Power-law decay of the spatial correlation function in exciton-polariton condensates

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We create a large exciton-polariton condensate and employ a Michelson interferometer setup to characterize the short- and long-distance behavior of the first order spatial correlation function. Our experimental results show distinct features of both the two-dimensional and nonequilibrium characters of the condensate. We find that the gaussian short-distance decay is followed by a power-law decay at longer distances, as expected for a two-dimensional condensate. The exponent of the power law is measured in the range 0.9–1.2, larger than is possible in equilibrium. We compare the experimental results to a theoretical model to understand the features required to observe a power law and to clarify the influence of external noise on spatial coherence in nonequilibrium phase transitions. Our results indicate that Berezinskii–Kosterlitz–Thouless-like phase order survives in open-dissipative systems.

Quantum well excitons | semiconductor microcavities

The spatial correlation function quantifies the coherence properties of a system (1). In a 3D Bose-condensed gas, long range order is observed, and the correlation function decays toward a plateau at large distances (2, 3). In the homogeneous 2D Bose gas (4), however, no long range order can be established (5). Instead, Berezinskii–Kosterlitz–Thouless (BKT) theory of the equilibrium interacting gas predicts a transition to a low-temperature superfluid phase, which shows a power-law decay of the correlation function (6, 7). Unfortunately, it is frequently hard to directly measure this, and only very recently (8) was indication of the power-law decay of coherence seen in a 2D atomic gas. It has been theoretically predicted (9, 10) that power-law decay of coherence survives in the nonequilibrium problem, and it is this prediction that the current experiment sets out to test.

Exciton polaritons are short-lived quasiparticles formed in a semiconductor quantum well strongly coupled to a planar microcavity (11). Each one is a superposition of a quantum well exciton and a microcavity photon, and they behave as 2D bosons below the Mott density. Above a threshold particle density, condensation is observed (12). Due to the nonequilibrium nature of polariton condensation, understanding its coherence properties is quite revealing regarding the different roles of fluctuations in the equilibrium and nonequilibrium problems.

Previous measurements on polariton condensates have demonstrated coherence at large distances but were limited by large experimental uncertainties (13) or highly disordered samples (14, 15), and the long-distance behavior could not be fully extracted. Recently, the correlation function at large distances was studied in 1D condensates confined in a quantum wire (16) and in a valley of the disorder potential (17). In ref. 16, the data was energy-resolved so that excited states were filtered out, while in ref. 17 a rare area on the sample was chosen in which a single mode condensate is seen. The purpose of both those experiments was to investigate how long the coherence length of a spectrally isolated 1D condensate state can be. We, on the other hand, are interested in the functional form of the correlation function in a 2D condensate and how the excitations populated by the pumping and decay processes can modify it.

With our setup, we can measure values of $g(1)(r)$ as low as 0.02, so we can reliably extract the long-distance behavior. We find that, although true thermal equilibrium is not established, an effective thermal de Broglie wavelength can still be defined from the short-distance gaussian decay of $g(1)(r)$. Furthermore, $g(1)(r)$ at long distances $r$ decays according to a power law, in analogy to the equilibrium BKT superfluid phase. The exponent of the power-law decay is, however, higher than can be possible within the BKT theory. We apply a nonequilibrium theory (9, 10) to identify the source of the large exponent. We argue that, although the spectrum is modified due to dissipation, the exponent would still have the equilibrium value if the spectrum was thermally populated. If, on the other hand, a white noise source acts on the system and induces a flat occupation of the excited states, the exponent can have a large value, proportional to the noise strength. We therefore conclude that the pumping and decay processes, which introduce a nonthermal occupation of the excited states, can be responsible for the large value of the exponent.

Results

In our study, we use a weak-disorder GaAs-based sample, the same one as in our recent experiments (18). The condensate is generated nonresonantly by the multimode laser, which creates free electron-hole pairs at an excitation energy approximately 100 meV above the lower polariton (LP) energy. Carriers suffer multiple scatterings before reaching the LP energy, so coherence is established spontaneously in the condensate and cannot be inherited from the laser pump. The laser is continuously on and replaces LPs that leak out of the microcavity at a ps rate. We are interested in the limit of the homogeneous 2D polariton gas. For this purpose, we employ a setup based on a refractive beam shaper that forms a large laser excitation spot with uniform intensity. There is no confining potential on our sample. Because of the short lifetime, however, the condensate density follows the photon density of the excitation spot, so we can create circular condensates with almost flat density and diameters ranging from 14 μm to 44 μm (see ref. 18 and SI Appendix). LP luminescence in the steady state is observed through a combination of a long-pass filter and a microcavity setup to characterize the short- and long-distance behavior of the first order spatial correlation function.
and a band-pass interference filter, which reject scattered laser light without distorting the LP spectrum.

We confirmed that the sample disorder potential is weak in two ways (see SI Appendix). First, the lineshape of the luminescence at low excitation power is Lorentzian, which is characteristic of a homogeneously broadened line. Second, we measured a 2D map of the disorder potential with resolution approximately 1 μm and found that its spatial fluctuations are indeed weaker than the homogeneous broadening and also much weaker than the energy shift due to polariton–polariton interactions. Therefore, we can ignore the sample disorder in our experiment. The condensate is still localized in space, though, following the shape of the laser excitation spot.

The first order spatial correlation function is defined as

$$g^{(1)}(r_1, t_1; r_2, t_2) = \frac{⟨ψ_1^†(r_1, t_1) ψ_2(r_2, t_2)⟩}{⟨ψ_1^†(r_1, t_1) ψ_1(r_2, t_2)⟩}$$

where $ψ_1^†$ and $ψ_i$ are the creation and annihilation field operators at space-time point $(r_i, t_i)$. To measure this function, we built a Michelson interferometer setup. A schematic is shown in Fig. 1A. It includes a mirror in one arm, and a right angle prism in the other. We overlap the condensate real-space image with its reflection of the prism. This allows us to measure

$$g^{(1)}(x, −x; t) = ⟨g^{(1)}(x, y, t + τ; −x, y, t)⟩,$$

where $⟨⟩$ denotes time average. In this experiment, we are mainly interested in interference at $τ = 0$, so when the time argument is not mentioned explicitly, we imply $τ = 0$.

We repeat the procedure explained in Fig. 1 for every pixel, so that we measure the phase difference between the two interfering images in addition to the correlation function across the whole spot. Representative data are shown in Fig. 2. Recording both these quantities allows us to identify useful signal from systematic or random noise. Because the prism displaces the beam that is incident on it, the images from the mirror and the prism are focused on the camera from different angles, so the two phase fronts are tilted with respect to each other. As a consequence, we expect to measure a constant phase tilt. This is the case in Fig. 2B, in which the laser power is above threshold and a condensate has formed. We conclude that our measurement of the correlation function in Fig. 2D is reliable over this whole area. On the other hand, at a pump rate below threshold, only short-range correlations exist. Fig. 2C shows that in this case the phase difference is measured correctly only over a small area around the center. The measured values of $g^{(1)}(x, −x)$ outside this area are not reliable and give an estimate of our measurement uncertainties. As is clear from Fig. 2C, the experimental error can be suppressed down to 0.01.

Phase maps such as those in Fig. 2 have been used to identify localized phase defects—namely, quantum vortices (19). The data of Fig. 2B show that such localized defects are not present in our sample. At points with large fringe visibility (near $x = 0$ μm), fringes are perfectly parallel, whereas defects that appear for large $|x|$ could be due to a numerical uncertainty in the measurement of the local phase due to the small fringe visibility. In any case, localized stationary phase defects cannot influence $g^{(1)}(r)$, because it is their motion that destroys spatial correlations and not their mere presence. It has been found that vortices appear in large disorder samples (19), when a direct external perturbation is introduced (20), before the condensate reaches its steady state (21), or when the condensate moves against an obstacle (22, 23). None of these conditions is satisfied in our experiment. On the other hand, we have found that, under the same conditions as the current experiment, mobile bound vortex pairs appear spontaneously due to the special form of the pumping spot and the pumping and decay noise (18). In ref. 18, we found that a single mobile bound vortex–antivortex pair is visible in a small condensate. In the current experiment, we probe larger condensate sizes, so it is likely that several vortex pairs are present at the same time. Mobile bound vortex pairs are in general invisible in time-integrated phase maps, like the one in Fig. 2B, and they are consistent with a power-law decay of $g^{(1)}(r)$.

Fig. 3 shows the short-distance dependence of $g^{(1)}(x, −x)$ for the same pumping power as in Fig. 2A and C. Every dot in Fig. 3A corresponds to one pixel on the camera, and the $x$ axis is its distance from the axis of reflection (slightly tilted with respect to the columns of the charge-coupled device array). Data at distances $|x| > 1$ μm is noise, because the measured phase in this area is random (Fig. 2A). At shorter distances, we can measure $g^{(1)}(x, −x)$ reliably, and we find that the correlation function has a

![Fig. 1](https://www.pnas.org/cgi/doi/10.1073/pnas.1107970109 Roumpos et al.)
gaussian form. This is the same functional dependence as for a thermalized Bose gas when the temperature is sufficiently high or the density sufficiently small (2, 4). In that equilibrium case, the width of the gaussian decay is proportional to the thermal de Broglie wavelength. Although our nonequilibrium system is quite different than the thermalized Bose gas, we will use this analogy to define a thermal de Broglie wavelength. For an insufficiently thermalized system, it is quite possible that excitations in different energy profiles of the high-energy states involved in producing the condensation threshold is at approximately 55 mW. The measured $g^{(1)}(x, -x)$ corresponding to $A$ and $B$, respectively, averaged over the $y$ axis inside the excitation spot area of 19-μm radius. Blue circles are experimental data. The continuous red and dashed yellow fitting lines are explained in Figs. 3 and 6, respectively.

Fig. 2. Phase map measured for laser power (A) below and (B) above the threshold power $P_{th}$. The prism in the Michelson interferometer is oriented horizontally. The schematics on the top right of $A$ and $B$ show the orientation of the two interfering images. (C) and (D) Measured $g^{(1)}(x, -x)$ corresponding to $A$ and $B$, respectively, averaged over the $y$ axis inside the excitation spot area of 19-μm radius. Blue circles are experimental data. The continuous red and dashed yellow fitting lines are explained in Figs. 3 and 6, respectively.

Power decay with no obvious threshold, analogous to the theory of equilibrium noninteracting 2D Bose gas as the particle density is increased (4). We performed the same experiment for two orthogonal prism orientations as shown in the legend of Fig. 3B. In one case we measured $g^{(1)}(x, -x)$, whereas in the other case we measured $g^{(1)}(y, -y)$. We found that $\lambda_{eff}$ is shorter along the $y$ axis and attribute this difference to a small asymmetry of the laser pumping spot. The occupation of excited states (which determines $\lambda_{eff}$) depends on the spatial overlap with the laser pumping spot, so states of equal energy are not always equally populated. This asymmetry shows that $\lambda_{eff}$ is simply related to the cryostat temperature and depends on the spatial and energy profiles of the high-energy states involved in producing this correlation length. We also note that the resolution limit of our imaging setup is approximately 1 μm, hence the measurement of $\lambda_{eff}$ at small pumping power is resolution-limited.

It is known that an ideal autocorrelation measurement with a Michelson interferometer provides the same information as an ideal measurement of the spectrum. In particular, $g^{(1)}(x, -x; t)$ is the Fourier transform of the power spectrum in momentum space $S(k, \omega)$ (24). However, systematic noise in measurement of $S(k, \omega)$ currently makes the direct measurement of $g^{(1)}(x, -x; t)$ the only way to reliably extract $\lambda_{eff}$ of Fig. 3B as well as the power-law decay at long distances to be explained later. The Fourier transform relationship between $g^{(1)}(x, -x; t)$ and $S(k, \omega)$ is illustrated in Fig. 4. The measured $g^{(1)}(x, -x; t)$ at very low pumping power is shown in Fig. 4A. At time delay $t = 0$, it has a gaussian form as a function of $x$, but for increasing $t$ it broadens and acquires a multipeak structure. This unusual space-time dependence is reproduced by the numerical Fourier transform (Fig. 4C) of measured $S(k, \omega)$ (Fig. 4B). As explained in SI Appendix, measurement of the time dependence of $g^{(1)}(x, -x; t)$ is limited by inhomogeneous broadening due to time-integrated data, so it cannot provide an estimate of the homogeneous dephasing time. At long distances, the behavior of the correlation function at zero time delay $t = 0$ is no longer gaussian. We found that it is
influenced by the edge of the condensate. In Fig. 5, we plot the measured momentum-space spectrum $S(k_x, k_0)$ for very low pumping power. As explained in the text, $g^{(1)}(x, t)$ is the Fourier transform of $S(k_x, k_0)$. (C) Fourier transform of the experimental data shown in B. The result indeed reproduces accurately A. In B and C, the data is plotted in linear color scale in arbitrary units.

We note that the condensate size is slightly smaller than the pump laser spot radius (3–4 μm smaller from each side for a large spot). Because of the repulsive interaction between polaritons, and between polaritons and reservoir excitons, the large density of the condensate and reservoir creates an antitrapping potential that pushes LPs away from the center. This effect is stronger for a gaussian or a very small pumping spot and in long-lifetime samples (16, 25), whereas in the present experiment it only influences LPs that are close to the edge.

In the case of a large condensate, we should recover the limit of (infinitely large) homogeneous polariton gas. Therefore, we consider a pumping spot radius $R_0 = 19$ μm. In Fig. 6A, we plot the correlation function $g^{(1)}(\Delta x)$ versus $\Delta x$ as the pumping power is increased. Only short-range correlations exist for small pumping power, whereas above the condensation threshold of approximately 55 mW (4.8 kW/cm²), substantial phase coherence appears across the whole spot. The functional form of the long-distance decay is measured to be a power law over about one decade, as can be seen in Fig. 6B, in which we plot the data at one specific laser power. We fit the data to a function $g^{(1)}(\Delta x) = (\lambda_p/\Delta x)^{\alpha}$ and plot the exponent $\alpha$ as a function of pumping power in Fig. 6C. It is found to be in the range 0.9–1.2. $\lambda_p$ is a parameter with units of length and is not related to $\lambda_{\text{eff}}$, which is plotted in Fig. 3B*

It has been claimed that a criterion for polariton condensation is the appearance of a second threshold as the pumping power is increased (26, 27). The state after the first threshold was called a “polariton BEC,” “polariton laser,” or “polariton condensate,” whereas the state after the second threshold has not been fully understood yet. It might be a Bardeen–Cooper–Schrieffer (BCS) crossover (28, 29), photon BEC (30), or photon laser (26). This double threshold behavior has been observed in micropillar structures (26), and using a stress trap (27). In the supplementary information of ref. 18, we also reported the observation of double threshold using the same sample and excitation conditions as in the present experiment. We found that in our sample the window of intensities between the two thresholds is not very wide, and can only be witnessed using a flat laser excitation spot. This spot creates a uniform polariton density over a large area, as opposed to the more common gaussian spot, where the density changes a lot across the pumping spot.

Finally, we repeated the same measurement of $g^{(1)}(x, \sim x)$ using an identical sample at a temperature of 200 K. Because of the small binding energy, the GaAs excitonic effect is weak at this temperature. Also, the lasing energy was well above the bandgap. Therefore, only standard photon lasing was possible. In this case, we only found exponential decay of the correlation function and no power law. The details of this measurement are reported in SI Appendix. This suggests that the interactions of the strongly coupled exciton-polaritons are essential in the observation of the reported phenomena.

*See SI Appendix for a discussion of $\lambda_p$ data at different detunings, the orthogonal prism orientation, as well as for time-resolved data.
Discussion
In ref. 31, it was found that excitation with a low-noise single mode laser revealed the formation of multimode condensation, and the different condensate modes could be spectrally separated. The authors of ref. 31 argued that, if one wants to measure the intrinsic linewidth of polariton condensates, single mode laser excitation and energy-resolved data are required. Indeed, it has been shown (32) that the temporal coherence properties can be understood based on the idea that laser intensity noise introduces population fluctuations, which modulate the interaction energy accordingly, leading to decoherence. However, it is not clear how pump and decay noise influences spatial coherence. Under single mode laser excitations, and when the lowest-energy state is spectrally isolated, long coherence lengths can be observed (15–17, 33). In the current experiment, we are interested in how robust spatial correlations are when excitations are included. We study the “worst case” scenario of multimode laser excitation, which gives broader spectra compared to single mode laser. However, as shown in SI Appendix, single mode laser excitation gives similar results in energy-integrated data. We note that laser phase noise cannot be an issue in our experiment, because the laser energy is approximately 100 meV above the LP energy, so the generated quasiparticles suffer multiple scatterings before forming the condensate.

Focusing on the multimode laser excitation case, let us now explore the interpretation of the power law that we observe and consider what it means for the properties of the nonequilibrium polariton condensate. In particular, we discuss under what conditions a power-law decay should be seen and what may control the value and pump power dependence of the observed exponent. As has been discussed previously (9, 10, 34), power-law decay of spatial correlations are not an artifact of equilibrium condensates but survive more generally in a nonequilibrium condensate. Because the power-law decay at long distances arises from the long wavelength collective modes, this statement is not trivial, because dissipation can modify the spectrum at long wavelengths (9, 10, 34).

Let us first recall the results that would apply if one were to consider an equilibrium interacting 2D Bose gas. In this case, the exponent is given by \( a_0 = 1/n_s \lambda^2 \leq 1/4 \), where \( n_s \) is the superfluid density and \( \lambda \) is the thermal de Broglie wavelength \( \lambda = \sqrt{2\pi\hbar^2/mk_BT} \). The restriction \( a_0 < 1/4 \) occurs because increasing temperature has two effects: It excites long wavelength phase fluctuations, which are responsible for the power-law decay, and it can also excite vortex pairs. The maximum value of \( a_0 \) occurs at the transition when vortex–antivortex pairs unbind, so that vortices would proliferate, and cause the BKT transition to a phase with short-range correlations. The observation here of a power law \( a_0 > 1/4 \) implies that effects beyond thermal equilibrium are required to explain the data; i.e., there is noise that excites phase fluctuations without leading to vortex proliferation. In addition, because the equilibrium exponent \( a_0 \propto 1/n_s \), one would expect the exponent to decrease with pump power, as the condensate density increases; the absence of such a decrease again implies effects beyond thermal equilibrium are relevant and suggests that pumping noise is indeed affecting the observed exponent.

Whereas the existence of power-law decay in a nonequilibrium condensate was discussed previously, the value of the exponent and its pump power dependence were not given in those previous works. Using the formalism described in refs. 9 and 10, the exponent can be found by calculating

\[
g_t(\Delta \gamma) \propto \exp \left[ \int \frac{d^3 k}{(2\pi)^3} \left( 1 - e^{i \Delta \gamma f(k)} \right) \right] \tag{4}
\]

where \( f(k) = \int \frac{d\omega}{2\pi} \mathcal{W}(D^K_{\phi \phi} - D^R_{\phi \phi} + D^A_{\phi \phi}) \) and \( D^K_{\phi \phi}(k, \omega) \) are the Keldysh, retarded, and advanced Green’s functions for phase fluctuations. The advantage of writing the correlation function in this formal way is that it allows one to disentangle the effects of changes to the spectrum of long wavelength excitations from the effects of how this spectrum is populated.

The retarded and advanced Green’s functions are independent of how the spectrum is occupied, and following quite general arguments (9, 10, 34) we can prove that they have poles describing the low energy spectrum \( \omega_k \approx -i\gamma + \sqrt{\epsilon_k(k + 2\mu) - \gamma^2} \) where \( \epsilon_k = h^2k^2/2m \) is the long wavelength polariton dispersion, \( \mu \) the chemical potential (or blueshift) and \( \gamma \) is the linewidth. Despite the modification of the equilibrium spectrum introduced by \( \gamma \), it nevertheless remains the case that if this spectrum is occupied thermally (i.e., if the Keldysh Green’s function is chosen to obey the equilibrium fluctuation dissipation theorem), one finds

\[
f_{\text{thermal}}(k) \approx \frac{1}{n_s} \int \frac{d\omega}{2\pi} \frac{4\gamma k_BT}{\omega^2 + 2\gamma \omega - 2\mu \epsilon_k} \approx \frac{mk_BT}{n_s h^2 k^2}. \tag{5}
\]

which is independent of \( \gamma \) and matches the equilibrium form of \( f(k) \). Thus, despite the modifications to the long wavelength spectrum, a sufficiently thermalized polariton condensate has the equilibrium exponent.

In order to explain the larger exponent observed, and the flat dependence on pump power, we consider a crude model of a system with excess pumping noise as an opposite extreme to the thermalized case. We thus consider a case where the occupation of excitations is set by a Markovian noise source of strength \( \zeta \). Namely, we take the inverse Keldysh Green’s function to be energy independent. This differs significantly from thermal noise correlations, which are frequency dependent, and diverge at the chemical potential. The measured spectra shown in SI Appendix are broad and their linewidth increases as the pumping power is increased. This occupation of excited states could be induced by an energy-independent noise source whose strength increases with the pumping power. In this case, the function \( f(k) \) is given by

\[
f_{\text{noise}}(k) \approx \frac{1}{n_s} \int \frac{d\omega}{2\pi} \frac{\zeta (\mu^2 + \gamma^2)}{\omega^2 + 2\gamma \omega - 2\mu \epsilon_k} \tag{6}
\]

which, despite the changed occupation spectrum, still yields a power-law decay. The exponent becomes \( a_0 = (m\zeta/2\hbar^2 N_m^{-1}) (\mu^2 + \gamma^2)/2\mu \). Because this has the form \( a_0 \propto \zeta n_s \), then if the noise strength and polariton density both increase with the pump power, then this would explain the absence of a \( 1/n_s \) decrease of the exponent, as seen on Fig. 6C.

One point not addressed so far regards the process of vortex proliferation in a noisy nonequilibrium condensate. As pump noise increases, it is likely eventually to lead to proliferation of vortices, and a transition to a state with only short-range correlations, just as occurs at high temperatures in equilibrium.

In conclusion, the measured power-law decay of the correlation function suggests that some form of the BKT superfluid phase survives in nonequilibrium condensates; namely, phase fluctuations are excited but no vortices. The large value of the exponent implies that, in the current experiment, this ordered phase is more robust against external noise than would be expected in equilibrium, in which equipartition holds. We conjecture that the main noise source is pump and decay noise, which create a nonthermal occupation of excited states, and apply a nonequilibrium theory to show that a power-law decay with a large exponent is possible in an open system with excess noise. One may anticipate that sufficient noise could induce vortex proliferation and a transition to short-range coherence. This

\[^5\text{See SI Appendix for further details.}\]
fascinating possibility remains an open question for future studies.

Materials and Methods

Our GaAs-based sample shows a Rabi splitting of $2\Delta \Omega_{\text{Rabi}} = 14$ meV and LP lifetime of $\tau_L = 2 - 4$ ps near photon-exciton detuning $\delta = 0$, where the data presented in this paper is taken. From the curvature of the measured energy versus momentum dispersion at low pumping power, the LP effective mass was found to be $m^* = 9.5 \times 10^{-5} m_e$ at this detuning, where $m_e$ is the electron rest mass. The sample is the same as in our recent experiments (18, 33), and the experimental setup is very similar to ref. 18. We pump the system with a multimode Ti:Sapphire laser operated in the continuous wave mode, combined with a chopper that creates 0.5-ms pulses at 100-Hz repetition rate. All powers quoted in the text and SI Appendix refer to the unchopped laser beam. We employ a commercial refractive beam shaper to generate a flat-top pumping profile of varying size. The Michelson interferometer consists of a 50-50 nonpolarizing cube beamsplitter, a dielectric mirror in the first arm, and an uncoated glass right angle prism in the second one. The position of the prism is controlled by a combination of a translation stage and a piezoelectric actuator.

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