Formation of a macroscopically extended polariton condensate without an exciton reservoir

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We report a record-size polariton condensate of a fraction of a millimeter. This macroscopically occupied state of macroscopic size is not constrained to the excitation spot and is free from the usual complications brought by high-energy reservoir excitons, which strongly alter the physics of polaritons, including their mobility, energy distribution and particle interactions. The density of this trap-free condensate is lower than 1 polariton/µm², reducing the phase noise induced by the inter-polariton interaction. Experimental findings are backed up by numerical simulations using a hydrodynamic model which takes into account both the polariton expansion and the phonon-assisted relaxation towards the lowest energy state. These results propel polariton condensates at the fundamental level set by their cold-atomic counterparts by getting rid of several solid-state difficulties, while still retaining their unique driven/dissipative features.

Under suitable conditions, light-matter interaction can be strong enough to drive the coherent exchange of energy between photonic and electronic modes \[^1\]. This is the paradigm of microcavity exciton-polaritons: quasiparticles created by the strong coupling between the photonic mode of a microcavity and the excitonic transition of a semiconductor quantum well \[^2\]. Polaritons manifest their composite nature with a combination of photonic and excitonic properties \[^3\]. Thanks to their photonic component, polaritons can ballistically propagate in the plane of the microcavity with velocities up to a few percent of the speed of light \[^4\]. On the other hand, the exciton component results in strong optical nonlinearities and induces an energy renormalization of the polariton dispersion at high densities \[^5\]. This energy shift can be much larger than the linewidth and is at the foundation of most polaritonic effects and applications \[^6\]–\[^8\]. As bosonic quasiparticles, polaritons experiment final-state stimulated scattering, which results, above a density threshold, in a laser-like emission without population inversion, a collective phenomenon that is explained in the framework of Bose-Einstein condensation \[^9\]–\[^11\]. A unique feature of polariton condensates is their driven/dissipative nature, in which the steady state is reached through a dynamical balance of pumping and dissipation.

Polariton condensates have been experimentally observed in different materials, both inorganic \[^12\]–\[^15\] and organic semiconductors \[^16\]–\[^17\] and thanks to their light mass, condensation is achieved also at room temperature \[^18\]. However, differently from their atomic counterpart, these condensates suffer from dephasing and density fluctuations induced by the interactions with the exciton reservoir, effectively resulting in multimode condensates \[^19\]–\[^21\]. The exciton reservoir also acts as a trapping mechanism, if the polariton lifetime is too short, confining the condensation process within the region of the excitation spot \[^22\]–\[^24\]. Moreover, polariton condensation is often localized in potential minima caused by imperfections of the sample structure, yielding to a fragmentation of the phase coherence \[^26\]–\[^30\]. All these aspects of polariton condensates are not welcomed as they blur the fundamental character of the phenomenon by disrupting it with technical impediments. These become obstacles for prospective applications with polaritons, such as investigating out-of-equilibrium phase transitions or to implement simulation and related devices \[^31\]. On the other hand, confinement of polaritons in one dimensional structures provides a striking evidence of the mechanism of expulsion and acceleration of polariton condensates far from the exciton reservoir \[^32\]–\[^35\]. In two-dimensional (2D) structures, interferences from scattering potentials, effects of laser-induced confinement and the wedge in the microcavity thickness, add additional difficulties in achieving extended and uniform condensates, even in samples with long polariton lifetimes \[^36\]–\[^39\].

In this work, we use a high quality 2D microcavity without spatial inhomogeneities, and observe the forma-
tion of a condensate that extends much beyond the laser spot region. Photoluminescence measurements are employed to demonstrate the expansion of polaritons from the excitation spot and the subsequent relaxation into the lowest energy level at the bottom of the polariton dispersion. The extended condensate is formed thanks to two main ingredients: the high homogeneity of the sample, which avoids localization effects, and the long radiative lifetime (∼100 ps), that allows the propagating polariton to relax into the ground state. Remarkably, condensation occurs at low densities (∼0.1 polariton/μm²), without the presence of the exciton reservoir, and covers an area of more than 0.03 mm². The energy-resolved spatial profiles obtained by solving a theoretical model, which combines the hydrodynamics of the expanding polaritons with their energy relaxation, reproduce the experimental results and confirm that, above a threshold power, phonon-mediated scattering into the lowest energy mode is effective in forming an extended 2D polariton condensate.

The sample used in this study is a high quality-factor 3/2 λ GaAs/AlGaAs planar cavity containing 12 GaAs quantum wells placed at three anti-node positions of the electric field. The front (back) mirror consists of 34 (40) pair of AlAs/Al₀.₂Ga₀.₈As layers. The Rabi splitting is of 16 meV and it is excited close to the zero detuning condition. Photoluminescence measurements are performed under non-resonant excitation with a low-noise, narrow-linewidth Ti:sapphire laser with stabilized output frequency to reduce the fluctuations in the exciton reservoir. The laser is focused on the sample in a spot with a Gaussian intensity profile of FWHM = 20 μm. The sample emission is collected and imaged on the entrance slit of a streak camera coupled to a spectrometer in order to measure the time-, energy- and space-resolved polariton dynamics.

Under nonresonant excitation, a high density of excitons accumulate within the region of the pumping spot, inducing a blueshift of the polariton energy proportional to their repulsive interaction strength. Outside the optically pumped area, the density of uncoupled excitons decreases quickly in space, due to the small exciton mobility (2–5 microns), and the polariton energy recovers the linear regime. The Gaussian profile of the exciting beam is roughly reproduced by the potential landscape, evidenced in Fig. 1(a) by a dashed, white line. In Fig. 1(a), the emitted intensity is energy- (vertical axis) and spatially- (horizontal axis) resolved along one direction passing through the center of the excitation spot. The high-energy polaritons sitting at the top (4 meV above the bottom energy) and formed at the center of the laser spot, expands radially outwards with a large in-plane wavevector (k = 2.2 μm⁻¹), as can be seen in the cross-section of the lower polariton dispersion (LPB, energy distribution in momentum space) shown in Fig. 1(b). The macroscopic occupation of the lowest energy mode (k=0), visible at the bottom of the LPB in Fig. 1(b), corresponds to the condensation outside of the spot region in Fig. 1(a). At the same time, also lower energy states along the whole dispersion (k ≤ 2.2 μm⁻¹) are occupied and expand. The high spatial homogeneity of the sample grants a uniform expansion of the polariton gas, as shown in the two-dimensional, energy-filtered space map shown in Fig. 1(c).

In order to study the polariton dynamics, the steady state, populated through the continuous wave (CW) pump laser, is perturbed by focusing an additional 100 fs pulsed beam on top of the CW laser (both lasers are tuned to nonresonantly excite the system at the first minimum of the mirrors’ stop band). The repetition rate of the pulsed beam is slow enough to allow the system to recover its steady state condition before the arrival of the next pulse. The evolution of the additional polaritons injected by the pulse is recorded with a time resolution of 10 ps.

By extracting the space-time images at different energies, as shown in Fig. 2(a) for E = 2.6 meV, the expansion velocity as a function of energy can be estimated from the slope of the emission intensity. These velocities are compared in Fig. 2(b) to the group velocities calculated from the LPB dispersion and the difference is indicated by the red-filling region, showing that the effect of relax-
The bare propagation of low-speed polaritons. This is the first time that such a behavior (fluid passing a defect) is recorded for a continuum of states, suggesting that, at higher densities, a new class of experiments on polaron superfluidity could be performed on expanding clouds free from reservoir artifacts and involving macroscopic distances. Remarkably, the polaron density outside of the pumped region, at the bottom of the LPB, manifests a nonlinear increase as a function of the pumping power for extremely low density values, as shown in Fig. 4(a). At the same time, the formation of the bottom condensate is marked by a narrowing of the linewidth, as shown in Fig. 4(b). The low density of the extended condensate dwindles the effects of polaron-polariton interaction, reducing the intrinsic dephasing of the condensate and making this configuration appealing both for applications and for future investigations on phase transition dynamics in polaron systems. In Fig. 4(c), the ratio between the density in the lowest energy state and the whole expanding cloud is compared at different excitation powers, showing a nonlinear increase at the condensation threshold.

A theory joining hydrodynamics and relaxation of an expanding and relaxing condensate can be developed by combining the mean field description of the dynamics given by the Gross-Pitaevskii equation (GPE) with the rate equations that account for stimulated scattering due to the interaction with a phonon
bath [33,43]. In order to obtain a simplified differential equation describing the steady-state polariton distribution in energy and space, we adapt the recent approach that merges both components of the dynamics at a level of the description that involves only the polariton density [44]. We study the dynamics outside of the pumping spot, without the presence of an exciton reservoir and with low polariton densities, allowing to ignore the nonlinear term, so that the GPE reads

\[ \hbar \partial_t \psi(r) = \left( E_0 - \frac{\hbar^2}{2m} \nabla^2 - \frac{\hbar^2 v^2}{2} \right) \psi(r) \]

where \( E_0 \) is the energy corresponding to the bottom of the lower branch, \( \gamma \) is the decay rate and we have assumed cylindrical symmetry. A continuity equation, relating spatial and temporal fluxes, is derived from the GPE for each polariton energy through spectral expansion:

\[ \frac{\partial}{\partial t} n_\omega \bigg|_{\text{exp}} = -\nabla \cdot (n_\omega v_\omega) - \gamma n_\omega \]  

where \( v_\omega \) is the velocity field of the condensate [45].

On the other hand, a rate equation is written for phonon-mediated stimulated scattering, coupling the different spectral modes:

\[ \frac{\partial}{\partial t} n_{\omega'} \bigg|_{\text{relax}} = -\int \left( W_{\omega',\omega} + W_{\omega',\omega} \right) n_\omega n_{\omega'} d\omega' \]  

where \( W_{\omega',\omega} \) is the scattering rate from a state with energy \( \omega \) to another with energy \( \omega' \). Combining the two, and taking the steady state solution, we derive a differential equation for the polariton density in energy and space, \( \partial_t n_\omega(r) \bigg|_{\text{relax}} + \partial_t n_\omega(r) \bigg|_{\text{exp}} = 0 \), that takes into account the relaxation and radiative lifetime. The equation is numerically solved outside of the pumping spot, with \( \sigma \) the spot radius and \( r = 0 \) the center of the spot, assuming as boundary initial condition for each energy the experimentally obtained polariton density at \( r = \sigma \), and then solving for \( r > \sigma \). The results of the calculation for two different conditions, with and without the presence of a phonon bath, are shown in Fig. 5(a) and Fig. 5(b) respectively. While Fig. 5(a) reproduces the formation of an extended polariton condensate at the bottom of the dispersion, in Fig. 5(b) the small polariton velocity dominates the dynamics close to \( k = 0 \) and precludes the observation of an extended 2D polariton condensate.

In conclusion, the formation under non-resonant pumping of an extended 2D polariton condensate at the bottom of the LPB has been experimentally demonstrated. The polariton energy under the laser spot is blueshifted with respect to the lowest energy outside of the laser spot. This produces an expanding flow of polaritons at the energy of the blue-shift, but also a continuum of states along the dispersion, which are formed through relaxation and which expand with decreasing velocities for decreasing energies. Thanks to the high quality of the sample, we are able to observe the relaxation until the bottom of the LPB. Above a threshold density, a condensate forms at \( k = 0 \) at the lowest available energy, covering a spatial region larger than 0.03 mm² around the pumping spot. These results are well reproduced by a simple theoretical model that captures the physics of expansion and relaxation of higher energy polaritons. Our findings provide the closest realization so far of a textbook Bose–Einstein condensate in a solid-state matrix, with macroscopic sizes and dilute, tunable densities. Such systems should allow to truly take advantage of polariton condensates for fundamental research of out-of-equilibrium quantum dynamics.
Acknowledgments

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See details in the Supplemental Material.
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Supplemental Material

Polariton Lifetime

In order to determine the polariton lifetime, time-resolved measurements are performed using a Ti:sapphire laser delivering 100 fs pulses with a 82 MHz repetition rate. This beam resonantly injects polariton inside the cavity by tuning the frequency and angle of incidence of the laser in order to match the LPB close to the bottom energy, \( E_{k=0} = 772.85 \text{ nm} \), but slightly shifted in angle to allow the filtering out of the reflected light. In this way, the bottleneck effect is limited and the polariton lifetime can be extracted from the polariton emission. This is visible from the exponential decay shown in Fig. S1. From the exponential decay in time of the population it is possible to extract a lifetime of 100 ps.

![Figure S1](image)

Figure S1: (a) Time resolved emission taken in a small bound of \( k = 0 \) and with the laser pump at \( E_{k=0} = 771 \text{ nm} \). (b) Cross Section of the time resolved decay emission in the region within the dashed, red lines in panel (a). The exponential decay fitting gives a decay of 100 ps (red line).

Time-Resolved Dynamics

In Fig. S2, four snapshots are shown with the emission intensity as a function of energy (vertical axes) and space (horizontal axes) at different delay times (42 ps, 66 ps, 77 ps and 104 ps after the arrival of the non-resonant pulse in Fig. S2(a), Fig. S2(b), Fig. S2(c) and Fig. S2(d), respectively). Polaritons at high energy quickly expand and relax into lower energy states. The expanding flow is apparent in the space-energy emission map shown in Fig. S2(b), and it is more effective for the lower energy states (Fig. S2(c)-(d)). An accumulation points, induced by the pulse spot width being slightly larger than the CW one, allows the observation of a characteristic "whiplash" of relaxation in the space-energy map. Eventually, at longer times, polaritons relax into the bottom of the LPB, as shown in Fig. S2(d).

Theoretical description

We consider a mean field description of the dynamics of the condensate wave function \( \psi(r) \) given by the Gross-Pitaevskii equation [S2]

\[
\frac{i\hbar}{\partial t} \frac{\partial \psi(r)}{\partial t} = \left\{ E_0 - \frac{\hbar^2}{2m} \nabla^2 r + \frac{i\hbar}{2} \left[ R[n_R(r)] - \gamma \right] + V(r) \right. \\
+ \frac{\hbar g}{2} |\psi(r)|^2 \left. \right\} \psi(r) \\
(S1)
\]

Here, \( R[n_R(r)] \) describes a coupling to the excitonic reservoir \( n_R(r) \), that has its own dynamics:

\[
\dot{n}_R(r) = P(r) - \gamma_R n_R(r) - R[n_R(r)]|\psi(r)|^2 \\
(S2)
\]
In this work we are interested on the expansion and relaxation dynamics outside the excitation spot. The excitonic interaction with the reservoir produces a repulsive potential that can be described by $V(r)$ and that tends to expel the polaritons from the spot where they have been created: we will study the dynamics of these polaritons outside the spot and, due to the low densities, ignore the nonlinearity. Therefore, in the our region of interest, $|r| > \sigma$ ($\sigma$ being the size of the spot, i.e., the region where $n_R(r)$ and $V(r)$ are zero) the hydrodynamical description of the polariton flow is given by:

$$i\hbar \frac{\partial \psi(r)}{\partial t} = \left\{ E_0 - \frac{\hbar^2}{2m} \nabla^2 r - i\hbar \frac{\gamma}{2} \right\} \psi(r). \quad (S3)$$

Since the mechanism that creates these polaritons take place for $|r| < \sigma$, the polaritons created by the pumping terms in Eq. (S1) must be accounted by an appropriate boundary condition in Eq. (S3).

Let’s consider now a time-dependent spectral expansion of the wavefunction as follows:

$$\psi(r,t) = \sum_\omega \psi_\omega(r,t)e^{i\omega t} \quad (S4)$$

where we assume that $\psi_\omega(r,t)$ evolves in time much slower than the oscillations given by $e^{i\omega t}$. Next, we write $\psi_\omega(r,t)$ in terms of density and phase:

$$\psi_\omega(r,t) = \sqrt{n_\omega(r,t)}e^{i\phi_\omega(r,t)} \quad (S5)$$

By plugging this ansatz in Eq. (S3) and taking the imaginary part, we find:

$$i\hbar \sum_\omega e^{i(\omega t + \phi_\omega)} \left\{ \frac{\partial}{\partial t} \sqrt{n_\omega} + i(\omega + \frac{\partial}{\partial t} \phi_\omega) \sqrt{n_\omega} \right\}$$

$$= -\frac{\hbar^2}{2m} \sum_\omega e^{i(\omega t + \phi_\omega)} \left[ \nabla^2 (\sqrt{n_\omega}) + 2i \nabla \phi_\omega \nabla (\sqrt{n_\omega}) \right]$$

$$+ i\sqrt{n_\omega} \nabla^2 \phi_\omega - (\nabla \phi_\omega)^2 \sqrt{n_\omega} - \frac{\hbar}{2} \sum_\omega e^{i(\omega t + \phi_\omega)} \sqrt{n_\omega} \quad (S6)$$
Now, we equate the terms in the sum with the same $\omega$ and take the imaginary part of this equation to get:

$$\frac{\partial}{\partial t} \sqrt{n_\omega} = -\frac{\hbar^2}{2m} (2i \nabla \phi_\omega \nabla (\sqrt{n_\omega}) + i \sqrt{n_\omega} \nabla^2 \phi_\omega) - \frac{\hbar}{2} \gamma \sqrt{n_\omega}$$ (S7)

which we can rewrite as an energy-resolved continuity equation:

$$\frac{\partial}{\partial t} n_\omega \bigg|_{\text{exp}} = -\nabla (n_\omega \mathbf{v}_\omega) - \gamma n_\omega$$ (S8)

where

$$\mathbf{v}_\omega = \frac{\hbar}{m} \nabla \phi_\omega.$$ (S9)

The suffix in the partial derivative accounts for the fact that this equation only describes the hydrodynamics of expansion of the polariton fluid. We now describe the variation in $n_\omega$ due to the relaxation to lower energy modes mediated by phonon scattering.

A rate equation is usually written in $k$ space, and if we take into account only stimulated scattering, it reads as [S1]:

$$\frac{\partial}{\partial t} n_k \bigg|_{\text{relax}} = -\sum_{k'} |W_{k,k'} n_k n_{k'} + (k \leftrightarrow k')|$$ (S10)

If the scattering is mediated by a phonon bath, Fermi’s golden rule gives the following transition rate in $k$ space:

$$W_{k,k'} \approx \frac{L_z (\chi k' k \Delta \tilde{E}_{k,k'})^2}{h \rho V u^2 q_z} B^2(q_z)|D_e - D_h| \times$$

$$|n_{\text{ph}}(\omega_{k'} - \omega_k)| \theta(\Delta \tilde{E}_{k,k'} - |k - k'|)$$ (S11)

where $u$ is the longitudinal sound velocity, $L_z$ is the quantum well width, $V$ is the crystal volume, $\rho$ is the mass density of the solid, $\Delta \tilde{E}_{k,k'} = |\omega_{k'} - \omega_k|/hu$, $q_z$ is, for a given in plane momentum change $k - k'$, the momentum in the $z$ direction that must be taken by the phonons for the scattering process to conserve energy $q_z = \sqrt{\Delta \tilde{E}_{k,k'}^2 - |k - k'|^2}$, $|n_{\text{ph}}(\omega)|$ is the absolute of the phonon density and $B(q)$ is given by:

$$B(q) = \frac{8\pi^2}{L_z q (4\pi^2 - L_z^2 q^2)} \sin \left(\frac{L_z q}{2}\right).$$ (S12)

Since the experiment is performed on a polar symmetry, we can assume that $n_k$ depends only on the module of $k$:

$$n_k = n_k$$ (S13)

and we can perform the sum in $k'$ in [S10] going to the continuum limit $\sum_{k'} \to \frac{S}{(2\pi)^2} \int dk_z dk_y = \frac{S}{(2\pi)^2} \int k dk d\theta$

$$\frac{\partial}{\partial t} n_k \bigg|_{\text{relax}} = -\frac{S}{(2\pi)^2} \int k' \left| W_{k,k'} n_k n_{k'} + (k \leftrightarrow k') \right| dk' d\theta$$ (S14)

and writing:

$$W_{k,k'} = \int_0^{2\pi} d\theta W_{k,k'}$$ (S15)

where we integrated the angular dependence of the scattering rate, entering in the expression of $W_{k,k'}$ from $q_z = \sqrt{\Delta \tilde{E}_{k,k'}^2 - k^2 - k'^2 + 2kk' \cos(\theta)}$, we get:

$$\frac{\partial}{\partial t} n_k \bigg|_{\text{relax}} = -\frac{S}{(2\pi)^2} \int k' \left| W_{k,k'} + W_{k',k} \right| n_k n_{k'} dk'$$ (S16)
Since the energy of the polaritons depends only on the modulus of the momentum, \( \omega_k \approx \frac{\hbar^2 k^2}{2m_{LP}} \), we can write \( n_k \) as a function of energy \( n_\omega \), and using \( d\omega = \frac{\hbar^2 k}{2m_{LP}} dk \), write:

\[
\left. \frac{\partial}{\partial t} n_\omega \right|_{\text{relax}} = -\int (W_{\omega,\omega'} + W_{\omega',\omega}) n_\omega n_{\omega'} d\omega'
\]  

(S17)

where \( W_{\omega,\omega'} = 2m_{LP}S/\left(\hbar^2 4\pi^2\right)W_{k(\omega),k'(\omega')} \). Finally, we write a joint equation for the time evolution of \( n_\omega \) by joining both the hydrodynamics of expansion of Eq. (S8):

\[
\frac{dn_\omega}{dt} = \frac{\partial}{\partial t} n_\omega \bigg|_{\text{exp}} + \left. \frac{\partial}{\partial t} n_\omega \right|_{\text{relax}} = -\frac{1}{r} \frac{\partial}{\partial r} (rn_\omega v_\omega) - \gamma n_\omega - \int (W_{\omega,\omega'} + W_{\omega',\omega}) n_\omega n_{\omega'} d\omega' 
\]  

(S18)

where we wrote the velocity as a vector field with radial component only \( v_\omega = v_\omega u_r \). Our next approximation will be to assume that the velocity will be given by \( v_\omega \approx \frac{1}{\hbar} \frac{\partial \omega_k}{\partial k} \).

To study the spatial profile resolved in energies, we will look now for steady state solutions of equation (S18) setting the time derivative to zero and getting the following integro-differential equation in real space:

\[
\frac{\partial n_\omega(r)}{\partial r} = -\frac{n_\omega(r)}{r} + \frac{1}{v_\omega} \left[ -\gamma n_\omega(r) - \int (W_{\omega,\omega'} + W_{\omega',\omega}) n_\omega n_{\omega'} d\omega' \right] 
\]  

(S19)

This we solve numerically by writing the integral again as a discrete sum:

\[
\frac{\partial n_\omega(r)}{\partial r} = -\frac{n_\omega(r)}{r} + \frac{1}{v_\omega} \left[ -\gamma n_\omega(r) - \sum_{\omega'} \left( \tilde{W}_{\omega,\omega'} + \tilde{W}_{\omega',\omega} \right) n_\omega n_{\omega'} \right] 
\]  

(S20)

with

\[
\tilde{W}_{\omega,\omega'} = \kappa \left( \frac{\chi_\omega \chi_{\omega'} \Delta \tilde{E}_{\omega,\omega'}}{|e^{\tilde{E}_{\omega,\omega'}} - 1|} \right)^2 \int \frac{B(q_z)^2}{q_z} d\theta 
\]  

(S21)

where \( \Delta \tilde{E}_{\omega,\omega'} = |\omega' - \omega|/(\hbar u) \), and

\[
\kappa = \frac{2m_{LP}S}{\hbar^2 (2\pi)^2 \hbar \rho V u^2} |D_e - D_h| \Delta \omega' 
\]  

(S22)

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