Polariton-condensation effects on photoluminescence properties in a CuBr microcavity

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Abstract. We have investigated the photoluminescence (PL) properties in a CuBr microcavity with HfO$_2$/SiO$_2$ distributed Bragg reflectors at 10 K from the viewpoint of cavity-polariton condensation. From the excitation-power dependence of PL spectra detected at an in-plane wave vector of $k_{//}=0$, we found that the PL characteristics of the lower polariton branch markedly change with a threshold nature: a drastic increase (decrease) in the intensity (band width). In addition, the PL energy exhibits a large blueshift, ~10 meV, around the threshold excitation power, reflecting strong polariton-polariton interactions. The blueshifted PL energy is far below the bottom energy of the cavity photon. In addition, the estimated density of photogenerated electron-hole pairs at the threshold excitation power is two orders lower than the Mott-transition density for the formation of electron-hole plasma. The above results consistently demonstrate the occurrence of the cavity-polariton condensation.

1. Introduction

Exciton polaritons in semiconductor microcavities, the so-called cavity polaritons, are bosonic quasiparticles resulting from the strong coupling between excitons and cavity photons. The bosonic properties have attracted much attention in Bose-Einstein condensation [1-6] and polariton lasing from polariton condensates without population inversion [7-9]. Recently, wide-gap semiconductors such as GaN [4, 7, 8], ZnO [6, 9-12], and copper halides (CuCl, CuI, and CuBr) [13-15] have been adopted as the active layers of microcavities because the excitons have large binding energies leading to high thermal stability of the cavity polariton: This is advantageous in applications of microcavities. In our previous works on copper halide microcavities [13-15], we demonstrated giant Rabi splitting energies of the order of 100 meV and room-temperature stability of the cavity polaritons. Especially, in a CuBr microcavity, we found that there is negligible population of the cavity polaritons in the bottleneck region under a weak excitation condition [15]. This fact suggests that the relaxation process of the cavity polaritons in the lower polariton branch (LPB) is hardly affected by a bottleneck effect, which is favorable for the cavity-polariton condensation.

In the present work, we have focused on the polariton-condensation effects on PL properties in a CuBr microcavity with HfO$_2$/SiO$_2$ distributed Bragg reflectors (DBRs) at 10 K. With an increase in excitation power, the intensity (width) of the PL band of the LPB at $k_{//}=0$ abruptly increases (decreases) with a threshold nature accompanying with a large blueshift, ~10 meV, due to polariton-polariton interactions. These phenomena reflect the cavity-polariton condensation. We discuss the stability of the cavity-polariton condensation from aspects of the Mott transition for the formation of...
electron-hole plasma and a change from the strong coupling to the weak coupling due to the large blueshift of the LPB energy.

2. Experimental details
The sample of a CuBr microcavity with HfO$_2$/SiO$_2$ DBRs was prepared on a (0001) Al$_2$O$_3$ substrate. The thickness of the CuBr active layer was $\lambda$, which is given by $\lambda = \lambda_{EX} / \sqrt{\epsilon}$, corresponding to an effective resonant wavelength of the lowest-lying exciton, where $\lambda_{EX}$ is the resonant wavelength of the $Z_1$ exciton, 418.5 nm [15], and $\epsilon$ is the background dielectric constant, 4.06 [16] in CuBr: $\lambda=208$ nm. The bottom and top DBRs consisted of 17.5 and 12.5 periods, respectively, and each DBR was terminated by the HfO$_2$ layer. The HfO$_2$ and SiO$_2$ layers were fabricated by rf magnetron sputtering. The CuBr active layer, which corresponds to the cavity in this case, the so-called bulk cavity, was grown by vacuum deposition in 2×10$^{-6}$ Pa. The quality factor of the microcavity was estimated to be $Q=810$. It was confirmed from X-ray diffraction patterns that the crystalline CuBr layer is just oriented along the [111] direction. The details of the sample preparation were described in Ref. [15].

In the PL measurements, the excitation light source was a mode-locked Ti:sapphire laser with a pulse duration of 110 fs and a repetition rate of 76 MHz. The energy of the fundamental laser light was doubled for the excitation: 3.351 eV that is higher than the LPB energy at an in-plane wave vector $k_0=0$ by ~0.45 eV. The incident angle of the excitation light was normal to the sample surface, $\theta=0^\circ$, and the spot diameter on the sample surface was ~30 $\mu$m. The detection angle of the PL was fixed to $\theta=0^\circ$, which corresponds to detecting the PL at $k_\parallel=0$. A spread of the detection angle was ±2°. The PL spectra were detected with a cooled charge couple device attached to a 32-cm single monochromator with a resolution of 0.15 nm. In addition, angle-resolved reflectance spectra were measured to characterize the cavity-polariton dispersions. All the optical measurements were performed at 10 K.

3. Results and discussion
At first, we describe the fundamental properties of the cavity polaritons. In a CuBr crystal, there exist three kinds of excitons labeled $Z_6$, $Z_{1,2}$, and $Z_3$ in order of energy. The $Z_6$ exciton peculiar to CuBr corresponds to a triplet exciton, the oscillator strength of which is markedly enhanced by mixing between the triplet and singlet excitons [17]. The $Z_{1,2}$ ($Z_3$) exciton is attributed to the degenerate heavy-hole and light-hole excitons (the split-off-hole exciton). Thus, the strong coupling between the $Z_6$, $Z_{1,2}$, and $Z_3$ excitons and cavity photon produces four cavity-polariton branches: the LPB, middle polariton branch 1 (MPB1), MPB2, and upper polariton branch (UPB). Figure 1 shows the incident angle dependence of the energies of observed reflectance dips due to the LPB, MPB1, and MPB2, which are indicated by the solid circles, open circles, and solid squares, respectively. The solid curves indicate the results of the analysis of the cavity-polariton dispersions with use of a phenomenological Hamiltonian for the strong coupling between the three excitons and cavity photon: The details of the analysis are described in Ref. [15]. The dashed horizontal lines and dashed curve show the exciton energies [15] and calculated cavity-photon dispersion, respectively. The bottom energy of the cavity photon is $E_0=2.977$ eV, and the detuning from the $Z_6$-exciton energy is $\Delta=+13$ meV. The Rabi splitting energies were estimated to be 33, 114, and 89 meV for the $Z_6$, $Z_{1,2}$ and $Z_3$ excitons, respectively.

Hereafter, we discuss the PL properties in the CuBr microcavity. Figure 2 shows the PL spectra at various excitation-power densities, where the reflectance spectrum at $\theta=0^\circ$ is depicted on the top for reference. At the lowest excitation-power density of 0.4 $\mu$J/cm$^2$, the PL-peak energy just agrees with the reflectance-dip energy of the LPB. This demonstrates that the PL band originates from the LPB at $k_\parallel=0$. With an increase in excitation-power density, the PL-peak energy exhibits a remarkable blueshift due to polariton-polariton interactions [1-3, 6-9, 18]. At the excitation-power density of 6.0 $\mu$J/cm$^2$, the intensity and width of the PL band suddenly increases and decreases, respectively, in comparison with those at 5.2 $\mu$J/cm$^2$. The drastic changes of the PL properties are discussed below.

Figures 3(a) and 3(b) show the integrated intensity and full width at half maximum (FWHM) of the LPB-PL band, respectively, at $k_\parallel=0$ as a function of excitation-power density. It is evident that the PL
The intensity exhibits a threshold-like increase at 6.0 \( \mu \text{J/cm}^2 \). The dashed line in Fig. 3(a) indicates the fitted result below the threshold, which exhibits almost linear excitation-power-density dependence of the PL intensity: 

\[
I_{PL} \propto P^{1.2}
\]

The narrowing of the FWHM just correlates with the threshold-like increase of the PL intensity. These facts indicate a drastic increase of the cavity-polariton population in the LPB at \( k_{//}=0 \) and population shrinkage in momentum space; namely, the cavity-polariton condensation occurs [1-6]. The extent of PL-band narrowing is not so marked. This seems to be due to the relatively low quality factor and/or coexistence of condensates and non-condensates.

Figure 3(c) shows the peak energy of the LPB-PL band at \( k_{//}=0 \) as a function of excitation-power density. The blueshift is considerably larger than those in the previous reports, a few meV [1-3, 6-9]; namely, the polariton-polariton interaction in the CuBr microcavity is relatively strong. The highest PL-peak energy is 2.925 eV, which is far below the cavity-photon energy at \( k_{//}=0 \): \( E_0=2.977 \text{ eV} \). This fact indicates that the strong coupling is fully kept in spite of the large blueshift, which is owing to the giant Rabi splitting energy. According to Ref. [18], the blueshift of the PL energy is approximately proportional to the cavity-polariton density at \( k_{//}=0 \) in the LPB. It is evident from Fig. 3(c) that the blueshift remarkably increases around the threshold excitation-power density, 6.0 \( \mu \text{J/cm}^2 \). This behavior reflects a drastic increase of the cavity-polariton density at \( k_{//}=0 \), which is consistent with the

**Figure 1.** Incident angle dependence of the energies of observed reflectance dips (solid circles, open circles, and solid squares). The solid curves indicate the results of the analysis of the cavity-polariton dispersions.

**Figure 2.** PL spectra at various excitation-power densities, where the reflectance spectrum at \( \theta=0^\circ \) is depicted on the top for reference.

**Figure 3.** (a) Integrated intensity, (b) FWHM, and (c) peak energy of the LPB-PL band at \( k_{//}=0 \) as a function excitation-power density. The solid lines are guides for the eye. The dashed line in (a) indicates the fitted result below the threshold.
excitation-power-density dependence of the PL intensity shown in Fig. 3(a). We have to consider the Mott transition in a high density regime. According to Ref. [19], the Mott-transition density is given by

\[ n_M = \frac{(2/9)^{3/4}}{\pi a_B^3} \]

in the framework of the random phase approximation, where \( a_B \) is the exciton Bohr radius, 1.25 nm in CuBr [16]; \( n_M = 2.7 \times 10^{19} \text{ cm}^{-3} \). Taking account of an absorption coefficient of CuBr, 8.2 \( \times 10^4 \text{ cm}^{-1} \) [20] at the excitation energy, the density of photogenerated electron-hole pairs is estimated to be \( \sim 4.4 \times 10^{17} \text{ cm}^{-3} \) at the threshold excitation-power density, which is two orders lower than the Mott-transition density. Thus, the Mott transition is fully negligible in the present case.

4. Conclusions

We have investigated the polariton-condensation effects on the PL properties in the CuBr microcavity. The excitation-power dependence of the intensity and width of the LPB-PL band at \( k_z = 0 \) exhibits the threshold-like behavior: a drastic increase (decrease) in the intensity (width). A large blueshift of the PL-peak energy, \( \sim 10 \text{ meV} \), due to the polariton-polariton interaction correlates with the excitation-power dependence of the PL intensity. The blueshifted energy of the LPB PL at the threshold excitation power is \( \sim 50 \text{ meV} \) lower than the bottom energy of the cavity photon. This indicates that the strong coupling is fully kept. The estimated density of photogenerated electron-hole pairs at the threshold excitation power is two orders smaller than the expected Mott-transition density. Consequently, the PL properties described above consistently demonstrate the occurrence of the cavity-polariton condensation in the CuBr microcavity.

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