Coherence properties and luminescence spectra of condensed polaritons in CdTe microcavities

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We analyse the spatial and temporal coherence properties of a two-dimensional and finite sized polariton condensate with parameters tailored to the recent experiments which have shown spontaneous and thermal equilibrium polariton condensation in a CdTe microcavity [Kasprzak et al., Nature 443, 409 (2006)]. We obtain a theoretical estimate of the thermal length, the lengthscale over which full coherence effectively exists (and beyond which power-law decay of correlations in a two-dimensional condensate occurs) of the order of 5µm. In addition, the exponential decay of temporal coherence predicted for a finite size system is consistent with that found in the experiment. From our analysis of the luminescence spectra of the polariton condensate, taking into account pumping and decay, we obtain a dispersionless region at small momenta of the order of 4 degrees. In addition, we determine the polariton linewidth as a function of the pump power. Finally, we discuss how, by increasing the exciton-photon detuning, it is in principle possible to move the threshold for condensation from a region of the phase diagram where polaritons can be described as a weakly interacting Bose gas to a region where instead the composite nature of polaritons becomes important.

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I. INTRODUCTION

Since the first observation of polariton bosonic stimulation under non resonant excitation obtained in 1998 in a CdTe microcavity1, there has been a considerable interest in realising a polariton Bose-Einstein condensate (see, e.g., the recent review2 and references therein). This search has been mainly motivated by the expected high transition temperature of polaritons due to their very light effective mass3,4,5. A spontaneous and thermal equilibrium polariton condensate has been very recently achieved in a CdTe microcavity6. This was soon followed by similar effects in a GaAs microcavity7,8. Other recent progress in polariton condensation includes studying trapped polaritons in a GaAs structure under stress9 and room temperature polariton lasing in GaN10.

The microcavity polariton system is finite, two-dimensional, decaying, and interacting, and therefore is expected to differ from the Bose-Einstein condensation of ideal three-dimensional bosons. Much theoretical work involving different modelling has been done to evaluate the phase diagram for microcavity polaritons11,12,13,14,15 — for a comprehensive review on the use of and comparison between different models see Ref.2. Already the results obtained in2,12, and adapted to the CdTe experiment15, have shown a very good agreement between the experimental estimate of the critical density for condensation obtained from the occupation data — at a given temperature and detuning conditions — and the theoretical estimate obtained from the lower polariton blue-shift (see the online supplementary information of Ref.14). Recently, a direct comparison between experimental and theoretical phase boundaries for condensation of microcavity polaritons in CdTe under different conditions of cryostat temperature and detuning have been performed in Ref.13. In that work, it was shown that for a steady state situation, in the presence of pumping and decay, although the polaritons may display a thermal population, the presence of pumping and decay may yet have noticeable effects. In particular the small discrepancies between the experimental data and the equilibrium theoretical estimates for a closed system can be attributed to the effects of pump and decay.

In fact, it has already been shown14,15 that quantum condensation is indeed possible in strongly dissipative systems with continuous pump and decay and that it shares several features with quantum condensation in a closed system at equilibrium. In particular, the mechanism of condensation, connected with the chemical potential reaching the lower polariton (LP) mode, is exactly the same in closed systems at equilibrium and in open systems with pump and decay. In the latter, the role of the chemical potential is played by the energy at which the non-thermal distribution diverges. However, it has been shown14,15 that the presence of pump and decay can significantly modify the coherence properties of the condensate. In particular, the power-law decay of temporal and spatial correlations, caused by the 2D nature of the polariton system, can be strongly modified by the presence of dissipation. Finally, it has been shown15 that the finite size of the system additionally modifies the temporal coherence properties and in particular it leads to a crossover from a power-law decay of temporal coherence in an infinite system to an exponential decay in a
finite system.

As well as coherence measurements, changes to the photoluminescence spectrum can also provide evidence for quantum condensation. The photoluminescence spectrum reflects the structure of the normal modes of the microcavity system, weighted (in thermal equilibrium) by the bosonic occupation factor. The structure of collective modes changes dramatically when microcavity polaritons condense: The lower and upper polariton modes, which are the eigenmodes of the system in the non-condensed regime, are now replaced by two new eigenmodes. In particular, the lower polariton is replaced by the collective (Goldstone) mode and its quadratic dispersion evolves to a linear dispersion at small momenta in the closed equilibrium system, while it becomes diffusive at small momenta in the open system with pump and decay.

In this paper we discuss the consequences of these general results for the properties of coherence and luminescence spectra of microcavity polaritons in the specific conditions of the CdTe experiments. In particular, by considering the closed system, we give estimates of the thermal length, the lengthscale over which full coherence effectively exists (and beyond which power-law decay of correlations in a two-dimensional condensate occurs), and find that its value, of the order of 5nm, is consistent with the measurements of the spatial decay of the first order coherence reported in. We also consider effects due to pump and decay, and calculate the luminescence spectra for the conditions of the CdTe experiments and in particular analyse the size of the diffusive region as well as the size of the linear regime in the luminescence. The non-equilibrium formalism, which accounts for the effects of pump and decay, also allows us to calculate the homogeneous polariton linewidth as a function of the pump power. Finally, in Ref. it has been shown that the experimental data for the phase boundary lie close to the crossover region between a Berezinskii-Kosterlitz-Thouless (BKT) transition of structureless bosons and a regime where instead the boundary is determined by the long-range nature of the polariton-polariton interaction. In this paper we show how the increase of the exciton-photon detuning, which results in a higher transition density at a given temperature, moves the transition (at a given temperature) from the BKT weakly interacting Bose boundary to a regime where the composite nature of polaritons becomes important.

The paper is organised as follows: in Sec.II we analyse coherence properties on the basis of an equilibrium theory for a closed system and in Sec. III we discuss how non-equilibrium and finite size effects modify these results. Finally, in Sec. IV we analyse the influence of the exciton-photon detuning on the polariton phase diagram.

II. COHERENCE PROPERTIES AT EQUILIBRIUM

The coherence properties of the condensed polariton system are described by the functional form of the first order coherence as a function of time and space:

\[ g^{(1)}(t; r) = \frac{\langle \psi^\dagger(r, t)\psi(0, 0) \rangle}{\sqrt{\langle \psi^\dagger(0, 0)\psi(0, 0) \rangle \langle \psi^\dagger(r, t)\psi(r, t) \rangle}}, \] (1)

where \( \psi(r, t) \) is the photon field. To determine the characteristic length- and time-scales in \( g^{(1)}(t; r) \), we must therefore evaluate the correlation function in the numerator of Eq. (1), which can be achieved by finding the normal modes — i.e. collective quasi-particle excitations — of the system. We will therefore start our analysis by determining the spectrum of the collective excitations above the polariton condensate, making use of a Bose-Fermi model — tailored to the parameters characterising the CdTe microcavity — which takes into account the composite nature of polaritons, the quantum well disorder, and the non-linearities associated with exciton-photon interactions (for more details on the model see Refs. 12,13).

A. Collective modes of the polariton condensate

The spectra of the collective modes of a polariton condensate can be evaluated by considering the second order fluctuations above the mean-field approximation (see, e.g. Refs. 5,12). One can show that, when the system condensates, the lower and upper polariton modes are not any longer the eigenmodes of the problem and are instead replaced by new collective modes: The lower polariton becomes a linear (Goldstone) mode at low momenta, while two new branches appear below the chemical potential, which are seen as gain in the spectral weight, as shown. Note that Fig. 1 shows the incoherent emission which is the product of the spectral weight times the Bose occupation factor \( n_B(\omega) \). The observation of these predicted features in the photoluminescence would provide very strong evidence for polariton condensation. However, the emission from below the chemical potential is suppressed at large angles (see Ref. 13), while the PL emission from modes far above the chemical potential is suppressed exponentially by the thermal occupation of these modes, making such features hard to observe. In addition, the strong emission from the condensate at zero momentum and the frequency corresponding to the chemical potential, which is not shown in Fig. 1, might mask
FIG. 1: Contour plot of the spectral weight \( W(\omega, p) \) as a function of energy \( \omega - \omega_0 \), where \( \omega_0 \) indicates the bare cavity photon energy, and emission angle \( \theta = \sin^{-1}(c|p|/\omega_0) \) for a closed equilibrium system at temperature \( T = 19 K \), detuning \( \delta = +6 \, \text{meV} \) and a fixed density \( n = 6.1 \times 10^8 \, \text{cm}^{-2} \) (the mean-field critical density for condensation for these values of temperature and detuning is given by \( n_c = 6 \times 10^7 \, \text{cm}^{-2} \)). The horizontal dashed (green) line marks the value of the chemical potential. The location of the peak for the upper branch Goldstone mode is explicitly plotted (red). Its fit to a linear dispersion is plotted in Fig. 2.

both the linear behaviour of the collective modes at low momenta and the emission from below the chemical potential.

Nevertheless, the analysis of the linear behaviour of the Goldstone mode can give important information on the coherence properties of the condensate. In particular, the slope of the linear mode is the sound velocity of the condensate, \( c_s \). As it will be shown in the next section, the sound velocity together with the temperature determines the lengthscale, the thermal length, for the decay of the spatial coherence.

In Fig. 2 we plot the upper branch of the Goldstone mode, obtained from evaluating the locations of the corresponding peaks as shown in Fig. 1 for increasing values of the polariton density \( n \) ranging from the non-condensed to the condensed regime. As shown in the picture, the extension of the linear behaviour grows with increasing density up to approximately 8° at the highest density considered. At the same time, the slope of the linear mode and therefore the sound velocity increases with increasing density.

B. Condensate emission — decay of spatial and temporal coherence

In the condensed state, because there is no restoring force for global phase fluctuations, the amplitude of phase fluctuations at low momenta can be large. However, there is a restoring force for amplitude fluctuations, and so except near the transition, it is valid to assume these to be small. Taking the phase fluctuations to all orders and the amplitude fluctuations to second order, one can calculate the first order coherence function:

\[
g^{(1)}(t; r) = [1 + \mathcal{O}(1/\rho_0)] \exp[-f(t, r)].
\]  

(3)

In this expression, the terms of order \( \mathcal{O}(1/\rho_0) \) arise from including density fluctuations, but are not relevant in describing the long distance behaviour. This form asymptotes to the well-known power law decay of correlations at large times and distances, for which:

\[
f(t, r) = \eta \log \left( \frac{\sqrt{c_s^2 t^2 + r^2}}{\beta c_s} \right).
\]  

(4)

Here, \( c_s \) is the velocity of the Goldstone mode, \( \rho_0 \) the mean-field estimate of the condensate density and \( \eta = m k_B T/(2\pi \rho_0) \). The logarithmic dependence on the spatial coordinate \( r \) reflects the expected BKT decay of correlations for a two-dimensional system. Note that since the asymptotic form of the spatial decay is power-law, and not exponential, there is strictly no characteristic lengthscale of the decay, so the coherence length is not a well defined quantity. However, there does exist a lengthscale — the thermal length — which describes the distances at which the long range asymptotic behaviour begins to be relevant; below this lengthscale, correlations
are short range, and approximately independent of distance. This thermal length is given by:

$$\xi_T = 2\pi \frac{c_s}{k_B T}. \quad (5)$$

Substituting the values of the velocity of the Goldstone mode $c_s$ determined in Section II A we obtain thermal lengths given by $\xi_T = 3.13\,\mu m$, $4.83\,\mu m$, and $5.92\,\mu m$ respectively for polariton densities of $n = 6.1 \times 10^8\,cm^{-2}$, $1.51 \times 10^9\,cm^{-2}$, and $2.59 \times 10^9\,cm^{-2}$ for $T = 19K$ and a detuning $\delta = +6\,meV$. The dependence of the thermal length on the density is shown in Fig. 3. Experiments on coherence in CdTe reported a contrast of interfer-ence fringes up to $5\%$ below the transition and up to $45\%$ above the transition. The $45\%$ contrast has been measured up to a distance of roughly $6 \,\mu m$ after which the contrast drops to $30\%$ up to a distance of roughly $12\,\mu m$. Our estimates of the thermal length, of the order of $5\,\mu m$, are consistent with these measurements. Unfortunately due to a very large spatial inhomogeneity of the condensate caused by large photonic disorder it was not possible in that experiment to examine a functional dependence of the decay of the correlation as a function of the distance. Spatial coherence has also been recently measured in GaAs systems, such systems appear to show less spatial inhomogeneity, and so might allow fuller investigation of the decay of correlations at long distances. However, the data presented in Ref. shows measurements of $g^{(1)}(0:\mathbf{r})$ at only six different separations, ranging between $1.3\,\mu m$ and $8\,\mu m$. According to the analysis in that paper, the data are adequately described over that range by modelling the system as a degenerate Bose gas, without considering any changes to the dispersion of the bosons. Such a model in effect a model of non-interacting Bosons, and so does not describe power law decay of correlations.

\section{III. Influence of Pump, Decay and Finite Size on Coherence Properties}

Since the effects of pump and decay in the polariton system are large compared to other relevant energy scales, such as temperature (one can, e.g., compare the homogeneous linewidth of polaritons, of roughly $1\,meV$ with the characteristic temperature $\sim 20K \sim 1.7\,meV$), we expect pump and decay to modify the coherence properties of the polariton condensate. We have addressed these issues by looking at the spontaneous condensation for a system, coupled to external baths, representing the pumping and decay mechanisms. We have shown that, even when the polariton system is characterised by a thermal distribution, the presence of pumping and decay significantly modify the spectra of collective excitations. In particular, the low energy phase modes become diffusive at small momenta, leading to correlation functions — and thus condensate line-shape — that differ both from an isolated equilibrium BEC and from those for phase diffusion of a single laser mode. Here we give estimates of the size of the diffusive region for conditions close to those of the CdTe experiments.

\subsection{A. Collective modes of the polariton condensate in presence of pump and decay}

In the normal state the fluctuation spectrum shows the usual polariton branches, which are now also homogeneously broadened due to pumping and decay. In the condensed state, however, the collective modes have the following energy $\omega$ vs. momentum $p$ form:

$$\omega = -ix \pm i\sqrt{x^2 - c_s^2 p^2}, \quad (6)$$

and are thus diffusive, rather than dispersive for $p \leq x/c_s$. Here, the parameter $x$ is a non-linear function of the pumping and decay strength and determines the linewidth of the Goldstone mode. Similarly to an equilibrium picture, the structure of the collective modes will be reflected in the PL which is a product of the spectral weight and the occupation function. In Figure 4 we plot the PL spectra for the parameters characterising the recent experiments on CdTe. It can be seen that in its low frequency and momentum part, the main feature of the spectrum is a flat region followed by a dispersive mode which then approaches the LP spectrum. As in the closed system, the coupling of particle- and hole-like bosonic excitations means that in the condensed state, the same mode structure can also be seen below the chemical potential. The spectral weight of this mode is however much weaker and so it is less visible in the luminescence in Fig. 4. Our numerical analysis shows that the range of the flat region depends mainly on the photon decay rate; but also depends weakly on the pump parameter $\gamma$ such that the flat region should increase slightly with increasing pump power which is consistent with the experiment. For the photon decay rate of $1\,ps$
(\sim 0.49\text{meV}) the size of the flat region extends up to
around 5 degrees (depending on the coupling strength \(\gamma\)).
There is also an alternative explanation that the
observed flattening above the condensation transition is
mainly due to the finite size effects. Note that since the
size of the flat region here is of similar order to the range
over which the Goldstone mode is linear (see Figure 3),
it is therefore possible that, at least close to the transition,
the dispersion will cross directly from diffusive to
quadratic, without a linear part.

B. Decay of temporal coherence

Similarly to an equilibrium picture, in order to exam-
ine the coherence properties of a condensed system, one
needs to determine the field-field correlation functions
including the phase fluctuations to all orders and to
determine the analogue of expression (3) for a dissipative
system. This approach has several advantages. Firstly it
naturally includes both condensate and non-condensate
luminescence in the same formalism, and (in the finite
system) provides a linewidth for the condensed part.
Secondly it recovers the correct power-law behaviour of oc-
cupation of modes in momentum space when integrated
nonduly it recovers the correct power-law behaviour of oc-
system) provides a linewidth for the condensed part. Sec-
luminescence in the same formalism, and (in the finite
s includes both condensate and non-condensate

where \(\eta'\), in distinction to \(\eta\) in Eq. 4, depends on \(x\) as well as temperature and condensate density, and \(\xi_c\)
is a characteristic length scale for the non-equilibrium
occupation function of polaritons, given by \(\xi_c \propto c_s/E\),
where \(E\) is a characteristic energy scale of the polaritons’
distribution. Thus, there is still power-law decay, but
due to pumping and decay the powers for temporal and
spatial decay do not match. In the case of systems with
strong pumping and decay, but where the distribution
function is close to thermal, as in the recent experiments
on CdTe, then \(E \simeq k_B T\) and so

Therefore, if the system, despite the presence of pump
and decay, is able to thermalize, the thermal length is not
strongly affected by the presence of pump and decay and
its expression coincides with that of a closed system.

In order to understand the temporal coherence mea-
surements which show exponential decay, it turns out to
be necessary to address the influence of finite size on the
expression 5. A detailed analysis of this can be found in reference 12. Summarising, in the finite condensed
system the energy level spacing is given by \(\Delta E = c_s/R\) [note
this differs from the single particle level spacing relevant
for the uncondensed regime \(\Delta_{s,p} = 1/(2mR^2)\)]. With
this level spacing it has been shown 12 that the function
which controls the decay of coherence can be approxi-
mately written as

\[
f(t, r) = \begin{cases} \frac{\eta'}{2} \log \frac{c_s t}{x} 
& \text{if } r \approx 0, \ t \to \infty, \\
\eta' \log \frac{r}{\xi_c} & \text{if } r \to \infty, \ t \approx 0. \end{cases}
\]  

(7)

In this expression, we have used the assumption of a ther-
mal distribution to write \(k_B T \) for the high energy cutoff.
c_s/\xi_c as in Eq. (5). The first term, which dominates at large times, gives exponential decay of correlations whereas the second term gives the power-law decay characteristic for infinite 2D systems. The relative importance of these two terms depends on the system size and temperature (i.e. how deep the system is into the condensed regime). If the system is large or is close to the phase boundary i.e where $k_B T \gg \Delta_\phi = c_s/R$ the second term (i.e power-law decay) dominates at short times and it crosses to exponential decay only at later times. Rearranging this condition it translates to $R \gg 2\pi c_s/k_B T = \xi_T$ which says that the system size is much larger than the thermal length. However if the system is small or deep inside the condensed region and so $k_B T \ll \Delta_\phi = c_s/R$ the phase fluctuations associated with 2D nature of the condensate are frozen out and the first term dominates giving an exponential decay at all times. This conditions translates to $R \ll 2\pi c_s/k_B T = \xi_T$. Thus spatial coherence over the whole system size implies the exponential decay of temporal coherence (however not vice-versa).

### IV. EXCITON PHOTON DETUNING

In this last section we discuss some aspects of the polariton phase diagram and the possibility of experimentally exploring different parts of it. As already mentioned in the introduction, there has been a recent investigation of the direct comparison between experimental and theoretical phase boundaries for condensation of polaritons in a CdTe microcavity. This work has shown that the current experimental data for the phase boundary lie close to the crossover between a BKT transition of structureless bosons (low density part of the diagram shown in Fig. 6) and a regime where instead the phase boundary is characterised by the long-range nature of the polariton-polariton interaction and where therefore the composite nature of polaritons matters (a region where the dependence of the critical temperature on the density is slower than linear). Those results therefore suggest that polariton condensation departs from the weakly interacting boson picture.

In that experiment, however, it has proven particularly difficult to change substantially the effective temperature of polaritons by changing that of the lattice (i.e. the cryostat temperature): Because of the short polariton lifetime, the polariton temperature is decoupled from that of the lattice. It has therefore not been possible to explore parts of the phase diagram other than the crossover region by means of changing temperature. However, if the effective temperature does not change much, then one can shift the boundary from the structureless boson to the long-range interaction part of the phase diagram, by changing the detuning between the photon and the exciton (see Fig. 6).

The change to the phase boundary due to detuning
can be simply explained in the low density (weakly interacting Bose gas) limit, as the increase of the polariton effective mass with the detuning — in this regime one can show that $k_B T \propto n/m_{\text{pol}}$ and neglecting exciton dispersion one may write, $m_{\text{pol}} = 2m_{\text{photon}}/[1 - \delta/\sqrt{\delta^2 + \Omega_R^2}]$, in which $m_{\text{photon}}$ is the photon mass, and $\Omega_R$ the Rabi-splitting at zero detuning, and zero-density. In the high density, long-range interaction part of the phase diagram the decrease of the critical temperature with the detuning at a fixed density is due to two mechanisms: a loss of coherence in the system as, when increasing the detuning, the lower polariton becomes less photon-like; and the decrease of the effective exciton-photon coupling, which controls the critical temperature at higher densities. In Fig. 6 we show that in order to see a significant move of the condensate threshold from the low-density to the high-density part of the phase diagram, one has to go at least to positive detunings larger that 20meV. Unfortunately, this has proven to be a challenge experimentally, and current experiments in CdTe allow one to reach a maximum detuning of roughly 12meV\textsuperscript{13}.

V. CONCLUSIONS

To summarise, we have analysed the spatial and temporal coherence properties of a two-dimensional, finite, and decaying condensate with parameters tailored to the recent experiments on CdTe microcavities. We have shown that the theoretical estimate of the thermal length (over which there is no decay of coherence) of up to 6 $\mu$m, and the exponential decay of temporal coherence are consistent with those found in experiment. We have also estimated the size of the dispersionless (flat) region in the PL — a manifestation of the diffusive nature of the Goldstone mode — to be around 5 degrees. This result suggests that the flattening of the polariton dispersion above the transition may be attributed to the dispersionless nature of the Goldstone mode. Since at current experimental conditions the linear part of the dispersion is of a similar size to the diffusive part it is likely that the flat region will cross directly to the quadratic dispersion and that the linear part will not be visible. In order to see linear dispersion, it would be necessary to decrease the size of the diffusive regime, which would require an improvement in the quality of the cavity mirrors. Finally we analyse the dependence of the phase diagram on exciton-photon detuning and suggest that going to higher positive detunings might provide a means of exploring different parts of the phase diagram.

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