pumping intensity.

Figure 3 shows emission spectra obtained by collecting emissions from only the center region of the excitation volume to eliminate effects of spatial variation of exciton densities. The measurement was done under experimental conditions similar to that of Fig. 1(a) but with a 5 μm aperture placed at the center of *the image* of the excitation spot (the laser spot size is estimated to be 130 μm). As shown in Fig. 3, just below the threshold pumping intensity lower cavity-polariton emissions are one order of magnitude smaller than emissions from the bare cavity resonance. In comparison, at similar pumping intensities emission spectra for the whole excitation volume feature nearly equal contributions from the lower cavity-polaritons and the bare cavity resonance [see Fig. 1(a)]. The residual emission from the lower cavity-polaritons near the threshold pumping intensity shown in Fig. 3 is due to temporal variation of exciton densities as we discuss below.

Measurements discussed so far have used output from a mode-locked Ti:Sapphire laser and the time interval between successive pulses (13 ns) is large compared with the exciton or carrier recombination time (of order 1 ns). Under these conditions, both the density and the distribution of electronic excitations in the sample are a function of the time. In particular, there are temporal regions where the density of excitons falls below n_s even when the peak exciton density is above n_s. This is especially true for sufficiently long delays after an excitation pulse. Therefore, emission spectra obtained with pulsed excitations can still feature contributions from both bare cavity resonance and cavity polaritons even when exciton densities are spatially uniform.

Figure 4(a) shows emission spectra obtained with off-resonant cw excitation and with a 5 μm aperture at the center of the image of the excitation spot to eliminate both spatial and temporal variations of excitation densities. Emerging of the bare cavity resonance and correspondingly the saturation of the lower cavity-polariton emission at densities far below $n_{\rm th}$ is clearly observed. Near and below the threshold pumping intensity, emissions from the lower cavity polaritons are now reduced to more than two orders of magnitude smaller than emissions from the bare cavity resonance. Note that, experimentally, it is difficult to compare excitation levels of cw and pulsed pumping and we have used the threshold input intensity as a reference. To further illustrate effects of spatial variations of exciton densities, Fig. 4(b) also shows emission spectra obtained by placing a 25 μm aperture at the edge of the image of the excitation spot. In this case, the persistence and a continued linear increase of the lower cavity-polariton emission even at intensities above the threshold pumping intensity is observed. These results clearly demonstrate that cavity polaritons vanish at a density far below $n_{\rm th}$ and that the persistence of lower cavity-polariton emissions at the threshold pumping intensity shown

in Fig. 1 is due to emissions from spatial or temporal regions where the exciton density is below n_s.

Finally, we discuss briefly mechanisms that prevent the realization of nonequilibrium condensation of cavity polaritons. Theoretically, quantum statistical effects of polaritons were predicted to be effective when the interparticle distance is small compared with the polariton thermal de Broglie wavelength, which implies that at very low temperature bosonic effects can become important at exciton densities as low as $10^7/\mathrm{cm}^2$ in GaAs QW microcavities. Significant buildup of cavity-polaritons near $k=0$, however, can be prevented by the extremely short polariton life time (of order 1 ps) along with the long polariton-acoustic phonon scattering time (of order 100 ps or longer [10]). Another mechanism that can prevent the condensation from occurring is exciton localization. For typical QW structures, interface fluctuations can localize excitons in local minima of the confinement potential [11–13]. These localized excitons behave as fermions rather than bosons. Experimental realization of nonequilibrium condensation of cavity polaritons therefore requires the use of nearly perfect quantum heterostructures where effects of exciton localization are negligible even at extremely low exciton densities.

In conclusion, we have shown that cavity polaritons play no role in laser emission from GaAs QW microcavities at low temperature when the cavity is nearly resonant with the excitons. The threshold density is much higher than the saturation density at which normal mode splitting collapses and cavity polaritons vanish. The presence of emission doublets near the lasing threshold, which was previously taken as an evidence for laser emission from cavity polaritons, is the result of spatial and/or temporal variations of exciton densities within the excitation volume.

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