Photon Emission as a Source of Coherent Behavior of Polaritons

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We show that the combined effect of photon emission and Coulomb interactions may drive an exciton-polariton system towards a dynamical coherent state, even without phonon thermalization or any other relaxation mechanism. Exact diagonalization results for a finite system (a multilevel quantum dot interacting with the lowest-energy photon mode of a microcavity) are presented in support of this statement.

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The effectiveness of phonon relaxation in systems of excitonic polaritons has been widely discussed recently [1] in connection to the possible Bose-Einstein condensation of polaritons [2]. Because of the very small polariton lifetime, of the order of picoseconds, and the small polariton-phonon scattering cross section [3], there is a common belief that phonons alone cannot account for thermalization of the polariton gas to the lattice temperature (the phonon bottleneck). However, the (time-resolved and angle-resolved) observed emission from decaying polaritons [4] suggests that, for a strong enough pumping pulse and a moderate positive detuning, the occupation probabilities of polariton states can be fitted to a Bose-Einstein distribution with an effective temperature even lower than the lattice temperature. The question is, thus, what is the source of coherence in such a situation?

In the present Letter, we give an answer to the question above showing that phonon relaxation is not the only mechanism forcing the polariton system to reach a coherent state. Indeed, in our computations we show, for a finite system, that the emission of photons is also a source of coherence of polaritons leading, under certain conditions, to a high occupation probability for the ground state.

We consider the model of Ref. [5], in which a multilevel quantum dot interacts with the lowest-energy photon mode of a microcavity. The Hamiltonian describing the system is the following:

$$H = \sum_i \{T_i^{(e)} e_i^\dagger e_i + T_{\bar{i}}^{(h)} h_{\bar{i}}^\dagger h_{\bar{i}}\} + \frac{\beta}{2}\sum_{ijkl}\langle ij||kl\rangle e_i^\dagger e_j^\dagger e_l e_k + \frac{\beta}{2}\sum_{\bar{i}\bar{j}\bar{k}\bar{l}}\langle \bar{i}\,\bar{j}\,||\bar{k}\,\bar{l}\rangle h_{\bar{i}}^\dagger h_{\bar{j}}^\dagger h_{\bar{l}} h_{\bar{k}} - \beta\sum_{i\bar{j}k\bar{l}}\langle i\bar{j}||k\bar{l}\rangle e_i^\dagger h_{\bar{j}}^\dagger h_{\bar{l}} e_k + (E_{\rm gap} + \hbar\omega) a^\dagger a + g\sum_i \{a^\dagger h_{\bar{i}} e_i + a e_i^\dagger h_{\bar{i}}^\dagger\}. \quad (1)$$

We include 12 single-electron and 12 single-hole (two-dimensional, harmonic oscillator) levels in Eq. (1). We assume that both electrons and holes are confined in a small spot of a quantum well, in such a way that the single-particle spectrum is almost flat:

$$T_i^{(e)} = E_{\rm gap}, \qquad T_{\bar{i}}^{(h)} = 0. \quad (2)$$

The Coulomb coupling constant β is modified at will in order to study its effects on the dynamics. $\langle ij||kl\rangle$ are matrix elements of the Coulomb interaction between harmonic oscillator states. The parameter $\hbar\omega$ gives the detuning of the photon energy with respect to the (bare) pair energy, and g is the photon-matter coupling strength. We consider only states with total angular momentum $L = 0$, which are maximally coupled to the photon mode.

The Hamiltonian [Eq. (1)] preserves the polariton number

$$N_{\rm pol} = a^\dagger a + \sum_i (h_{\bar{i}}^\dagger h_{\bar{i}} + e_i^\dagger e_i)/2 \quad (3)$$

and leads to an excitation gap (the gap from the ground state to the first excited state of the system with a fixed $N_{\rm pol}$ number) roughly proportional to $\sqrt{N_{\rm pol}}g$.

Because of the small, but finite, transparency of the microcavity mirrors, photons are continuously emitted. In order to study the evolution of the system due to successive photon emission events, we first consider the transition probabilities:

$$P_{fi} \sim |\langle\psi_f|a|\psi_i\rangle|^2, \quad (4)$$

where ψ_i is a state with polariton number $N_{\rm pol}$, and ψ_f is a state with $N_{\rm pol} - 1$. The probabilities should be normalized:

$$\sum_f P_{fi} = 1. \tag{5}$$

We take an initial distribution of weights $\rho_i^{(10)}$ for the states with $N_{\rm pol} = 10$. After the emission of one photon, the weight of the $N_{\rm pol} = 9$ state ψ_f is

$$\rho_f^{(9)} = \sum_i P_{fi}\rho_i^{(10)}. \tag{6}$$

Proceeding in this way, we compute $\rho_f^{(8)}$, $\rho_f^{(7)}$, etc. This "dynamics" captures the basics of the Liouville equation for the density matrix and, even more, could be appropriate for the description of an experiment in which the number of photons leaving the cavity is continuously measured.

We draw in Fig. 1 the resulting occupation probabilities after the emission of 5 photons. Figure 1(a) shows the initial distribution in the 10-polariton system. Note that the x axis represents the 10-polariton states. $x = 1$ refers to the ground state, $x = 2$ to the first excited state, etc. Figures 1(b)–1(d) show the probabilities of the 5-polariton states for different choices of the parameters β and $\hbar\omega$. The photon-matter coupling strength g was taken equal to 3 meV, corresponding to an excitation gap of around 10 meV in the 10-polariton system. This is a strong coupling regime.

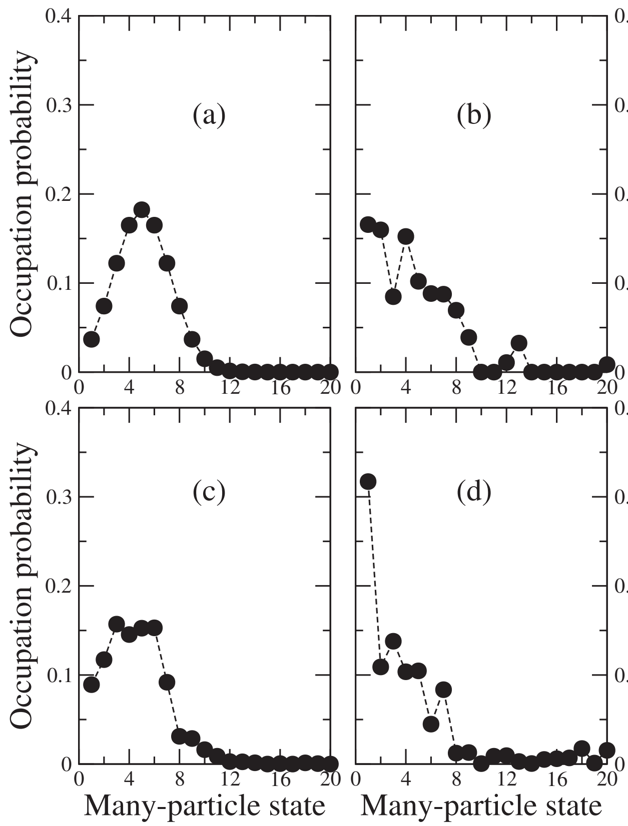

FIG. 1. Occupation probabilities of the first 20 states in the 10-polariton system [initial distribution, (a)] and in the 5-polariton systems after the emission of 5 photons: (b) $\beta = 0$, $\hbar\omega = 5$ meV; (c) $\beta = 2$ meV, $\hbar\omega = -3$ meV; (d) $\beta = 2$ meV, $\hbar\omega = 5$ meV.

Figure 1 shows that a positive detuning leads to an enhancement of the ground-state occupation probability. If Coulomb interactions are turned on, the ground-state occupation reaches 30% or higher. However, for negative detuning, the shape of the initial distribution is preserved. This fact, coming only from dynamical considerations, could be in the basis of the observed polariton coherence in Ref. [4].

These qualitative findings are confirmed by the numerical solution of the Liouville equation for the density matrix [6,7]:

$$\frac{d\rho_{fi}}{dt} = \frac{i}{\hbar}(E_i - E_f)\rho_{fi} + \kappa\sum_{j,k}\langle\psi_f|a|\psi_j\rangle\rho_{jk}\langle\psi_k|a^\dagger|\psi_i\rangle - \frac{\kappa}{2}\sum_{j,k}\langle\psi_f|a^\dagger|\psi_j\rangle\langle\psi_j|a|\psi_k\rangle\rho_{ki} - \frac{\kappa}{2}\sum_{j,k}\rho_{fj}\langle\psi_j|a^\dagger|\psi_k\rangle\langle\psi_k|a|\psi_i\rangle, \tag{7}$$

where $\rho_{fi} = \langle\psi_f|\rho|\psi_i\rangle$. The energy eigenvalues E_i and transition amplitudes $\langle\psi_f|a|\psi_i\rangle$ come from the exact diagonalization of the Hamiltonian [Eq. (1)] for $N_{\rm pol}$ ranging from 1 to 10. The parameter κ accounts for photon losses through the cavity mirrors ($\hbar\kappa \approx E_{\rm gap}/Q$, where Q is the cavity quality factor). In our calculations, we take $\kappa = 0.1\ {\rm ps}^{-1}$. As in the previous example, the initial (mixed) state is a distribution of weights $\rho_{ii}^{(10)}$ for the states of the 10-polariton system. In the simulations, we take 20 states ($i = 1, \ldots, 20$) in each sector with fixed $N_{\rm pol}$.

A sample of the results is shown in Fig. 2, where the weights $\rho_{ii}^{(N{\rm pol})}$ for the state i (x axis) and polariton number $N_{\rm pol}$ (y axis) are represented as circles of areas proportional to $\rho_{ii}^{(N{\rm pol})}$. The distribution of panels in the figure is similar to Fig. 1. That is, in the left upper corner the initial ($t = 0$) distribution is drawn. The mean number of polaritons is 10 at $t = 0$. For the other three panels, whose β and $\hbar\omega$ parameters are the same as in Fig. 1, the running time has been chosen in such a way that the mean number of polaritons is 6.

Besides the same qualitative feature noticed above, i.e., enhancement of ground-state occupations when both positive detuning and Coulomb interactions are combined, Fig. 2 reveals that high ground-state occupation (as compared with the rest of the states in the same $N_{\rm pol}$ sector) is a common characteristic of states with low polariton numbers, i.e., those that have radiated the most.

We may define a total ground-state occupation as

$$\rho_{\rm gs}^{(\rm total)} = \sum_{N{\rm pol}=1}^{10} \rho_{11}^{(N{\rm pol})}. \tag{8}$$

From our numerical solution, it follows that $\rho_{\rm gs}^{(\rm total)} \approx 0.25$,

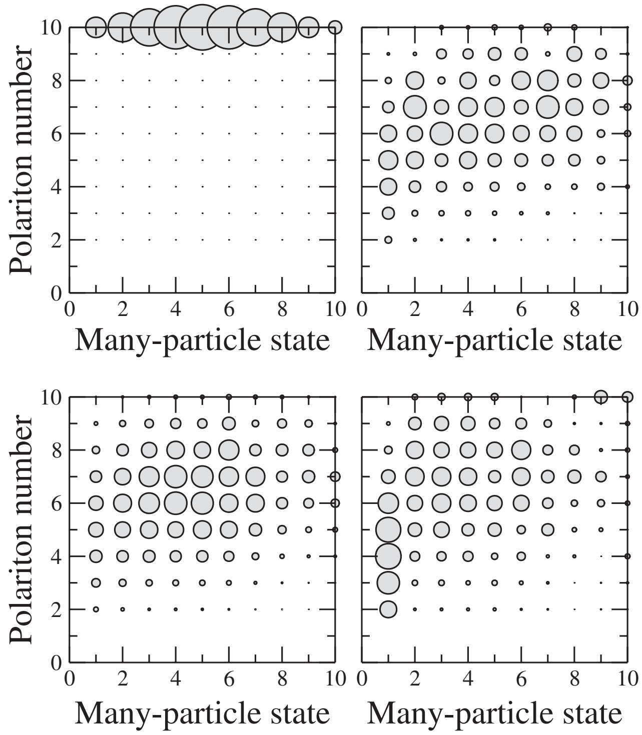

FIG. 2. The diagonal elements $\rho_{ii}^{(N\text{pol})}$ (occupations) as functions of i (x axis) and N_{pol} (y axis). The areas of the circles are proportional to $\rho_{ii}^{(N\text{pol})}$. The left upper panel corresponds to the initial distribution, whereas the other three correspond to the same sets of parameters as in Fig. 1. The running time is such that the mean polariton number is around 6 in each case.

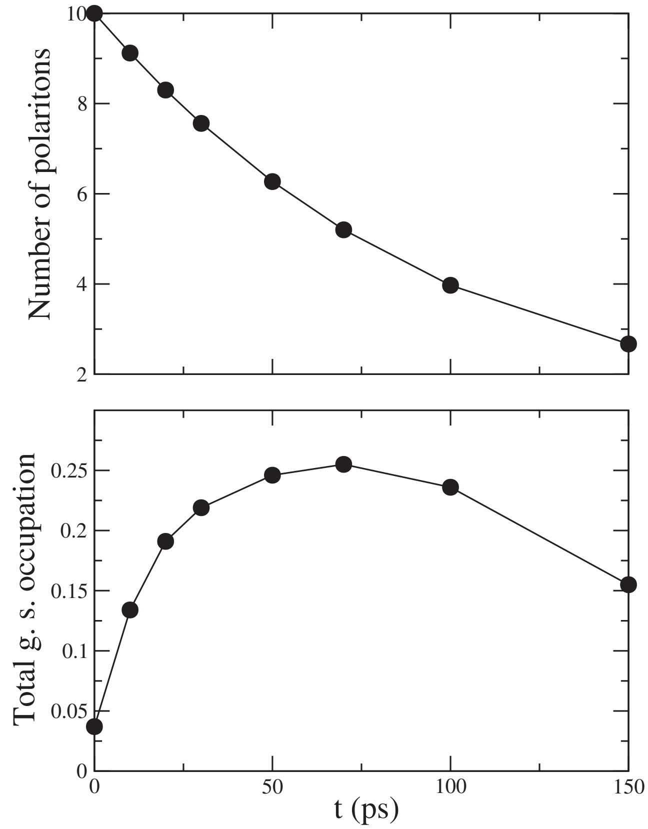

FIG. 3. The mean number of polaritons (upper panel) and the total ground-state occupation, defined in Eq. (8), as functions of time. The system constants are $\hbar\omega = 5$ meV, $\beta = 2$ meV.

0.11, and 0.07 for the sets of parameters used in Figs. 1 and 2, i.e., $\beta = 2$ meV, $\hbar\omega = 5$ meV; $\beta = 0$, $\hbar\omega = 5$ meV; and $\beta = 2$ meV, $\hbar\omega = -3$ meV, respectively.

It is interesting to draw the time evolution of $\rho_{\text{gs}}^{(\text{total})}$. This result is shown in Fig. 3 along with the time dependence of the mean number of polaritons. The system constants are $\beta = 2$ meV, $\hbar\omega = 5$ meV. At $t = 0$, we have $\langle N_{\text{pol}}\rangle = 10$, $\rho_{\text{gs}}^{(\text{total})} = 0.036$. In around 20 ps, i.e., when $\langle N_{\text{pol}}\rangle \approx 8$ or two photons have been emitted, the total ground-state occupation rises to nearly 0.2. From $t = 20$ to 70 ps ($\langle N_{\text{pol}}\rangle$ from 8 to 5), the total occupation still increases, reaching a maximum value of 0.25. Afterwards, the total occupation decreases. This later decrease is also a dynamical result, not connected with the increasing occupation of the vacuum state. Indeed, for $t = 150$ ps, the weight of the vacuum state is only 0.16.

In conclusion, we have extracted energy eigenvalues and transition amplitudes from exact diagonalization calculations in order to solve the Liouville equation for the density matrix of a decaying polariton system. The results show that the emission of photons is an additional source of coherence.

Further work is needed in order to conceptually clarify the role of detuning and Coulomb interactions in the dynamical coherence. We can, nevertheless, add some qualitative arguments for the system with a large number of particles based on the simplified picture of weakly interacting composite quasiparticles (polaritons), adequate for the present strong coupling regime. In Ref. [4], it was argued that a small positive detuning increases the excitonic component of the polariton wave function, leading, in this way, to higher polariton-phonon and polariton-polariton scattering rates. These mechanisms act at the "horizontal level" in Fig. 2. We, on the other hand, stress the role of polariton decay, i.e., the "vertical" transitions. The decay of noninteracting polaritons could not, of course, be the source of coherence because the ground-state population is not increased in this way. But events in which two polaritons, one with momentum k and the second with $-k$, transform into a zero-momentum polariton and a zero-momentum photon could. These events could be stimulated by the occupation of the ground state. The photon should carry the excess energy of the pair, a fact that is apparent in the observed blueshift of the $k = 0$ emission at the initial stages of the decay process [4]. The role of Coulomb interactions is evident if we imagine the event in two steps: first (virtual) polariton-polariton scattering leading to two $k = 0$ states and then decay of the off-mass-shell polariton into a photon. This picture also helps to understand the dependence on detuning. A positive detuning increases the scattering rate, as argued above.

The results should be extended to larger systems in order for their relevance to the experiment reported in Ref. [4] to be more evident. Perhaps this could be possible within the stochastic dynamics framework [8]. An interesting question is whether the total ground-state occupation may reach still higher values with an increasing number of particles. On the other hand, preliminary calculations based on a mean-field (BCS) dynamics [9], similar to the semiclassical spin dynamics of the Dicke model [10] but including Coulomb interactions and photon losses, show that the emission of photons through leaky modes of the cavity, in spite of its incoherent nature, helps establish a dynamical coherent state. Other mechanisms, such as phonon relaxation, acting at the horizontal level in Fig. 2, could drive the system toward a quasiequilibrium state.

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