Observation of a laserlike transition in a microcavity exciton polariton system

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We present experimental evidence of spontaneous buildup of coherent exciton polariton population in a microcavity, i.e., an exciton polariton laser. The laser phase transition was confirmed by the increased differential quantum efficiency and decreased linewidth of the lasing polariton mode due to onset of final-state stimulations and the decreased differential quantum efficiency of a nonlasing polariton due to onset of gain clamping at threshold. The exciton polariton laser is distinctly different from an optical laser because of its density-dependent scattering mechanism. The rate equations, taking into account phonon-assisted polariton emission and the polariton-reservoir exciton scattering rate, explain the measurement results well. [S1050-2947(96)51109-1]

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The indistinguishability and final-state stimulation in bosonic systems create many interesting physical phenomena, such as lasing, superconductivity, and Bose-Einstein condensation [1]. For a massive bosonic field, quantum statistics become important when the thermal de Broglie wavelength exceeds the interparticle spacing. One possible approach to generate a coherent population of nonequilibrium excitons is to dress an exciton with an electromagnetic vacuum field in a microcavity. The relevant quasiparticles in such a strongly coupled system are exciton polaritons [2]. Due to their light mass, GaAs quantum-well (QW) microcavity exciton polaritons with nearly zero in-plane wave number have a large de Broglie wavelength of 7μm (compared to 0.07 μm of a bare exciton) at 4 K [3], and it is relatively straightforward to obtain polariton densities with much smaller interparticle spacings than the thermal de Broglie wavelength. Due to their light mass (small density of final states), GaAs QW microcavity polaritons have also a smaller scattering rate by acoustic phonon absorption [3]. An exciton is not a pure boson and GaAs QW bare excitons are subject to exciton-exciton interaction and phase-space-filling effects at interparticle spacings smaller than 0.3 and 0.03 μm, respectively [4]. These critical values for undesirable scatterings are also improved by dressing excitons with an electromagnetic vacuum field. Therefore, at reasonably high exciton polariton densities below the Mott density, an exponential growth of exciton polariton occupancy due to final-state stimulation without scattering effects is expected. In this Rapid Communication we report experimental evidence of spontaneous buildup of coherent exciton-polariton population (laserlike behavior of microcavity exciton polaritons) based on the many-body polariton-phonon interaction.

The GaAs/Al$_x$Ga$_{1-x}$As microcavity sample is grown by molecular-beam epitaxy and has 19 (30) pairs of Bragg reflectors on top (bottom) and a 20-nm quantum well at the center of the λ cavity. The cavity buffer layer is tapered in one direction so that the cavity resonance energy varies with sample position, while the exciton energy is constant. The sample is cooled to 4.2 K and is off-resonantly excited above the band at 767 nm by a mode-locked Ti:sapphire laser (pulse width of 200 fs with repetition rate of 76 Mhz). Created electron-hole pairs form hot excitons with large in-plane momenta $k_\|$ that relax by multiple acoustic phonon emissions to populate the two exciton-polariton states at $k_\| \simeq 0$ (Fig. 1) [5]. It is this phonon emission rate for the last phonon into one of the two exciton-polariton states that is enhanced by final-state stimulation.

Figure 2(a) shows double anticrossing characteristics associated with heavy-hole (λ_{HH}=813.6 nm) and light-hole (λ_{LH}=811 nm) polaritons. The resonances are taken from fitting reflectivity spectra with superposition of Lorentzians. We have observed a Rabi splitting of 3.2 meV (2 meV) for the heavy- (light-) hole exciton in the reflectivity spectra. Calculation using a transfer-matrix formalism shows that this corresponds to an oscillator strength of 28×10^{-5} Å^{-2} (11×10^{-5} Å^{-2}), in agreement with the theoretical value of 30×10^{-5} Å^{-2} (13×10^{-5} Å^{-2}) for a 20-nm quantum well [6]. Figure 2(b) shows the anticrossing in the angular

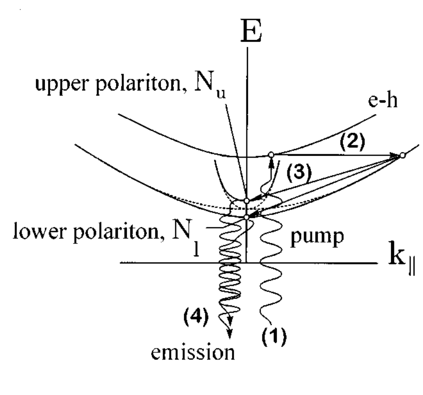

FIG. 1. Schematic energy flow in the microcavity exciton-polariton laser, which consists of (1) nonresonant excitation of electron-hole pairs, (2) formation of hot excitons, (3) exciton relaxation into two polariton states via phonon emissions, and (4) photon leakage from the cavity. Dashed lines denote dispersion with no exciton-photon coupling.

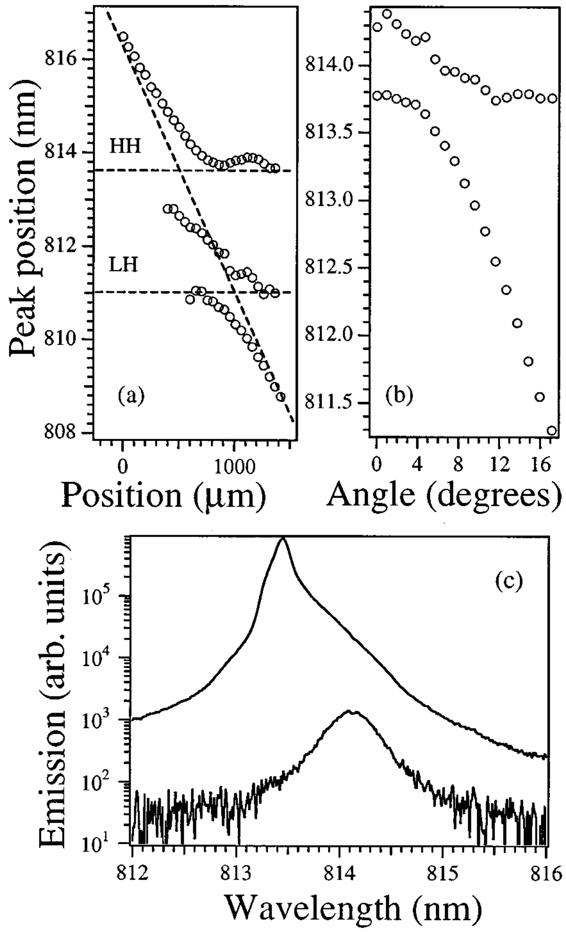

FIG. 2. (a) Dispersion characteristics for polariton energies taken from reflection spectra at different adjacent sample positions. (b) Anticrossing behavior of heavy-hole (HH) exciton polaritons measured from angular PL. (c) Emission spectrum of HH exciton polaritons at an excitation power of 0.005 and 0.2 mW/μm^2.

photoluminescence (PL) [7] at a high excitation of 0.26 mW/μm^2 above the threshold. Emission spectra of the heavy-hole polaritons with $k_{\parallel}\simeq 0$ for below and above the threshold are shown in Figure 2(c). The particular data [Fig.2(b)] show that the exciton-polariton splitting is decreased to $\sim$1.5 meV at high pumping; this is caused primarily by the fact that increased exciton density leads to a decrease in exciton oscillator strength [7]. We believe that we are at a density where the exciton oscillator strength is reduced because of exciton-exciton interaction. *Yet the polariton still exists, as shown by the anticrossing behavior in the angular resolved PL above threshold.* At low pump power, the emissions from the upper and lower polaritons increase linearly with pump, and the intensity difference can be described by thermal equilibrium Boltzman distribution of the two states [8], as shown in Fig. 2(c). Above a certain threshold, the population of the upper polariton starts to increase nonlinearly, leading to a deviation from thermal equilibrium distribution. In general, a polariton state can be expressed as the superposition of exciton and photon states $|\text{polariton}\rangle = a|\text{exciton}\rangle + b|\text{photon}\rangle$. These particular data were taken at a sample position where the upper polariton (higher energy branch) is a photonlike state, $|b|>|a|$, and the lower polariton (lower-energy branch) is an excitonlike state, $|b|<|a|$. When an excitation spot is shifted to an oppositely detuned position, we also observe the nonequilibrium buildup of the photonlike state (lower polariton in this case).

For the buildup of the upper polariton, the emission intensity and linewidth of the two polariton peaks obtained by fitting the spectrum with two Lorentzians are shown in Figs. 3(a) and 4(a). At a pump power density of 0.05 mW/μm^2, the differential quantum efficiencies of the upper and lower polariton emissions increase and decrease, respectively. These nonlinear behaviors resemble the onset of stimulated emission of a lasing mode and gain clamping of a nonlasing mode at above threshold in an optical laser. The pump power density of 0.05 mW/μm^2 corresponds roughly to the average excitation of an exciton-polariton density of 10^9–10^{10} cm^{-2} [9]. At this pump power density, the spectral linewidth of the upper polariton starts to decrease inversely proportionally to pump power. Note that the empty cavity linewidth is about 0.5 meV and the upper polariton linewidth decreases to below 0.2 meV. The result suggests the formation of phase coherence in the upper polariton population and the existence of random-walk phase diffusion due to spontaneous emission into the upper polariton (like the Schawlow-Townes linewidth for an optical laser). On the other hand, the spectral linewidth of the lower polariton continuously increases with increasing pump. The spectral linewidth of the lasing upper polariton at high pump rates also deviates from a conventional inverse pump power dependence. These results suggest the existence of polariton density-dependent dephasing and loss mechanism in our system. Similar results are observed using a cw pump light.

In the entire pump power region up to 0.5 mW/μm^2, we observe a decrease of exciton-polariton normal-mode splitting from 3.2 to 1 meV. Despite the decreased splitting, anticrossing behavior is still visible at pump power five times above threshold [Fig. 2(b)]. Similar results for the buildup of coherent population and decreased linewidth at a lower polariton taken at an adjacent sample position is shown in Figs. 3(b) and 4(b). In this case, the intensity of the upper polariton is too small to allow accurate extraction of linewidth, since the emission of the upper polariton is smaller by about a Boltzmann factor below threshold. We have performed the same experiment on a sample of the same structure but without the top mirror and found that the emission intensity is always sublinear and that the linewidth increases with increasing pump power.

In order to understand why the nonequilibrium buildup of exciton polaritons occurs at the photonlike branch instead of the excitonlike branch, we must consider the loss and gain of the two polariton modes. For finite exciton-cavity detuning, the photonlike polariton has a smaller effective mass than the excitonlike polariton. Thus the photonlike polariton has a larger thermal de Broglie wavelength λ_T and smaller polariton-reservoir exciton scattering rates, which leads to a larger net gain. Since λ_T is bigger and the effective scattering length is smaller, larger phase-space density can be achieved for the photonlike branch. This is the reason why a photonlike polariton [the upper polariton in the case of Fig. 3(a) and the lower polariton in the case of Fig. 3(b)] always wins to lase.

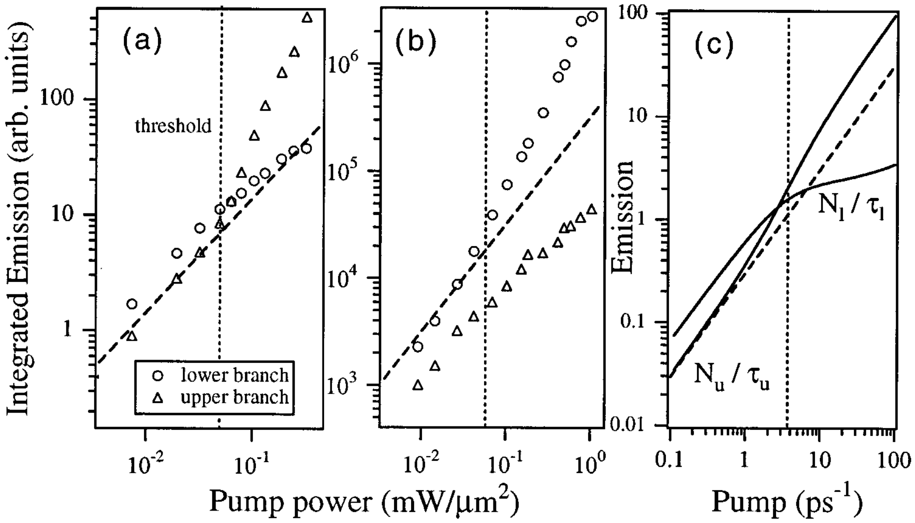

FIG. 3. Observed emission intensities of the upper and lower exciton polariton vs pump power density for the cases where nonlinear buildup occurs at the (a) upper polariton energy and (b) lower polariton energy. (c) Theoretical emission intensities of the upper and lower exciton polaritons vs pump rate for case (a) using the parameters: $\tau_r = 50$ ps, $\tau_u = 5$ ps, $\tau_l = 1$ ps, $C_{u(l)} = 0.3(0.25)$ ps^{-1}, $n = 2$, and $D = 0.001$ ps^{-1}. Vertical dashed line marks the location of threshold.

The exciton-polariton laser system is distinctly different from the optical laser system because of its density-dependent dephasing and loss mechanisms. There are three loss mechanisms that determine polariton lifetime: cavity leakage, acoustic phonon absorption, and polariton-reservoir exciton scattering, which originates from the exciton-exciton collision and leads to a broadening of the excitonic linewidth at high pump rate. In a conventional optical laser, the photon lifetime is independent of photon density inside the cavity. The polariton-reservoir exciton scattering is responsible for our observation of the deviation from the (inverse) power dependence of the linewidth of the lasing polariton in Figs. 4(a) and 4(b) with increasing pump.

To describe the dynamics of the polariton laser system, we use the rate equations in the three-level approximation (Fig. 1) in which both phonon-assisted polariton emission and polariton-reservoir exciton scattering are taken into account. Since the light-hole exciton is more than $k_B T$ away from the heavy-hole polariton levels, we shall neglect the presence of the light-hole transition in our simple rate equation analysis. The populations at the upper and lower polaritons, N_u and N_l, and at the reservoir excitons, N_r, obey [3]

$$\frac{dN_{u(l)}}{dt} = -\frac{N_{u(l)}}{\tau_{u(l)}} + C_{u(l)}[N_r(N_{u(l)}+1)(n+1) - N_{u(l)}(N_r+1)n] - D(N_l+N_u+N_r)N_{u(l)}, \quad (1)$$

$$\frac{dN_r}{dt} = P_r - \frac{N_r}{\tau_r} - C_l[N_r(N_l+1)(n+1) - N_l(N_r+1)n] - C_u[N_r(N_u+1)(n+1) - N_u(N_r+1)n] + D[(N_l+N_u+N_r)N_u + (N_l+N_u+N_r)N_l], \quad (2)$$

where $\tau_{u,l,r}$ are the decay rates of upper and lower polaritons and reservoir excitons, P_r is the pump rate to the reservoir, and n is the mean phonon population at the energy difference between the reservoir and the polariton. In writing (1) and (2), we neglect interband phonon scattering between the upper and lower polariton branch, which is small. $C_{u(l)}$ and D represent phonon-assisted polariton emission and polariton-reservoir exciton scattering coefficients, respectively. The polariton-exciton scattering rate introduces

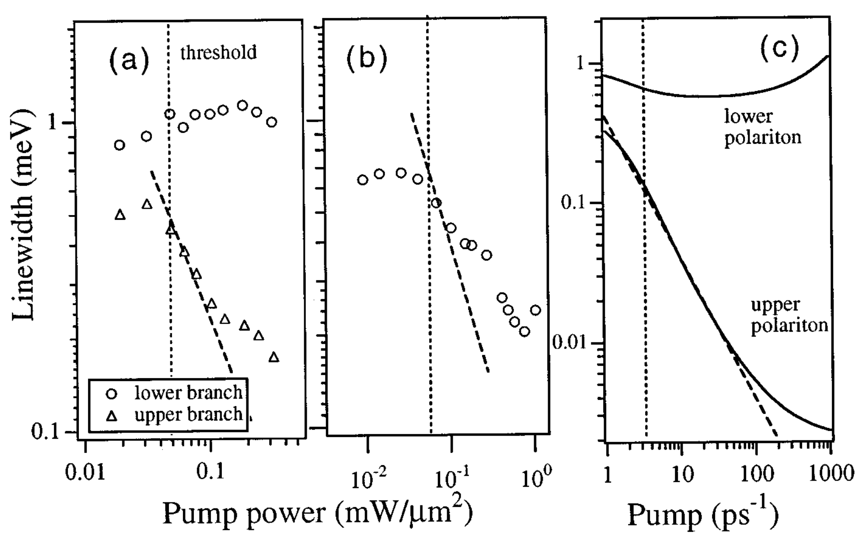

FIG. 4. Observed spectral linewidths of the upper and lower exciton polaritons vs pump power density for the cases where the nonlinear linewidth decrease occurs at the (a) upper polariton and (b) lower polariton (long dashed lines denote $1/P$ dependency). (c) Theoretical spectral linewidths of the upper and lower exciton polaritons vs pump rate for case (a), using same numerical parameters as in Fig. 3.

population-dependent loss and decreases net stimulated emission gain. Using (1), we calculate the linewidth of the polariton to be

$$\Delta\nu_{u(l)}=\frac{1}{2\pi}\left[\frac{1}{\tau_{u(l)}}+D(N_l+N_u+N_r)-C_{u(l)}(N_r-n)\right]. \tag{3}$$

For large pump, the linewidth for the lasing polariton due to spontaneous polariton emission is a constant

$$\Delta\nu_{\text{sat}}=\frac{n+1}{2\pi}\frac{CD}{C-D}\simeq\frac{n+1}{2\pi}D, \tag{4}$$

where $C=C_{u(l)}$ if the lasing polariton is the upper (lower) polariton. Figures 3(c) and 4(c) show the theoretical emission intensities and spectral linewidths of the two polaritons versus pump rate (for the case where the upper branch lases) using physical parameters of our system in (1)–(3). The parameters $C_{u(l)}$ are calculated from deformation-potential interaction [3]. The values for $\tau_{u(l)}$ are extracted from experimental reflection spectra, and D is estimated from Ref. [10]. The fact that polaritons are interacting particles limits the amount of linewidth narrowing. The (elastic) polariton-reservoir exciton scattering, which is not included in the present rate equation model, introduces an additional dephasing mechanism and further enhances the linewidth [11]. The observed linewidth of the lasing lower polariton [Fig. 4(b)] features such a distinctly different behavior from an optical laser. We define the threshold pump in analogy to that of an optical laser, i.e., the condition that the unsaturated gain equals a loss. From (1) and (2) and neglecting polariton-reservoir exciton scattering, the threshold reservoir exciton population $N_{r,\text{th}}$ and pump P_{th} are found to be

$$\frac{1}{\tau_{u(l)}}=C_{u(l)}(N_{r,\text{th}}-n), \tag{5}$$

$$P_{\text{th}}=\left[\frac{1}{\tau_{u(l)}}+(C_u+C_l)(n+1)\right]N_{r,\text{th}} \tag{6}$$

when the lasing mode is the upper (lower) branch. In order to obtain the net stimulated emission gain, the reservoir exciton population N_r must exceed the phonon population. This is the population inversion condition for a bare exciton boser [11]. In the limit of zero temperature ($n=0$) and zero-exciton-reservoir decay ($1/\tau_r=0$), the threshold pump rate becomes equal to $P_{\text{th}}=(C_u+C_l)/C_{u(l)}\tau_u$. This is analogous to the threshold condition for a microcavity laser with a spontaneous-emission coupling efficiency $\beta=C_{u(l)}/(C_u+C_l)$ [12].

Comparison of Figs. 3 and 4 shows that the three-level rate equation model qualitatively describes the experimental results. The threshold for the exciton-polariton system as described by (6) is different from that of an optical laser [11]. The dependence of phonon population n agrees qualitatively with experimental observation of the temperature-dependent threshold between 4 and 50 K. Experimentally, we observe an increase of threshold with increasing temperature. Similar threshold and linewidth behaviors are also observed by near-resonance and off-resonance cw excitation. The experimental result for saturated linewidth reduction shown in Fig. 4(a) was bigger than the theoretical prediction shown in Fig. 4(c). The difference is attributed to the neglect of the polariton-reservoir exciton elastic scattering effect [11,13], inhomogeneous polariton distribution [14], and index fluctuation due to nonequilibrium electron-hole pairs [15]. In conclusion, we present experimental evidence of a microcavity exciton-polariton laser and compare its characteristics with those of an optical laser. Our results can be described by a three-level rate equation model that includes both (1) phonon emission or absorption and (2) polariton scattering.

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