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Dynamical instability of a non-equilibrium exciton-polariton condensate

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By imaging single-shot realizations of an organic polariton quantum fluid, we observe the long-sought dynamical instability of non-equilibrium condensates. Without any free parameters, we find an excellent agreement between the experimental data and a numerical simulation of the open-dissipative Gross-Pitaevskii equation, which allows us to draw several important conclusions about the physics of the system. We find that the reservoir dynamics are in the strongly nonadiabatic regime, which renders the complex Ginzburg-Landau description invalid. The observed transition from stable to unstable fluid can only be explained by taking into account the specific form of reservoir-mediated instability as well as particle currents induced by the finite extent of the pump spot.

\textit{Introduction.} Semiconductor microcavities are one of the most versatile systems for realizing and studying quantum fluids of light \cite{1}. As a result of the strong coupling between light and matter modes at resonance, new excitations called exciton-polaritons emerge. These hybrid quasiparticles are coherent superpositions of the semiconductor exciton with microcavity photons \cite{2-4}. Although their low effective mass has been touted as an advantage for realizing equilibrium polariton condensates, the driven-dissipative nature of polariton systems—a result of the short particle lifetime—plays an important role in the condensation process. Even out of equilibrium, several phenomena related to Bose-Einstein condensation can be observed in microcavities at elevated temperatures \cite{5-8}. The physics of such non-equilibrium condensates is attractive for both fundamental research and potential applications. Nonlinearity due to strong exciton-mediated interactions gives rise to fascinating physical properties such as superfluidity \cite{9,10} and solitons \cite{11,12}. Recently, nonlinearities have also been demonstrated in organic semiconductors at room temperature \cite{13-15}. This can be attractive for realizing low-cost room-temperature devices based on non-equilibrium condensates such as interferometers \cite{16} or polariton circuits \cite{17-19}.

Despite these remarkable developments, the physics of non-resonantly pumped polariton condensates is still not completely understood. Condensation requires external optical \cite{5} or electronic \cite{20,21} pumping, which creates a reservoir of high-energy electronic excitations. The energetic relaxation of these excitations due to interaction with the environment enhanced by bosonic stimulation can then lead to a macroscopic occupation of the low-lying polariton ground state. This complicated process has been modeled theoretically within various approximations \cite{22-27}. A particularly useful description is based on the phenomenological open-dissipative Gross-Pitaevskii equation (ODGPE) \cite{28}. Its application is widespread due to the simplicity of this description, the limited number of free parameters required and its success in reproducing experimental results. We note that a similar model was also used to describe non-equilibrium condensates in atom laser systems \cite{29}.

Since the introduction of the ODGPE model \cite{28} it was realized that for certain parameters it predicts a peculiar instability of the condensate due to the interaction of polaritons with the reservoir of uncondensed excitons. Since to date there was no experimental evidence of this instability, its physical relevance was unclear. Some authors have suggested that the instability is an artifact that would disappear when energy relaxation in the condensate was properly accounted for \cite{24,30,31}. Many theoretical studies have swept this problem under the rug by imposing the adiabatic assumption of a fast reservoir response, either indirectly by choosing a reservoir lifetime much shorter than typical lifetime of an exciton, or directly by using a simplified complex Ginzburg-Landau (CGLE) description with no separate reservoir degree of freedom \cite{32,33}. In this regime, the model becomes instability free \cite{34}.

Here, we demonstrate that the reservoir-induced instability is a real phenomenon that can occur in non-equilibrium polariton condensates. We confirm this by measuring single-shot realizations of the condensate emission from an oligofluorene-filled microcavity. The results are compared to the predictions of the ODGPE model without using any free parameters. Previous measurements of first-order spatial correlations in this system hinted at the possible breakdown of the stable condensate model \cite{35}. Excellent agreement between experiment and numerical modeling allows us to determine that the lifetime of the reservoir indeed places the system in the unstable and strongly nonadiabatic regime, where the simplified CGLE-like description does not pro-
vide a reliable description of system dynamics. We also demonstrate the transition from a stable to an unstable condensate with increasing pump power. This behavior opposes the previously reported stability criterion obtained for continuous wave pumping [36]. We explain this seeming contradiction as resulting from a competition between reservoir-induced instability, finite condensate lifetime under pulsed excitation, and stabilizing effect of particle currents. Finally, we discuss the relevance of our results to the stability and coherence of inorganic polariton condensates.

**Model.** We model the exciton-polariton condensate using the two-dimensional stochastic ODGPE for the wavefunction \( \psi(r,t) \) coupled to a the rate equation for the polariton reservoir density, \( n_R(r,t) \) [23, 28]

\[
\begin{align*}
 id\psi & = \left[ -\frac{h}{2m^*}\nabla^2 + \frac{g_C}{\hbar} |\psi|^2 + \frac{g_R}{\hbar} n_R + \frac{1}{2} \right] \left( Rn_R - \gamma_C \right) d\psi + dW, \\
\frac{\partial n_R}{\partial t} & = \left( \gamma_R + |\psi|^2 \right) n_R - k_0 n_R^2, \\
\end{align*}
\]

where \( P(r,t) \) is the exciton creation rate due to the pumping pulse, \( m^* \) is the effective mass of lower polaritons, \( \gamma_C \) and \( \gamma_R \) are the polariton and exciton dissipation rates, \( R \) is the rate of stimulated scattering to the condensate, \( g_C \) an \( g_R \) are the polariton-polariton and polariton-reservoir interaction coefficients, respectively, and \( k_0 \) is the bimolecular annihilation rate. The latter is specific property of organic semiconductors, but it does not qualitatively affect the calculation results. The quantum noise \( dW \) can be obtained within the truncated Wigner approximation [23] as Gaussian noise with correlations \( \langle dW(r) dW^*(r') \rangle = \frac{2}{\Delta x^2} (Rn_R + \gamma_C) \delta_{r,r'} \) and \( \langle dW(r) dW(r') \rangle = 0 \) where \( \Delta x \) is the lattice constant of the discretized mesh.

The cavity under consideration is shown schematically in Fig. 1 and composed of a single thin film of 2,7-bis(9,9-di(4-methylphenyl)-fluorene-2-yl)-9,9-di(4-methylphenyl)fluorene (TADF) sandwiched between two DBRs composed 9 pairs of alternating Ta2O5/SiO2. It was excited impulsively at high energy (3.22 eV), with a laser repetition rate of 100 Hz. For each laser shot, a time-integrated image of the condensate emission was recorded using a CCD camera (Thorlabs BC106-VIS). Further details are given in Refs.[15, 35]. All of the model parameters were obtained independently in previous measurements and are summarized below Fig. 2. In Eq. (2), we assume that the pump pulse length \( \tau_{\text{pulse}} = 250 \text{ fs} \) is short enough that \( P(r,t) \) as a \( \delta(t) \) function in time. We neglect the effect of static disorder of the sample. This assumption will be justified by comparison with experimental data.

**Instability of the condensate.** As previously reported, the condensate solution is prone to dynamical instabilities for a certain range of parameters [28]. The physical origin of this instability is the repulsive interaction \( g_R \) between the condensate and reservoir excitons, which can lead to phase separation of these two components. For the parameters of our system, the instability is predicted to occur for all continuous pump powers up to \( P = (g_R \gamma_C)/(g_C \gamma_R) P_{\text{th}} \approx 3000 P_{\text{th}} \) [36]. This greatly exceeds the range of pump powers accessible here and in any other organic microcavity for typical values \( \gamma_C \) and \( \gamma_R \). Many inorganic microcavities also fall within the instability regime.

Single-shot real space images of the condensate photoluminescence are shown in Fig. 2(a)-(e) for varying dimensions of the Gaussian pump. These were taken at a pump power \( P = 2P_{\text{th}} \) for Fig. 2(a)-(d) and \( P = 2.5P_{\text{th}} \) for Fig. 2(e), where \( P_{\text{th}} \) is the condensation threshold. The corresponding ODGPE calculations for the same powers, where only the shape of the pump is varied are shown below in Fig. 2(f)-(j). The agreement is remarkable given that no free parameters were used in the modeling. Note that both the experiment and calculation are time-averaged over the duration of the condensate emission for each pulse. The exact size and orientation of the patterns varies randomly from shot to shot both in experiment and simulations, but these remain qualitatively the same. This shows that disorder does not play an important role in determining the final condensate profile. The experiment and calculation highlight that the instability is more pronounced for large spatial pump sizes (or flat-top, which is not shown). In contrast, the smallest condensate size is only slightly affected by the instability.

The excellent agreement between the experiment and theory allows us to draw some important conclusions about the physics of the system. The parameters of the model indicate that the dynamics are not only in the unstable, but also strongly nonadiabatic regime, i.e. the reservoir \( n_R(r,t) \) does not quickly follow the changes in the condensate density \( |\psi(r,t)|^2 \). The adiabatic regime...
is attained only when three independent analytical conditions are fulfilled simultaneously [34]. Here, all three conditions are violated. In particular \( g_{Ri,R} \approx 4(\gamma_{R} + R|\psi|^2) \), which means that the reaction time of the reservoir is 40 times slower than the response time of the condensate due to interactions with the reservoir.

The breakdown of the adiabatic approximation suggests that CGLE-like models based on a single equation cannot reliably describe the dynamics of the system. This is due to the fact that they do not incorporate the reservoir as a separate degree of freedom. We demonstrate this by numerically modeling the single stochastic Gross-Pitaevskii equation (SGPE) [31, 37] for the same parameters. As shown in Fig. 3(a), this model gives a poor agreement with experimental data. The coefficients of the SGPE equation were determined using the correspondence formulas derived in [34]. The effective interaction between polaritons turns out to be attractive, which is due to the reservoir-mediated attraction in the unstable regime. Nevertheless, the instability does not give rise to multiple domains, but rather to condensate collapse with no spatial symmetry breaking. We also verified that SGPE simulations with an explicit repulsive interaction coefficient are not able to reproduce the experimental patterns [38], see Fig. 3(b).

**Transition from stable to unstable condensate.** For a large size of the pump spot, we generally observe the instability independently of the pump pulse power, see Fig. 2. However, in the case of a small size (e.g., Fig. 2(e, j)), a transition from a stable to an unstable condensate is seen with increasing pump power [38]. When the power of the pump pulse is less than about 1.8 \( P_{th} \), a single condensate is formed, while for pump powers above this value, the instability results in appearance of two or more domains. Note that this stable region is precisely where first-order spatial coherence measurements were previously reported. Stability was a necessity due to the extraction procedure, which requires fitting several interferograms as a function of phase delay. Any shot-to-shot fluctuations would consequently wash out the fringe visibility. Finally, via numerical simulations, we observe that the transition from stable to unstable is not abrupt and that there is some shot-to-shot variation along the boundary.

The observed power dependence is in contradiction with the previously predicted transition from unstable to stable condensate with increasing continuous wave pumping [36, 39]. To understand this effect we developed a theoretical model of condensation dynamics under impulsive excitation. The main elements that determine the stability of the condensate are the unstable dynamical Bogoliubov spectrum the condensate, the density current of polaritons flowing away from the center of the pumping spot, and the finite lifetime of the condensate. The competition of these processes can explain the existence of
stable or unstable condensate for various pumping conditions, and allows for the calculation of the transition region.

The evolution of the system can be divided into several stages shown in Fig. 4. At the arrival of the ultrashort pulse, the reservoir density is fixed according to the Gaussian profile of the pump, while the wavefunction \( \psi \) contains only random fluctuations inherited from the Wigner noise. Due to the finite size of the pump spot, a polariton current from the center of the pump spot is created by the reservoir-induced potential. This favors the formation of a single condensate, as the most central fluctuation “spills over” and repels the other domains outside. However, for large pumping powers a single condensate is not formed, and the system evolves to a fragmented state, while some flow of polaritons from the center is still visible. We attribute this effect to the existence of unstable Bogoliubov modes which break up the condensate. These unstable modes are always present, but for low pumping powers the instability is too weak to develop during the lifetime of the condensate. The condensate lifetime is understood here as the FWHM duration of the emission from the condensate, see Fig. 4.

We calculate the Bogoliubov instability timescale in the local-density approximation, i.e. neglecting the spatial inhomogeneity of the pump. We find that in the case of a pulsed pump, the dynamical Bogoliubov spectrum can be qualitatively different from the one in the case of continuous pumping, which was considered in previous reports [28, 36, 40, 41], due to the absence of the pumping term after the arrival of the pulse. Instead of examining stability about a steady-state, we consider small fluctuations around the homogeneous state with a polariton and reservoir density evolving in time

\[
\psi = \psi_0 e^{-\frac{\beta_C}{\hbar} t + \frac{\beta_R}{\hbar} t} \left( 1 + \sum_k \left[ u_k e^{-i(\omega_k t - k \mathbf{r})} + u_k^* e^{i(\omega_k t - k \mathbf{r})} \right] \right),
\]

\[
n_R = n_R^0 e^{\beta_R t} \left( 1 + \sum_k \left[ w_k e^{-i(\omega_k t - k \mathbf{r})} + c.c. \right] \right),
\]

where \( \omega_k \) is the frequency of the mode with the wavenumber \( \mathbf{k} \), and \( u_k, v_k, w_k \) are small fluctuations. In the above \( \beta_C, \beta_R \) are condensate and reservoir density growth or decay rates, that need to be taken into account in the case of pulsed excitation. The excitation spectrum is described by the eigenvalue problem \( \mathbf{L}_k \mathbf{U}_k = \hbar \omega_k \mathbf{U}_k \), where \( \mathbf{U}_k = (u_k, v_k, w_k)^T \) and

\[
\mathbf{L}_k = \begin{pmatrix}
g_C n_p + \epsilon_k & g_C n_p & \left( \frac{\hbar}{2} R^{2D} + g_R \right) n_R^0 \\
-g_C n_p & g_C n_p - \epsilon_k & \left( \frac{\hbar}{2} R^{2D} - g_R \right) n_R^0 \\
-i\hbar R^{2D} n_p & -i\hbar R^{2D} n_p & -i\hbar (P/n_R^0 + k_b n_R^0)
\end{pmatrix}
\]

and \( \epsilon_k = \hbar^2 k^2/2m^* \), \( n_p = |\psi_0|^2 \), \( \mu = n_p g_C + g_R n_R^0 \), \( \beta_C = R^{2D} n_R^0 - \gamma_C \), \( \beta_R = P/n_R^0 - n_p R^{2D} - \gamma_R \). In the special case of a stationary state under CW pumping the above matrix is equivalent to the one considered before [28]. In the pulsed case, we have \( P = 0 \) after the arrival of the pulse, and consequently \( \beta_R < 0 \), i.e. the decay of the reservoir density.

Fig. 5 shows the comparison between the timescales of the Bogoliubov instability and condensate lifetime. It turns out that the Bogoliubov timescale \( \tau_B \) is longer than the condensate lifetime \( \tau_C \) for \( n_0/n_{th} \approx 1.8 \), where \( n_0 \) is the maximum of \( n_R(x,t) \) and \( n_{th} \) is the threshold value of \( n_0 \) for condensate formation, see Fig. 5. Above this value these timescales become comparable. This is in very good agreement with the observed threshold for domain formation. The similarity between the timescales \( \tau_B \) and \( \tau_C \) above \( n_0/n_{th} \approx 1.8 \) is explained by the similar magnitude of all nonlinear coefficients (\( R, g_C, g_R \)) in Eq. (1). At high pumping, the maximum density of the condensate \( |\psi|^2 \) becomes comparable to \( n_0 \), and all non-linear energy scales have similar order of magnitude; in particular, the spontaneous scattering rate \( \Gamma |\psi|^2 \), which depletes the reservoir and influences the lifetime.

Relevance to inorganic condensates. The vast majority of exciton-polariton condensates are realized in inorganic semiconductors, where properties are slightly different from the organic case considered here. Nevertheless, the parameters of inorganic samples also place them in the unstable regime. In particular, independent measurements indicate a reservoir lifetime in the hundreds of picoseconds [42], which suggests that it should not be treated adiabatically. In several experiments, however, the ”bottleneck” region plays the role of the reservoir and the relaxation kinetics may need to be considered. In contrast, organic microcavities have short enough polariton lifetimes that single-step relaxation processes from the reservoir can be considered to be dominant. Meanwhile, instabilities in inorganic condensates may also be obscured because there are no single-shot reports due to the lower polariton densities in these samples. Averaging over tens of pulses already washes out clear signatures of the domain formation. An alternative to single-shot experiments is the measurement of spatial correlation functions in which the signatures of domains can persist. Indeed, this is observed in the microcavity considered here as a reduction in the first-order spatial coherence [35].

In conclusion, we have demonstrated for the first time the reservoir-mediated instability of a non-equilibrium exciton-polariton condensate. Excellent agreement between the experiment and theory suggests that models with reservoir treated as a separate degree of freedom should be used to describe these systems. Under pulsed excitation, we find that various timescales determine stability boundary including the finite condensate duration, the reservoir-induced instability and particle currents due to repulsive exciton-polariton interactions.
FIG. 4: The development of the condensate instability for the small Gaussian spot at power $P = 1.6P_{th}$. The left panel shows the imaginary part of the frequency of the unstable Bogoliubov branch calculated with (4) at peak polariton density. The middle panel shows the simulated evolution of the condensate and reservoir particle number, with the condensate lifetime $\tau_c$ defined as the FWHM duration of the condensate emission. Right frames (a)-(c) show snapshots of density profiles at the three chosen instants of time. The outgoing flow of polaritons is responsible for the creation of a single condensate. At high pumping, the Bogoliubov instability breaks up the condensate, as shown in frames (b)-(c).

FIG. 5: Comparison between the characteristic time scales that are responsible for the development of the instability according to numerical simulations. Polariton domains are formed only when the Bogoliubov instability time $\tau_B$ becomes comparable to the lifetime of the condensate emission $\tau_c$, which is determined as in Fig. 4. At lower density, a single symmetric condensate is created. Averaged over 5 realizations.

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