Intrinsic Decoherence Mechanisms in the Microcavity Polariton Condensate

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The fundamental mechanisms which control the phase coherence of the polariton Bose-Einstein condensate (BEC) are determined. It is shown that the combination of number fluctuations and interactions leads to decoherence with a characteristic Gaussian decay of the first-order correlation function. This line shape, and the long decay times (~150 ps) of both first- and second-order correlation functions, are explained quantitatively by a quantum-optical model which takes into account interactions, fluctuations, and gain and loss in the system. Interaction limited coherence times of this type have been predicted for atomic BECs, but are yet to be observed experimentally.

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The recent observations of polariton condensation in semiconductor microcavities have provided a new, solid state system for the study of Bose-Einstein Condensate (BEC) phenomena [1,2]. The Bose-condensed state exhibits characteristic properties including massive occupation of the ground state [1–3] and long range spatial coherence [1], which due to the very small mass of polaritons (10^{-5} of the free electron mass) occur at high critical temperatures of 20–30 K [4]. Like atomic BECs, the polariton condensate is a mesoscopic system containing a few hundred particles, so number fluctuations are important. The fundamental mechanisms limiting the coherence times of such mesoscopic condensates have been discussed in the case of atomic BECs [5], but have yet to be observed experimentally [6].

In this work, we demonstrate that the phase coherence time of such systems is intrinsically limited by the combined effect of number fluctuations and interactions in the coherent state. Since the polaritons interact with each other, the number fluctuations lead to fluctuations in energy which cause the coherence to be lost, giving a characteristic Gaussian decay of the first-order correlation function ($g^{(1)}(\tau)$) which we observe experimentally. These effects have not been observed previously for polariton condensates, because of dominating pump laser noise. Our measurements, using a noise-free diode laser, reveal very long coherence times up to ~150 ps, up to 30 times longer than those deduced previously [1,2], making it realistic to study other condensate phenomena, such as Josephson oscillations, in polariton systems.

It is notable that the polariton condensate differs from an ideal BEC in that it is not in equilibrium. Polaritons decay, as externally emitted photons, with a lifetime ~1.5 ps, so the steady-state population has to be maintained by a pump laser. This continual replacement of particles is a unique aspect of the polariton condensate, and has to be accounted for in any theoretical treatment of the system. Here, we present a model which shows that the decoherence mechanism discussed above, which is essentially a property of an equilibrium condensate, can be observed despite the short lifetime. This is possible because the effective time for the replacement of polaritons is lengthened by stimulated scattering terms in the gain, which dominate above threshold. The lengthening is demonstrated directly by experimental measurements of the second-order $g^{(2)}$ coherence function, which show a decay time of ~150 ps. Using only experimentally derived parameters as input, our quantum-optical model explains simultaneously the time dependence of both $g^{(1)}$ and $g^{(2)}$ functions, which we show have very different physical origin.

The sample employed here is a piece from the CdTe-based microcavity wafer used in Ref. [1] where BEC with extended spatial coherence was reported. Figure 1 shows spectra from the bottom of the lower polariton (LP) branch, excited nonresonantly with a semiconductor diode laser free from intensity fluctuations on a 0.1–1 ns time scale, below and above the threshold for condensation. Above threshold the spectrum breaks up into a series of lines with ~0.2 meV separation, which are more than 30 times narrower than that below threshold. The marked narrowing and the accompanying superlinear increase of the intensity with excitation power [Fig. 1 (inset)] are characteristic of the formation of a macroscopically occupied polariton condensate. We note that the resolved spectral structure and the narrow linewidths (long coherence times) are not observable under multimode laser excitation [1,2], where as we argue later, laser intensity fluctuations obscure the quantum properties of the polariton BEC.

To understand the origin of the coexisting condensates revealed by the high spectral resolution, spectrally and

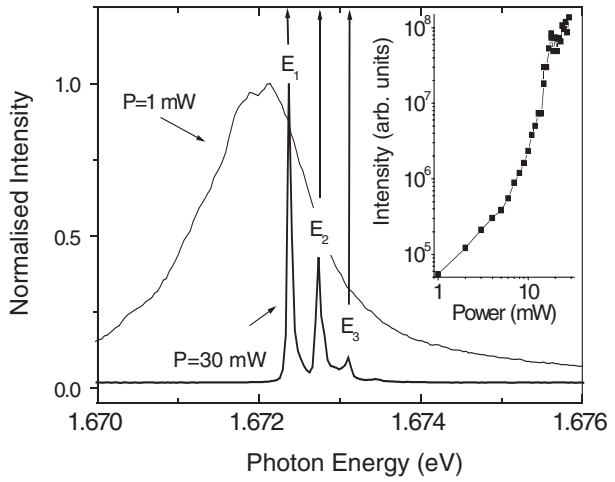

FIG. 1. Spectra corresponding to polariton emission from the bottom of the LP branch for detection angle $\Theta = \sim 0°$ at excitation powers $P = 1$ mW and 30 mW, respectively, below and above the threshold ($P_{th} \sim 10$–15 mW) for condensation. Below threshold, the linewidth is broad (FWHM ~ 1.5 meV), determined by the polariton lifetime ($\sim$1.5 ps). By contrast, above threshold the emission consists of a number of very narrow lines with energy separations of $\sim$0.2 meV and linewidth of 0.06 meV (limited by the spectrometer resolution: the true linewidths (decay times) are obtained from the first-order correlation measurements). The inset shows the strongly nonlinear variation of the intensity of mode E_1 with power.

spatially resolved images were obtained below [Fig. 2(a)] and above [Figs. 2(b)–2(d)] threshold. Below threshold the emission has the Gaussian profile of the excitation laser. By contrast, above threshold each mode has a distinct spatial pattern, with intensity maxima separated by $\sim$2–3 μm.

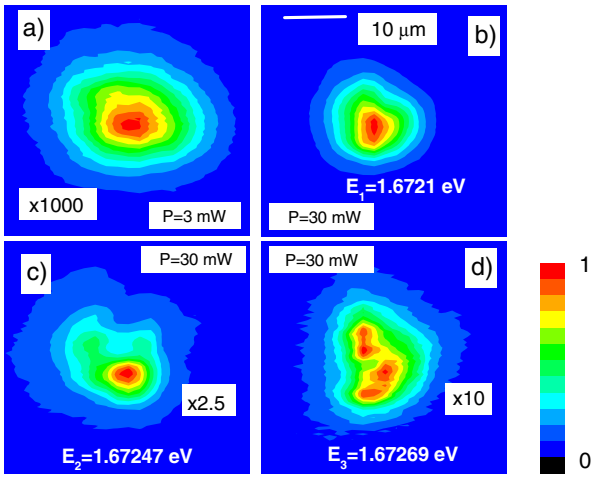

FIG. 2 (color online). (a)–(d) Spectrally resolved, spatial images ($\sim$2 μm resolution) of the polariton emission for $P =$ 3 mW (a) and $P = 30$ mW (b)–(d). The detection energies E_1 to E_3 for images (b) to (d) are given on the images and correspond to detection at the energies of the individual, resolved modes of Fig. 1.

The modes nevertheless show strong spatial overlap. Also the intensity of the modes decreases going to higher energy in the spectrum. We attribute the coexistence of the overlapping condensed states to weak spatial fluctuations ($\sim$0.2–0.5 meV) in the cavity mode energy [1,7] which result in localized polariton modes at different energies [8]. Above threshold condensation into each localized mode occurs, and the modes become spectrally resolved due to the accompanying strong narrowing.

Having achieved spectral resolution of localized states, we investigate the time dependence of the phase coherence of the strongest individual condensate mode using a Mach-Zehnder interferometer. Figure 3(a) shows the decay of the first-order correlation function $g^{(1)}(\tau)$ for a single mode above threshold. It is found to have a Gaussian form with a coherence time $\tau_c^{(1)}$ of $\sim$120 ps. In Fig. 3(b), the variation of this phase coherence time is plotted as a function of the intensity of the emission, which is proportional to the number of particles in the condensed state [9]. The coherence time first grows rapidly with increasing intensity, from $\sim$1.5 ps around threshold, until it saturates to a value of $\sim$120–150 ps [10], at a pump power approximately 2 times threshold. The higher energy modes are found to exhibit coherence times close to that of the most intense mode within experimental error.

More information about the character of the condensate was obtained from measurements of the second-order (in-

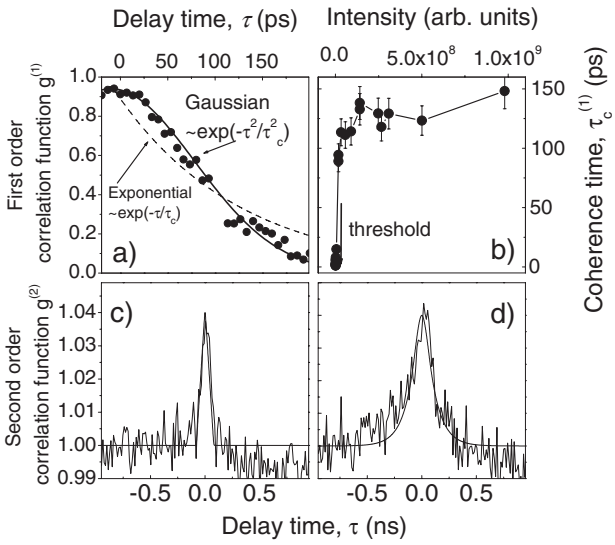

FIG. 3. (a) Variation of $g^{(1)}$ first-order correlation function versus delay time for a single condensate mode above threshold. (b) Phase coherence time $\tau_c^{(1)}$ of the condensate mode versus emission intensity showing a strong increase at threshold followed by saturation at powers approximately a factor of 2 above threshold. (c)–(d) Measured $g^{(2)}$ second-order correlation function versus delay time at powers $P = 6$ and 30 mW, below (c) and a factor of 2 above (d) threshold, respectively. The full lines are fits with values for $g^{(2)}(0)$ and the coherence time, $\tau_c^{(2)}$ of 2 and 1.5 ps below threshold and 1.1 and 100 ps above.

tensity) correlation function $g^{(2)}(\tau)$ using a Hanbury Brown–Twiss (HBT) setup [11]. The $g^{(2)}$ results, which provide essential parameters for the modelling of the coherence below, are presented in Figs. 3(c) and 3(d) for two pump powers, below and a factor 2 above threshold. In both cases photon bunching is observed with the measured $g^{(2)}$ $(g^{(2)}_{\mathrm{meas}}(\tau))$ having a maximum at $\tau = 0$ of ~ 1.04. However, the decay times $(\tau_c^{(2)})$ are different: ~ 40 ps (detector resolution limited) below and ~ 100 ps above threshold, respectively. Using an expression relating $g^{(2)}_{\mathrm{meas}}(\tau)$ to the true value of $g^{(2)}(0)$, which takes into account the detector efficiency, the resolution time of the detectors (~ 40 ps) and $\tau_c^{(2)}$ [11], and assuming the expected value of $g^{(2)}(0) = 2$ for a thermal state below threshold, we calibrated the efficiency of the detectors. The true value of $g^{(2)}(0)$ above threshold is then found to be $\sim 1.1 \pm 0.015$, slightly above 1, the prediction for a coherent state. This value is consistent with values in Ref. [12], but it should be noted that the present studies eliminate any fluctuations from multimode excitation.

We now discuss our model for the first and second-order coherence treating initially an isolated, equilibrium BEC, then introducing the nonequilibrium character. Consider an isolated state, with a Gaussian probability distribution for the number of polaritons $P(n)$ expected, for example, for a coherent state. This is characterized by its mean $\bar{n}$ and variance (fluctuations in particle number) σ^2. The state evolves under a nonlinear Hamiltonian $H = \kappa a^+ a a^+ a$, describing the polariton-polariton interaction. To find $g^{(1)}(\tau)$, we evaluate $\langle a^+(\tau) a(0)\rangle$, which corresponds to removing a polariton at time $t = 0$ and putting it back at $t = \tau$. The phase change is τ times the difference in energy of states with n and $n-1$ polaritons, that is $\kappa\tau(n^2 - (n-1)^2) \sim 2\kappa\tau n$. Averaging over the probability function $P(n)$ gives

$$g^{(1)}(\tau) \sim \int dn P(n) \exp(2i\kappa\tau n) \sim \exp(-2\kappa^2\sigma^2\tau^2) \quad (1)$$

which has the Gaussian form observed experimentally in Fig. 3(a).

We obtain the variance σ^2 from the second-order correlation function, $g^{(2)}(0) = 1 + (\sigma^2 - \bar{n})/\bar{n}^2$. Using the measured $g^{(2)}(0)$ of 1.1, and estimating $\bar{n} \sim 500$ from the emission intensity, we obtain $\sigma^2 = 25\,500$. As shown below, κ is estimated to be 2×10^{-5} ps^{-1} from the blueshift (0.5 meV) from below to above threshold. This gives a decay time $\tau_c \sim 220$ ps, close to the experimental value of ~ 150 ps. An equilibrium model including number-fluctuations and interactions thus explains both the $g^{(1)}(\tau)$ line shape and the quantitative values of the decay time.

The coherent mode is, of course, not isolated, as it is not in equilibrium; polaritons are lost by external emission at a rate determined by the cavity linewidth γ, and are replaced from the reservoir of particles in other modes [13]. This disrupts the Hamiltonian evolution on a time scale τ_r, which depends on the loss rate, but is generally slower than γ^{-1}, because the stimulated component of the scattering into the mode exactly replaces the particle which is lost. This time scale is obtained most directly from the decay of $g^{(2)}(\tau)$, that is $\tau_c^{(2)} \sim 100$ ps. It is this slowing down which allows us to see the equilibrium condensate physics, despite the short cavity lifetime.

To make these considerations more quantitative, we have solved a generalization of a model for atom lasers with interactions [5]. We treat the case where the occupation of the mode is less than or comparable to the saturation value n_s. The population dynamics are given by a master equation of the form

$$\dot{P}_n = \gamma n_c \left[\frac{n}{n + n_s} P_{n-1} - \frac{(n+1)}{(n+1) + n_s} P_n \right] + \gamma[(n+1)P_{n+1} - nP_n], \quad (2)$$

where n_c is a measure of the pumping strength. Above threshold, the steady-state P_n has a Gaussian form with mean $\bar{n} = n_c - n_s$ and variance n_c. Solving this model, we find approximate expressions for the correlation functions

$$g^{(2)}(\tau) = 1 + \frac{n_s}{\bar{n}^2} \exp(-\bar{\gamma}\tau), \quad (3)$$

$$|g^{(1)}(\tau)| \approx \exp(-2\kappa^2 n_c \tau^2) \exp(-\gamma\tau/2\bar{n}) \quad (\bar{\gamma}\tau \leq 1), \quad (4a)$$

$$\approx \exp(-4\kappa^2 n_c \tau/\bar{\gamma}) \exp(-\gamma\tau/4\bar{n}) \quad (\bar{\gamma}\tau \gg 1), \quad (4b)$$

where the decay rate $\bar{\gamma} = \bar{n}\gamma/n_c$ is much slower than that of the bare cavity mode.

It is the slowed decay of $g^{(2)}$, with decay rate $\bar{\gamma} = \bar{n}\gamma/(\bar{n} + n_s)$, which determines that we are in the early time regime $\bar{\gamma}\tau \leq 1$ for $g^{(1)}$ as given by Eq. (4a). In this regime, the first factor in $g^{(1)}$ is identical to the Gaussian expression for an equilibrium condensate of Eq. (1), since the variance $\sigma^2 = n_c$. The second factor, $\exp(-\gamma\tau/2\bar{n})$, corresponding to a Schawlow-Townes phase diffusion, is much slower. From the measured $g^{(2)}(0)$ of 1.1, we obtain $n_s = 25\,000$ which predicts $\bar{\gamma}^{-1}$ to be ~ 100 ps when $\bar{n} \sim 500$, in very good agreement with the experimental decay

of $g^{(2)}$ (100 ps). The deduced value of $\bar{\gamma}$ means that we are in the regime $\bar{\gamma}\tau \leq 1$ long enough to see almost the entire Gaussian decay of $g^{(1)}$ with the decay time of 220 ps, shown above to be in quantitative agreement with experiment. Using experimentally determined input parameters and our modeling, we thus explain quantitatively the decay times of both $g^{(1)}$ and $g^{(2)}$, corresponding to very different physical mechanisms.

In addition to the exchange of particles above between the condensate and the reservoir, there is a direct interaction through the nonlinear Hamiltonian, which means that noise in the reservoir will cause energy fluctuations and decoherence. This effect explains the short coherence times and large linewidths obtained previously using multimode laser excitation. For such a laser, we measure intensity fluctuations of ~15–20% on a nanosecond time scale; comparable variations in the reservoir population will be expected. The interaction with the reservoir population causes a blueshift of the mode energy to ~0.5 meV above the low power peak [Fig. 1]. The fluctuations will thus be 20% of this, ~0.1 meV, which translates into a coherence time ~20 ps, consistent with the experimental results under multimode excitation (~6 ps).

Even with a noise-free excitation laser, there will be thermal fluctuations in the reservoir population, which can cause decoherence. If the mean reservoir population is N_r, the blueshift of the mode due to the interactions is $\sim 4\kappa N_r$. Estimating $N_r \sim 10^4$ using the pump power, we deduce $\kappa \sim 2 \times 10^{-5}$ ps^{-1}. Assuming the reservoir modes are independent, the variance in the population will be $\sim N_r$, so the arguments leading to Eq. (1) give a coherence time of $(8N_r\kappa^2)^{-1/2} \sim 180$ ps. We conclude that decoherence due to the thermal fluctuations is unlikely to obscure the intrinsic effects of interactions between the polaritons in the mode.

In conclusion, we show that study of the polariton condensed state in semiconductor microcavities reveals new physics of interacting BECs. We demonstrate the roles of interactions and fluctuations in determining the phase coherence and the lengthening of the second-order correlation decay by stimulation. A quantum-optical model is presented which explains both these properties using a treatment of a driven interacting condensate. These results show that the polariton condensate exhibits properties characteristic of an equilibrium BEC, even though it is exchanging particles with its environment. The true coherence properties of the polariton condensate reported here were obscured in previous work by fluctuations in excitation intensity of the multimode laser sources employed.

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