Quantum Condensation from a Tailored Exciton Population in a Microcavity

P. R. Eastham and R. T. Phillips

1Department of Physics, Imperial College London, London SW7 2BW, U.K.
2University of Cambridge, Cavendish Laboratory, Cambridge CB3 0HE, UK
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An experiment is proposed, on the coherent quantum dynamics of a semiconductor microcavity containing quantum dots. Modeling the experiment using a generalized Dicke model, we show that a tailored excitation pulse can create an energy-dependent population of excitons, which subsequently evolves to a quantum condensate of excitons and photons. The population is created by a generalization of adiabatic rapid passage, and then condenses due to a dynamical analog of the BCS instability.

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There is great interest in the possibility of quantum-condensed phases of solid-state quasiparticles, such as excitons, polaritons, and magnons. Such phases are characterized by the presence of a quantum state whose population scales with the size of the system, and hence is much larger than one — macroscopic occupation. This is seen in recent experiments on Bose-Einstein condensation (BEC) of polaritons and polariton lasing. In these experiments, a semiconductor microcavity is excited at high energies, and a macroscopic population of low-energy polaritons emerges following relaxation and inelastic scattering. These condensates appear spontaneously, from states without macroscopic occupations. This differentiates them from the microcavity parametric oscillator experiments, where resonant pumping of polaritons leads directly to a macroscopic occupation.

The aim of this paper is to show how microcavities could be used to access condensation, even in the absence of relaxation or inelastic scattering. We propose an experiment on a microcavity containing an ensemble of quantum dots, where the exciton decay times are many tens or hundreds of picoseconds. We demonstrate that this experiment could be faster than these decay times, so that energy relaxation and inelastic scattering would be negligible. Nonetheless, we shall show that a condensate develops. In contrast with a laser, this condensate is formed from part-matter, part-light quasiparticles. In contrast with the microcavity parametric oscillator, it develops from a state with no macroscopic occupations, in the absence of the pump laser. And whereas relaxation is essential to obtain an equilibrium BEC or polariton lasing, in our approach condensation occurs due to an instability of the coherent quantum dynamics. Our proposal implements, in a solid-state system, the type of dynamical condensation predicted in quenched Fermi gases.

The first stage in our proposed experiment is the creation of a population of excitons in the quantum dots. We propose using a chirped laser pulse, which sweeps up through part of the inhomogeneously-broadened exciton line. As shown by the demonstration of Rabi oscillations and density-matrix tomography, excitons in quantum dots are discrete two-level systems, which can therefore be manipulated using laser pulses. The proposed pulse implements adiabatic rapid passage, which is a well-established technique for populating discrete states. It extends the technique, by controlling the pump spectrum to create an energy-dependent exciton population.

The second stage occurs after the pump pulse has passed. It is the coherent quantum dynamics of the system, starting from the exciton population created by the pump. The pump is chosen such that this population is similar to a Fermi distribution, with a sharp upper step. The system is described by a model similar to that which describes pair condensation in atomic gases and superconductors. We therefore expect that a population with the form of a Fermi distribution could condense, due to a dynamical version of the BCS instability.

We now turn to the theoretical demonstration of this proposal. For simplicity, we suppose that the pump is circularly polarized, so we may consider only one of the polarization states of the excitons. We model the quantum dots as a set of two-level systems, each describing the presence or absence of an exciton of the pump polarization in a given localized dot state. These localized excitons are coupled to the electromagnetic field by the dipole interaction. Since the exciton states in the dots are spatially separated, we neglect the non-radiative interactions between the different two-level systems. The appropriate Hamiltonian is then the generalized Dicke model.

We label the dot states with an index $i$, so that $E_i$ is the energy of an exciton in the $i$th dot state; this state is localized at $r_i$, with dipole-coupling strength $g_i$. The area density of dots is $n$, so that if $g_i = g$ and $E_i = E$ the vacuum Rabi splitting at resonance is $2g\sqrt{n}$. The state of a dot is specified by the Bloch vector $\langle \sigma \rangle$, where $\sigma_i^z = \sigma^z - i\sigma^y$ is the exciton annihilation operator, and $\sigma^z$ the inversion. The inversion is related to the occupation of the dot $n_i$ by $n_i = (\sigma^z + 1)/2$, and is $-1(+1)$ for an unoccupied (occupied) dot.
Dicke models have previously been used to describe polariton condensation in equilibrium [2, 16, 17], and in a dissipative open system [18]. Here we are concerned with the opposite limit, of timescales short compared with the relaxation times. The dynamics therefore obeys the Heisenberg equation. Since we are concerned with condensation phenomena, involving large photon numbers, we treat the field classically. However, we retain the full quantum dynamics of the dots, and hence the possibility of an incoherent population of excitons. In this approximation, the Heisenberg equation gives

\[ i\dot{\psi}_k = \omega_k \psi_k + \frac{1}{2} \int g P_k(E, g) dEdg + F_{k,t}, \]  

(1a)

\[ i\dot{P}_{k,t}(E, g) = EP_{k,t}(E, g) - 2g \sum_{k'} D_{k-k', t}(E, g) \psi_{k', t}, \]  

(1b)

\[ i\dot{D}_{k,t}(E, g) = g \sum_{k'} (\psi_{k', t}^* P_{k+k', t}(E, g) + P_{k'-k, t}(E, g) \psi_{k', t} - D_{k-k', t}(E, g) \psi_{k', t}). \]  

(1c)

\[ \psi_k \] is the normal-mode amplitude for an electromagnetic field mode, with in-plane wavevector \( k \) and energy \( \omega_k \), and \( F_{k,t}(t) \) is the (classical) pump. \( P_{k,t}(E, g) \) and \( D_{k,t}(E, g) \) are collective variables describing the polarizations and inversions of the dots. They are defined as

\[ P_k(E, g) = \frac{1}{4\pi} \sum_i (\sigma_i^z) e^{-ikr_i}, \]  

(2)

\[ D_k(E, g) = \frac{1}{4\pi} \sum_i (\sigma_i^z) e^{-ikr_i}, \]  

(3)

where the sums run over the states with \( E \to E + \delta E \) and \( g \to g + \delta g \).

Eqs. (1a–1c) generalize the Maxwell-Bloch equations [19], to allow for the distribution of energies and dipole-coupling strengths in the dots. Thus the inversion and polarization become distribution functions: \( D_0(E, g) \delta \delta g \) is the inversion due to states with energies \( E \to E + \delta E \) and couplings \( g \to g + \delta g \).

The fields \( P_{k,t}, \psi_{k,t}, \) and \( F_{k,t} \) have been normalized such that their square magnitudes are particle numbers per exciton state. A condensate is characterized by a macroscopic occupation number, \( \langle \sigma_i^z \rangle \sim -1 \), and there are many exciton states distributed over the active area of the sample. Thus the sum in (3) is strongly peaked near \( k = 0 \), and we approximate the initial conditions as \( D_{k,t}(E, g) = \delta_k D_{0,t}(E, g) \). The dynamics is then that of a continuous medium due to motional narrowing, with the short-range spatial structure of the exciton states averaged out on the long scales of the photons.

We consider a plane-wave pump, at a high angle where the excitons lie outside the stop-bands of the mirrors. The field acting on the dots at this wavevector, \( k_p \), may then be taken as the driving field. Anticipating our analysis of the condensation, we retain only one other mode of the field, specifically the confined cavity mode with \( k = 0 \). This reduces (1) to

\[ i\dot{\psi}_0 = \omega_0 \psi_0 + \frac{1}{2} \int g P_0(E, g) dEdg, \]  

(4a)

\[ i\dot{P}_0(E, g) = EP_0(E, g) - 2g D_0(E, g) \psi_0, \]  

(4b)

\[ i\dot{P}_p(E, g) = EP_p(E, g) - 2g D_0(E, g) F_p, \]  

(4c)

\[ i\dot{D}_0(E, g) = g (F_p^* P_p(E, g) + P_p^* (E, g) F_p) + \psi_0^* P_0(E, g) + P_0^* (E, g) \psi_0, \]  

(4d)

corresponding to an ensemble of two-level systems, interacting with two modes of the field. \( P_p \) is the polarization at the pump wavevector, and \( F_p \) the driving field.

We have simulated our proposed experiment by solving (4) numerically, with \( N = 4500 \) two-level systems. Results are shown in Figs. 1–3. These results focus on a model with a single coupling strength \( g \), and a Gaussian distribution of exciton energies with variance \( \sigma^2 \). The pump is a linearly-chirped Gaussian,

\[ 2g F_p(t) = \frac{S}{\sqrt{2\pi\tau^2}} e^{-i(\nu_0 + \alpha t/2)t^2} e^{-t/(2\tau^2)}, \]  

(5)
normal modes of the pumped population. Linearizing (1) gives
demonstrates condensation of excitons and photons. Finite, reflecting the fact that the pump laser does in-
finite, reflecting the fact that the pump laser does in-
pulse, to the dynamics. In the first stage, during the pump
first to Fig. 1, we see that there can indeed be two stages
states, this does not qualitatively change our results.
containing interfacial quantum dots. Though there is a
of the pumping stage. We therefore argue that the pump
produce an incoherent population of excitons. Further-
tive temperature 
more, as shown in Fig. 2, this population has a sharp
ties (shading), and energy of the 

two, like that of the Fermi function at a low effective
upper step, like that of the Fermi function at a low effec-
tory, (7a) has a solution below the step in the exciton

due to the step in the Fermi distribution; in the same

tion created by the pump. In a superconductor, there is a

tion of such pulses (inset). The dotted curve is an equilibrium

ty mode. Fig. 1. In fact, (6) predicts instabilities for

 instability gives the exponential growth of the

fastest-growing instability is at k ≈ 0. This mode grows, it suppresses the gain for the others. It will thus be
dynamically selected, and we therefore neglected cavity
modes with k ≠ 0 in the simulations.

For the parameters used here, the instability predicted by (7a) corresponds to the BCS instability in a superconductor. This can be seen by considering (6) for a single coupling strength, close to an instability. The eigenenergies λ′ k and growth rates λ′′ k then obey

$$\frac{\omega_k' - \lambda_k'}{g^2} = -\mathcal{P} \int \frac{D_0(E, g) dE}{E - \lambda_k},$$  \hspace{1cm} (7a)

$$\lambda_k'' = \pi g^2 \text{sgn}(\lambda_k') D_0(\lambda_k') - \gamma.$$  \hspace{1cm} (7b)

(7b) describes the growth or decay of the normal mode, with the first term the gain/loss from the excitons, and the second the loss due to the cavity decay γ = −3(ωk).

(7a) is the Cooper equation of the BCS model [22]. The term corresponding to the usual pairing interaction is g^2/(λ_k - ω_k), which we recognize as the effective interaction between excitons, mediated by the cavity modes. Here it is an attractive interaction, as required for BCS, since the excitons lie below the photons. The term corresponding to the Fermi distribution is the exciton population created by the pump. In a superconductor, there is a solution to the Cooper equation below the Fermi energy, due to the step in the Fermi distribution; in the same way, (7a) has a solution below the step in the exciton occupation. (7b) shows that this mode experiences gain, and hence can become unstable.

FIG. 1: Electromagnetic fields and polarizations as functions of time, for the simulation described in the text. Dotted: pump field |P_p| (left axis). Dot-dashed: polarization |P_p| at pump wavevector, integrated over dot energies and couplings (left axis). Solid: cavity field |ψ_0| at k = 0 (right axis). Dashed: polarization |P_p| at k = 0 (right axis). Inset: spectrum |ψ_0(ω)|^2 during the shaded region of the main plot (solid curve). The pumped population (dashed curve), exciton energy distribution (shading), and energy of the k = 0 cavity mode (arrow) are shown for comparison.

FIG. 2: (Color online) Simulated exciton inversion profile immediately after pumping, showing the population created by the Gaussian pump pulse (solid line), and by a superposition of such pulses (inset). The dotted curve is an equilibrium exciton distribution with fitted temperature $\hbar/(4.2k\tau)$; this is 0.6 K for $\tau = 3$ ps. Arrows mark the energy of the k = 0 cavity mode.
To confirm the origin of the condensation, we show in the inset to Fig. 1 the spectrum of the $k = 0$ field, and in Fig. 3 the evolution of the population during condensation. As expected, the condensate is at a frequency below the step in the exciton population (Fig. 1); this leads to hole-burning there (Fig. 3). Reducing the center frequency of the chirp, $\nu_0$, we find that the condensation stage in Fig. 1 disappears, as the effective pairing interaction decreases below its critical value. The long-time limit of the simulations is then just the population of excitons. We have also confirmed that the phase of the pump $\tilde{g}$ is irrelevant, by plotting the phase associated with the power spectrum in the inset to Fig. 1. This plot is identical in simulations with different pump phases, but the same random noise.

Since our condensate occurs on timescales short compared with the relaxation times, it will not be in equilibrium. This leads to phenomena absent for an equilibrium condensate. In Fig. 1 for example, we see ringing oscillations (corresponding to those predicted for atomic gases [9, 10, 11, 12]; unrelated oscillations have been predicted for non-interacting two-level systems, driven by a chirped laser pulse [15]). Such a population can condense, even in the absence of relaxation or inelastic scattering. Since the dynamics is slow enough to create a controlled inversion profile within the inhomogeneous line, such that the couplings can be increased so that the instability occurs after the pump pulse, but before the excitons decay. Other systems described by generalized Dicke models, such as Fermi gases, SK dots in microcavities, or Josephson junction arrays, could be considered.

To conclude, we have proposed and analyzed a new approach to quantum condensation in a solid-state system. The key is the chirped pumping [4], which we have shown can create an energy-dependent population in the exciton line. Such a population can condense, even in the absence of relaxation or inelastic scattering. Since our approach uses the spectrum of the pump to tailor the population, it would be possible to pump other initial states (inset to Fig. 3). Thus our technique could be used more generally, to explore the quantum dynamics of many-particle systems from controlled initial conditions.

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