Ballistic propagation of exciton–polariton condensates in a ZnO-based microcavity

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Abstract. We report on the observation of macroscopically coherent states of exciton–polaritons in a ZnO-based bulk planar microcavity up to 250 K. Excitation power-dependent photoluminescence investigations show a clear threshold behaviour and corresponding spectral narrowing of the emission for negative detunings, revealing clear signatures of a Bose–Einstein condensate. For positive detunings, no condensation occurred but the emission from an electron–hole plasma was detected. Above threshold interscattering phenomena of condensate polaritons between roughly equidistant energy levels have been observed. As a special feature we found ballistic propagation of the condensate in the pump-induced potential landscape, making these ZnO-based microcavities promising candidates for applications based on polariton transport. This effect is caused by strong repulsive interactions in our system, leading to an immense blueshift of the condensate emission and hence to pronounced dynamic effects.

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1. Introduction

Exciton–polaritons are fascinating quasi-particles with bosonic character arising from the strong coupling of excitons with photons in a microcavity (MC). They allow for the occupation of a coherent ground state, the result being a so-called dynamic Bose–Einstein condensate (BEC), thus circumventing the lasing requirement of strong pumping for population inversion \[1\]. First observations of polariton condensation have been made in CdTe- and GaAs-based systems but for a very limited temperature range. A breakthrough was achieved in GaN-based MC showing room temperature polariton lasing \[2\] 4 years ago and later in organic semiconductor MC \[3\]. Even more promising—for devices operating at or above room temperature—is ZnO due to its high exciton binding energy of about 60 meV. Within this material system we observed condensation of exciton–polaritons. Very strong repulsive effects were found, leading to rather large free propagation lengths of several \(\mu\)m of the exciton–polariton condensate and corresponding wave vectors. Thus such MCs are predestined for devices based on polariton transport \[4\].

2. The sample and the experimental setup

2.1. Fabrication

The sample has been grown by pulsed laser deposition using a KrF excimer laser at 248 nm with a pulse length of 25 ns and an energy density of about 2 J cm\(^{-2}\). The bottom distributed Bragg reflector (DBR) was deposited at \(T = 650°C\) and was made of 10.5 layer pairs yttria stabilized zirconia (YSZ) and alumina on a single-side polished \(c\)-plane sapphire substrate rotating at 9 rpm, starting and ending with YSZ. The oxygen background pressure was \(2 \times 10^{-2}\) mbar and \(2 \times 10^{-3}\) mbar for YSZ and alumina, respectively, in order to achieve a low surface roughness of \(R_{\text{rms}} \approx 1\ \text{nm}\) (root mean square) for the DBR. In a second step the ZnO-cavity layer with
Figure 1. (a) AFM image of the cavity surface (ZnO layer) before deposition of the protecting capping layer made of YSZ, (b) $2\theta-\omega$ scan of the MC showing $c$-plane orientation of the cavity and (c) transmission electron microscopy (TEM) bright-field image of the half MC (capped with YSZ and annealed) structure, showing the textured structure and columnar growth of the ZnO-cavity layer. The diffraction pattern in the inset was taken from the encircled region (yellow) in order to estimate the tilt of the ZnO crystallites with respect to the $c$-plane orientation.

an optical thickness of about half a medium wavelength was grown at $T = 150^\circ C$ without substrate rotation, yielding an amorphous but smooth film with thickness gradient. This cavity layer was then capped with the first layer of the top DBR (YSZ), to prevent roughening during the subsequent annealing process at $T = 900^\circ C$ in 700 mbar oxygen for 30 min. After this crystallization step the top DBR was finished consisting of a total of 10.5 layer pairs, identical to the bottom DBR.

2.2. Structural properties

Atomic force microscopy (AFM) and x-ray diffraction (XRD) measurements have been carried out to investigate the structural properties of the ZnO thin film. An AFM image of an area of $2 \mu m^2$ reveals the surface to be rather smooth (figure 1(a)), reflected by the low rms roughness of $R_{rms} = 1.9 \text{ nm}$. The XRD $2\theta-\omega$ scan in figure 1(b) clearly shows the preferential $c$-plane orientation of the ZnO cavity. To further investigate the crystal structure, TEM cross sections have been prepared from a forerunner sample grown under nominally identical conditions. The bright-field images reveal columnar growth of the ZnO crystallites (figure 1(c)). All layers, DBR layers as well as cavity, are textured, the grain size inside YSZ being much larger than inside $Al_2O_3$. The ZnO layer contains the largest grains of about 100 nm in diameter. From the selected area diffraction pattern (see the inset in figure 1(c)) the maximum tilt angle of the
ZnO crystallites can be estimated to be ±12°, confirming the preferential c-plane orientation obtained from XRD data.

2.3. Continuous wave excitation

Due to the wedge-shaped form, our MC allows access to a wide range of detunings $\Delta$ between the cavity and exciton modes ($\Delta = (-70 \cdots +50)$ meV) at $T = 10$ K. Angular-resolved non-resonant cw-photoluminescence (PL) experiments on this sample with a quality factor of about 1000 show the typical features of strong coupling with a maximum coupling strength of about 43 meV ($\Omega_{\text{Rabi}} \approx 80$ meV) at $T = 10$ K slightly decreasing towards room temperature [5]. An excitation power-dependent cw-PL series gave indications that depending on the temperature negative to slightly positive detunings are favourable for achieving condensation [6]. At high excitation powers, heating of the structure started, which manifested itself in a slight redshift of the polariton branch.

2.4. Pulsed excitation

Starting from this knowledge, PL experiments have been carried out using non-resonant and pulsed excitation. For this, the sample was excited using a pulsed Nd:YAG laser at 266 nm with a repetition rate of 8.52 kHz and a pulse length of 500 ps. The laser light is focused onto a spot area of about 10 $\mu$m$^2$. The emitted light was detected confocally and guided to a monochromator with a spectral resolution of 400 $\mu$eV. The so-called Fourier images are recorded with a charge-coupled device camera after imaging the PL signal from the Fourier plane onto the entrance slit of the monochromator, so that the spectral PL intensity distribution versus wave vector $k$ (angular range ±33°) is detected.

3. Exciton–polariton condensation

3.1. Threshold

Typical threshold behaviour of polariton condensation can be seen in figures 2(a) and (b) exemplarily for $T = 130$ K and a detuning of $\Delta \approx -40$ meV. Below a threshold power density of $P_0 \approx 37$ W cm$^{-2}$, the characteristic dispersion of a lower polariton branch (LPB) is observed. The emission of the upper polariton branch is self-absorbed in the ZnO-cavity layer [5, 7]. Above threshold, a nonlinear increase of the emission intensity is observed. The FWHM of the emission decreases more than one order of magnitude at threshold and then slightly increases as a function of pump power. Also the expected blueshift in energy is observed (figure 2(c)). The dispersion of the LPB was fitted with the model for the cavity mode described in [8] and using the coupling strength ($V = 43$ meV) determined from cw measurements in order to determine the energetic position of the uncoupled cavity photon mode and the corresponding detuning. The emission from the polariton condensate is situated well below the cavity mode at $E_{\text{Cav}}(k=0) = 3.325$ eV (see figure 2(c)).

In figure 3(a) Fourier images for selected temperatures are presented for $P = 1.1 P_0$, showing condensation up to $T = 250$ K. The emission from the condensate typically appears spot-like exactly on the LPB or is distributed dispersionless at horizontal lines in $k$-space strictly bounded by the LPB dispersion, thus photon lasing can be excluded. This distinct $k$-space
Figure 2. (a) From the left to the right: Fourier images in a linear colour scale below ($P = 0.4 P_0$), slightly ($P = 1.1 P_0$) and clearly ($P = 1.25 P_0$) above threshold for $T = 130$ K and a detuning of $\Delta \approx -40$ meV with $P_0 = 37$ W cm$^{-2}$, showing the strong increase in PL intensity and the peak narrowing of the condensate emission. The scaling factors are indicated in the figures. (b) Integrated intensity of the condensate and (c) the corresponding blueshift of the emission energy and full-width at half-maximum (FWHM) of the emission. The cavity mode energy is $E_{\text{Cav}}(k_{\parallel} = 0) = 3.325$ eV and the exciton energy is $E_X = 3.365$ eV.

distribution of the emission will be discussed in detail in the next section. The dependence of the threshold power density on temperature and detuning is depicted in figure 3(b). The threshold increases with increasing temperature and less negative detuning due to the decreasing optical trap formed by the LPB. This confirms results from cw studies of the exciton–polariton occupation behaviour in this MC [6]. Comparing these data with the phase diagram for GaN-based MC [9, 10], we find that the available detuning range on our sample allows access to the
thermodynamic relaxation regime, approaching the border to the kinetic regime and therewith the optimal detuning of $\Delta < -50$ meV at $T = 10$ K.

### 3.2. Formation of an electron–hole plasma

At low pump power and negative detuning, polariton condensation can be clearly observed (figure 4(b)). At very high pump power—above the exciton saturation density of $n_X^{\text{sat}} = 1 \times 10^{19}$ cm$^{-3}$—a trace from another emission process arises between 3.30 eV and 3.31 eV (figure 4(c)), characterized by dispersionless lines, distributed in the entire $k$-space. In contrast to that, for positive detunings (figures 4(d)–(f)), no condensation of exciton–polaritons was observed, analogous to observations in GaN-based MC [9, 10] and in contradiction to theoretical predictions for ZnO [11]. In this case the MC instantaneously shows the above described emission appearing at high pump power for negative detunings. We attribute this emission to the formation of an electron–hole (e–h) plasma, which is typically located in the energy range of about 3.31 eV and which is the standard gain mechanism for lasing in ZnO [12]. We explain these dispersionless lines by 3D photonic confinement, where the grains in the cavity layer—which are certainly present as visible in TEM images (figure 1(c))—provide this additional confinement, similar to a random laser [13]. The e–h plasma emission band shifts to lower energies with increasing pump power due to band gap renormalization [12]. The threshold of the emission from the e–h plasma is decreasing with increasing temperature and detuning (not shown here). This is explained by the less effective relaxation of polaritons into the condensate with increasing temperature and detuning, which leads already at lower pump powers to a sufficiently high carrier density in the reservoir for the formation of an e–h plasma.
Figure 4. Fourier images in a linear colour scale for $T = 130$ K and $\Delta \approx -40$ meV for (a)–(c) and $\Delta \approx +40$ meV for (d)–(f). Below threshold, in (a) and (d), emission from the LPB is seen. Above threshold, (b) shows polariton condensation, whereas in (e) the formation of an e–h plasma emission band at about 3.31 eV is observed. At high excitation, (c) shows both condensate and very weak e–h plasma emission whereas in (f) strong e–h plasma emission, is detected. In all figures, $P_0 \approx 37$ W cm$^{-2}$ is the threshold power density for polariton condensation in the negative detuning case. The scaling factors are indicated in the figures.

4. Dynamic effects

4.1. Repulsive interactions and relaxation

The most striking feature in all of our Fourier images—showing a condensate—is that at threshold the condensate energy $E_C$ exhibits an exceptionally large blueshift with respect to the dispersion minimum $E_0$ of the LPB. This indicates the presence of large repulsive interaction potentials in our MC. To more closely analyse this fact, the derivation given by Wouters et al [14] following the Gross–Pitaevskii formalism was used: the repulsive interactions of polaritons of the condensate with each other and with excitonic states including reservoir and bound states as well as the repulsion due to the laser pump create an antitrapping potential which consists of the following three terms [14]:

$$\Delta E = \hbar (g \rho + g_E n_E + G P)$$

$$= \hbar \left( \frac{g}{\gamma_C} + G \right) P + \left( \frac{g_E}{\gamma_E} + \frac{g}{\gamma_C} \right) P_0$$

with $\rho$ being the condensate density, $n_E$ the density of the excitonic states and $P$ the pumping rate. The respective interaction constants are named $g$, $g_E$ and $G$. The rates $\gamma_C$ and $\gamma_E$ determine...
the linear loss of the condensate and the effective relaxation of the excitons, respectively. At the condensate threshold, where $P = P_0$, 
\[ \frac{\Delta E}{P_0} = \hbar \left( \frac{g_E}{\gamma_E} + G \right) \] 
(3)
holds. The strength of the interaction with excitonic states and the laser pump as a function of increasing temperature is decreasing and slightly decreasing with the detuning as visualized in figure 5.

Taking into account the evolution of $\Delta E$ as a function of pump power, a linear approximation of equation (2) to the experimental data yields the two terms in round brackets as slope and offset (not shown here). Our results show that $g_E/\gamma_E - g/\gamma_C \approx g_E/\gamma_E + G$, and we conclude that the repulsive excitonic Coulomb interaction $g_E/\gamma_E$ is the dominating contribution for the observed blueshift of the condensate states described by the Gross–Pitaevskii formalism. The decrease of $g_E/\gamma_E$ with increasing temperature (and detuning) can be understood by increasing thermal radiative and/or non-radiative losses of excitons. Assuming the potential is provided only by reservoir excitons, their density can be estimated within an order of magnitude via [15–17] 
\[ n_{\text{res}} \approx \frac{\Delta E}{15 E_X^b a_B^3} \] 
(4)
with the free exciton binding energy $E_X^b = 60 \text{ meV}$ and the Bohr radius $a_B = 1.8 \text{ nm}$. Density values at threshold range from one to half an order of magnitude below the exciton saturation density $n_{\text{sat}}^X = 1 \times 10^{19} \text{ cm}^{-3}$ [15]. Therefore we conclude that additional excitonic states such as excitons localized within a disorder potential or on impurities or dark excitons play a distinct role and that the threshold reservoir density is overestimated by equation (4). Note that donor bound excitons play an important role in ZnO [12]. At this point, the significant role of disorder in our sample should be elucidated further. Studies concerning the Bose glass phase transition towards superfluidity would be very interesting, as discussed in [18].

Figure 5. Interaction terms with excitonic states and the pump-induced potential as a function of temperature and detuning deduced from $\Delta E$ at threshold and above (showing that excitonic states dominate our system).
Above threshold, more and more dispersionless lines of condensate states, exhibiting roughly equidistant energy spacing, appear within the dispersion of the LPB. With increasing pump power, relaxation of condensate polaritons into lower levels can be observed (figures 6(a)–(d)). This effect was explained by the picture of polaritons relaxing down the strongly inhomogeneous pump potential by steps of energy $\Delta E$ (see figure 2(a) in [19]). The model established in [19] shows that the net scattering rate from a mode at $\omega'$ to a mode at $\omega$ can be expanded:

$$r(\omega, \omega') = \kappa \times (\omega' - \omega).$$  \hspace{1cm} (5)

Comparing this with the rate given by the linear loss rate times an effective spot area (the area of the excitation spot is the same for our case and in [19]), the relaxation constant $\kappa$ can be deduced. To access experimental values, the Fourier images have been summed over all wave vectors $k$ (figure 6(e)) shown exemplarily for $T = 130$ K and $\Delta \approx -40$ meV. Doing so, relaxation constants for some $T$ and $\Delta$ can be estimated being about five times larger compared...
Figure 7. Condensate forming at the edge of the LPB dispersion at $\hbar \omega_C$ for (a) $(T, \Delta) = (10 K, -50 \text{ meV})$, (b) $(T, \Delta) = (10 K, -30 \text{ meV})$ and (c) $(T, \Delta) = (70 K, -45 \text{ meV})$ slightly above threshold with threshold power densities of $P_0 \approx 5, 50$ and $40 \text{ W cm}^{-2}$, respectively. Panel (a) is strikingly similar to figure 1(e) in [14]. Emission from non-condensed lower polaritons is superposed on the sharp and intense emission from condensed polaritons. Scaling factors are indicated in the figures.

to $\kappa \approx 0.01 \mu\text{m}^2$ in GaAs wires [19], which despite the shorter lifetime in ZnO allows for the observation of these scattering processes.

4.2. Propagation of exciton–polariton condensates

In the following, several characteristic effects emerging in the $k$-space distribution (e.g. shown in figure 3(a)) at different temperatures and detunings will be explained. There are two opposite characteristics emerging in the spectra: for low temperature and very negative detuning, the formation of a condensate starts symmetrically at a finite $k$ and $-k$ of the LPB with a corresponding energy $E_C$. This emission is supposed to be present for the entire elastic $k$-circle, but due to our experimental setup spectra are recorded along a $-k \ldots k$-line only (see section 2.4). For higher temperatures and less negative detuning, spot-like emission on dispersionless lines is situated in between the LPB dispersion. Superposed on each of these cases, emission from non-condensed polaritons, following the LPB dispersion, is detected.

The symmetric formation of the condensate at the edge ($k$ and $-k$) of the LPB (figure 7) is caused by ballistically propagating condensate polaritons. This effect has been discussed in detail by Wouters et al [14] for CdTe-based systems who applied a mean field theory in the Gross–Pitaevskii formalism. It arises when the pump spot size at threshold is smaller than the propagation length of the condensate polaritons. The strong potential due to the repulsive interactions between excitons in the reservoir accelerates the condensate polaritons away from the centre of the pump spot with a Gaussian intensity distribution and they can propagate ballistically at a wave vector $k_{\text{max}} = k(E_C)$. In this model, the appearance of the condensate
at finite \( k \)-vector can be described by means of a propagation length \( L \), which is given by [14]

\[
L = \frac{2}{\gamma C} \sqrt{\frac{2(E_C - E_0)}{m_{\text{eff}}}} \tag{6}
\]

with \( m_{\text{eff}} \) being the effective mass of the LPB. The polariton lifetime was taken from the FWHM of the observed condensate state. The effective mass was determined from the curvature of the LPB at \( k = 0 \) for the respective detuning. The influence of the change of the effective mass as a function of \( k \) on the propagation length should be negligible, as in the discussed \( k \)-range we stay in a predominantly photonic regime. We found the maximum value for \( T = 10 \text{ K} \) to be \( L \approx 7 \mu \text{m} \). The propagation length decreases with increasing temperature and detuning, on the one hand due to a decrease in the lifetime and an increase in \( m_{\text{eff}} \) caused by a more excitonic character of the polaritons and on the other hand due to decreasing \( \Delta E \). These huge values obtained are in the same order of magnitude as the propagation lengths in GaAs wires, which can be estimated from the data in [20]. This fact is due to a trade-off of the larger energy offset generated by the huge exciton densities in our case and the higher lifetime, arising from the quite perfect MC structure in their GaAs wires. In our cavity the large interaction potential seems to naturally provide such dynamic condensates. This interesting effect can be exploited for polariton transport applications in devices with the future vision of establishing polariton neurons [4].

According to [14], for a pump laser spot larger than the characteristic propagation length of the condensate polaritons their emission should be distributed at \( E_C \) in the entire region within the LPB dispersion showing maximum intensity for \( k = 0 \) and decreasing towards the edge of the LPB dispersion. But in our case the spot-like emission in between the LPB can be explained by photonic disorder, which we of course expect from the TEM results of the reference sample and which lead to interference of the emission from different spatial positions.

Superposed on the sharp emission, non-condensed polaritons are detected. Their experimentally observed behaviour clearly follows the expectations from theoretical considerations [14]: when the threshold power is quite low, non-condensed polaritons are following the linear regime dispersion because most of them are outside of the high-density region located in the very centre of the excitation spot (see figure 7(a)). When the threshold power is higher (see figures 7(b) and (c)), more and more non-condensed polaritons are located in grains with different higher densities, which leads to various blueshifts and therefore to a broadening of the dispersion towards higher energies (see figure 7(b)). However, for increasing \( k \) respective \( E \), this broadening effect decreases as non-condensed polaritons are more and more restricted to states on the linear regime dispersion, because they are faster and are therefore spending most of their lifetime in the outer low-density region.

5. Summary

In conclusion, we have observed dynamic BEC of exciton–polaritons in a ZnO-based MC for negative detunings up to \( T = 250 \text{ K} \). For positive detuning, only emission from an e–h plasma was detected. For very negative detunings, condensate polaritons propagate ballistically with propagation lengths of several \( \mu \text{m} \) in the pump-induced potential. The corresponding large \( k_{\text{max}} \) is naturally provided by the large blueshift already present at threshold caused predominantly by
repulsive excitonic Coulomb interaction. This unique property makes ZnO-based MC promising candidates for polariton transport e.g. in waveguides. The presence of disorder in our MC could serve for studies of the Bose glass phase [18]. We further could observe efficient relaxation mechanisms between different condensate states in our sample.

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