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Experimental evidence for nonequilibrium Bose condensation of exciton polaritons

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We observe dramatic changes in the near-field and far-field emission from a semiconductor microcavity excited by a pulsed and nonresonant optical pump with varying power. Above a threshold pumping power, light is emitted by a single quantum state lying at the bottom of the lower exciton-polariton band. Its intensity increases exponentially with the pump power and its linewidth becomes narrower than the cavity mode width. Near-field spectroscopy shows that the stimulated emission comes from several bright spots in the cavity plane, but no diffraction-induced angular broadening of the emission is observed. This is direct evidence for spontaneous formation of a nonequilibrium Bose condensate of coherent exciton polaritons with their wave function sharply peaked at structural imperfections.

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Bose-Einstein condensation[1] is a rare and intriguing physical phenomenon observed at ultralow temperatures in superconductors, superfluids, and atomic gases.[2,3] Recently, exciton polaritons[4] have been proposed as candidates for Bose-Einstein condensation at very high temperatures (up to 300 K and even more in wide-band-gap semiconductor and organic microcavities[5]). The exciton polaritons are two-dimensional quasiparticles of Fabry-Perot-type microcavities with embedded quantum wells, which result from the coupling between excitons (electron and hole pairs bound by Coulomb interaction) in the quantum wells and photon modes of the microcavity. Optical confinement in microcavities helps to achieve *the strong coupling regime*, when a characteristic anticrossing of the exciton and photon bands takes place, and two exciton-polariton dispersion branches are formed.[6] Having extremely light effective masses, the polaritons may condense at the bottom of their lower dispersion branch if they thermalize quickly enough.[7] Bose-Einstein condensation is defined as a phase transition of a bosonic system in thermal equilibrium towards a *coherent* state characterized by a long-lived order parameter (average complex amplitude of the optical field). Our system is characterized by a finite lateral size determined by the experimental conditions (e. g., the size of the optical beam used to pump the microcavity) for which a quasi-Bose-Einstein condensation[5,8] can take place at temperatures up to 150 K.

The quest for Bose-Einstein condensation of exciton polaritons began at the end of the 1990s when bosonic stimulation in the energy relaxation and scattering of polaritons was observed experimentally under nonresonant[9] and resonant excitation.[10] The theoretical argument in favor of the condensation of polaritons is their extremely light effective mass (of the order of 10^{-4} of the free electron mass). On the other hand, the extremely short polariton lifetime, typically on the order of 1 ps, is a major obstacle for the achievement of a fully thermalized polariton population. In 2002, Deng *et al.*[11] reported a noticeable variation of the second-order coherence $g_2(0)$ in the emission from a III-V compound semiconductor microcavity with an increase of pumping in the strong-coupling regime. This experiment showed that the statistics of exciton polaritons in their ground state changes from thermal-like to coherentlike as their concentration increases, which is an important step towards Bose-Einstein condensation.[12] Very recently the spontaneous buildup of a coherent phase of polaritons has been demonstrated in a II-VI compound semiconductor microcavity.[13] However, this phase transition takes place for polaritons with in-plane wave vectors $k_{\parallel} > 0$, and thus is not a Bose-Einstein condensation.

Our present paper reports a far-field and near-field photoluminescence study of exciton polaritons in a II-VI semiconductor microcavity under pulsed and nonresonant optical pump. By spectroscopic imaging in k space, we show that above a critical *stimulation threshold* the polaritons are accumulated in a single quantum state ($k_{\parallel}=0$ state) in our cavity, forming a nonequilibrium condensate. By means of near-field spectroscopy we investigate the wave function of the condensate and show that it is strongly inhomogeneous and is peaked around a few shallow potential traps formed by in-plane disorder in the cavity. This behavior of the condensate is reproduced theoretically with use of the Gross-Pitaevskii equation.

We have studied a CdTe/CdMgTe microcavity sample containing four quantum wells and characterized by a *vacuum field Rabi splitting* (i.e., splitting between the two exciton-polariton modes at the anticrossing) of 13.2 meV.[14] Electron-hole pairs were excited by linearly polarized light at 1.74 eV provided by a Ti:sapphire laser operating in the pulsed mode (at 80 MHz rate). The exciting laser spot diameter was about 20 μm on the sample surface. Both near-field and far-field time-integrated photoluminescence (PL) spectra were detected as a function of pumping power. For planar microcavities, photons emitted along a given direction θ are due to the recombination of polaritons with the in-plane wave-vector $k_{\parallel}=k_0 \sin(\theta)$ (where k_0 is the wave vector of the emitted light). Thus imaging the far-field emission onto the monochromator entrance slit allows us to probe the population distribution of the polaritons along their in-plane dispersion curve. In our setup, the angular resolution is about 1° for

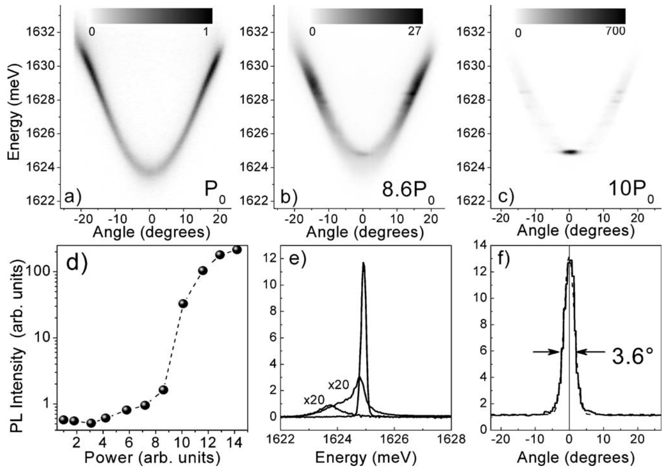

FIG. 1. (a) to (c): Gray-scale two-dimensional images of the far-field polariton PL intensity. Vertical and horizontal axes correspond to the emission energy and emission angle, respectively. PL intensity increases from white to black (see horizontal scale bars). Each map is obtained from direct observation of the Fourier plane. The excitation pumping was (a) $P_0=500\ \mathrm{W/cm^2}$, (b) 8.6 P_0, just below threshold, and (c) 10 P_0, just above threshold. (d) Spectrally integrated intensity of the emission from the polariton ground state $k_{\parallel}=0$ ($\theta=0$) normalized to the pumping power, as a function of pumping power. (e) PL spectra of the ground state, corresponding to these three pumping powers P_0, 8.6 P_0, and 10 P_0, normalized to the pumping power. (f) Angular profile of the emission at 1624.9 meV for the highest pumping power 10 P_0.

the far-field emission. All the measurements were done at 7.3 K and for a detuning of −5 meV between the bare exciton and photon modes.

Figures 1(a)–1(c) show intensity maps of the angle-resolved far-field PL from the lower polariton band (LPB) measured at three different values of pumping. At low pumping, light is mostly emitted from the so-called "bottleneck" region of the polariton dispersion curve. This can be explained as follows. Electron-hole pairs are created by the nonresonant pump, and very quickly form an incoherent exciton-polariton reservoir. Exciton polaritons from this reservoir relax efficiently along the excitonlike part of the LPB by interaction with optical and acoustic phonons. At the bottleneck region, the LPB becomes photonlike, characterized by an effective mass four orders of magnitude lighter than the exciton mass and a short lifetime (a few picoseconds). In the low pumping regime, the relaxation time in this region is longer than the radiative lifetime. Thus polaritons do not reach the ground state at $k_{\parallel}=0$, but remain near the bottleneck. The emission is quite broad both in energy and angle as shown in Fig. 1(a).

Increasing the pumping power speeds up the relaxation kinetics due to the onset of the polariton-polariton scattering mechanism.[5] One can see that the distribution of the polariton population shifts to lower energies while remaining broad in energy and wave vector in this regime [Fig. 1(b)]. The intensity emitted by the ground state increases linearly with the pumping in this intermediate regime [Fig. 1(d)] that shows that polariton-polariton scattering plays only a weak role and that the relaxation is dominated by interactions with phonons or residual free carriers.[6]

At stronger pumping, the average occupation of the lowest energy polariton state reaches unity. From this point, scattering of polaritons to the ground state becomes much more effective due to the bosonic final state stimulation effect. The corresponding pumping value will be referred to as the *stimulation threshold*. Above this threshold (about $5\ \mathrm{kW/cm^2}$ for the present experimental conditions), the rate for polariton-polariton scatterings, with one polariton going towards the ground state, is enhanced by a factor N_0+1 where N_0 is the ground-state population. This leads to an avalanchelike process that results in an exponential increase of the ground-state population [Fig. 1(d)]. The exciton-polariton distribution is now strongly peaked at the bottom of the LPB, approaching the equilibrium Bose-Einstein distribution function [Fig. 1(c)].

The emission spectrum above the stimulation threshold is shown in Fig. 1(e). The pumping power was only 15% higher than in the previous case, but the emission intensity has jumped by a factor of 50 as shown in Fig. 1(d). This implies that the ground-state population is at least $N_0\approx 50$ for this pumping power. The spectrum shape is also drastically different from those seen at lower pumping. The emission peak is extremely narrow both in energy and wave vector, and corresponds to the lowest-energy state, at the LPB minimum. Its linewidth is about 0.2 meV, 10 times narrower than the bare cavity mode, as shown in Fig. 1(e). This remarkable spectral narrowing indicates that, in the stimulation regime, the coherence time of the polariton population in the ground state is longer than the polariton lifetime. The full angular width of the emission is 3.6° as shown in Fig. 1(f). It corresponds approximately to the diffraction limit of our excitation spot size.

The strong intensity and extremely narrow linewidth of the emission peak strongly suggest that the detected light is emitted from a *single quantum state*, at the bottom of the LPB, i.e., from a Bose condensate of coherent exciton polaritons. The critical temperature for Bose condensation of exciton polaritons is governed by the value of the Rabi splitting, the effective mass of the polaritons, their concentration and the lateral size of the system (given by the exciting spot diameter). Simple estimates (4–8) show that under our experimental conditions the critical temperature is about 100 K, far above the actual temperature of our system (7.3 K). Thus, formation of the condensate is indeed possible above the stimulation threshold. It should be noted that the temperature of the polariton population in the present paper is not well defined due to the use of pulsed excitation pump. Therefore the spontaneous appearance of a phase coherent condensate observed here cannot be assigned to the Bose-Einstein condensation that is an equilibrium phase transition.

It is interesting to note that the bottom of the LPB is blueshifted by about 1.2 meV with increase of pumping. This shift, which is a factor of 6 larger than the broadening of the ground state, is due to the repulsive interaction between polaritons.[15] It is given by $E_{\mathrm{shift}}\approx VN$, where V is the matrix element of polariton-polariton interaction, and N is the total number of polaritons in the system. $V\approx 6xE_b a_b^2/S$, where E_b and a_b are the exciton binding energy and Bohr radius, S is the area of the sample, and x is the excitonic fraction of the lowest energy polariton state. Taking $x=1/3$, $E_b=25$ meV,

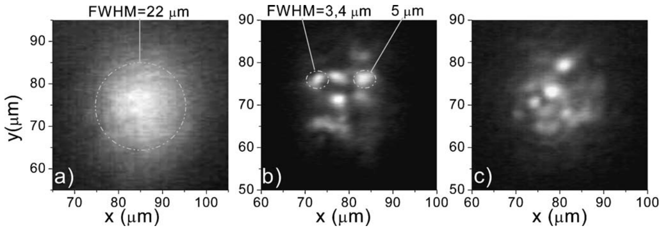

FIG. 2. Gray-scale two-dimensional images of the near-field polariton PL for different pumping powers. Vertical and horizontal axes are in-plane coordinates on the sample surface. PL intensity increases from black to white. The pumping power used was $P_0 = 500\ \mathrm{W/cm^2}$, below threshold, in (a), and $10\ P_0$, above threshold, in (b) and (c). Images (b) and (c) were taken from two different areas on the sample. FWHM stands for full width at half maximum of the PL intensity profile.

and $a_b = 30$ Å, we deduce the polariton density in our system at stimulation threshold to be $N/S \approx 2.7 \times 10^{11}\ \mathrm{cm^{-2}}$, which corresponds to $6.7 \times 10^{10}\ \mathrm{exciton/cm^2}$ in each of our four quantum wells. This is far below the CdTe exciton screening density of $4.8 \times 10^{11}\ \mathrm{cm^{-2}}$,[9] and thus one can conclude that the microcavity is still in the strong coupling regime. Another evidence of the strong coupling regime is the energy of the stimulated emission peak, which is 10 and 5 meV below the bare exciton resonance and the bare cavity modes, respectively.

Bose condensation of exciton polaritons in k space should be reflected by a macroscopic phase coherence of their wave function in real space. We have obtained this decisive proof by imaging the condensate wave function using near-field PL spectroscopy. Figs. 2(a)–2(c) show spatially resolved PL spectra taken at pumping powers (a) below the stimulation threshold and (b), (c) above it. At weak excitation, the emission has a Gaussian profile in the plane. Its size of 20 μm is close to the size of the excitation laser spot. Above threshold, the spatial profile of the emission is drastically modified. A small number of bright spots appear, each of them having a size of about 3 μm. By moving the laser spot over the sample, it could easily be seen that this marked inhomogeneous spatial distribution of the emission is induced by some structural disorder, e. g., fluctuations in the cavity width and/or Bragg mirror reflection coefficients. As shown in Fig. 2(c) the same near-field emission pattern is qualitatively reproduced at another place on the sample. This is very different from a previous report of polariton near-field emission in a GaAs microcavity.[16] It was found that the emission spot at stimulation threshold reduces drastically from 20 μm to a single spot of 4 μm around the center of the pump beam, which has been interpreted as a result of the Gaussian intensity profile of the pump beam. In our case, stimulated emission comes from several spots of various sizes (3–5 μm) randomly distributed within the Gaussian excitation spot; thus it can be concluded that polaritons are localized around some kind of traps.

Surprisingly, spatial localization of polaritons is not accompanied by any angular broadening of the far-field emission spectrum. While each 3-μm-size light source should have an angular width of at least 10°, the overall angular width remains within 3.6° [Fig. 1(f)]. This observation rules out the hypothesis of fragmentation of the condensate into different localization areas. It also rules out a statistical mixture of coherent states having random phases (the so-called "randomly phased coherent state"). Both of these would lead to a broad emission in k space. Thus, only a macroscopic phase coherence extended over the set of emitting spots can explain the narrow emission width. Moreover, since the emission line is much narrower than the cavity mode, all spots appear to emit light at the same frequency.

Clearly, the near-field and far-field data confirm the formation of a single coherent polariton state having an inhomogeneous wave function peaked at different spots. This wave function emerges spontaneously from the incoherent exciton-polariton reservoir, breaking the symmetry of the system to establish its own phase. The space dynamics of the condensate wave function moving in the inhomogeneous in-plane potential of the cavity can be modeled by the Gross-Pitaevskii equation[2,3]

$$i\hbar \frac{\partial \langle \psi(\vec{r},t) \rangle}{\partial t} = \left(-\frac{\hbar^2}{2m}\Delta + U(\vec{r}) + VS\left|\langle \psi(\vec{r},t)\rangle\right|^2 - i(D_S + D_V) \right) \times \langle \psi(\vec{r},t) \rangle, \qquad (1)$$

where m is the polariton mass, S is the system surface, $U(\vec{r})$ is the in-plane disorder potential, $\psi(\vec{r},t)$ is the condensate wave function, and $\langle \psi(\vec{r},t) \rangle$ is its quantum mechanical average, representing the order parameter for the Bose condensation of exciton polaritons. With respect to the standard Gross-Pitaevskii equation,[2,3] Eq. (1) includes the term describing decay of the order parameter (the last term in the brackets). The quantity $D_S/\hbar$ is the dephasing rate induced by the spontaneous scattering of polaritons toward the ground state. It is proportional to the ratio between the spontaneous scattering and the stimulated scattering toward the ground state.[17,18] It is approximately given by $D_S \approx \Gamma_0 / 2(N_0+1)$, where Γ_0 is the radiative broadening of the polariton ground state and can be neglected if $N_0 \gg 1$.[17] The quantity $D_V/\hbar$ is the rate of dephasing induced by interactions of the condensate with the reservoir of polaritons in excited states. We performed calculations using a value of 0.2 meV, corresponding to the width of the stimulated emission line, for the parameter $D_V + D_S$. We introduced five shallow potential traps in $U(\vec{r})$ to reproduce the experimental situation. Their depths ranged between 0.3 and 0.7 meV and their size was 3 μm. As the initial condition, we chose a Gaussian wave function with a full width at half maximum (FWHM) of 11 μm for the condensate density, which corresponds to the experimentally observed angular width of 3.6°, and $\int d\vec{r} |\langle \psi(\vec{r},0) \rangle|^2 = N_0$.

Figures 3(a)–3(c) show two-dimensional plots of the calculated condensate density $|\langle \psi(\vec{r},t) \rangle|^2$ at $t=0$, $t=6$ ps, and $t=30$ ps, respectively. One can see the buildup of sharp peaks at the potential traps and a ringlike distribution, which indicate possible formation of vortices pinned to the traps. The vortices are specific for systems of interacting bosons and can be observed in condensates of exciton polaritons. Fig. 3(d) shows the time-averaged $|\langle \psi(\vec{r},t) \rangle|^2$, which can be directly compared with the experimental near-field emission

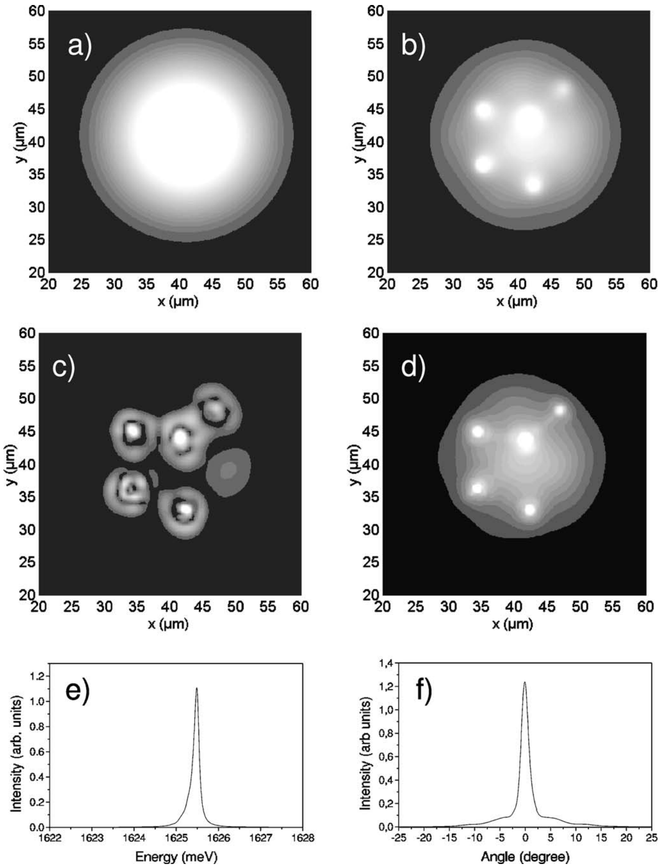

FIG. 3. (a) to (d) Gray-scale two-dimensional plots of the condensate density $|\langle\psi(\vec{r},t)\rangle|^2$ at (a) $t=0$, (b) $t=6$ ps, (c) $t=30$ ps, and (d) the time average over 100 ps. The density increases from black to white. (e) The PL spectrum of the condensate calculated from $\int_r |\langle\psi(\vec{r},\omega)\rangle|^2 d\vec{r}$, where $\langle\psi(\vec{r},\omega)\rangle$ is the time-Fourier transform of $\langle\psi(\vec{r},t)\rangle$. (f) Angular profile of the condensate emission as deduced from $\int_r |\langle\psi(\vec{k},t)\rangle|^2 d\vec{r}$, where $\langle\psi(\vec{k},t)\rangle$ is the space-Fourier transform of $\langle\psi(\vec{r},t)\rangle$.

spectra [Figs. 2(b) and 2(c)]. The experimental observation that the emission pattern comes mainly from the potential traps is quite well reproduced by the calculations, despite the Gaussian shape of the condensate wave function at the beginning. Localization of the wave function at different traps does not induce any substantial broadening in energy and wave vector space, as seen in the theoretical Figs. 3(e) and 3(f). For stronger dephasing, the wave function would be less localized around the traps, while its broadening in energy and wave vector would increase. These broadening effects would be expected also for deeper potential traps due to stronger localization of the condensate.

In conclusion, we have observed dramatic changes in the far-field and near-field emission of a microcavity optically pumped in the strong coupling regime. Just above the stimulation threshold the polariton population in k space is redistributed and becomes sharply peaked at the lowest energy state of the lower polariton band. The population of this ground state increases exponentially with the pump power, and its PL linewidth is reduced by one order of magnitude with respect to the bare cavity mode. The angular width of the PL is found to correspond to the inverse excitation spot size, even though near-field spectroscopy shows that the stimulated emission comes from five or six much smaller separate spots localized in the sample plane. This proves that light is emitted by a single macroscopic quantum state (condensate) having a spatially structured wave function. We estimate the corresponding number of polaritons in the condensate to be about 50 just above threshold. Modeling based on the Gross-Pitaevskii equation has reproduced the localization of the wave function of the polariton condensate at different potential traps in the plane of the sample without any additional energy and angular broadening. Vortices are possibly formed by the condensate and pinned by the traps. We believe that these observations represent unambiguous experimental evidence of a nonequilibrium Bose condensation of exciton polaritons in a semiconductor microcavity. Equilibrium Bose-Einstein condensation is still an open issue that could be clarified by future experiments using cw pumping to probe the thermal distribution of the polariton population.

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