Finite momentum condensation in a pumped microcavity

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We calculate the absorption spectra of a semiconductor microcavity into which a non-equilibrium exciton population has been pumped. We predict strong peaks in the spectrum corresponding to collective modes analogous to the Cooper modes in superconductors and fermionic atomic gases. These modes can become unstable, leading to the formation of off-equilibrium quantum condensates.

We calculate a phase diagram for condensation, and show that the dominant instabilities can be at a finite momentum. Thus we predict the formation of inhomogeneous condensates, similar to Fulde-Ferrel-Larkin-Ovchinnikov states.

I. INTRODUCTION

The appearance of order at an equilibrium phase transition is a central concept in many areas of physics, from condensed matter to the physics of the early universe. Recently there has been considerable interest in the more general problem of ordering far from thermal equilibrium, motivated by the possibility of quantum quench experiments in cold atomic gases.\textsuperscript{3,4} In a quench the parameters of the system are rapidly switched from a disordered to an ordered phase, and the disordered state forms the initial conditions for a dynamics with the new parameters. An interesting regime is that of coherent relaxationless dynamics, which can lead to the formation of non-equilibrium order including crystallization,\textsuperscript{3} condensation, and ferromagnetism.\textsuperscript{2}

Among condensed matter systems, semiconductor microcavities are promising candidates for studying such quench dynamics. The non-equilibrium dynamics of microcavities has attracted considerable interest both experimentally\textsuperscript{5–9} and theoretically\textsuperscript{10–14} with recent experiments demonstrating regimes where the low-energy quasiparticles, polaritons, form a condensate. More recently, an experiment has been proposed to implement a quantum quench\textsuperscript{15} by rapidly preparing a microcavity in a non-condensed initial state. The coherent dynamics of this non-condensed state is predicted to lead to a form of non-equilibrium order including crystallization,\textsuperscript{2} condensation, and ferromagnetism.\textsuperscript{3}

In this paper, we first calculate the optical spectra of a semiconductor microcavity into which a non-equilibrium exciton population has been pumped. We predict strong peaks in the spectrum corresponding to collective modes analogous to the Cooper modes in superconductors and fermionic atomic gases. These modes can become unstable, leading to the formation of off-equilibrium quantum condensates.

II. MODEL

We consider an experiment, proposed in Ref. 15, on a set of localized exciton states in a planar semiconductor microcavity. Such excitonic states could be realized in practice using either highly disordered quantum wells (where excitons are localized by disorder) or quantum dots. The proposed experiment involves two stages which are separated in time and can be regarded as independent. In the first stage, the localized states are driven by a chirped laser pulse. This pulse creates an energy-dependent population in the inhomogeneously broadened exciton line by adiabatic rapid passage (ARP). For certain populations a second stage may then occur, where the population evolves into a non-equilibrium condensate due to the photon-mediated interactions between the excitons.

In this paper, we first calculate the optical spectra of a microcavity a short time after it has been prepared in a non-condensed state, i.e., immediately after the “quench”. We find that the collective mode responsible for condensation is directly observable in these spectra. We use this analysis to calculate a phase diagram for the non-equilibrium condensation, and show that the condensation generally occurs at a finite momentum. While we focus on a microcavity containing quantum dots, our analysis is based on the Maxwell-Bloch equations. These describe a wide variety of coupled light-matter systems, implying a broad relevance of our work.

The remainder of this paper is structured as follows. In Sec. I we briefly review the proposed quench experiment, and outline our model. In Sec. II we present absorption spectra of the system. In Sec. III we discuss the phase diagram and the possibility of finite momentum condensation, and in Sec. IV we discuss the connections to FFLO and the role of nonlinear terms. Section V summarizes our conclusions. Finally, the appendix contains a brief treatment of the preparation of non-condensed initial states by optical pumping.
We consider time scales short compared with the exciton lifetime, which is at least 100 ps\cite{21} and treat the electromagnetic field using a mean-field approximation. In this approximation the photon creation and annihilation operators are replaced with their expectation values, and hence become c-numbers. The resulting equations of motion are linear in the remaining operators, so that we may take their expectation values without further approximation. The resulting dynamics obeys the generalized Maxwell-Bloch equations:

\begin{align}
\dot{\psi}_k &= \omega_k \psi_k + g \int P_k \, dE + f_k + F\delta_{k-p}, \\
\dot{P}_k &= E P_k - g \sum_{k'} D_{k-k'} \psi_{k'}, \\
\dot{D}_k &= 2g \sum_{k'} (P^*_{k'-k} \psi_{k'} - P^*_{k'+k} \psi^*_{k'}). 
\end{align}

Here \( \psi_k \) is the complex amplitude of the microcavity mode with in-plane wavevector \( k \) and energy \( \omega_k \) (\( h = 1 \)). It is related to the expectation value of the photon annihilation operator by \( \psi_k = (\langle \hat{\psi}_k \rangle) / \sqrt{N} \), where \( N \) is the total number of localized states. This normalization is convenient when dealing with condensation, since macroscopic occupation corresponds to a finite \( \psi_k \) in the thermodynamic limit \( N \to \infty \). We allow for the finite lifetime of the photon modes by taking \( \Im \omega_k = -\gamma \). \( f_k \) is introduced to allow us to calculate the linear response. \( F \) is an externally applied pump field, with wavevector \( p \), that is used to create the non-equilibrium population.

The coupling \( g = g_i \sqrt{n} \) in Eqs. (1) is related to the dipole coupling strengths of the localized states, \( g_i \), and their area density \( n \). To simplify the notation we have taken \( g_i \) to be the same for all states; the extension to a non-equiprobable distribution is straightforward. In the dipole gauge

\[ g_i = d \sqrt{\frac{E_i}{2\epsilon_0 c w}}, \]

where \( d \) is the matrix element of the dipole operator \( e^\hat{r} \) between the zero-exciton and one-exciton states, and \( E_i \) their energy difference. \( w \) is the effective width of the cavity, which arises from the normalization of the cavity mode functions.

\( P_k(E) \) is the collective polarization of the ensemble at wavevector \( k \) due to states with energy in a small interval near \( E \),

\[ P_k(E) \delta E = \frac{1}{N} \sum_i \langle \hat{\sigma}^- \rangle e^{-ik \cdot r}, \]

where the prime indicates that the sum runs over states with exciton energies between \( E \) and \( \delta E \). \( D_k(E) \) is the collective inversion, defined in a similar way with \( \hat{\sigma}^+ \) replacing \( \hat{\sigma}^- \).

In the following we shall be concerned with large \( N \), and wavevectors which are small compared with the inverse spacing of the localized states. In these limits we may approximate sums over dot positions, such as those in Eq. (5), by

\[ \frac{1}{N} \sum_i e^{-ik \cdot r_i} \approx \nu(E) \delta_{k,0} \delta E + O \left( \frac{1}{\sqrt{N}} \right). \]

Here \( \nu(E) \) is the distribution of localized states in energy, normalized to one. Thus \( N\nu(E) \delta E \) is the number of terms in the primed sum, Eq. (6). For \( k \neq 0 \) the phasor sum is a 2D random walk, producing the \( O(1/\sqrt{N}) \) corrections\cite{22}.

The approximation of Eq. (6) corresponds to replacing the response of the disordered dielectric with its homogeneous average response, so that the wavevector is well-defined. This is similar to the linear dispersion model which has been extensively used for inorganic microcavities\cite{23}. The \( O(1/\sqrt{N}) \) corrections describe Rayleigh scattering from density fluctuations in the dielectric. They are generally small corrections because the scatterers are dense, so that on long wavelengths the medium appears homogeneous. The corrections can become important at very small or large wavevectors\cite{24,25} for modes whose group velocity becomes very small. In this case a long lifetime is required for the wavevector to be well-defined, so that even weak scattering or absorption destroys the quasi-propagating modes. Here, however, we are concerned with modes that have a significant dispersion due to their photon component. Furthermore, the lifetime of these modes is massively enhanced by resonant gain from the populated excitons. Thus the leading approximation of Eq. (6) will capture the physics at the experimentally relevant wavevectors.

### III. ABSORPTION SPECTRA

In the experiment \( F \) is a chirped pulse, which creates a non-equilibrium population of excitons using ARP. In the appendix we demonstrate this explicitly, using a model pulse, Eq. (A5), for which analytical solutions to the dynamics exist. Following the pulse the exciton states are populated with a distribution given by Eq. (A6) and the fields and polarizations are negligible \( \psi_k \approx 0 \), \( P_k \approx 0 \).

To establish the optical properties of the microcavity immediately after the pump pulse we find the response to a weak probe \( f_k \). The susceptibility can then be found from the induced electromagnetic field \( \delta \psi_k = \sum_{k'} \int \chi_{k,k'}(t-t') f_k(t') \, dt' \). If the system is stable then \( \psi_k \) and \( P_k \) are small (of order \( f_k \)) for all times, whereas if it is unstable they are only small soon after the pump pulse. In both regimes we may neglect terms above first order in \( \psi_k \) and \( P_k \). Eq. (3) then gives \( D_k = 0 \), so the non-equilibrium population is constant. Fourier transforming the linearized Eqs. (1) and (2) gives

\[ \omega \delta \psi_k = \omega_k \delta \psi_k - g^2 \sum_{k'} \int \frac{D_{k-k'} \delta \psi_{k'}}{\omega - E} \, dE + f_k. \]


The pumping populates the states independently of their position, within the pump spot. Thus the sum in $D_{k-k'}$ is strongly peaked near the forward scattering direction $k = k'$, as discussed above [Eq. (6)]. Neglecting the smaller off-diagonal scattering terms, we obtain a diagonal response function

$$\chi_k(\omega) = \frac{1}{\omega - \omega_k + g^2 \int \frac{D_0(E)}{\omega - E} dE}.$$  \hspace{1cm} (8)

The absorption coefficient of the microcavity follows from the susceptibility $\chi_k(\omega)$

$$A(\omega) = -2 \lim_{\epsilon \to 0} \text{Im} \chi(\omega + i\epsilon),$$  \hspace{1cm} (9)

where the infinitesimal $\epsilon$ appears due to causality, and can be physically understood as a small damping constant for the excitons. The sign is such that $A(\omega) > 0$ corresponds to absorption of energy by the system. Thus from Eq. (8) we obtain

$$A(\omega) = \frac{\gamma - g^2 \pi D_0(\omega)}{(\omega - \omega_c + g^2 \gamma P \int \frac{D_0(E)}{\omega - E} dE)^2 + (\gamma - g^2 \pi D_0(\omega))^2}.$$  \hspace{1cm} (10)

When the dots are unoccupied $D_0(\omega) < 0$ and the empty exciton states contribute to absorption. For energies where there are occupied exciton states $D_0(\omega) > 0$, describing gain due to the population.

In general the response, Eq. (10), peaks near the zeroes of $G(\omega)$, which are at the energies of the normal modes. These energies differ from the energy of the cavity resonance due to the coupling to the exciton states. For an unpopulated state the condition $G(\omega) = 0$ recovers the usual polariton energies of the Lorentz oscillator model but in general the spectrum differs due to the presence of the non-equilibrium population. The modes have a lifetime determined by the second factor in the denominator, with contributions from the cavity losses and the resonant mixing with the band of exciton states. As expected it is damping which controls the overall strength of the absorption, so that the damping factor $H(\omega)$ also appears in the numerator.

The only dependence of the spectra, Eqs. (8) and (10), on the wavevector is in the energy of the cavity mode, $\omega_c = \Re(\omega_k) \approx \omega_0 + |k|^2/(2m)$. We therefore show results as functions of $\omega_c$, which corresponds experimentally to both the incident probe angle and the cavity width.

Figure 1 illustrates absorption spectra obtained from Eq. (10) for both a pumped and unpumped exciton line. These spectra are valid at all times if condensation does not occur (see later), but only soon after the pump pulse if it does. We have taken a Gaussian model for the inhomogeneously-broadened exciton line, with standard deviation $\sigma$, and measure energies relative to the center of the line. We choose the duration $\tau$ of the pump pulse as our unit of energy, and have taken

$$g = 13/\tau, \gamma = 1.5/\tau, \sigma = 15/\tau.$$  \hspace{1cm}

These parameters, with $\tau = 3$ ps, are reasonable for a microcavity containing strongly disordered quantum wells. As discussed in the appendix the pump creates a population equivalent to a Fermi function with temperature $1/(\pi k_B \tau)$, and Fermi energy $\mu$ dictated by the chirp and center frequency of the pumping pulse. We have taken a Gaussian pulse with standard deviation $\sigma$. These spectra are valid at all times if condensation does not occur (see later), but only soon after the pump pulse if it does. We have taken a Gaussian model for the inhomogeneously-broadened exciton line, with standard deviation $\sigma$, and measure energies relative to the center of the line. We choose the duration $\tau$ of the pump pulse as our unit of energy, and have taken $g = 13/\tau, \gamma = 1.5/\tau, \sigma = 15/\tau$. These parameters, with $\tau = 3$ ps, are reasonable for a microcavity containing strongly disordered quantum wells. As discussed in the appendix the pump creates a population equivalent to a Fermi function with temperature $1/(\pi k_B \tau)$, and Fermi energy $\mu$ dictated by the chirp and center frequency of the pump pulse.
the pulse; we choose a pulse for which $\mu = -12.5/\tau$.

The lower panel of Fig. 1 shows the expected result for an unpopulated microwavcavity. There is a pronounced peak in the absorption at the cavity mode energy, which broadens as the cavity mode is tuned through the excitons. There is some suggestion of an anticrossing near resonance, i.e., a polariton splitting, but since the inhomogeneous broadening is relatively large compared with the coupling this is a weak effect. The top panel shows that the population dramatically changes the absorption spectrum. For these parameters it leads to a range of probe frequencies for which $\gamma < g^2 \pi D_0(\omega)$, and the absorption coefficient, Eq. (10), becomes negative. This occurs when the gain from the populated exciton states exceeds the losses, so that there is a net gain for the probe beam. Moreover, we see a pronounced additional peak in the absorption spectrum, which first appears near the upper edge of the population as the cavity mode energy is decreased. As the cavity energy is decreased still further this peak moves down through the region of gain, before the spectrum finally reverts to one dominated by the unperturbed cavity mode.

This additional peak in the absorption spectrum is analogous to the Cooper pairing mode in a superconductor or Fermi gas, that gives rise to the Cooper instability. The analogy can be seen by noting that the normal-mode condition $G(\omega) = 0$ contained in Eq. (10) is the Cooper equation, as discussed for this system in Ref. 13. The non-equilibrium exciton population corresponds to the Fermi distribution, while the photon-mediated interaction between excitons corresponds to the pairing interaction between the electrons. As in a superconductor the sharp step in the population leads to collective modes generated by the pairing interaction. Fig. 1 shows that, for reasonable parameters, these collective modes give rise to strong features in the spectra.

It is interesting to compare the spectra of Fig. 1 with the predictions for an equilibrium condensate in the same model. In that case the condensation opens a gap in the single particle spectrum, which is the analog of the Cooper gap of the superconductor. Inside this gap is a collective mode, which is the analog of the Cooper mode or phase mode of the superconductor. The features visible in Fig. 1 arise from the non-equilibrium generalization of the collective mode (which is a different spectral feature than the gap). It is clear from Fig. 1 that it is the collective mode which dominates the spectrum. Thus, although the single particle features may be affected by condensation, this would have little effect in practice. It may be possible to isolate the single particle spectrum in a Rayleigh scattering experiment, as has been proposed for equilibrium condensates.

IV. PHASE DIAGRAM

The normal modes of the system, with frequencies determined by $G(\omega) = 0$, have decay rates $\tilde{H}(\omega)$. If a normal mode frequency lies in the $H(\omega) < 0$ region produced by the non-equilibrium population it will be unstable, growing exponentially to give a state with a highly populated mode, i.e., a condensate. The condition for the onset of such an instability gives a non-equilibrium phase diagram, which is shown for our chosen parameters in Figs. 2 and 3.

Fig. 2 shows the phase diagram assuming that only a single cavity mode, of energy $\omega_c$, is relevant. The dotted line shows the phase boundary for equilibrium condensation in the same model with temperature and chemical potential corresponding to the pumped population. We see that one sheet of the non-equilibrium phase boundary extends the equilibrium result to allow for the cavity damping. Whereas in equilibrium the presence of the collective mode is sufficient to create an instability, in the open system condensation only occurs if the gain at the energy of the collective mode overcomes the cavity loss. Thus the collective mode can exist even in the normal state (see Fig. 1), and the damping pushes the transition to larger couplings. In addition, we see that there is a lower limit on $\omega_c$ in Fig. 2. This lower threshold is a purely dynamical effect, not present in the equilibrium case. Below it there is a bosonic collective mode at an energy well below that of the populated states (see Fig. 1). Although this mode would be occupied in equilibrium it is far out of resonance with the excitons. As a result, it is not occupied dynamically, and the uncondensed state is metastable. A similar metastable region has been predicted in quenched atomic gases.

Figs. 1 and 2 show that, for a given $g$ and $\gamma$, the
condensation instability occurs over a range of $\omega_c$. In a microcavity different values of $\omega_c = \omega_0 + |k|^2/2m$ correspond to either changing the cavity width, which varies the detuning $\omega_0$, or considering modes at a different wavevector $k$. As such, a range of unstable $\omega_c$ implies that for a fixed cavity detuning there can be instabilities at many wavevectors, with different growth exponents $|H(\omega)|$. At short times after the population has been created the mode with the highest growth exponent will dominate. Figure 4 shows that as $\omega_0$ is lowered this dominant mode occurs at $k \neq 0$, implying a condensate with finite momentum, and a spatially inhomogeneous order parameter. Thus the full phase diagram, allowing for the continuum of in-plane modes, takes the form shown in Fig. 3.

The phase diagram of Fig. 3 can be understood physically by noting that the condensation is a result of the gain-loss criterion, $H(\omega) > 0$.

Thus we see that the condensate can be formally represented as a coherent state of fermions, pairing with a finite total momentum. While in this respect the state is similar to FFLO, there are other differences. For example, in Eq. (11) the relative wavefunction of the pair is independent of momentum, and the pairing is entirely local. In a general FFLO state there is a momentum dependent pairing function, describing Cooper pairs of finite size.

Because the growth exponent depends only on $|k|$ the condensate emission at short times will cover a circle of in-plane wavevectors, giving a cone of emitted light.
However at later times the nonlinear terms neglected in Eq. (7) will break the degeneracy, selecting a spatial form for the condensate. In equilibrium such interactions favor condensate structures consisting of a pair of antipodal wavevectors \((k, -k)\), or more complex structures such as face-centered cubes. Here the nonlinearity corresponds to the depletion of the exciton population by the growth of the condensate. This will reduce the gain for collective modes of similar energies, suggesting that a single plane wave (Fulde-Ferrell) state may be favored. Although these nonlinearities determine a particular form for the condensate it is unlikely they will lead to a homogeneous state, so we do not treat them in detail here.

It is interesting to note that finite-momentum polarization condensates have been observed although in a different experimental protocol to that considered here. In these cases there is continuous pumping and relaxation, and a spatial structure imposed by a pump and trap. The mechanisms leading to this finite momentum condensate have yet to be established, and are likely different from those discussed here. Nonetheless, these experiments demonstrate that microcavities could support exotic ordered states that have proved elusive in equilibrium.

VI. SUMMARY

We have calculated the linear response of a microcavity with a non-equilibrium population of excitons. The population produces new collective modes, which are analogs of the Cooper pairing mode in superconductors. We have shown that these modes are visible as peaks in the optical spectra. By considering the growth exponents of these collective modes we have found a phase diagram for the dynamical condensation. In a microcavity with a continuum of in-plane wavevectors there can be multiple unstable modes of different wavevectors. For some parameters the dominant (and, for sufficiently negative detuning, only) instabilities can occur at a non-zero wavevector. In these regimes the microcavity will develop a condensate with spatial structure, signaled by coherent emission at an angle to the cavity normal.

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Appendix A: Analytical pump solution

Ref. 15 gives the results of numerical simulations of Eqs. (1-3), driven by a linearly-chirped Gaussian pump pulse. These simulations show that there are parameter regimes in which the dynamics separates into a fast pumping stage, followed by a slower condensation stage. In this appendix we present an approximate analytical solution to Eqs. (1-3) which gives the population and (negligible) polarization at the end of the pump pulse. This solution forms the starting point for the dynamics discussed in the body of the paper.

The full numerical solutions in Ref. 15 show that the only significant polarization during the pumping is at the pump wavevector \(p\). Moreover, this polarization can be seen to be small compared with the applied pump field \(F\). Thus during the pumping we may neglect the second term in Eq. (1) for all wavevectors. With this approximation, Eqs. (1-3) reduce to an ensemble of independent two-level systems, driven by a field \(\psi_0^\alpha\) which is the externally applied field \(F\) filtered by the cavity response. For pumping at high angles, outside the stop band of the mirrors, \(\psi_0^\alpha\) is proportional to the pump pulse. Thus Eqs. (1-3) become the Bloch equations

\[
\begin{pmatrix}
\dot{P}_x^p \\ \dot{P}_y^p \\ \dot{D}_0
\end{pmatrix} = \begin{pmatrix}
0 & E - \Delta(t) & 0 \\ -E + \Delta(t) & 0 & g\Omega(t) \\ 0 & -g\Omega(t) & 0
\end{pmatrix}\begin{pmatrix}
P_x' \\ P_y' \\ D_0'
\end{pmatrix},
\]

where we have defined

\[
\psi_0^p = \Omega(t)e^{-i\int t'\Delta(t)dt'},
\]

\[
P_0^p = P_0 e^{i\int t'\Delta(t)dt'},
\]

\[
P' = \frac{1}{2}(P_x - iP_y).
\]

Note that the collective polarizations are at the pump wavevector, while the collective inversion is spatially uniform.

For a model pump pulse of the form

\[
g\Omega(t) = \frac{\Omega_0}{\tau} \text{sech} \frac{t - t_0}{\tau},\]

\[
\Delta(t) = \frac{\alpha}{\tau} \tanh \frac{t - t_0}{\tau} + \nu_0,
\]

Eq. (A1) has an analytical solution. The form of the population \(D_0\) at times \(t \gg \tau\) after the pulse is:

\[
\frac{D_0(E)}{\nu(E)} = 2 \frac{\cosh^2 \frac{\pi \alpha}{2} - \cos^2 \frac{\pi \sqrt{\frac{\nu_0}{2} - \alpha^2}}{2}}{\cosh \left[ \frac{\pi}{2} \left( (E - \nu_0)\tau - \alpha \right) \right] \cosh \left[ \frac{\pi}{2} \left( (E - \nu_0)\tau + \alpha \right) \right]} - 1.
\]
In the limit \( \Omega_0 > \alpha \gg 1 \) the distribution becomes

\[
\frac{D_0(E)}{\nu(E)} = 2n_F(E - \mu_+ - \nu_0)(1 - n_F(E - \mu_+ - \nu_0)) - 1,
\]

where \( n_F(E) \) is a Fermi distribution with temperature \( 1/(k_B\pi\tau) \) and the chemical potentials \( \mu_+ = \nu_0 \pm \frac{E}{\tau} \). If the density of states \( \nu(E) \) is sufficiently small at energies below \( \mu_- \) then this lower edge is irrelevant. The occupation function \( D_0(E) \) is then equivalent to an equilibrium Fermi distribution with \( \mu = \mu_+ \). In this paper we consider parameters where this applies, choosing \( \nu_0 = -30/\tau, \Omega_0 = 18 \) and \( \alpha = 17.5 \).

Since the dynamics during the pumping, Eq. (A1), involves only \( P_p \), the polarization at any other wavevector \( P_{k \neq p} \) remains zero. For the polarization at the pump wavevector, the analytical solution gives a window of energies \( \sim \tau \) in which there is a non-zero polarization after pumping. However, in the absence of an external field and with \( \psi_p \approx 0 \) as is the case after pumping, the subsequent evolution of the polarization is free. As a result, the total polarization \( P_p = \int P_p(E)dE \) decays by free induction decay, and so may be neglected after a time of order \( \tau \). The numerical work of Ref. 15 showed that for suitable parameters the preparation of this state, including the free induction decay of the remnant polarization, finishes before the dynamics discussed in the main body of this paper takes place.

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