Interaction between a high-density polariton phase and the exciton environment in semiconductor microcavities

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Spontaneous polariton coherence is observed following creation of a cool exciton distribution in a GaAs-based semiconductor microcavity. Interactions between the high density polariton state and high-k excitons result in a blueshift in energy and are shown to limit the temporal coherence of the polariton phase to 10–20 ps. The effect of exciton-polariton interaction is also revealed in two beam experiments where the coherence of the polariton optical parametric oscillator is strongly suppressed by the creation of a reservoir population. These interactions are likely to be important in any polariton high density phase created under conditions of nonresonant excitation.

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There is significant current interest in the study of exciton-polariton systems which undergo transitions into high density macroscopically occupied states in semiconductor microcavities.[1] The transition can be achieved at much higher temperature than for excitons or atoms due to the very small polariton mass. Although at low excitation densities polariton relaxation is strongly inhibited by bottleneck effects,[2] condensation does occur for higher excitation densities when inter-particle scattering becomes important.[3] The formation of a high density state at the bottom of the lower polariton (LP) branch with occupancy $N \sim 10^3 - 10^4$ has been observed[4] for conditions of resonant excitation of the LP branch in a GaAs microcavity. This effect is due to direct polariton-polariton pair scattering from the pump to signal and idler states with $k \sim 0$ and $k \sim 2k_p$ and arises from optical parametric oscillator (OPO) behavior.[5] Recently, condensation to the ground state of the LP branch was reported in a CdTe-based MC under nonresonant excitation, accompanied by the observation of spatial coherence of the $k=0$ polaritons.[6] By contrast, the small exciton binding energy of GaAs-based microcavities has been an obstacle to the observation of a high density polariton phase for nonresonant excitation, since collapse of exciton-photon strong coupling occurs before a high polariton density is achieved.[7,8]

In this work we observe macroscopic occupation of the $k=0$ polariton state by resonant excitation of the UP branch in a GaAs microcavity. We demonstrate that the occupation arises from incoherent relaxation from a cool exciton population, the creation of which is found to be a crucial condition in such cavities for the observation of spontaneous polariton coherence from the $k=0$ LP states. The short coherence time and energy renormalization of the stimulated polariton emission we observe are attributed to the interactions of the polariton phase with the exciton reservoir. The strong influence of interactions on the coherence is further confirmed in two-beam experiments: we investigated the coherence time of the OPO emission occurring for resonant excitation of the LP branch[4] using additional weak laser excitation into the UP branch. A reduction of the coherence time by a factor of 2–3 due to interactions with the reservoir excitons is observed. These results further suggest the importance of interactions between the Bose-condensed polariton state recently reported in II-VI systems and the exciton reservoir from which condensation occurs.[6]

The structure of the GaAs microcavity (MC) studied is identical to that of Ref. 4. Several spatial regions across the sample were investigated at $T \sim 5$ K, all with Rabi splitting $\Omega \sim 6$ meV and near zero detuning between exciton and cavity modes, which have energy of ~1.4535 eV at $k=0$. A tunable cw Ti-sapphire laser beam was focused to ~30 μm on the sample at an angle of incidence of $\theta_p = \sim 3° - 29°$ to achieve excitation of polariton photoluminescence. Spectrally resolved images of the near and far-field polariton emission were recorded using a standard imaging setup. First order correlation functions were measured using a Mach-Zehnder interferometer.[9]

Figure 1(a) shows the dependence of the polariton emission intensity on energy and emission angle (θ), for nonresonant excitation at an energy ~100 meV above the LP branch, at a low excitation power of 1 mW. The (θ,E) images exhibit the familiar near-parabolic dispersions of both the upper (UP) and lower (LP) polariton branches. The UP linewidth is ~1.5 meV, much larger than that of the LP

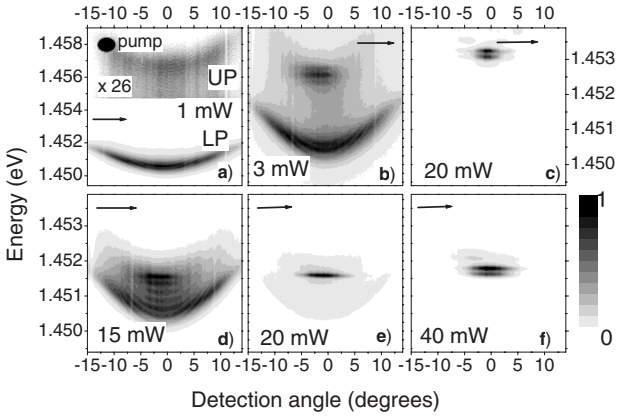

FIG. 1. Two dimensional images of the far field emission for nonresonant excitation above the GaAs band gap (a)–(c) and for resonant excitation into the upper polariton branch (d)–(f) at different excitation powers. Vertical and horizontal axes correspond to the emission energy and emission angle (θ). The PL intensity in each image is normalized. The arrow shows the energy of the bare photon mode.

[full width at half maximum (FWHM) ~0.35 meV] due to efficient scattering of upper polaritons by the quantum well disorder potential to the reservoir of excitons with high in-plane k vector.[10] Increasing power results in significant broadening and blue-shift of the polariton emission. At powers of ~3 mW, emission emerges at higher energy (note the different energy scales between Figs. 1(a)–1(e)), with maximum in intensity around $\theta \sim 0$ [Fig. 1(b)]. With further increase of the excitation power the emission transforms into a narrow line (FWHM ~0.1 meV) blueshifted to $E = 1.4532$ eV by 2.7 meV with a pronounced maximum in k space at $\theta \sim 0$ [Fig. 1(c)], while its intensity exhibits thresholdlike superlinear behavior, indicating the onset of stimulation. Above threshold the emission [Fig. 1(c)] occurs at an energy close ($\Delta \sim 0.3$ meV) to the energy of the bare photon mode (~1.4535 eV, shown by the arrow) and is attributed to the stimulated emission of photons in the *weak* coupling regime, where the excitonic absorption resonance is bleached due to phase space filling and screening.[11,7]

By contrast, the nonlinear behavior is markedly different for excitation resonant with the UP branch at pump angles, θ_p in the range 3°–29°. Figures 1(d)–1(f) show (θ, E) images at different powers for $\theta_p \sim 13°$ [the UPB resonant excitation pump position for Figs. 1(d)–1(f) is indicated on Fig. 1(a)]. With increasing power the LP emission broadens and shifts to higher energies. At an excitation power of 15 mW a narrow peak appears at 1.4515 eV. With further increase of power this emission occurs in the normal direction ($\theta \sim 0$) and shifts to higher energies by ~0.3 meV, and its intensity increases superlinearly. However, in contrast to the case of nonresonant excitation [Fig. 1(c)] the stimulated emission occurs at an energy which is lower than that of the uncoupled photonic mode by $\Delta = 1.7$ meV. Since Δ is larger than the halfwidths (HWHM) of the uncoupled exciton and photon modes (~0.7 meV) and of the LP branch (~0.17 meV), we conclude that the observed stimulated emission occurs in the strong coupling regime, and arises from a macroscopically occupied polariton state, which is populated by stimulated relaxation.[12] To further verify this point we performed photoluminescence excitation (PLE) measurements of the emission at $\theta = 0$ by tuning the energy of the laser between the lower and the upper polariton branches.

Figure 2(a) (solid squares) shows the variation of the intensity of the LP emission at $\theta = 0$ (energy 1.4504 eV), as a function of the laser energy E_l at low excitation power of 2 mW below the threshold for stimulated emission. The PLE spectrum shows two well-defined peaks at energies of 1.458 and 1.4515 eV, which correspond to the UP and LP states at $\theta = 13°$, respectively. Increasing the power to 40 mW, above the threshold for stimulated emission, the PLE spectrum still contains two distinct resonances corresponding to the LP and the UP branches [Fig. 2(b)], showing that the system remains in the strong coupling regime. As E_l is decreased, a monotonic decrease of the energy of the stimulated emission, E_s (open circles) first occurs.[13] However, at $E_l < 1.454$ eV an abrupt jump of E_s to higher energies occurs, and the intensity of the stimulated emission at $\theta = 0$ increases significantly by ~50 relative to the case of excitation into the UP branch. Such behavior arises when resonance is approached with the LP branch; under such conditions efficient polariton-

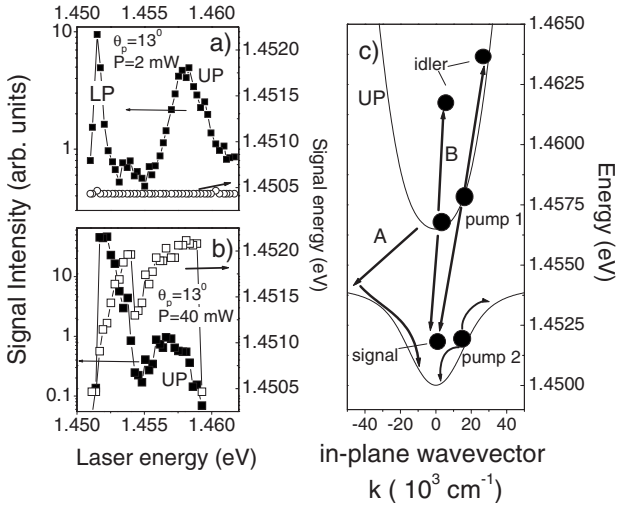

FIG. 2. (a) Intensity (solid) and energy (open) of polariton emission at $\theta = 0$ as a function of excitation energy for resonant excitation into the UP branch for excitation power of 2 mW, below threshold, (b) at 40 mW, above threshold. (c) Schematic diagram of relaxation processes for resonant excitation into the UP branch (pump 1), by processes A, relaxation via the reservoir and B pair scattering. Pump 2 corresponds to resonant excitation of the LP branch in the two-beam experiment.

polariton pair scattering from the pump to the LP ground state switches on.[4]

The stimulated emission at ~1.7 meV below the bare cavity mode for the case of resonant pumping of the UP branch [Figs. 1(e) and 1(f)] can thus be explained in terms of bosonic stimulation of polariton relaxation to the ground state, which results in the formation of a macroscopically occupied polariton state. The number of particles at $k = 0$ at $P = 60$ mW is estimated to be 50–100 taking into account the total intensity of the polariton emission and polariton lifetime (~5 ps). As shown below the properties of this high density phase are determined by the interaction with exciton environment and are different from the OPO case.

There are two main relaxation channels which may lead to the formation of the macroscopically occupied state at $\theta = 0$. First, quasielastic scattering of the upper branch polaritons by the quantum well disorder potential will result in efficient population of the exciton reservoir at high-k vector. These excitons will subsequently relax towards the ground state of the LP branch as a result of exciton-exciton scattering[3] or via long lived localized excitons[14] (channel A, Fig. 2). Secondly, polariton-polariton pair scattering may occur directly from the pump to the LP branch and corresponding UP states with energy and momentum conservation [channel B, Fig. 2(c)].[15] For the latter mechanism a rapid increase in the threshold for stimulation by a factor of ~1000 is predicted by semiclassical theory[16] of the OPO with increasing excitation angle (θ_p) from 3° to 30° as shown in Fig. 3(a) (open squares). This theory takes into account the polariton linewidths, excitonic contents and the phase matching conditions for the states involved in the scattering process. The rapid increase in the threshold is predicted due to the strong reduction of the excitonic component of the idler and pump UP states from ~50% to ~2–10% with increasing θ_p.

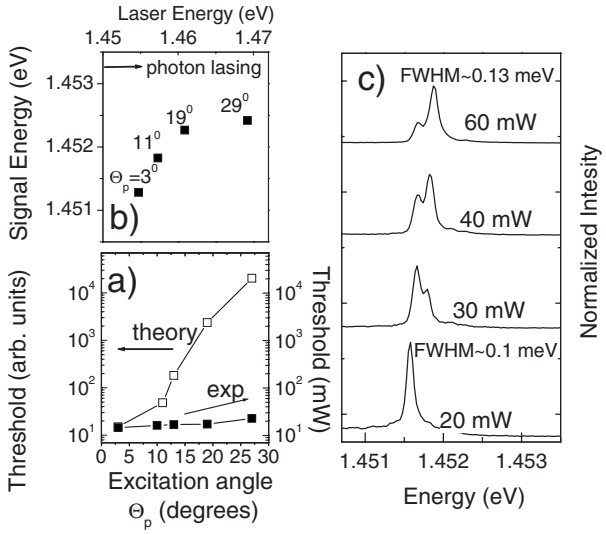

FIG. 3. (a) Calculations of the threshold for onset of stimulated parametric scattering as a function of the excitation angle (open) of the UP branch, compared with experiment (solid). (b) Energy of signal emission versus resonant excitation energy and excitation angle. (c) Spectra of polariton emission at powers above threshold at $\theta=0$.

In order to reveal the dominant polariton relaxation mechanism (process A or B), the polariton emission intensity was studied for varying θ_p into the UP branch. Figure 3(a) (solid) shows that the experimental threshold power P_{th} for stimulated emission is insensitive to angle, increasing slightly from 15 to 23 mW with θ_p from 30° to 29°. This is in marked contrast to the theoretical dependence expected for pair scattering from the pump (process B), indicating that the stimulated polariton emission occurs predominantly due to relaxation via the reservoir (process A). The contribution of the process B occurs only above threshold and is very small ($<5\%$).

In this context, we note that Deng *et al.* recently reported the formation of a high density polariton phase under pulsed resonant excitation of lower branch excitonlike polaritons at high $k\sim 5\times 10^4$ cm^{-1}.[17] Although the observation was explained as the result of polariton thermalization and Bose condensation, direct polariton-polariton pair scattering (OPO) to the LP ground state is likely in this case to lead to the macroscopic occupation due to the high excitonic content of the pump and idler states [process B in Fig. 3(a)].[18]

In Fig. 3(b) it is shown that the signal energy E_s increases (by 1.2 meV) with increasing excitation angle θ_p, with simultaneous change of the pump energy to maintain resonant excitation into the UP. Nevertheless, even at high θ_p the energy of the stimulated emission is still ~1.1 meV below the energy of the bare photon mode (~1.4535 meV), demonstrating again that strong coupling is retained.

The blueshift of the stimulated LP emission and the red shift of the UP states at high power [Fig. 2(b)] arise from a decrease of the oscillator strength of excitons (and hence a decrease of the Rabi splitting) due to exciton-polariton interactions and Coulomb screening. The effective temperature of the exciton distribution depends on the excitation energy, since exciton relaxation and life times have comparable magnitude.[19] Thus increase of θ_p, and thus excitation energy E_l, results in a greater concentration of high energy, high k excitons, which scatter more effectively with other excitons and polaritons, resulting in greater decrease of the Rabi splitting. Furthermore for E_l more than ~10 meV above the exciton energy ($\theta_p > \sim 20°$), free carriers can be created, leading to a further effective screening channel. The expected increase of exciton screening with E_l thus accounts for the increase of the signal energy with E_l (and θ_p) in Fig. 3(b) and also in Fig. 2(b) at fixed θ_p.

For E_l above the GaAs band gap, by contrast, a strongly nonequilibrium distribution of free hot carriers and excitons is formed, that leads to a strong signal blueshift by ~2 meV at threshold [Fig. 1(b)]. With further increasing excitation power by ~50–100 % a rapid transition into the weak coupling regime [Fig. 1(c)] occurs. We conclude that creation of a cool exciton distribution using resonant excitation into the UP branch is crucial to retain strong coupling and to observe spontaneous coherence in LP ground states in such GaAs cavities.

Further evidence for the role of interactions between the polariton phase and the reservoir of excitons is obtained from study of the temporal coherence of the polariton stimulated emission. Figure 3(c) shows polariton emission spectra above the stimulation threshold, as a function of excitation power. At low power of 20 mW, the emission consists of a single peak with FWHM of 0.1 meV (corresponding to a coherence time $\tau_c\sim 20$ ps), a factor of 2 larger than the spectrometer resolution. With further increase of power the emission evolves into a double mode structure,[20] with the linewidth of each mode also becoming broader, ~0.13 meV (~15 ps). This polariton coherence time (τ_c) is much smaller than that observed for the signal emission of the OPO (~200–500 ps) obtained for resonant excitation into the LP where population of the exciton reservoir is much smaller[9] than for the case of UP branch pumping, strongly suggesting the important role of interactions of polaritons at $k=0$ with reservoir excitons for the case of excitation into the upper branch.

In order to further demonstrate the effect of such interactions on the coherence properties, a two beam experiment was performed [Fig. 2(c)]: one beam (pump 2) is used to resonantly excite the LP branch at $\theta=12°-15°$, resulting in efficient OPO emission at $\theta=0°$; the other beam (pump 1) is used to resonantly excite the UP branch, resulting in efficient population of the reservoir. Figure 4(a) shows the signal emission spectrum (solid) excited by pump 2 only, for an excitation power of 35 mW above the stimulation threshold. When additional low power (2 mW) excitation with pump 1 is added, a small reduction of intensity by 30% is found (open).[21]

However, a much stronger effect on the coherence time is found, measured using a Mach-Zehnder interferometer as in Ref. 9. Figure 4(b) (solid) shows the signal emission intensity for pump 2 excitation, as a function of the phase shift between the two arms of the interferometer, at a delay time of 230 ps. The visibility of the interference fringes is ~25%, from which a τ_c of 190 ps is deduced. By contrast, with additional pump 1 illumination the visibility (open) is less than 10%, indicating τ_c to be in the range from 40 to 70 ps.

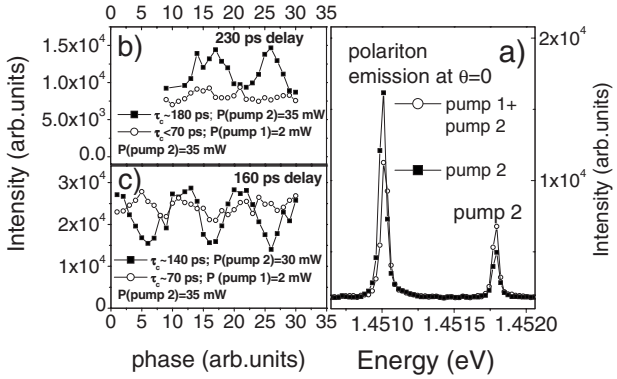

FIG. 4. (a) Spectra of polariton emission for resonant excitation with pump 2 only (solid) [Fig. 2(c)] and simultaneous excitation with pumps 1 and 2 [Fig. 2(c)]. Interference fringes as a function of the phase variation between the two beams of the Mach-Zehnder interferometer at delay times of 230 ps (b) and 160 ps (c) (one beam solid, two beams open symbols).

Since the coherence time of a macroscopically occupied state depends on the number of particles we also performed a two beam experiment where we varied the intensity of the pump to maintain constant signal intensity, and hence $k=0$ occupancy. Figure 4(c) shows the interference with (open) and without (solid) additional excitation to the UP at pump 2 power of 35 (open) and 30 mW (solid), respectively. In this case we also observed a reduction of the signal coherence time down to ~70 ps. The above experiments clearly demonstrate the major effects of interaction between the high density polariton state at $\theta=0$ and the exciton reservoir on the macroscopic properties of the polariton phase.

In conclusion, the formation of a macroscopically occupied polariton state has been reported, initiated by incoherent relaxation from a cool exciton reservoir. Strong polariton-exciton interactions lead to significant energy renormalization and limit the temporal coherence of the polariton stimulated emission to 10–20 ps. Similar coherence times of ~10 ps were observed for the polariton condensate in CdTe microcavities under nonresonant excitation conditions.[6] We conclude that polariton condensates excited nonresonantly do not exist in isolation but rather interact strongly with their quasiparticle environment.

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