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Preparation and properties of ZnSe/(Zn, Cd)Se multi-quantum-well microcavities for room temperature polariton emission

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Abstract

Due to their large oscillator strengths, ZnSe microcavities with (Zn, Cd)Se quantum wells are particularly suited for investigation of the photon–exciton coupling behaviour in semiconductors. We have observed a strong coupling between the excitonic and photonic modes in a ZnSe microcavity with four (Zn, Cd)Se quantum wells and distributed Bragg mirrors of ZnS and YF_3. A very large Rabi splitting $\hbar\Omega > 40$ meV was observed in temperature dependent photoluminescence investigations.

1. Introduction

Semiconductor quantum wells as parts of microcavities allow the study of the interconversion between excitons and photons, which in the strong coupling regime is manifested as a Rabi splitting into two resonance features of the optical spectrum. The coupling between excitons and photons generates quasi-particles known as polaritons. These particles have some unusual properties, which open the possibility of developing new kinds of efficient light emitters or quantum processors [1, 2].

For room temperature applications the Rabi splitting must exceed the spectral broadening of the excitons by interaction with phonons and the broadening of the cavity mode. Since the Rabi splitting is proportional to the oscillator strength, this condition is excellently satisfied with ZnSe based quantum structures.

After the first observation of a polariton splitting in a Fabry–Perot microcavity [3, 4], the strong coupling regime of quantum well excitons was studied by various spectroscopy methods for III–V and II–VI semiconductor microcavities [5–8]. Kelkar *et al* [9] reported a Rabi splitting of 17.5 meV at 70 K and 10 meV at 175 K in a (Zn, Mg)(S, Se) microcavity with three (Zn, Cd)Se quantum wells as the resonant medium and dielectric Bragg mirrors of SiO_2/TiO_2. André *et al* [10] have investigated CdTe multi-quantum-well structures enclosed in

(Cd, Mg)Te/(Cd, Mn)Te semiconductor Bragg mirrors showing Rabi splitting energies between 12 and 26 meV at 4 K as a function of the number of quantum wells.

Recently Saba *et al* [11, 12] have measured parametric polariton amplification up to 220 K in CdTe based multi-quantum-well microcavity structures. The experimental observations demonstrate that the polariton amplification cut-off temperature is directly related to the exciton binding energy. ZnSe and CdSe based materials combine a large Rabi splitting energy and a high exciton binding energy of about 40 meV. Therefore these compounds are particularly suited for microcavity applications such as in polariton amplification devices.

In this paper, we report a large Rabi splitting at room temperature, which was realized in a ZnSe microcavity containing four strained (Zn, Cd)Se quantum wells as the resonant medium. We used dielectric ZnS and YF_3 distributed Bragg mirrors (DBRs). Temperature dependent photoluminescence measurements reveal clear evidence of the strong coupling between the photonic mode (PM) and the excitonic mode (EM) in our microcavities, yielding a Rabi splitting $\hbar\Omega_{\mathrm{Rabi}} > 40$ meV at room temperature.

2. Experimental set-up

ZnSe/(Zn, Cd)Se multi-quantum-well (MQW) structures were grown on (001) GaAs by molecular beam epitaxy (MBE) at $T = 310\,^{\circ}\mathrm{C}$. The nominal layer thickness of the sample was about 195 nm, which is identical to the optical length of the 528 nm emission in (Zn, Cd)Se quantum wells. A cavity size gradient across the sample of about 2 nm mm^{-1} sample length was realized by placing the substrate at an appropriate angle with respect to the effusion cells. Four (Zn, Cd)Se quantum wells with a cadmium content of $x = 0.34 \pm 0.02$ and a thickness of $d = 7$ nm were placed near the antinodes of the standing wave in the microcavity. After the growth of the microcavity an eightfold stack of ZnS and YF_3 DBRs was deposited by thermal evaporation on top of the MQW structures. Then the GaAs substrate was removed by wet etching and the microcavity structure was completed by a sixfold stack of DBRs on the back.

The sample structure was characterized by high resolution x-ray diffraction (HRXRD) in triple-axis mode and simulation of the x-ray data by dynamic diffraction theory [13]. The photoluminescence was measured using a HeCd laser at $\lambda = 325$ nm with an excitation density of approximately 1 W cm^{-2}.

3. Results and discussion

Figure 1 shows a cross-section drawing of our microcavity structure. Four (Zn, Cd)Se quantum wells enclosed in ZnSe barriers represent the resonant medium. The cadmium content $x = 0.34$ and the well size $d = 7$ nm were obtained from high resolution x-ray diffraction. During MBE growth the sample was placed at an appropriate angle with respect to the effusion cells to generate a cavity length gradient in the structure, which is schematically indicated in figure 1.

The room temperature quantum well photoluminescence (PL) was measured at $E_{\mathrm{QW}} = 2.3$ eV (539 nm) with a FWHM of 27 meV. With the finite barrier model and the structural parameters obtained by means of x-ray diffraction, we calculated a quantum well transition of $E_{\mathrm{QW}} = 2.34$ eV (530 nm), which is in good agreement with the experimental data.

An analysis of the strain status of the microcavity structures was carried out, measuring the $(\bar{2}\bar{2}4)$ reciprocal space map by means of high resolution x-ray diffraction before mirror deposition and substrate removal. The results are shown in figure 2. It was found that the (Zn, Cd)Se quantum wells are fully strained on ZnSe, whereas the ZnSe barriers are partially relaxed with respect to the GaAs substrate.

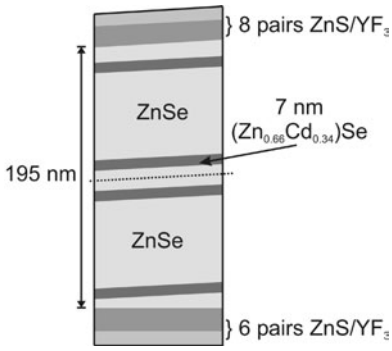

Figure 1. A cross-section drawing of the microcavity structure. Four (Zn, Cd)Se quantum wells embedded in ZnSe barriers represent the resonant medium. The cadmium mole fraction $x = 0.34$ and quantum well width $d = 7$ nm were obtained by x-ray measurements. The cavity length gradient allows the tuning of the cavity mode.

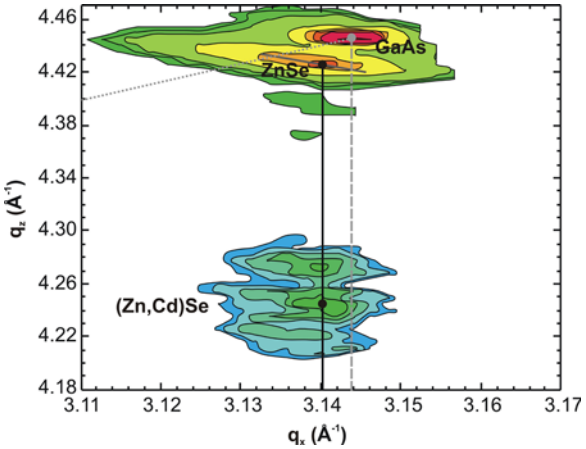

Figure 2. A $(\bar{2}\bar{2}4)$ reciprocal space map of the sample before mirror deposition. The ZnSe barrier layers are partially relaxed with respect to the GaAs substrate (black vertical line, dashed grey line). The (Zn, Cd)Se quantum wells are lattice matched on the ZnSe barriers.

(This figure is in colour only in the electronic version)

For the investigation of the photon–exciton coupling in this structure we measured temperature dependent PL between 270 and 330 K near the resonance of the PM and EM. In these experiments the cavity mode energy (PM) was kept constant, while the variation of the temperature leads to a shift of the quantum well emission energy (EM) into the resonance.

The PL spectra measured at different temperatures are shown in figure 3. With the temperature increasing from 270 to 330 K the QW emission energy shifts to lower energy by about 1 meV K^{-1}. This energy is indicated by the black arrows for each spectrum in figure 3. Therefore the EM approaches a fixed PM (dashed line, given by the geometrical cavity length of about $L_C = 200.5$ nm) and reaches the resonance condition at about 300 K. The luminescence peaks show a clear anticrossing behaviour. The minimal energy difference between the two peaks of about $\hbar\Omega_{\text{Rabi}} = 44$ meV is the Rabi splitting energy.

The dots in figure 4 show the peak energies of the polariton luminescence versus temperature. The full curves in figure 4 are calculated using a model of the polariton dispersion

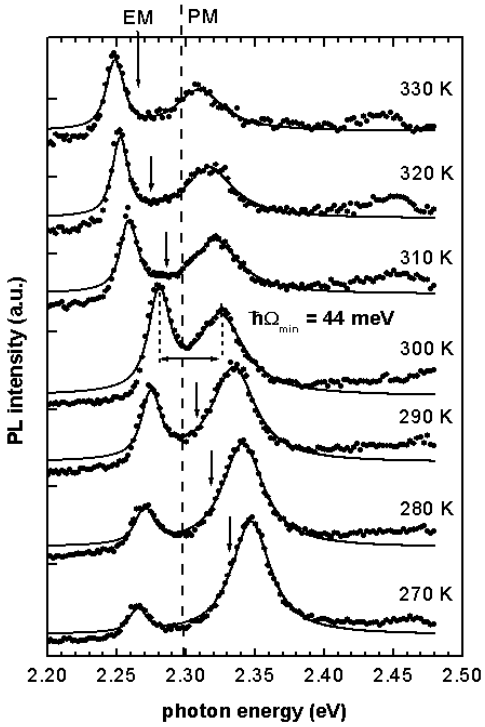

Figure 3. PL spectra of the microcavity near the resonance condition measured between 270 and 330 K (dots). The full curves represent Lorentzian fits. The PM is fixed at $E_P = 2.298$ eV (dashed line) and the EM, indicated by the arrows, shifts to lower energy with increasing temperature. The experimental data show a clear anticrossing behaviour of the polariton luminescence peaks. A minimum Rabi splitting of $\hbar\Omega_{min} = 44$ meV was detected at 300 K.

given by André *et al* [10]. The polariton dispersion $E_{pol\pm}$ is given by

$$E_{pol\pm} = \tfrac{1}{2}(E_X + E_P) \pm \tfrac{1}{2}[\hbar\Omega_{Rabi}^2 + (E_X - E_P)^2]^{1/2}, \tag{1}$$

where E_X and E_P are the uncoupled dispersions of the EM and the PM, respectively, and $\hbar\Omega_{Rabi}$ is the Rabi splitting energy. The parameters which were used in the calculation are $E_X = 2.627 - 1.1 \times 10^{-3} T$ (eV) for the temperature dependence of the QW emission (between 270 and 330 K) and $E_P = 2.298$ eV for the cavity mode energy, respectively. The fitting procedure yields $\hbar\Omega_{Rabi} = 45$ meV, which is in agreement with our experimental results and confirms the existence of the strong coupling at room temperature in our microcavity structure.

The square of the Rabi splitting energy $\hbar\Omega_{Rabi}$ in a microcavity is proportional to the number of quantum wells N_{QW}, the oscillator strength f_{osz} and the inverse of the effective cavity length L_C [14]. Kelkar *et al* [9] measured the low temperature Rabi splitting of a $4\times$ λ-cavity containing three ZnSe/$(Zn_{0.76}Cd_{0.24})$Se quantum wells. Using their value of 17.5 meV and considering an effective cavity length of about 800 nm calculated in our structure, we obtain approximately $\hbar\Omega_{Rabi} = 32$ meV. The Rabi splitting energy in our sample is significantly larger, indicating that the oscillator strength in our sample is larger than in the samples discussed in [9]. The difference may be due to an increased exciton binding energy and improved quantum well confinement in our sample, since the cadmium mole fraction is higher than in the sample used by Kelkar *et al*.

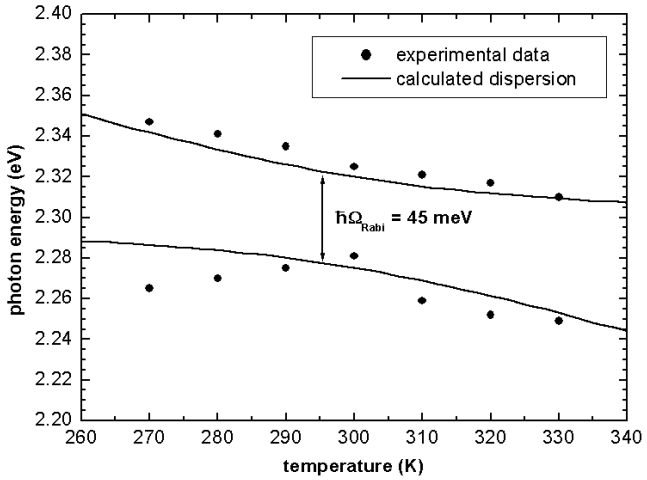

Figure 4. Polariton luminescence peak energies as a function of temperature (dots). The curves are the calculated polariton dispersion branches $E_{\mathrm{pol}+}$ and $E_{\mathrm{pol}-}$ with a Rabi splitting energy of $\hbar\Omega_{\mathrm{Rabi}} = 45$ meV.

Further, the 'splitting-to-linewidth ratio' is an important factor for achieving strong coupling. The Rabi splitting energy must exceed the averaged linewidth of the PM and EM:

$$\tfrac{1}{2}(\Delta E_{\mathrm{X}} + \Delta E_{\mathrm{P}}) < \hbar\Omega_{\mathrm{Rabi}}. \tag{2}$$

In the case of our microcavity we calculated an averaged linewidth of about 22 meV. This results in a 'splitting-to-linewidth ratio' of about two and the condition given by equation (2) is sufficiently fulfilled.

4. Conclusion

$ZnSe/(Zn_{0.66}Cd_{0.34})Se$ MQW cavity structures, which were covered by six pairs of ZnS and YF_3 DBRs on top and eight pairs of the same material on the back, were grown.

X-ray diffraction shows the high quality of the interfaces of the fully strained quantum wells, whereas the ZnSe barrier layers are partially relaxed. Photoluminescence investigations at room temperature reveal a clear quantum well transition at $E_{\mathrm{X}} = 2.3$ eV with a FWHM of 27 meV.

The polariton emission of the microcavity was investigated by cavity detuning. The EM was shifted into resonance with the PM via the temperature dependence of the semiconductor band gaps. Photoluminescence was measured between 270 and 330 K. The spectra show clear anticrossing behaviour of the polariton emission, yielding a Rabi splitting energy of about 44 meV. The temperature dependent polariton luminescence is in agreement with theoretical calculations. Further investigations such as absorption measurements are required to confirm the large oscillator strength in the quantum wells.

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