Cavity Polariton Condensate in a Disordered Environment

Martin Thunert, 1, * Alexander Janot, 2, * Helena Franke, 1 Chris Sturm, 1 Tom Michalsky, 1 María Dolores Martín, 3, 4 Luis Viña, 3, 4, 5 Bernd Rosenow, 2 Marius Grundmann, 1 and Rüdiger Schmidt-Grund 1

1 Institut für Experimentalphysik II, Universität Leipzig, 04103 Leipzig, Germany
2 Institut für Theoretische Physik, Universität Leipzig, 04103 Leipzig, Germany
3 Departamento de Física de Materiales, Universidad Autónoma de Madrid, Madrid 28049, Spain
4 Instituto de Ciencia de Materiales "Nicolás Cabrera"; Universidad Autónoma de Madrid, Madrid 28049, Spain
5 Instituto de Física de la Materia Condensada, Universidad Autónoma de Madrid, Madrid 28049, Spain

(Dated: December 31, 2014)

We report on the influence of disorder on an exciton-polariton condensate in a ZnO based bulk planar microcavity and compare experimental results with a theoretical model for a non-equilibrium condensate. Experimentally, we detect intensity fluctuations within the far-field emission pattern even at high condensate densities which indicates a significant impact of disorder. We show that these effects rely on the driven dissipative nature of the condensate and argue that they can be accounted for by disorder-induced phase fluctuations, which occur even for increasing condensate densities realized in the regime of high excitation power. Thus, dynamical effects strongly suppress the stabilization of the condensate against disorder fluctuations, contrary to what is expected for equilibrium condensates in the high density limit. Numerical simulations based on our theoretical model reproduce the experimental data.

I. INTRODUCTION

The observation of a macroscopically coherent quantum state of exciton-polaritons, a so-called polariton Bose-Einstein condensate (BEC), 1, 2 has opened an active and challenging research field. Exciton-polaritons (for brevity polaritons) are mixed light-matter excitations in a microcavity (MC). 3, 4 At finite quasi-particle density, several fascinating phenomena like superfluidity 5, 6 and the formation of quantum vortices 7, were discovered. This allows for numerous novel applications like optical parametric oscillators 8, polariton lasers 9, 10 and logical elements 11–15, which are usually restricted to low temperatures. However, polariton BECs at room-temperature were observed recently in MCs based on wide band gap materials like GaN 16–18 and ZnO 19–21 or organic materials 22, paving the way for technological applications. At the moment, experiments are significantly affected by disorder 16,23,24, and a thorough understanding of the impact disorder has on experimental observables in a polariton BEC is called for.

In contrast to conventional BECs, occurring for example in cold atom systems, polaritons have a finite life time, which gives rise to peculiar properties of the condensate. Nonetheless, there remain similarities in the absence of disorder, for instance the quasi-long range order of a two-dimensional polariton condensate 25–28 and its superfluid behavior 5, 6, 29, 30, which are both theoretically expected and experimentally observed. However, recent theoretical studies have revealed exciting differences between equilibrium and non-equilibrium condensates 31–35. For example, it is predicted that even the presence of weak disorder leads to an exponentially decaying correlation function and vanishing superfluidity. 35

We study the impact of disorder on a two-dimensional polariton BEC in a ZnO based MC. For this purpose, the $k$-space intensity distribution as a function of excitation power, or rather condensate density, is analyzed. We observe significant disorder effects even at high condensate densities. For an equilibrium BEC such behavior is unexpected since an increasing density should screen the disorder potential. 36 Therefore, the observed persistence has to rely on the non-equilibrium nature of a polariton BEC. A similar observation was made for a one-dimensional condensate. 37, 38

A polariton BEC is a steady state out of equilibrium where losses are compensated by external excitation. In the presence of disorder, spatial fluctuations of the condensate phase are induced. 35 If the phase fluctuations on length scales comparable to the condensate size, spatial correlations and phase rigidity are strongly reduced. We will show that this leads to significant traces of disorder in the experimentally observed intensity distribution. Furthermore, we experimentally demonstrate that the ratio of the condensate correlation length to the condensate size is independent of the condensate density and hence of the excitation power. Consequently, the condensate stabilization against disorder fluctuations with increasing excitation power is strongly suppressed, as confirmed by our experimental results. In order to quantify our analysis we reproduce the experimental data by numeric simulations.

The paper is organized as follows: In Sec. II we introduce our theoretical model. We discuss the disorder impact on a homogeneously and inhomogeneously excited condensate for a quasi-equilibrium (weak gain and loss) and dynamical (strong gain and loss) condensate, respectively. Furthermore, we provide a general argument that explains our experimental findings. These are presented in Sec. III. In Sec. IV the theoretical predictions are confirmed by comparing experimental data to theoretical simulations. The summary and conclusion can be found
II. THEORETICAL PREDICTIONS

A. Model

A phenomenological description of the dynamics of the polariton condensate wavefunction $\Psi(\vec{x}, t)$ is given by an extended Gross Pitaevskii equation (eGPE)\textsuperscript{39,40}

$$i\hbar \partial_t \Psi = \left( -\frac{\hbar^2}{2m} \nabla^2 + V(\vec{x}) + U |\Psi|^2 \right) \Psi + i \left( R(\vec{x}) - \Gamma |\Psi|^2 \right) \Psi,$$

(1)

where $m$ is the effective mass of the lower polariton branch, $V$ an external potential and $U > 0$ an onsite interaction constant. The function $R(\vec{x})$ represents the gain of particles from a reservoir of non-condensed polaritons, while the term linear in density, $\Gamma |\Psi|^2$, leads to gain saturation with $\Gamma$ as gain-depletion constant. Since the propagation of the reservoir polaritons can be neglected, the spatial shape of $R(\vec{x})$ is set by the Gaussian excitation spot, namely

$$R(\vec{x}) = h \gamma_c \left( \frac{P}{P_{th}} e^{-\vec{x}^2/\xi_p^2} - 1 \right).$$

(2)

The parameter $\gamma_c$ denotes the condensate decay rate (inverse life time $\gamma_c = 1/\tau$). The ratio $P/P_{th}$ is the excitation power versus its value at condensation threshold $P_{th}$, and $\xi_p$ is the waist size of the Gaussian pump spot.

Because of interactions, the condensate energy is blueshifted by $n_0 U$ where $n_0$ is the mean condensate density set by the balance of gain and loss (see Eq. (C4)). Both kinetic and interaction energy in Eq. (1) determine the healing length, $\xi \equiv \hbar/\sqrt{2m \alpha n U}$.

The disorder environment is described by a random external potential $V(\vec{x})$. We choose $V$ as Gaussian-distributed, delta-correlated variable with correlation length $\xi_V$ and strength $V_0$, and introduce an effective dimensionless disorder parameter,

$$\kappa \equiv \frac{\xi_V V_0}{\xi n_0 U}.$$ \hspace{1cm} (3)

An analysis of the gain and loss terms in Eq. (1) allows us to define a ‘non-equilibrium parameter’

$$\alpha \equiv \frac{\Gamma}{U}.$$ \hspace{1cm} (4)

Its magnitude parametrizes the strength of gain and loss dynamics of the polariton BEC. In the limit $\alpha \to 0$ (keeping $n_0$ finite), the equilibrium mean field description for a BEC is obtained, cf. Eq. (C6).

For the sake of clarity, we will focus on single-mode steady-state solutions and therefore make the ansatz

$$\Psi(\vec{x}, t) = \Psi(\vec{x}) \exp(-i\omega t),$$

where $\hbar \omega$ is the condensate energy. However, in experimental realizations more than one condensate mode can exist. For any further details we refer to Appendix C.

B. Disorder Effects

1. Infinite condensate size

Before discussing a finite size polariton BEC we would like to consider a homogeneously excited condensate ($\xi_p \to \infty$, so that the reservoir function Eq. (2) is a constant). We will i) review disorder effects on an equilibrium condensate\textsuperscript{41}, and, ii) describe differences to a dynamical polariton BEC (driven dissipative condensate)\textsuperscript{35}.

With regards to equilibrium condensates i), the disorder potential attempts to pin the condensate into its minima. To this end it has to compensate the energy costs for density deformations (kinetic term in Eq. (1)). The balance of pinning and kinetic energy determines the density Larkin length $L_n \approx 2\pi \hbar^2/2m \xi_V V_0$\textsuperscript{41,42}, as the characteristic scale for density fluctuations. Since the smallest possible scale for density fluctuations is the healing length $\xi$, we identify the ratio $\xi/L_n \sim \kappa$, see Eq. (3), as the relevant parameter\textsuperscript{41} for the effective disorder strength, in support of a picture where disorder potential, kinetic energy, and interaction energy compete with each other. Due to the fact that $\xi \propto 1/\sqrt{n_0}$, the effective disorder strength $\xi/L_n \sim \kappa$ decreases with increasing density, and disorder effects will fade away in this limit.

For a non-equilibrium condensate ii), the interplay of disorder and the gain-loss mechanism results in random sources and sinks, such that random currents are generated, which induce condensate phase fluctuations. The phase-correlation length, over which the condensate phase typically varies by $2\pi$, is given by $L_\phi \approx \sqrt{2\pi L_n/\alpha}$. This scale can be obtained by a generalized Imry-Ma argument in the presence of the driving term Eq. (1), in the limit $\xi_p \to \infty$.\textsuperscript{35} The argument demands that a condensate current flowing out of (or into) a region of diameter $L_\phi$ is generated by an effective source (or sink) determined through an area average of multiple random sources and sinks. In contrast to an equilibrium condensate ($\alpha \to 0$ with $L_\phi \to \infty$), the phase fluctuations occurring in the case $L_\phi < \infty$ destroy the quasi-long-range order of the condensate. As a consequence of these phase fluctuations, the superfluid stiffness vanishes in the thermodynamic limit even for weak disorder, and a rigid response to an external perturbation is limited to a finite length scale, namely the superfluid depletion length $L_s \sim L_n/\alpha^2$.\textsuperscript{35}

2. Finite condensate size

From the analysis above we conclude that in a disordered environment a condensate of size $L_c \ll L_\phi$ will behave completely different from one of size $L_c \gtrsim L_\phi$. We emphasize that the scale $L_\phi$ is inherently related to the non-equilibrium nature of polaritons. In the following we discuss these two scenarios sketched schematically in Fig. 1.
that spatial correlations and phase rigidity are destroyed. A short analysis shows that the ratio $L_c / L_\phi \sim (V_0 \xi \xi \nu m / \hbar^2) (\Gamma / U)$ does not depend on the condensate density and, thus, is expected to be independent of excitation power. A similar conclusion holds for the ratio $L_c / L_s$. As a consequence, no condensate stabilization with increasing density is expected.

In order to make our analysis more quantitative, we have studied theoretically the excitation power dependence of the two-dimensional $k$-space intensity $I_P(k)$ which can be directly compared to experimental data. To this end, Eq. (1) was simulated for many disorder realizations (see Appendix D for details). We have extracted the expectation value, denoted by $\mu_P(k)$, and the variance, denoted by $\sigma_P^2(k)$, of the normalized intensity $I_P(k)$ by averaging over disorder configurations. We note that for a sufficiently large number of realizations, the disorder average restores radial symmetry, such that the expectation values $\mu_P(k)$ and $\sigma_P^2(k)$ depend on the magnitude $k = |k|$ of wavevector only.

In Fig. 2, the results for $\mu_P$ and $\sigma_P$ are shown for scenario I (left panels) and II (right panels). We find that the intensity $I_P$ vanishes for all wavevectors outside of the lower-polariton dispersion ($k \gtrsim \xi^{-1}$) and that its average value does not change qualitatively compared to a disorder free system (cf. Ref. 43). However, for a single snapshot (see Fig. 5 for a scenario II simulation) disorder breaks the radial symmetry and induces intensity fluctuations proportional to $\sigma_P$. For scenario I and for wavevectors $|k| \lesssim \xi^{-1}$, these fluctuations decay linearly.

For the quasi-equilibrium scenario (I) we expect strong phase-correlations, since $L_c \ll L_\phi$, and in this regime disorder induces mainly density fluctuations. As discussed above, the impact of disorder will decrease with increasing density, which should be directly observable by increasing the excitation power. At sufficiently high density the interaction potential (blueshift) will screen completely the disorder potential. Such kind of percolation transition is expected for a quasi-equilibrium polariton BEC.\textsuperscript{36}

For a driven dissipative condensate in a disordered environment, phase-fluctuations are induced as explained above. These can occur at a scale comparable to the condensate size $L_\phi \gtrsim L_c$ (scenario II), such that
Table I. Used simulation parameters as well as the ratio of density Larkin, phase correlation and superfluid depletion length to the condensate size $L_c$.

| scenario | $L_a/L_c$ | $L_b/L_c$ | $L_a/L_c$ | $\xi_P$ | $m$ | $\tau$ |
|----------|-----------|-----------|-----------|--------|-----|--------|
| I        | 10        | 20        | 1.8 $\mu$m | $4.4 \times 10^{-5} m_e$ | 0.1 ps |
| II       | 2.5       | 0.3       |           |        |     |        |

with inverse excitation power, in agreement with the expectation $\sigma^2 \sim k^2 \sim 1/P$ for $k \ll 1$. We note that regions with $k \approx \xi^{-1}$ show a high ratio $\sigma_P/\sigma_B$ (peaks in Fig. 2 lower left panel). In this $k$-region, the emission intensity is increasing very rapidly with excitation power (see Fig. 2 upper left panel), because of the repulsive potential hill created by the finite excitation spot. Thus, the increase of fluctuation strengths with excitation power for $k \approx \xi^{-1}$ is really due to the increase of emission power and does not yield information about the screening of the disorder potential for high condensate densities.

For scenario II, the stabilization with increasing excitation power is suppressed (see lower right panel of Fig. 2). Compared to scenario I, the decrease of $\sigma^2$ with increasing condensate density is weaker than $\sigma^2 \sim 1/P$. These findings agree well with our argument provided above.

### III. EXPERIMENT

In this section we discuss the experimentally observed behavior of the far-field photoluminescence (PL) emission pattern of a polariton condensate in a ZnO-based MC with pronounced structural disorder as a function of excitation power. The experimental setup is described in Appendix A. The MC consists of a half wavelength ZnO cavity, which simultaneously acts as active medium, showing a quality factor of about 1000 and a maximum coupling strength of about 45 meV ($\Omega_{Rabi} \approx 90$ meV) at $T = 10$ K. By using a wedge-shaped cavity, the detuning between the cavity mode energy and the excitonic transition energy strongly varies with the lateral sample position. Structural investigations (atomic force microscopy, X-ray diffraction, cross-sectional transmission electron microscopy) yield a smooth but polycrystalline cavity layer, exhibiting a low interface roughness of $R_{rms} = 1.9$ nm. Furthermore, the cavity layer is preferentially c-plane oriented and laterally textured, containing large grains aligned in the growth direction reaching from the bottom to the top (grain sizes ranging from 20 nm up to 120 nm). Further information about the sample properties can be found in Ref. 23. Due to the textured structure we suppose that an electronic disorder potential is primarily caused by depletion of carriers, e.g. aluminum donor bound excitons$^{44}$, due to interface band bending at grain boundaries$^{45}$. (see Appendix B for details).

Figures 3(a)-(d) show the excitation power dependence of the PL $k$-space emission pattern. We deduce a polariton effective mass of $m = 4.4 \times 10^{-5} m_e$ ($m_e$: free electron mass) from the dispersion of the lower polariton branch (LPB) (not shown here). A rough estimation of the polariton lifetime ($\tau = 0.1$ ps) is obtained from the spectral linewidth of the condensate emission. The condensation threshold density is $P_{th} = 39$ W cm$^{-2}$.

In all cases investigated here, the condensate emission is distributed dispersion-less at horizontal lines in $k$-space with maximum intensity between the LPB dispersion, which is visible in the far-field PL images (c.f. Fig. 3) for low excitation power $P \leq 4 P_{th}$. This indicates a weak expansion of the condensed polaritons due to the background potential induced by the excitation spot, whose size is similar or even larger than the polariton propagation length$^{23,44}$. For the lowest excitation power shown here, $P = 2 P_{th}$, the emission intensity from the condensed polaritons is in the same order as from the condensate preventing a clear delimitation. With increasing excitation power the BEC states undergo a blue-shift due to the increasing interaction potential. Furthermore, new BEC states appear and others disappear reflecting their dynamical character. Previous studies in the literature on this multimode behavior show that the emission from coexisting individual modes originates from different regions of the same condensate$^{46-48}$. However, other studies on polariton condensates in a disordered environment
found that long-range spatial coherence is still present for their energy-averaged emission\cite{14,49} indicating persistent correlations between different, possibly spatially separated condensate states.

For a wide range of excitation powers, condensate emission out of two energy ranges is observed, which are stable and energetically well separated. For further analysis we select only one of these energy channels. This is needed for comparison with theoretical calculations discussed in Sec. IV, which consider a single-mode condensate state. It should be noted that within each of these channels the emission of more than one single condensate state could be overlapped, which cannot be resolved experimentally.

Within these condensate states several randomly distributed emission spots are observable. This non-homogeneous emission pattern strongly differs from the smooth and ideally radial symmetric distribution, which would be expected for a disorder-free sample.\cite{43} These pronounced intensity fluctuations persist even for high excitation power, i.e. high condensate densities. We note that the constant sharp stripes in the $I_P(k)$ profile at a specified $k$ for all excitation powers are caused by imperfection of the setup, probably due to the microscope objective.

In the following, we focus on a single emission channel delimited by the white dashed lines in Fig. 3(b)-(d). These are defined by an energy range $\Delta E$ which corresponds to the excitation power dependent full-width at half-maximum of the condensate emission. $\Delta E$ is found to be overlapped, which cannot be resolved experimentally.

To investigate the temporal coherence properties of the condensate we used a Michelson interferometer in the mirror-retroreflector configuration to superimpose the PL emission of polaritons with opposite emission angles or wavevectors, respectively. Figures 4(a)-(c) show the images of the individual interferometer arms separately as well as the interferogram. The cw excitation power is $P = 100 \text{ kWm}^{-2} = 2 P_{th}$. For the individual arms we observe two horizontal lines with pronounced disorder induced intensity fluctuations, reflecting two individual condensate states at different energy levels. The image of arm 1 is not the mirror image of arm 2 as it would be expected. We relate this to a slight variation of the emission pattern to be caused e.g. by the geometry of the experimental setup or by temporal fluctuations due to the dynamical character of the condensate and thus different realizations of the BEC. The latter one is not present for the superimposed image, since in this case the individual interferometer arm images originate from the same emission process. The intensity of the interferogram appears to be modulated with a fixed frequency over the entire $k$-space. This is clearly seen by performing a Fourier transform (cf. Fig. 4(d)) of the exemplarily selected $I(k)$ profiles, highlighted by the white rectangle in Fig. 4(a)-(c). A significant peak at $z = 7.8 \mu m$ arises in the Fourier transformed interferogram, indicating mutual temporal coherence of the emission from polariton states with opposite wavevectors. We note that the peak position in the Fourier spectrum corresponds to the experimentally observed distance between consecutive intensity maxima, $\Delta k = 0.13 \mu m^{-1}$, within the $k$-space interferogram (cf. Fig. 4 determined by the geometry of the interferometer (see Appendix. A for details).

Due to the presence of an interference pattern, polaritons exhibiting the same energy are temporally coherent. Although the interference measurements were performed in $k$-space, qualitative results regarding the temporal coherence in real space can also be extracted indirectly. To reach this conclusion, we argue that polaritons with different $k$ values are from different spatial positions corresponding to their outward propagation by acceleration out of the excitation center. Thus, the emission of $k$ and $-k$ which we superimpose is assigned to $x$ and $-x$. Consequently, interferometry measurements show that the emission from a certain energy state arises from a temporally coherent condensate rather than of uncorrelated condensate fragments.\cite{50}

Summarizing, the experimental observations indicate a strong impact of disorder on the polariton BEC even at high excitation power well above the condensation threshold. As discussed in Sec. II the suppression of disorder effects with increasing condensate density is strongly hindered for a polariton BEC. Hence, we suppose that the interplay of gain-loss and disorder prevents a stabilization at high excitation power also in the experiment.
In order to quantify the agreement with our simulations, we restrict our experimental data to low intensities. This is possible with the assumptions from the model. For very high excitation power, we restrict ourselves to performing simulations for scenario I and II described earlier. We note that the model (see Eq. (1)) assumes a linear increase of the condensate energy with increasing condensate density (excitation power). Therefore, we restrict our experimental data set to this regime, namely $P = (4, \ldots, 22)P_{th}$. For $P < 4 P_{th}$, the integrated PL intensity of the investigated condensate state shows a typical super-linear slope for increasing excitation power, which is incompatible with the assumptions from the model. For very high excitation power $P > 22 P_{th}$ the emission from the investigated single-mode state partially coincides with other condensate modes which prevents a reasonable spectral line shape analysis.

For a typical disorder realization, a series of numerically obtained snap shots of the two-dimensional intensity distribution $I_P(\vec{k})$ for increasing excitation power is shown in Fig. 5. These images correspond to scenario II (dynamical polariton BEC with significant gain and loss). We clearly observe a disorder induced deviation from the ideally radial distribution for all excitation powers. This asymmetry does not converge to a symmetric intensity distribution while increasing $P = (4, \ldots, 22)P_{th}$. Such an asymmetry as well as its persistence is also observed experimentally, see Fig. 3, and thus agrees qualitatively with our simulations. We note that the experimental data represent the intensity distribution of one disordered sample, and correspond to a one-dimensional cut along a fixed axis crossing the origin of the two-dimensional $k$-space distribution.

For a more quantitative analysis we go beyond the single snap shot picture. To this end, we directly compare the experimental data with the numerically computed expectation value of the intensity distribution. We symmetrize the experimentally measured data $I_P(k) \rightarrow (I_P(k) + I_P(-k))/2$ with $k \geq 0$ and superimpose them with the numerical simulations. Since the condensate density and healing length are hard to extract from experimental data, we fix the scaling of the $x$- and $y$-axis by a least-square fit. In Fig. 6 we compare the experimental data with simulations of scenario II. We have excluded experimental data with low intensities ($I_P(k) < 0.25$) because they are strongly affected by a systematic artifact, which is present for $k \sim 3 \ldots 4 \mu m^{-1}$ for all excitation powers. In order to quantify the agreement between model and experiment we introduce the mean squared error (MSE) and the goodness-of-fit value $Q$ (see text) are plotted. For used parameters see Table I.

**IV. COMPARISON BETWEEN THEORETICAL MODEL AND EXPERIMENT**

We compare the experimental data with increasing excitation powers $P$. The black solid lines depict the expectation value whereas the gray band indicates the expected fluctuations (standard deviation). For low intensities we have excluded systematically biased data (gray crosses). For each $P$ the mean squared error (MSE) and the goodness-of-fit value $Q$ (see text) are plotted. For used parameters see Table I.
or GaN.

sates in microcavities based on other materials, e.g. CdTe

tions. We note that these findings may also explain the
citation power, well above the condensation threshold,
that the persistence of disorder effects even at high ex-
cavity is exposed to significant structural disorder, and
we conclude that the polariton condensate in the micro-
can be reproduced by numerical simulations. From this
even at high excitation power. This experimental finding
can be reproduced by numerical simulations. From this
we conclude that the polariton condensate in the micro-
cavity is exposed to significant structural disorder, and
that the persistence of disorder effects even at high ex-
citation power, well above the condensation threshold,
relies on the intrinsic non-equilibrium nature of polaritons.
We note that these findings may also explain the
observation of similar phenomena for polariton condens-
ates in microcavities based on other materials, e.g. CdTe
or GaN.16,47,48

V. SUMMARY AND CONCLUSION

In this work we have characterized a polariton condens-
ate in a disordered environment. Our theoretical analy-
sis shows that the magnitude of condensate phase fluctu-
ations, which are induced by the interplay of disorder and
gain-loss of particles, does not depend on the condensate
density and hence is independent of excitation power.
Thus, in contrast to an equilibrium condensate, the inher-
ent non-equilibrium nature of polaritons does not allow
a stabilization against disorder fluctuations with increas-
ing condensate density. To verify our prediction we have
analyzed experimentally the photoluminescence emission
of a ZnO based microcavity. Indeed, we find a lack of sta-
bilization with increasing density in terms of pronounced
intensity fluctuations within the $k$-space emission pattern
even at high excitation power. This experimental finding
can be reproduced by numerical simulations. From this
we conclude that the polariton condensate in the micro-
cavity is exposed to significant structural disorder, and
that the persistence of disorder effects even at high ex-
citation power, well above the condensation threshold,
relies on the intrinsic non-equilibrium nature of polaritons.
We note that these findings may also explain the
observation of similar phenomena for polariton condens-
ates in microcavities based on other materials, e.g. CdTe
or GaN.16,47,48

VI. ACKNOWLEDGEMENT

We acknowledge experimental support and fruitful dis-
cussions from the group of N. Grandjean at EPFL. AJ
is supported by the Leipzig School of Natural Sciences
BuildMoNa. This work was supported by Deutsche
Forschungsgemeinschaft through project GR 1011/20-2,
by Deutscher Akademischer Austauschdienst within the
project PPP Spain (ID 57050448) and by the Spanish
MEC MAT2011-22997.

MT and AJ contributed equally to this work. MT per-
formed the experimental part and AJ carried out the
theoretical analysis.

Appendix A: Experimental setup

In order to investigate the optical properties of the
polariton condensate, we applied two different photo-
luminescence configurations, which have in common a
non-resonant and pulsed excitation as well as a detection
of the far-field emission. The setup to investigate the dis-
order effects on the polariton distribution and their dy-
namics as a function of the excitation power is described
in Ref. 23. Here, the Gaussian excitation spot covers an
sample area of about 10 µm².

For the coherence measurements, the sample was ex-
cited via a frequency-tripled Ti:sapphire laser at 266 nm
(repetition rate: 82 MHz, pulse length ≈ 2 ps). The PL
signal of the Fourier plane was sent to a Michelson inter-
ferometer in the mirror-retroreflector configuration. The
retroreflector image is a centrosymmetric counterpart of
the mirror arm image. In the resulting interferogram we
superimposed the signal with wavevector $\vec{k}_||$ with that
of $-\vec{k}_||$. Interference maxima occur when the path differ-
ence between the individual beams, $\Delta L = c\Delta t$, is an
integer multiple of the PL emission wavelength, being $\Delta t$
the delay between the beams and $c$ the speed of light. With
the help of a streak camera the relative delay between
the two arms was set to zero for $k_|| = 0$.

Real space measurements with a sufficient spatial reso-
lution could not be performed due to a spherical aberra-
tion induced by the cryostat window. For the conditions
used in our experiments, namely the UV spectral range,
a window thickness of 1.5 mm and the large range of
collected emission angles of ±23°, the resulting spatial
distortion of the image is larger than structural fluctu-
ations that we would like to resolve. Consequently, the
distortion of the measured real space image prevents a
precise investigation of the spatial distribution of the lu-
minescence as well as spatially resolved correlation mea-
surements. Note, that far-field images are not affected by
the cryostat window, which causes a parallel beam shift
but does not change the angle of the transmitted rays.
Appendix B: Origin of disorder potential

Due to the dual light-matter nature of the polaritons, the effective disorder potential can be of photonic as well as electronic origin.

Photonic disorder can be caused by surface and interface roughness as well as thickness fluctuations within the MC structure. This leads to a spatial fluctuating cavity length and therefore to a variation of the cavity photon energy. Due to results of other ZnO-based MCs, a minimum potential strength of $V_C \geq 2 \text{ meV}$ can be expected. The corresponding correlation length $\xi_V$ is of the order of the photonic wavelength, of about 370 nm.

In the literature, usually electronic disorder is neglected. In contrast to this, we assume a strong influence of an electronic background potential caused by randomly distributed excitonic states which are accumulated within the bulk of grains or bound to impurities. This is supported by two facts: firstly, cross-sectional TEM analysis of a MC, that is fabricated under the same conditions, provides a granular structure of the investigated ZnO MC with grain sizes ranging from 20 nm up to 120 nm. Secondly, the slope of the polariton blueshift $\Delta E(P)$ is by a factor of about 8.5 larger for $P < P_{th}$ than above (cf. Fig. 7). This can be explained by assuming an additional electronic background potential $\Delta E_{eb}$, which may include localized states within a disorder potential or bound to impurities as shown in Ref. 23 and 56. Since the concentration of these electronic defects is finite, their contribution to the condensate blueshift saturates for a certain excitation power or rather condensate density. Thus, the further blueshift for $P > P_{th}$ is restricted to polariton-polariton interaction.

We assume that the condensate blueshift for small excitation power $\Delta E(P < 4 P_{th})$ is primarily caused by its interaction with aluminum donor bound excitons ($D^0$, $X$) and that $\Delta E$ scales linearly with its concentration. As mentioned in Sec. III, we suppose a depletion of bound excitons at grain boundaries and thus an accumulation of them within the grain bulk. According to the model described in Ref. 45 the grain boundaries act like two back-to-back Schottky barriers and the carrier flow between grains is driven by thermionic emission over the Schottky barrier. In general, the average height and width of these barriers can be determined from the temperature-dependent evolution of the hall mobility. Unfortunately, this was not possible for our MC due to low current values, below the resolution limit of 1 nA, for temperatures below 200 K caused by the small cavity thickness of about 100 nm as well as due to strong inhomogeneities of the current density, which may be caused by the cavity thickness gradient.

Assuming the mechanism of carrier depletion at grain boundaries to be the dominant one for the effective electronic disorder potential, its correlation length $\xi_V$ is similar to the grain size with values between 20 nm and 120 nm. This is about two orders of magnitude below the condensate size $L_c$, limited by the size of the pump spot and thus even lower than the assumed correlation length for photonic disorder of about 370 nm. Consequently, a trapping of the entire condensate within a minimum of the disorder potential can be excluded. We rather suppose that the disorder potential causes condensate density fluctuations and thus phase fluctuations due to the interplay of disorder and the non-equilibrium nature of the polariton condensate.

Appendix C: Details of the Model

A phenomenological description of the dynamics of the macroscopic polariton condensate wavefunction $\Psi(\vec{x}, t)$ is given by an extended Gross-Pitaevskii Equation (eGPE),

$$i\hbar \partial_t \Psi = \left( -\frac{\hbar^2}{2m} \nabla^2 + V(\vec{x}) + U |\Psi|^2 \right) \Psi + i \left( R(\vec{x}) - \Gamma |\Psi|^2 \right) \Psi .$$

The first part of the right hand side is the ordinary equilibrium GPE with $m$ as effective polariton mass of the lower polariton branch, $V(\vec{x})$ as external potential, and $U$ as repulsive onsite interaction potential. The second part models phenomenologically the gain and loss of the condensate. Here, $R(\vec{x})$ is a reservoir of particles and describes the gain of condensated polaritons. The non-linearity implements a density dependent gain saturation with $\Gamma$ as gain depletion parameter. The reservoir function $R(\vec{x})$ provides a simplified description of the complex gain process, including relaxation of high-momentum polaritons which are generated by incoherent excitation with an external laser beam. Since the non-condensated polaritons have a short lifetime compared to the lifetime of the condensate, we can safely neglect diffusion processes of these and relate the spatial extension of the reservoir to the Gaussian excitation profile of the laser beam, i.e.

$$R(\vec{x}) = \frac{\hbar \gamma_c}{P_{th}} \left( \frac{P}{P_{th}} e^{-\vec{x}^2/\xi_P^2} - 1 \right) ,$$

with decay rate $\gamma_c = 1/\tau$, where $\tau$ is the condensate life-time, and waist size $\xi_P$ of the laser beam. The parameter $P/P_{th}$ gives the excitation power normalized by its value at the threshold for first condensation. The disorder landscape is incorporated by the external potential $V(\vec{x})$. We use a $\delta$-correlated Gaussian distributed quenched disorder with vanishing mean value and variance,

$$\langle V(\vec{x}) \rangle = 0 , \quad \langle V(\vec{x}) V(\vec{y}) \rangle = V_0^2 \xi_P^2 \delta(\vec{x} - \vec{y}) ,$$

respectively. The average disorder strength is given by $V_0$ and its characteristic length is denoted by $\xi_P$.

In the case of a spatially homogeneous excitation, i.e. $\xi_P \rightarrow \infty$, Eq. (C1) is exactly the model used in Ref. 40. In
Ref. 39 and 43 the dynamics of non-condensed reservoir polaritons is considered explicitly. However, an expansion of this extended model in the ratio of condensate over reservoir density reproduces Eq. (C1).

In the following we will describe the model Eq. (C1) in more detail. The mean condensate density \( n_0 \) is found by averaging the second term of the right hand side of Eq. (C1) over the condensate area \( \Omega_c \approx \pi \xi_0^2 \) and demanding a balance of gain and loss,

\[
n_0 \equiv \frac{1}{\Omega_c} \int_{\Omega_c} |\Psi(\vec{x})|^2 = \frac{\hbar \gamma_c}{\Gamma} \left( \frac{1}{\Omega_c} \int_{\Omega_c} \frac{P}{P_{th}} e^{-\vec{x}^2 / \xi_0^2} - 1 \right)
\approx \frac{\hbar \gamma_c}{\Gamma} \left( \frac{P}{P_{th}} - 1 \right).
\]

(C4)

Since the interaction term in Eq. (C1) is proportional to the density, we find an energy blueshift of strength \( n_0 \theta_0 \). The healing length is extracted by comparing the kinetic energy term versus the interaction term in Eq. (C1),

\[
\xi \equiv \sqrt{\frac{\hbar^2/2m}{n_0 \theta_0}}.
\]

(C5)

Let us understand its physical relevance and assume, for example, a small region at which the condensate is forced to vanish, \( \Psi = 0 \), however remains unperturbed everywhere else. Now, the healing length is the distance over which the condensate density changes from zero to \( n_0 \). The blueshift and the healing length allow us to obtain a dimensionless eGPE (C1), namely

\[
i \partial_t \psi = (-\nabla^2 + \vartheta(\vec{x}) + |\psi|^2)\psi + i\alpha(g_R(\vec{x}) - |\psi|^2)\psi,
\]

(C6)

where density, length, energy and time are measured in units of \( n_0 \), \( \xi \), \( n_0 \theta_0 \) and \( \hbar/n_0 \theta_0 \), respectively. The ‘non-equilibrium’ parameter \( \alpha \) and the dimensionless reservoir function \( g_R \) are defined in Eq. (C8) and Eq. (C9), respectively. With \( \psi(x,t) \equiv \Psi(x,t) / \sqrt{n_0} \) we denote the dimensionless wavefunction and \( \vartheta(\vec{x}) = V(\vec{x}) / n_0 \theta_0 \) is the disorder potential relative to the blueshift with

\[
\langle \vartheta(\vec{x}) \rangle = 0, \quad \langle \vartheta(\vec{x}) \vartheta(\vec{y}) \rangle = \kappa^2 \delta(\vec{x} - \vec{y}).
\]

(C7)

We have introduced two important dimensionless parameters, namely an effective disorder strength and a ‘non-equilibrium’ parameter

\[
\kappa \equiv \frac{\xi_V V_0}{\xi n_0 \theta_0}, \quad \text{and} \quad \alpha \equiv \frac{\Gamma}{\Gamma_U}.
\]

(C8)

The first one can be understood as follows: Let us coarse grain the random potential disorder up to the healing length (assuming \( \xi_V \ll \xi \)). This process will renormalise the disorder strength, which now is inversely proportional to the square root of the number of uncorrelated patches, i.e. \( \sim 1/\sqrt{\xi/\xi_V} \). Now, if we compare its value at this scale, \( \xi_V V_0 / \xi \), with the blueshift \( n_0 \theta_0 \), the effective dimensionless disorder parameter \( \kappa \) is obtained.

The second parameter \( \alpha \) implements the non-equilibrium nature of polaritons. In the limit \( \alpha \to 0 \) (keeping \( n_0 \) constant) Eq. (C6) reduces to the standard equilibrium GPE, whereas, for \( \alpha \to \infty \) the condensate is dominated by gain and loss. Finally, the rescaled reservoir function is given by

\[
g_R(\vec{x}) = \left( \frac{P/P_{th}}{P/P_{th}} - 1 \right)
\]

with \( \theta_P \equiv \xi_P / \xi \). For a steady state solution (single-mode condensate) we make the ansatz

\[
\psi(x,t) = \psi(\vec{x}) e^{-i\omega t} = \sqrt{n(\vec{x})} e^{i\vec{\phi}(\vec{x}) - i\omega t},
\]

(C9)

where \( \omega \) is the condensate energy.

We emphasize that both blueshift and healing length depend on the excitation power \( P \) via \( n_0 \). Thus, \( \kappa \) and \( \theta_P \) depend on \( P \), too. For our analysis it is useful to identify energy and length scales which are excitation power independent, namely the line width energy \( \Gamma \equiv h\gamma_c \) and the quantum correlation length \( \xi_q \equiv \sqrt{\hbar/2m\gamma_c} \) (a non-equilibrium analgoum of the thermal de Broglie wavelength)\(^{24} \), so that \( \kappa \) and \( \theta_P \) become functions of \( \alpha, P \) and sample parameters (see Table 1).

Appendix D: Numerical Simulations and Comparison with Experiment

Numerical simulations – Computing the condensate wavefunction by solving the eGPE allows us to extract the real and \( k \)-space intensity distribution, which can all be measured in the experiment. We have

\[
I_P(\vec{k}) \equiv h\omega |\psi(\vec{k})|^2,
\]

(D1)

where the momentum space wavefunction is defined via a two-dimensional discrete Fourier transform \( \psi(\vec{k}) = (1/N^2) \sum \psi(\vec{x}) e^{-i\vec{k} \cdot \vec{x}} \), with \( \vec{x}, \vec{k} \) being elements of a discrete lattice with \( N \) lattice points in each spatial direction. We choose an appropriate set of simulation parameters extracted from the experiment (see Table I) and solve Eq. (C6) numerically. To this end we look for a steady state solution, see Eq. (C10), by solving the time evolution of the discretized wavefunction \( \psi(\vec{x},t) \). The latter is defined on a real-space square lattice with spacing \( a_L \).

We employ a variable order Adams-Bashforth-Moulton PECE algorithm\(^{51} \) to obtain the time evolution, where the lattice spacing is chosen to be equal to the healing length \( \xi \). First, we compute the steady state solution of the disorder-free system, \( \vartheta = 0 \). Then, we choose independent Gaussian distributed variables of vanishing mean and variance \( \kappa^2 \) for each lattice site and calculate the steady state solution of the disordered system. The time evolution of the disordered system is started from the disorder-free solution as initial condition. Thus, we obtain for each disorder realization the discretized two-dimensional wavefunction \( \psi(\vec{x}) \), and after Fourier transforming the two-dimensional \( k \)-space intensity \( I_P(\vec{k}) \).
Finally, we average over all disorder realizations and compute the expectation value and variance,

$$\mu_P(k) \equiv \langle I_P(\vec{k}) \rangle / \langle I_P(0) \rangle,$$

$$\sigma_P^2(k) \equiv \left( \langle I_P(\vec{k}) - \langle I_P(\vec{k}) \rangle \rangle \right)^2 / \langle I_P(0) \rangle^2,$$

respectively. Above, the bracket $\langle \rangle$ denotes an average with respect to the disorder realizations, and we have normalized the mean and variance by the intensity expectation value at $k = 0$. Since the excitation profile Eq. (9) is radial symmetric, Eqs. (D2, D3) are radial symmetric, too, assuming a sufficiently large number of disorder realizations.

Comparison with Experiment – The numerically obtained mean and variance of the $k$-space intensity can be directly compared with the experimental data (see Sec. III and Fig. 6), denoted by $I_{ex}(k_x)$ here. We note that these measurements represent a line-cut along a fixed axis (e.g. the $x$-axis) of the two-dimensional intensity distribution and are measured for one disorder configuration set by the disorder of the sample. We perform a spatial averaging step by symmetrizing the experimentally obtained intensity: $I_{ex}(k_x) \rightarrow (I_{ex}(k_x) + I_{ex}(-k_x))/2$ and $k_x \geq 0$. In order to quantify the agreement between experiment and theory we introduce the chi-square value,

$$\chi_P^2 = \sum_{k_j \geq 0} \left( \frac{I_{ex}(k_j)/a - \mu_P(bk_j)}{\sigma_P(bk_j)} \right)^2.$$

Since the condensate density $n_0$ and the healing length $\xi$ are hard to extract experimentally, we use two scaling parameters $(a, b)$ instead. Both are determined by a least-squares fitting procedure.

Estimating the goodness-of-fit value we extract the complement of the $\chi^2$-probability distribution function $F_{\chi^2}$, denoted by $Q = 1 - F_{\chi^2}(\chi_P^2)$. Roughly speaking $Q$ is the probability that the simulations agree with the experimental data. If $Q \ll 1$ the apparent discrepancies of model and data are unlikely to be random fluctuations. A probable conclusion is that the model is not specified correctly, or that the fluctuation strength $\sigma_P$ is underestimated. If $Q \lesssim 1$ it is likely that the model does agree with the data. Finally, we define the mean squared error: $\text{MSE} \equiv (1/N) \sum_{k_j} (I_{ex}(k_j)/a - \mu_P(k_j))^2$, which is a simple measure of how well the data match the simulated intensity distribution. The comparison of experimental data and numeric simulations of scenario I is shown in Fig. 8 and the comparison with simulations of scenario II was shown and discussed in the main part, see Fig. 6.
Figure 8. (color online) Experimental $I_p(k)$ distribution (blue diamonds) matched to numeric simulations of scenario I (quasi-equilibrium) for increasing excitation powers $P$. The black solid lines depict the expectation value whereas the gray band indicates the expected fluctuations (standard deviation). For low intensities we have excluded systematically biased data (gray crosses). For each $P$ the mean squared error (MSE) and the goodness-of-fit value $Q$ (see text) is given. For used parameters see Table I.

20 F. Li, L. Orosz, O. Kamoun, S. Bouchoule, C. Brimont, P. Disseix, T. Guillet, X. Lafosse, M. Leroux, J. Leymarie, M. Mexis, M. Mihailovic, G. Patriarche, F. Réveret, D. Solnyshkov, J. Zúñiga-Pérez, and G. Malpuech, Phys. Rev. Lett. 110, 196406 (2013).

21 Y.-Y. Lai, Y.-P. Lan, and T.-C. Lu, Appl. Phys. Express 5, 082801 (2012).

22 J. D. Plumhof, T. Stöferle, L. Mai, U. Scherf, and R. F. Mahrt, Nat. Mater. 13, 247 (2014).

23 H. Franke, C. Sturm, R. Schmidt-Grund, G. Wagner, and M. Grundmann, New J. Phys. 14, 013037 (2012).

24 A. Trichet, E. Durupt, F. Méard, S. Datta, A. Minguzzi, and M. Richard, Phys. Rev. B 88, 121401 (2013).

25 G. Roumpos, M. Lohse, W. H. Nitsche, J. Keeling, M. H. Szymanska, P. B. Littlewood, A. Löffler, S. Höfling, L. Worschech, A. Forchel, and Y. Yamamoto, Proc. Natl. Acad. Sci. U. S. A. 109, 6467 (2012).

26 A. Chiochetta and I. Carusotto, EPL (Europhysics Letters) 102, 67007 (2013).

27 R. Spano, J. Cuadra, G. Tosi, C. Antón, C. A. Lingg, D. Sanvitto, M. D. Martín, L. Viña, P. R. Eastham, van der Poel, M., and J. M. Hvam, New J. Phys. 14, 075018 (2012).

28 R. Spano, J. Cuadra, C. Lingg, D. Sanvitto, M. D. Martín, P. R. Eastham, M. van der Poel, J. M. Hvam, and L. Viña, Opt. Express 21, 10792 (2013).

29 M. Wouters and I. Carusotto, Phys. Rev. Lett. 105, 020602 (2010).

30 J. Keeling, Phys. Rev. Lett. 107, 080402 (2011).

31 L. M. Sieberer, S. D. Huber, E. Altman, and S. Diehl, Phys. Rev. Lett. 110, 195301 (2013).

32 L. M. Sieberer, S. D. Huber, E. Altman, and S. Diehl, Phys. Rev. B 89, 134310 (2014), 1309.7027.

33 U. C. Täuber and S. Diehl, Phys. Rev. X 4, 021010 (2014).

34 E. Altman, L. M. Sieberer, L. Chen, S. Diehl, and J. Toner, arXiv (2013), arXiv:1311.0876.

35 A. Janot, T. Hyart, P. R. Eastham, and B. Rosenow, Phys. Rev. Lett. 111, 230403 (2013).

36 G. Malpuech, D. Solnyshkov, H. Ouerdane, M. Glazov, and I. Shelykh, Phys. Rev. Lett. 98, 206402 (2007).

37 F. Manni, K. G. Lagoudakis, B. Pietka, L. Fontanesi, M. Wouters, V. Savona, R. André, and B. Deveaud-Plédran, Phys. Rev. Lett. 106, 176401 (2011).

38 P. Stepnicki and M. Matuszewski, Phys. Rev. A 88, 033626 (2013).

39 M. Wouters and I. Carusotto, Phys. Rev. Lett. 99, 140402 (2007).

40 J. Keeling and N. G. Berloff, Phys. Rev. Lett. 100, 250401 (2008).

41 T. Nattermann and V. L. Pokrovsky, Phys. Rev. Lett. 100, 060402 (2008).

42 Y. Imry and S.-K. Ma, Phys. Rev. Lett. 35, 1399 (1975).

43 M. Wouters, I. Carusotto, and C. Ciuti, Phys. Rev. B 77, 115340 (2008).

44 Aluminum is a common donor in ZnO layers and as a component of the used sapphire substrate and Bragg mirror layers it can easily diffuse into the ZnO cavity during the annealing process at high temperatures of $T \approx$ 900 °C.

45 J. W. Orton and M. J. Powell, Rep. Prog. Phys. 43, 1263 (1980).

46 D. Krizhanovskii, D. Sanvitto, A. Love, M. Skolnick, D. Whittaker, and J. Roberts, Phys. Rev. Lett. 97, 097402 (2006).

47 A. Baas, K. G. Lagoudakis, M. Richard, R. André, L. S. Dang, and B. Deveaud-Plédran, Phys. Rev. Lett. 100, 170401 (2008).

48 D. Krizhanovskii, K. Lagoudakis, M. Wouters, B. Pietka, R. Bradley, K. Guda, D. Whittaker, M. Skolnick, B. Deveaud-Plédran, M. Richard, R. André, and L. S. Dang, Phys. Rev. B 80, 045317 (2009).

49 M. Richard, J. Kasprzak, R. Romestain, R. André, and L. S. Dang, Phys. Rev. Lett. 94, 187401 (2005).

50 C. Antón, G. Tosi, M. D. Martín, Z. Hatzopoulos, G. Kontantinidis, P. S. Eldridge, P. G. Savvidis, C. Tejedor, and L. Viña, Phys. Rev. B 90, 081407 (2014).

51 W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery, Numerical Recipes: The Art of Scientific Computing (Cambridge University Press, Cambridge, UK, 2007).

52 The shoulder in the experimental data at $k \sim 3 \ldots 4 \mu m^{-1}$ is visible for all excitation powers and its shape and magnitude is independent on the power. Therefore we can conclude that this shoulder is caused by residual effect and does not belong to the condensate emission. Different options are possible, e.g. the emission of uncondensed polaritons occupying the LPB or artifacts from the experimental setup, e.g. transmission fluctuations from the microscope objective (q.v. Sec. III).

53 L. Orosz, F. Réveret, F. Méard, P. Disseix, J. Leymarie, M. Mihailovic, D. Solnyshkov, G. Malpuech, J. Zúñiga-Pérez, F. Semond, M. Leroux, S. Bouchoule, X. Lafosse, M. Mexis, C. Brimont, and T. Guillet, Phys. Rev. B 85, 121201 (2012).

54 V. Savona, J. Phys.: Condens. Matter 19, 295208 (2007).

55 G. Nardin, K. G. Lagoudakis, M. Wouters, M. Richard, A. Baas, R. André, L. S. Dang, B. Pietka, and B. Deveaud-Plédran, Phys. Rev. Lett. 103, 256402 (2009).
H. Franke, *PLD-grown ZnO-based Microcavities for Bose–Einstein Condensation of Exciton-Polaritons*, Ph.D. thesis, Universität Leipzig (2012).