The crossover between lasing and polariton condensation in optical microcavities

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We study a model of a photon mode dipole-coupled to a medium of two-level oscillators in a microcavity in the presence of dephasing processes introduced by coupling to external baths. Decoherence processes can be classified as pair-breaking or non-pair-breaking in analogy with magnetic or non-magnetic impurities in superconductors. In the absence of dephasing, the ground state of the model is a polariton condensate with a gap in the excitation spectrum. Increase of the pair-breaking parameter $\gamma$ reduces the gap, which becomes zero at a critical value $\gamma_{C1}$; for large $\gamma$, the conventional laser regime is obtained in a way that demonstrates its close analogy to a gapless superconductor. In contrast, weak non-pair-breaking processes have no qualitative effect on the condensate or the existence of a gap, although they lead to inhomogeneous broadening of the excitations.

There are two apparently distinct phenomena where quantum coherence has been observed on the macroscopic scale: the laser and Bose Einstein condensation (BEC) of massive particles.

The laser is a weak-coupling phenomenon: a coherent state of photons created by stimulated emission from an inverted electronic population due to the strong pumping. The polarisation of the medium is heavily damped and the atomic coherence is very much reduced. A coherent photon field, oscillating at bare cavity mode frequency, is the only order parameter in the system \[\text{\textcircled{1}}\].

Polaritons are strongly coupled modes of light and electronic excitations. A theoretically constructed polariton condensate is a mixture of coherent state of light and coherent state of massive particles in the media. It is characterised by two order parameters: the coherent polarisation and the coherent photon field and exhibit a gap in the excitation spectrum \[\text{\textcircled{2}}\].

The simplest models of both the laser and the polariton condensate start from similar equations of motion, but make different physical approximations. The laser is assumed to be far from equilibrium, and decoherence (dephasing) — arising from collisions, interactions with phonons, defects or impurities, and manifested by homogeneous broadening ($1/T_2$) of the electronic excitations — is imposed by hand. In contrast, the polariton condensate has been assumed to be in thermal equilibrium, and dephasing phenomena neglected.

In this letter, we show that a consistent inclusion of decoherence phenomena drives a crossover from the polariton condensate to the lasing regime. Using a model of two-level oscillators coupled to a degenerate optical cavity mode with coupling strength $g$, and with further coupling of the internal degrees of freedom to external baths (defined in the Hamiltonian below), we find the generic phase diagram of Fig. \[\text{\textcircled{3}}\], where a single pair-breaking parameter $\gamma$ encapsulates the strength of electronic dephasing. When the decoherence is weak, we find a strong-coupling regime with a polariton condensate and a gap in the excitation spectrum. The gap is robust against (weak) dephasing processes, a result that can be obtained only by the correct self-consistent treatment of decoherence processes. For larger decoherence, the gap is suppressed to zero at a critical value $\gamma_{C1}$ in a similar way to the effect of pair-breaking processes in a BCS superconductor, and the coherent polarisation is reduced. For low excitation densities coherence is completely suppressed at a higher value $\gamma_{C2}$. In the limit of high excitation densities (photon dominated) and large decoherence, we recover the traditional model of the laser.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Phase diagram. $p_{ex}$ is a density of total excitations in the microcavity, which is a sum of photons and electronic excitations. In this terminology the minimum $p_{ex} = -0.5$ corresponds to no photons and no electronic excitations in the system, while 0.5 would correspond to the maximum of electronic excitations in the absence of photons. The phase boundaries $\gamma_{C1}$ and $\gamma_{C2}$ are marked in Fig. \[\text{\textcircled{3}}\].
\end{figure}
The principal aim of this paper is to establish the correct theoretical framework to discuss the differences and similarities between a condensate and a laser, so we adopt a deliberately simplified model that cannot accommodate all the details of important physical systems. Nevertheless, in closing we attempt estimates of the relevant parameters for some relevant physical systems.

To obtained the phase diagram we have considered the following Hamiltonian: 

\[ H = H_0 + H_{SB} + H_B, \]

where \( H_0 = \omega_c \psi|\psi\rangle + \sum_j \epsilon_j (b_j^\dagger b_j - a_j^\dagger a_j) + \frac{g}{\sqrt{N}} (b_j^\dagger a_j \psi + \psi^\dagger b_j). \]

\( H_0 \) describes an ensemble of \( N \) two-level oscillators with an energy \( \epsilon_j \) dipole coupled to one cavity mode. \( b \) and \( a \) are fermionic annihilation operators for an electron in an upper and lower states respectively and \( \psi \) is a photon bosonic annihilation operator. The sum here is over the possible sites where an exciton can be present (different molecules or localised states in the disorder potential).

\( H_{SB} \) contains all interactions with an environment:

\[
H_{SB} = \sum_k g_k \xi_k d_k^\dagger d_k^\dagger + \sum_{j,k} [g_{j,k}^a (b_j^\dagger a_j^\dagger b_j + c_{j,k}^\dagger + c_{j,k}^\dagger)] + g_{j,k}^b (b_j^\dagger a_j^\dagger b_j + a_j^\dagger a_j^\dagger c_{j,k}^\dagger + c_{j,k}^\dagger)
\]

and the \( H_B \) is a Hamiltonian for the baths. The first term in \( H_{SB} \) gives rise to the decay of the photon field from the cavity. The second term describes an incoherent pumping of two-level oscillators. The third term contains all the processes which destroy the electronic excitations such as the decay to photon modes different to the cavity mode. These processes, apart from a dephasing effect, will cause a flow of energy through the system. However, in the steady state the total number of excitations, \( n_{ex} = \psi^\dagger \psi + \frac{1}{2} \sum_j (b_j^\dagger b_j - a_j^\dagger a_j) \), which is the sum of photons and excited two-level oscillators, is conserved.

Finally, the fourth and the fifth terms describe all the dephasing processes which do not change the total number of excitations in the cavity, for example collisions and interactions with phonons and impurities. They can be divided into a part, which acts in the same way (fifth term) and with an opposite sign (fourth term) on the upper and lower levels. These four terms contain all the essential groups of decoherence processes in the media.

Processes, described by the second, the third and the fourth terms in \( H_{SB} \), are analogous to the pair-breaking, magnetic impurities in superconductors and correspond to potentials which vary rapidly in space (on the length-scale of excitons) or in time. On the contrary the fifth term contains interactions which vary very slowly in space in comparison to the size of excitons and in time and are analogous to normal, non-magnetic, impurities. They do not have any pair-breaking effects and lead only to an inhomogeneous broadening of energies.

For very big pumping and photon decay rates the system would be out of equilibrium. However, if the system is pumped slowly, with a rate slower than the thermalisation rate equilibrium assumption can be justified. In this work we present only the influence of the decoherence on the system leaving the implications of a non-equilibrium system for the future work. This does not put any restrictions on the strength of the decoherence nor the density of excitations. The pair-breaking processes described by the fourth term in \( H_{SB} \) give exactly the same decoherence effects as the pumping and damping of two-level oscillators preserving equilibrium for any strength of the interaction. The ratio between the pumping and all the damping processes which can be arbitrary big even for small absolute values of both will be expressed in terms of the excitation density \( \rho_{ex} \).

Usually the baths are averaged out before the exciton - photon interaction is studied which leads to the well-known quantum Maxwell-Bloch (Langevin) equations for the photon field and polarisation. When there is a coherent polarisation this procedure is not valid (Langevin equations can be used to study the laser regime but not the polariton or intermediate regime). Instead, we must use the self-consistent Green’s function techniques similar to the Abrikosov and Gor’kov theory of gapless superconductivity. Decoherence processes are treated by perturbation theory about a BCS - like Green function for the interacting exciton-photon system. The major difference between our calculations and the Abrikosov and Gor’kov theory is that we have two order parameters, the coherent photon field and polarisation connected through the chemical potential, set by the excitation density. We consider two-level systems in contrast to free propagating electrons model used by Abrikosov and Gor’kov. Within the Markov approximation we can express the influence of the bath by a single parameter \( \gamma_l = g_l^2(0)^2 N_l(0) \) where \( l = 1, 2 \) correspond to the pair-breaking and non-pair breaking decoherence and \( N_l \) are densities of states for the respective baths. The details of the method are published elsewhere.

We first determine the ground state properties of the system with uniform energies, \( \epsilon_j = \epsilon \), for different densities in the presence of the pair-breaking decoherence. Figure 3 shows the ground state behaviour of the coherent fields and inversion. For small values of \( \gamma/g \), up to some critical value \( \gamma_{c1} \), \( \langle \psi \rangle \) is practically unchanged while for \( \gamma/g > \gamma_{c1} \) is damped quite rapidly with the increasing dephasing (see Fig. 3 - upper panel). This critical value of the decoherence strength, \( \gamma_{c1} \) is proportional to \( \rho_{ex} \), suggesting that for higher excitation densities the system is more resistant to the dephasing. At low excitation densities, where \( \rho_{ex} < 0 \), there is a second critical value of the decoherence strength, \( \gamma_{c2} \), where both coherent fields are sharply damped to zero.

The ratio of coherent polarisation to coherent field \( \langle P \rangle/\langle \psi \rangle \) (Fig. 3 - lower panel) for an isolated system, where \( \gamma = 0 \), depends on the excitation density. The condensate becomes more photon like as \( \rho_{ex} \) is increased due to the phase space filling effect. For finite \( \gamma \), at a given excitation density, this ratio decreases with increasing \( \gamma \) meaning that \( \langle P \rangle \) is more heavily damped than \( \langle \psi \rangle \).

To understand the behaviour of coherent fields we
study the excitation spectrum of the system (Fig. 3). As $\gamma$ increases the two quasi-particle peaks (Fig. 3 a) broaden, which causes the decrease in the magnitude of the energy gap (Fig. 3 b and c). Finally, precisely at $\gamma C_1$ (shown in Fig. 2), these two broadened peaks join together and the gap closes (Fig. 3 d). $\gamma C_1$ defines the phase boundary (see Fig. 1) between a gapped and gapless condensate at low densities and between a condensate and laser at high densities.

The laser operates in the regime of a very strong decoherence, comparable with the light-matter interaction itself. In the presence of such a large decoherence laser action can be observed only for a sufficiently large excitation density. The coherent polarisation in a laser system is much more heavily damped than the photon field and a gap in the density of states is not observed. Thus the laser is a regime of our system for very large $\rho_{ex}$ and $\gamma$. It has to be, however, pointed out that there is no formal distinction between a laser and a phase-coherent condensate of polaritons. In the laser, coherence in the media (manifested by the coherent polarisation) although small, is not completely suppressed so the laser can be seen as a gapless condensate with a more photon-like character. One of the possible distinctions between a polariton condensate and laser could be an existence of an energy gap in the excitation spectrum.

FIG. 2. Coherent photon field $\langle \psi \rangle$ (upper panel), inversion (middle panel) and ratio of coherent polarisation and photon field $\langle P \rangle / \langle \psi \rangle$ (lower panel) as functions of the pair-breaking decoherence strength, $\gamma/g$ for different excitation densities, $\rho_{ex}$. The coherent fields are rescaled by $\sqrt{N}$ and consequently the inversion by $N$. In this terminology the minimum $\rho_{ex} = -0.5$ corresponds to no photons and no electronic excitations. Inset: Comparison between the influence of a pair-breaking (solid line) and a non-pair-breaking (dotted line) decoherence on $\langle \psi \rangle$ (upper panel), inversion (middle panel) and $\langle P \rangle / \langle \psi \rangle$ (lower panel) for two values of $\rho_{ex}$.

FIG. 3. Density of states for $\rho_{ex} = -0.2$ and different decoherence strengths, $\gamma/g$ for a pair-breaking (solid line) and a non-pair-breaking (dotted line) decoherence processes. The solid line corresponds to a normal state in which coherent fields are suppressed.
The dotted lines in onset of Fig. 2 and in Fig. 3 show the influence of the non-pair breaking processes (the fifth term in $\rho$). We found that these processes have some quantitative influence on the coherent fields and the gap in the density of states but do not cause any phase transitions. Although the two quasiparticle peaks get very broad, the gap is only slightly affected. Even for much larger values of $\gamma$ than presented in Fig. 2 the gap is present until the coherent fields get completely suppressed. This type of processes give similar effect as the inhomogeneous broadening of energy levels.

Finally, we study an influence of the pair-breaking processes on the system of realistic, inhomogeneously broadened two-level oscillators. The coherent fields, $\gamma_{C1}$, $\gamma_{C2}$ and the energy gap are slightly smaller than in the uniform case but all the regimes and transitions are the same. The broadening of the density of states and the suppression of the energy gap can be observed as $\gamma$ is increased (Fig. 3).

![Density of States](image)

**FIG. 4.** Density of states for a Gaussian broaden case with $\sigma = 0.5$ for different values of the pair-breaking decoherence at $\rho_{ex} = -0.3$ (left panel) and $\rho_{ex} = 0.2$ (right panel).

General conclusions about the conditions and systems where polariton condensation is likely to be observed can be drawn from our theory. Systems with large dipole coupling $g$, and high densities but low decoherence are good candidates. Inhomogeneous broadening of energies have much weaker influence than a homogeneous dephasing. The large value of $g$ is particularly important as it defines the energy scale in this problem. Only the relative strength of the dephasing and inhomogeneous broadening with respect to $g$ influence the condensate. Thus systems with larger $g$ can allow larger values of decoherence and broadening. The second important factor is the excitation density. Since the gap is proportional to the amplitude of the coherent field the condensate is more robust at high densities. Unfortunately the increase in the density usually implies an increase in decoherence as the dephasing time ($T_2$) for most of materials increases linearly with density. An additional increase in dephasing comes from the increase in the pump intensity required to achieve higher densities. This could be partially overcome by improving the quality of the mirrors in microcavity systems.

To be more specific we consider in detail different materials. Among semiconductors CdTe, ZnSe and GaN are characterised by large values of $g$ (measured by the polariton splitting in the normal state). For CdTe, the gap was measured to be 29 meV [6] and the dephasing ($1/T_2$), at the exciton density of 0.05 per Bohr radius, to be 2.5 meV (0.08 g) [7]. In the absence of inhomogeneous broadening our theory predicts a gap $\Delta = 0.7g = 20.3$ meV. In the case of Gaussian broadening with $\sigma = 0.2g = 5.8$ meV reported in some structures the gap is still significant $\Delta = 0.08g = 2.32$ meV and it gets suppressed for broadening larger then 8.7 meV. Other promising candidates would be excitons in organics, where the reporting polariton splitting was as large as 80 meV [8], or dilute atomic gases in which the dephasing is measured to be around 1000 times smaller then the dipole coupling [9] and high excitations can be achieved without an increase in the density of the gas. GaAs — due to the small value of $g$ — is not the best material for the observation of the condensate. Polariton splitting in GaAs based structures is usually only around 9 meV [10] and the recently reported 20 meV splitting in 36 well structure [11] is probably approaching the absolute limit for these materials.

In this work we have developed a method to include decoherence effects in the systems with an energy gap in the excitation spectrum, where the widely used Langevin equations are not valid. We have studied an equilibrium, incoherently pumped system. However, in the same way it would be possible to examine the influence of an environment on coherently driven condensates. We have shown that in the presence of small decoherence the polariton condensate is protected by an energy gap in the excitation spectrum. The gap is proportional to the coherent field amplitude and thus the excitation density, so the condensate is more robust at high densities. As the decoherence is increased the gap gets smaller and finally is completely suppressed. If the polariton condensate is ever to be observed an increase in density without an increase in decoherence is necessary. Our method allows us to study different regimes in the cavity and thus to examine the crossover from a polariton condensate to a laser. The laser emerges from the polariton condensate at high densities when the gap in the density of states closes for large decoherence and thus is analogous to a gapless superconductor. Our work generalises the existing laser theories to include coherence effects in the media. When this coherence is included the generation of a coherent photon field without population inversion becomes possible.
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