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Coherence properties of the microcavity polariton condensate

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Abstract – A theoretical model is presented which explains the dominant decoherence process in a microcavity polariton condensate. The mechanism which is invoked is the effect of self-phase modulation, whereby interactions transform polariton number fluctuations into random energy variations. The model shows that the phase coherence decay, $g^{(1)}(\tau)$, has a Kubo form, which can be Gaussian or exponential, depending on whether the number fluctuations are slow or fast. This fluctuation rate also determines the decay time of the intensity correlation function, $g^{(2)}(\tau)$, so it can be directly determined experimentally. The model explains recent experimental measurements of a relatively fast Gaussian decay for $g^{(1)}(\tau)$, but also predicts a regime, further above threshold, where the decay is much slower.

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Introduction. – Microcavity polaritons are quasi-particles arising from the strong coupling between excitons and photons confined in planar cavity structures. The observation of coherent emission from a CdTe microcavity [1] has demonstrated that polaritons can form a new type of quantum condensate. As in other quantum condensates, such as atomic gases or superconductors, a key property is the existence of an order parameter, the local phase, which is correlated over large times and distances. The polariton condensate presents interesting theoretical challenges since, unlike these other systems, it is mesoscopic, typically consisting of a few hundred particles, and out of equilibrium, with pumping required to maintain the population against emission losses. In mesoscopic systems order parameters fluctuate [2,3], so the phase correlations decay. In this letter, we present a theory which shows that the source of the fluctuations is variations in the number of condensed particles, combined with polariton-polariton interactions: it is this dynamics that is responsible for the decoherence. Our theory shows that, under appropriate pumping conditions, existing microcavity structures should display much longer coherence times than currently measured, opening up opportunities for experiments manipulating the quantum state of the system.

The coherence of the polariton condensate can be quantified by the decay of the first-order coherence function, $g^{(1)}(\tau) \propto \langle a^\dagger(0)a(\tau) \rangle$, whose Fourier transform is

the emission spectrum. For the polariton condensate this function is directly revealed by coherence measurements on the optical emission [1,4,5]. In a condensate we expect long phase coherence times and thus a spectrally narrow emission above threshold. Recent experimental results [4] (see fig. 1) show that the decay time of $g^{(1)}(\tau)$ is ~ 150 ps. This is much longer than was originally believed [1,5], but short compared to a laser or atomic gas. Furthermore, the decay has a distinctive Gaussian form, and the decay time is approximately constant above threshold. The experiments also determine the intensity-intensity correlation function (second-order coherence function), $g^{(2)}(\tau)$, which reveals significant number fluctuations ($g^{(2)}(0) > 1$), decaying with a timescale ~ 100 ps.

In ref. [4], along with the experiments, we quoted the semiclassical results, eqs. (7) and (9) of this letter, which we showed to be compatible with the measured values of the coherence times. The discussion was limited to the case of slow number fluctuations, whose presence is directly evident in the experimental data. Here we show that this regime is achieved due to critical slowing down in the threshold region. At higher powers, where the critical slowing down disappears and fluctuations become faster, we predict that the phase coherence times will become significantly longer. This is shown to be a manifestation of the Kubo stochastic line-shape theory [6,7], in the motional narrowing limit. We also present numerical results for the threshold region, where mesoscopic effects are important and the semiclassical results break down.

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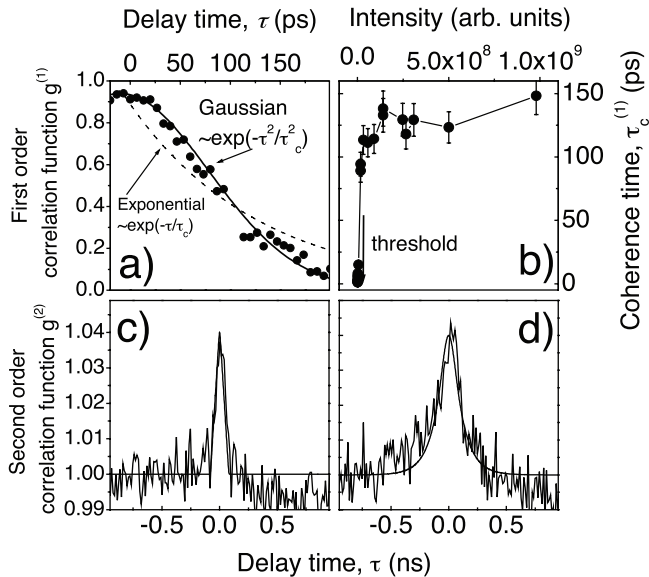


Fig. 1: Experimental coherence properties of the polariton condensate. (a) First-order coherence function $g^{(1)}(\tau)$ above threshold. (b) Dependence of the coherence time on pumping intensity. (c,d) Experimental intensity-intensity correlation function $g^{(2)}(\tau)$ below (c) and a factor of two above (d) threshold. Correcting for experimental factors gives $g^{(2)}(0) = 1.1$ for the emission above threshold. Reproduced from ref. [4].

This leads to a plateau in the variation of coherence time with condensate occupation (fig. 2b), in agreement with experiments (fig. 1b).

Rather than attempting a detailed microscopic model, our treatment is based on general considerations of interacting, open condensates. We argue that the observed slowing-down of number fluctuations near to threshold means that the pumping term that drives the system must be saturable, that is the pumping decreases as the population of the condensate grows. This, along with polariton-polariton interactions, and fluctuations due to pumping, is required in order to obtain agreement with the experiments. However, one or more of these features are missing from previous theories of condensate coherence [3,8–13]. Although a detailed description of the incoherent pumping process is beyond the scope of the present work, our results should provide a useful guide to the development of microscopic theories [8,10–14].

In the remainder of this paper, we briefly review the semiclassical treatment in the limit of slow number fluctuations [4]. We next explain how the Kubo stochastic line-shape theory [6,7] can be applied to give a general form for $g^{(1)}(\tau)$, accounting for both interactions and number fluctuations. This expression reduces to the observed Gaussian decay provided the number fluctuations are slow. We then develop a solution to a quantum-mechanical model of a pumped condensate, which has the same Kubo form for $g^{(1)}(\tau)$ in the semiclassical limit. Close to threshold this solution gives a slow decay for number fluctuations,

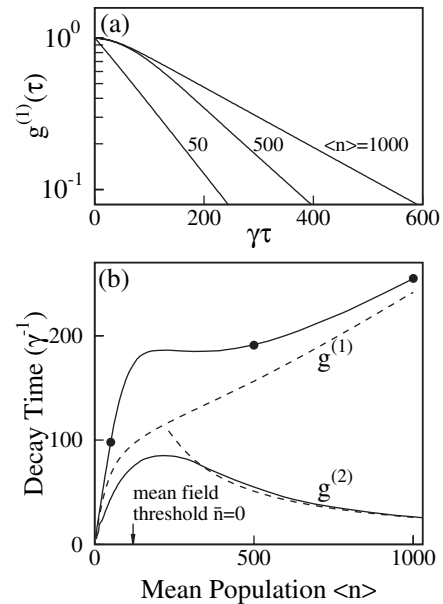


Fig. 2: (a) The decay of $g^{(1)}(\tau)$ for populations $\langle n \rangle = 50, 500$ and 1000 . On this logarithmic plot, a simple exponential is linear, while a Gaussian is quadratic. (b) Decay times for $g^{(1)}(\tau)$ and $g^{(2)}(\tau)$, as a function of population, obtained from the numerical solutions of eqs. (4) and (5) (solid lines). The non-linearity is $\kappa = 4 \times 10^{-5} \gamma$, and $n_s = 2.5 \times 10^4$, as derived from the experimental results. The dashed lines are from the analytic expressions, eqs. (7) and (9). The marked points indicate the values of $\langle n \rangle$ used in (a).

and hence explains the observed line-shape as well as the observed decay time. However, it also predicts that further above threshold number fluctuations will be much faster, and significantly longer coherence times should be obtained.

Static limit. – In our discussions we neglect spatial effects, which is justified in the CdTe system where the emission spot is strongly localised by disorder. We thus model the condensate mode as a single anharmonic oscillator, with Hamiltonian

$$H = a^\dagger a \omega_0 + \kappa (a^\dagger a)^2, \quad (1)$$

where ω_0 is the oscillator frequency, and κ the strength of the polariton-polariton interaction.

For a condensate of interacting particles, the interactions translate number fluctuations in the condensate into random changes to its energy, and so the coherence is lost. A similar effect, commonly termed “self-phase modulation”, was originally observed for laser beams propagating in a non-linear Kerr medium [15]. If we assume that the condensate has a Gaussian probability distribution for the number of polaritons, with variance σ^2 , it is straightforward to obtain $g^{(1)}(\tau)$ when the number fluctuations are sufficiently slow [4]. It has a Gaussian form

$$|g^{(1)}(\tau)| = \exp(-2\kappa^2 \sigma^2 \tau^2) = \exp(-\tau^2/\tau_c^2). \quad (2)$$

As detailed in ref. [4] we can obtain, directly from the measured data, values of $\sigma^2 \sim 25000$ and $\kappa \sim 2 \times 10^{-5} \text{ ps}^{-1}$, giving a decay time $\tau_c \sim 200 \text{ ps}$, in reasonable agreement with the experiments. The value for κ is consistent with theoretical estimates [16,17] of the interaction in a mode of linear size $\sim 5 \mu\text{m}$.

The picture described above is essentially static; it assumes that the time scale on which the number of polaritons changes, τ_r , is much longer than the coherence time τ_c , so the only relevant time evolution is caused by the action of the Hamiltonian. The obvious problem with this description is that the coherence time, τ_c , is much longer than the polariton lifetime, $\tau_0 \sim 2 \text{ ps}$, due to emission from the cavity. This suggests that the microcavity system may well not be in the quasi-static regime, and we need to consider the processes by which the number fluctuations occur in more detail.

Kubo approach. – The effect of introducing a time scale τ_r for fluctuations can be understood using the Kubo stochastic line-shape theory [6], which describes the decay of $g^{(1)}(\tau)$ for emission associated with a transition whose energy varies randomly in time. In our case, the random variations in energy are a consequence of the number fluctuations, with time scale τ_r determined by the pumping. When the random energies have a Gaussian distribution, the Kubo theory gives

$$|g^{(1)}(\tau)| = \exp\left[-\frac{2\tau_r^2}{\tau_c^2}(e^{-\tau/\tau_r} + \tau/\tau_r - 1)\right]. \quad (3)$$

For τ_r slow compared to the scale set by the variance of the random energy distribution (τ_c above), we are in the static regime and a Gaussian decay with lifetime τ_c is predicted. However, in the opposite regime, $\tau_r \ll \tau_c$, motional narrowing occurs, and the decay becomes a simple exponential, with lifetime $\tau_c^2/2\tau_r$, much longer than τ_c . If we naively take $\tau_r = \tau_0 \sim 2 \text{ ps}$, we would clearly be in the motional narrowing regime, giving an exponential function, with a very slow decay $\sim 10 \text{ ns}$. The measurement of the $g^{(1)}(\tau)$ decay thus shows that τ_r is, in fact, much longer than τ_0 , and must be comparable to, or greater than, τ_c . This prediction is very well confirmed by the measurement of the decay of $g^{(2)}(\tau)$, which directly determines the time scale for number fluctuations; the experimental decay time is $\sim 100 \text{ ps}$, similar to τ_c for the same condensate population.

The explanation for the slow decay of $g^{(2)}(\tau)$ comes from laser physics, where it is well known that number fluctuations are slowed close to the threshold. This can be explained using a simple classical model where the pumping provides a gain which is saturable, that is, has a dependence on the mode population. Above threshold, the linear part of the gain term exceeds the loss, so the population grows, and a non-linear saturation term is required to obtain a finite steady state. However, the response to small fluctuations, which is what the intensity

correlation experiment measures, depends only on the net linear gain. Thus near threshold, where the linear gain and loss are closely matched, fluctuations in the system relax very slowly. Haken [18] shows the close analogy of this behaviour to the critical slowing down of fluctuations in the vicinity of an equilibrium phase transition.

Quantum model. – To put these considerations on a more formal footing, we have developed a quantum model of the polariton condensate which can be solved analytically for $g^{(1)}(\tau)$ and $g^{(2)}(\tau)$. This model is a generalisation of one studied by Thomsen and Wiseman [9] in the context of atom lasers, extended to cover the full range of mode occupancies; their model only applies to the ‘far above threshold’ limit, where the gain is fully saturated and slowing of number fluctuations no longer occurs. The coherent mode is treated as an anharmonic oscillator with a Kerr non-linearity, eq. (1). This mode is coupled to a reservoir, using the master equation formalism for the density matrix ρ . Reservoir losses are offset by a standard laser-like saturable pump term [19]. We thus obtain equations for the population distribution, $P_n = \rho_{n,n}$, and the coherence, $u_n = \sqrt{n}\rho_{n-1,n}e^{-i\omega_0 t}$:

$$\begin{aligned} \dot{P}_n = \gamma n_c \left[\frac{n}{n+n_s} P_{n-1} - \frac{(n+1)}{(n+1)+n_s} P_n \right] \\ + \gamma [(n+1)P_{n+1} - nP_n], \end{aligned} \quad (4)$$

and

$$\begin{aligned} \dot{u}_n = \gamma n_c \left[\frac{n}{n+n_s - \frac{1}{2}} u_{n-1} - \frac{n+\frac{1}{2}}{n+n_s + \frac{1}{2}} u_n \right] \\ + \gamma [nu_{n+1} - (n - \frac{1}{2})u_n] + 2i\kappa nu_n. \end{aligned} \quad (5)$$

Here, $\frac{1}{2}\gamma = 1/\tau_0$ is the cavity decay rate, and n_c and n_s are parameters describing the pump process¹. n_c characterises the strength of the pumping, while n_s provides the saturation: for $n \gtrsim n_s$ the gain decreases. Physically, this corresponds to the depletion of the pump reservoir by the processes which populate the condensate. Far above threshold, where the mean occupation $\langle n \rangle \gg n_s$, this model becomes identical to ref. [9]. Pumping terms like this are required to give a finite condensate population, and hence should be derivable from microscopic kinetic theories. Similar gain saturation effects appear in some recent mean-field theories [16,20,21].

The steady-state solution of eq. (4) is

$$P_n^S \propto \frac{n_c^n}{(n+n_s)!} \approx \exp\left[-\frac{(n-\bar{n})^2}{2n_c}\right], \quad (6)$$

where the Gaussian form is valid when $\bar{n} = n_c - n_s \gg 1$. The variance of the population is $\sigma^2 = n_c$, so that number fluctuations in the threshold region, $\bar{n} \ll n_s$, are super-Poissonian. It is convenient to divide the threshold region

¹In the terminology of ref. [19], $\gamma = C$, $n_s = A/B$, and $n_c = A^2/B C$. We have neglected some terms $O(1/n_s)$, since $n_s \sim 10^4$ for our system.

into two: for $\bar{n} \gtrsim 3\sqrt{n_s} \sim 500$ the Gaussian form is valid for all n , because the non-physical $n < 0$ states are not significantly occupied, while for smaller values of \bar{n} these states have to be explicitly excluded. In the former case, the mean population $\langle n \rangle = \bar{n}$.

Equations (4) and (5) are easily solved numerically for populations of a few hundred particles. Figure 2b shows the decay times for the correlation functions obtained from these solutions, plotted as a function of the mean population. As the population increases, the $g^{(2)}$ decay time rises rapidly to ~ 100 ps, then decreases. The $g^{(1)}$ time also rises quickly, then flattens for a while before increasing again. Populations up to ~ 500 correspond to the experimental regime of fig. 1b, and the observed $g^{(1)}$ behaviour has a very similar form. In fig. 2a the actual decay of $g^{(1)}(\tau)$ is plotted, for three population values. For $\langle n \rangle = 50$, corresponding to the initial rise in coherence time, the form is a simple exponential. In the flat region, with $\langle n \rangle = 500$ the decay starts off Gaussian, before becoming exponential at longer delays; this is the near static behaviour of the self-phase modulation regime. The final rise occurs when the $g^{(2)}$ time shortens and motional narrowing sets in. This is evident in the decay curve for $\langle n \rangle = 1000$, which starts off with the same Gaussian as $\langle n \rangle = 500$, but much sooner becomes a slower exponential.

We now turn to deriving semiclassical analytic solutions to eqs. (4) and (5), which provide the dashed lines on the figure. These solutions are valid in the regime where $\bar{n} \gtrsim 3\sqrt{n_s} \sim 500$ and the Gaussian in eq. (6) applies without truncating the $n < 0$ part. Equation (4) for the population is independent of the non-linearity, so we can quote standard laser theory results [19] for the intensity correlation function:

$$g^{(2)}(\tau) = 1 + \frac{n_s}{\bar{n}^2} \exp\left(-\frac{\bar{n}}{n_c} \gamma \tau\right) = 1 + \frac{n_s}{\bar{n}^2} \exp(-\bar{\gamma} \tau), \quad (7)$$

where $\bar{\gamma} = \bar{n}\gamma/n_c$ is the slowed decay rate. This result fits the single experimental data point fairly well; using the experimental measurements $g^{(2)}(0) = 1.1$ and $\bar{n} = 500$ we obtain $n_s = 25000$ and $\bar{\gamma} = \gamma/50$. This gives a decay time of ~ 50 ps, in reasonable agreement with the measured 100 ps.

The first-order correlation function, $g^{(1)}(\tau) \propto \sum_n u_n(\tau) e^{i\omega_0 \tau}$, is obtained by solving eq. (5). The solution is required with an initial condition $u_n(0) = n P_n^S$, which is a similar Gaussian function to P_n^S , but with mean $\bar{n} + n_c/\bar{n}$. To obtain the correlation function, we follow the approach of Gardiner and Zoller [22]. Using a Kramers-Moyal expansion, the difference operators are converted into differentials, leading to a Fokker-Planck equation for u , which we now write as $u(n, t)$, with n a continuous variable. To deal with the appearance of n in the denominator of the pumping terms, we linearise around the mean value, writing $n = \bar{n} + n_c/\bar{n} + n'$. Thus

we obtain

$$\frac{\partial u}{\partial t} = 2i \left(\kappa + \frac{i\gamma}{4n_c} \right) n' u - \frac{\gamma}{2\bar{n}} u + \left(\gamma \frac{\bar{n}}{n_c} \right) \frac{\partial}{\partial n'} \left[n' u + \frac{1}{2} (2n_c) \frac{\partial u}{\partial n'} \right], \quad (8)$$

where we have omitted constant non-linear contributions to the oscillator energy, which can be absorbed in a renormalised ω_0 . This equation is solved in the Fourier domain [22] to give

$$|g^{(1)}(\tau)| = \exp \left[-\frac{4n_c \kappa^2}{\bar{\gamma}^2} (e^{-\bar{\gamma} \tau} + \bar{\gamma} \tau - 1) \right] \times \exp \left[\frac{n_c}{4\bar{n}^2} (e^{-\bar{\gamma} \tau} - \bar{\gamma} \tau - 1) \right]. \quad (9)$$

The first factor is just the Kubo expression, eq. (3), with $\tau_c^2 = 1/2\kappa^2\sigma^2$ and $\sigma^2 = n_c$, as before, and $\tau_r = 1/\bar{\gamma}$. This constitutes the main result of our treatment: we obtain Kubo type behaviour, with the fluctuation time τ_r given by the decay time of $g^{(2)}(\tau)$. The second term corresponds to the Schawlow-Townes decay, enhanced in the threshold region due to the finite amplitude fluctuations. It is generally much slower than the first, in the regime where the expression is valid.

Discussion. – Figure 2 shows very clearly the importance of the fluctuation time scale $\tau_r \propto 1/\bar{n}$ (for large \bar{n}) on the coherence time. When \bar{n} is increased τ_c only changes very slightly, in fact decreasing as $n_c = n_s + \bar{n}$ grows, but τ_r shortens rapidly. This pushes the system into the motional narrowing regime, where the lifetime $\tau_c^2/2\tau_r$ is proportional to \bar{n} . Note that we have to be careful treating this as a prediction of a linear relationship between coherence time and emission intensity for high powers; we have simply kept n_s constant and increased n_c . In the textbook laser model [19], n_s is indeed independent of pump power P , and $n_c \propto P$. Though pumping of the the polariton system is considerably more complex than this, it is likely that n_c increases more rapidly with P than n_s does. Thus the fluctuation decay time should decrease with stronger pumping, potentially taking the condensate into the motional narrowing regime. For this to be observable it would, of course, be necessary that other mechanisms should not take over and restrict the coherence as the number fluctuation effect is suppressed.

One surprising feature of the experiments is that very significant slowing down is still occurring at pump powers of twice the threshold value P_{th} . This is inconsistent with the simplest assumption, $n_c \propto P$, since then $\bar{\gamma}/\gamma = (1 - P_{th}/P)$ is close to one at these powers. A full understanding of this requires microscopic models of the pumping, but it may be explained by assuming that there are a limited number of reservoir states which can provide gain for the condensate, so the gain parameter n_c must become independent of P at high pumping. In a system where the maximum achievable gain only just exceeds the

loss, the reservoir must be almost full at threshold, so this saturation will extend the critical region to large values of P/P_{th} .

Instead of increasing the pump power, an alternative way of obtaining larger populations and hence longer coherence times may be to increase the size of the condensate. In the experiments the size is determined by the disorder, so this means finding larger emission spots. We present a simple argument for how the various time scales should vary with spot size, A , assuming constant density, that is $\bar{n} \propto A$. Since $\bar{n} = n_c - n_s$, this suggests $n_c \sim A^1$ and $n_s \lesssim A^1$, for large A . The polariton lifetime τ_0 is independent of A , while $\kappa \sim A^{-1}$. Together, this gives $\tau_c \sim \sqrt{A}$ and τ_r independent of A . Hence in the static regime the coherence time, which is then just τ_c , would grow as \sqrt{A} , but for sufficiently large A this will always take us in the motional narrowing regime, $\tau_c \gg \tau_r$, and the coherence time will be $\tau_c^2/\tau_r \sim A$. This argument shows that the decoherence is a mesoscopic phenomenon; as in other ordered phases such as ferromagnets and lasers, the finite condensate has a finite ergodic time, beyond which the order-parameter correlations disappear. However, in the thermodynamic limit $A \rightarrow \infty$, this ergodic time diverges, and true symmetry breaking can occur.

Conclusions. – In summary, we have shown that the available experimental results are well explained by a model of a microcavity polariton condensate as a pumped dissipative system where the main decoherence process is the combined effect of number fluctuations and inter-particle interactions. However, our model also predicts that a regime of motional narrowing should be accessible at higher pump powers, which would lead to much longer coherence times. This would make the polariton condensate much more suitable for experiments involving the manipulation of its quantum state, such as generating Josephson oscillations, ultimately leading to applications in quantum information processing.

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