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Temperature-Dependent Photoluminescence in Coupling Structures of CdSe Quantum Dots and a ZnCdSe Quantum Well *

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A coupling structure of CdSe quantum dots (QDs) and a ZnCdSe quantum well (QW) is fabricated by using the molecular-beam epitaxy technique. The effect of temperature on the photoluminescence (PL) of the structure is studied. The results reveal that the activation energy of exciton dissociation in the coupling QDs/QW structure is much higher than that of simple CdSe QDs, which is attributed to the exciton tunnelling from the QW to QDs through a thin ZnSe barrier layer. The results also reveal that the position and width of the emission band of the QDs vary discontinuously at certain temperatures. This phenomenon is explained by the QD ionization and exciton tunnelling from the QW to the QDs. It is demonstrated that the coupling structure significantly improves the PL intensity of CdSe QDs.

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Ⅲ-V Quantum-dot (QD) lasers have been extensively investigated in last decades because of their low threshold current density, high differential gain, and ultrahigh temperature stability.[1-3] In contrast, Ⅱ-Ⅵ QD laser that possesses shorter wavelength is difficult to achieve the same performance as Ⅲ-V QD lasers. One reason is that it is difficult to form high QD density in Ⅱ-Ⅳ semiconductors. The density of Ⅱ-Ⅵ QDs is usually lower than that of Ⅲ-V QDs by two orders of magnitude. Therefore, it is difficult to make high-performance Ⅱ-Ⅵ QD lasing when a simple Ⅱ-Ⅵ QD structure is used. Previous studies on Ⅲ-V semiconductors[4-9] revealed that the PL behaviour of QDs can be significantly improved by using coupling QDs/QW structures. However, little attention has been paid to the similar structure of Ⅱ-Ⅵ semiconductors.[10,11]

In this study, a coupling structure of CdSe QDs and a $Zn_{0.72}Cd_{0.28}Se$ quantum well (QW) with ZnSe as the barrier layer were fabricated. The CdSe/ZnSe system is a promising candidate for light emitting devices in blue-green regions since CdSe QDs can be easily formed on the ZnSe layer via self-organization. The effect of tunnelling and thermal dissociation of excitons on the recombination of excitons in the QDs is studied by measuring the photoluminescence of QDs/QW structures at different temperatures. The changes in PL intensities, PL peak positions and width of the CdSe QDs as a function of temperature are discussed. The results demonstrate that the light emission of the CdSe QDs can be significantly improved by exciton tunnelling processes from the ZnCdSe QW to CdSe QDs.

The CdSe QDs/ZnCdSe QW samples were grown by molecular-beam epitaxy (MBE). A 1-μm-thick ZnSe buffer layer, followed by ten periods of QDs/QW structures and a 120 nm ZnSe capping layer were epitaxy successively on a (100)-oriented GaAs substrate. The coupling QDs/QW structure consists of a self-assembled CdSe QDs layer (three monolayers), a ZnSe barrier layer (L_b) with three kinds of different thicknesses, a 5-nm-thick $Zn_{0.72}Cd_{0.28}Se$ QW layer and a ZnSe separation layer with the same thickness as the ZnSe barrier layer. An atomic force microscopy (AFM) image of the uncapped surface of the CdSe QD layer shows that the average height, base diameter and density of dots are 10 nm, 10 nm and ten dots per μm^2, respectively.[12]

The room-temperature (RT) PL spectra of the three samples were measured by a UV-Lamb micro Raman spectrometer under the excitation of 514.5 nm of an Ar^+ laser. The PL spectra of the samples at different temperatures were also measured under the excitation of 488 nm of an Ar^+ laser in the temperature range 80–300 K. The excited area is 20 μm^2. The temperature of the specimens was controlled by a cold-finger cryostat.

Figure 1 shows the room-temperature PL spectra of the three samples under the excitation of the 514.5 nm line, which is close to the absorption edge of the ZnCdSe QW. The intensity of the PL emission is normalized to that of the CdSe QDs. It can be seen

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that all spectra contain three peaks: the peak around 2.316 eV corresponds to the exciton recombination in the ZnCdSe QW,[13] the broad emission band centered at 2.05 eV arises from the exciton recombination in CdSe QDs, and the strong and sharp peak at 2.345 eV comes from the secondary longitudinal optical (LO) phonon Raman scattering line from ZnSe interface. The broadening of the CdSe QDs emission peak results from the variation of size and spatial distribution of the QDs. It is worth noting that with the decreasing thickness of the ZnSe barrier layer, the PL intensity of the QW decreases, whereas that of the QDs increases. The inset of Fig. 1 shows the ratio of the integrated intensity of the exciton emission of the CdSe QDs to that of the ZnCdSe QW, I_{QDs}/I_{QW}, as a function of L_b. It can be seen that I_{QDs}/I_{QW} increases significantly with the decreasing L_b. This strongly suggests a rearrangement of excitons between CdSe QDs and ZnCdSe QW via the exciton tunnelling from the QW to the QDs. The PL intensity of the CdSe QDs in sample A is significantly stronger than that of the simple CdSe QD structure. The improvement in PL can be attributed to the exciton collection and injection by the QW: when the carriers are thermally excited from QDs, they are recollected by the QW and injected back into the QDs, and this process is repeated until recombination occurs in QDs. Consequently, the QW plays a major role in confining the excitons in the QDs and improving the PL.

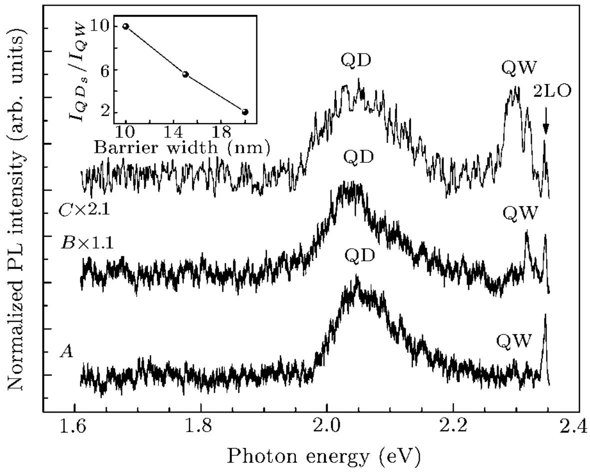

Fig. 1. Normalized PL spectra of samples A, B and C. The inset shows the ratio of the integrated intensity I_{QDs}/I_{QW} as a function of the barrier width L_b.

In order to determine the effect of temperature on exciton recombination processes in the coupling QDs/QW structure, the PL spectra of sample A were measured at different temperatures in the temperature range 80–300 K under the excitation of the 488 nm line of a cw Ar^+ laser, as shown in Fig. 2. Also, the broad strong emission peak around 2.0 eV comes from the CdSe QDs. The emission peaks located at 2.341 and 2.426 eV correspond to the emission from the CdSe wetting layer (WL) and the ZnCdSe QW, respectively. The three peaks at 2.451, 2.481, and 2.510 eV are ascribed to the LO phonon replica lines of ZnSe.

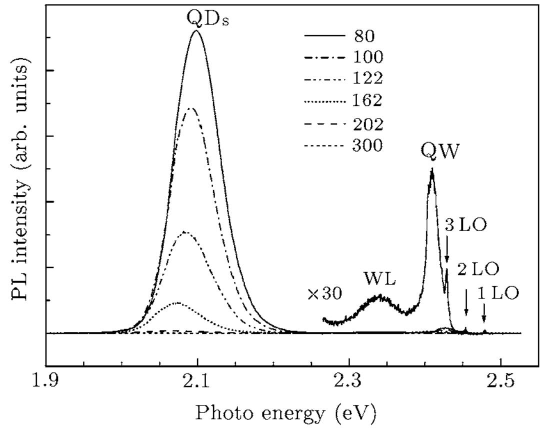

Fig. 2. PL spectra of sample A at different temperatures.

The integrated PL intensity of the CdSe QDs as a function of temperature is displayed in Fig. 3. For wide-gap II-VI semiconductors, the LO phonon can dramatically increase the exciton-tunnelling process,[14] thus increasing temperature could enhance the tunneling rate.[15] However, temperature can also strongly influence the stability of the excitons. The probability of thermal dissociation of excitons increases with temperature, which will hasten the escape of excitons from QDs and will recombine elsewhere radiatively or nonradiatively. Taking both the tunnelling and dissociation processes into account, the PL intensity can be expressed by[15]

$$I = \frac{A}{\exp\left(\dfrac{E_{LO}}{K_B T}\right) - 1} + \frac{B}{1 + C\exp\left(\dfrac{-E_A}{K_B T}\right)} + D. \quad (1)$$

The first term on the right-hand side of Eq. (1) represents the contribution from the exciton tunnelling, and the second term associates with the thermal dissociation. A, B, C, and D are constants, E_{LO} is the LO-phonon energy of the tunnelling assistance, E_A is an activation energy of thermal dissociation, and k_B is Boltzmann's constant. It can be seen that the theoretical prediction closely agrees with the experimental data (Fig. 3). The E_{LO} and E_A values calculated by data fitting are 33 and 71.4 meV, respectively. The value of E_{LO} is close to the LO phonon energy of the ZnSe barrier layer ($h_{\mathrm{ZnSe,LO}} = 31.7$ meV). However, the value of E_A obtained is much larger than that of the simple CdSe QDs alone.[16] The higher activation energy possibly associates with the exciton tunnelling from QW to QDs,[17,10] which increases the restriction of excitons in QDs.

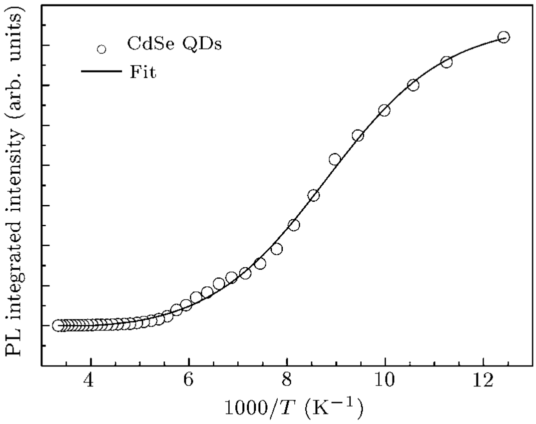

Fig. 3. Temperature dependence of PL-integrated intensity of the CdSe QD peak for sample *A*.

Figure 4 shows the peak energy and full width at half maximum (FWHM) of the PL band as a function temperature for the CdSe QDs in sample A. It can be seen that the peak energy shifts to lower-energy side with increasing temperature; and the shift of the peak position with temperature are discontinuous at certain temperatures (indicated by 1, 2, 3, 4 and 5). Meanwhile the discontinuous changes in the FWHM can also be seen in the similar temperature range. As temperature increases from 80 to 106 K (between points 1 and 2), those QDs with carriers of higher energy levels (contributing to the high-energy side of the broad PL peaks) are thermally ionized first. The free carriers are then thermally transported to QDs of lower energy states, in which they eventually recombine with each other, resulting in the increase of intensity of the red side of the broad PL band and in the decrease of the FWHM of the PL band. When temperature is above 106 K (between points 2 and 3), the PL peak is red shifted slightly with the increasing temperature, but the FWHM of the PL band increases rapidly. This phenomenon can be ascribed to the exciton injection from the QW to the QDs of higher energy levels. With the increasing temperature, the energy level difference ΔE between the QDs and the QW will change. In a certain temperature range, ΔE between some QDs and the certain energy state of the QW could equal to the LO-phonon energy of the ZnSe barrier layer, thus the tunnelling process is enhanced. With the further increase of temperature from 134 to 162 K (between points 3 and 4), the peak position shifts to red end and the FWHM decreases quickly. This process can be attributed to the energy level mismatch between the QW and QDs, thus the tunnelling process is reduced and the intensity on the blue side of the PL peak decreases.

Figure 5 shows the PL intensity of the ZnCdSe QW as a function of temperature. It is noted that the PL intensity of QW begins to increase at 134 K, and reaches a maximum value at 162 K. This phenomenon suggests that the excitons in the QW are accumulated in this temperature range. It is consistent with the above discussion. When temperature is higher than 162 K, the electro-phonon scattering becomes dominant; and the PL width increase with temperature.[18] The change in peak position with temperature can be attributed to electron-lattice interaction as described in the Varshni relation.[19]

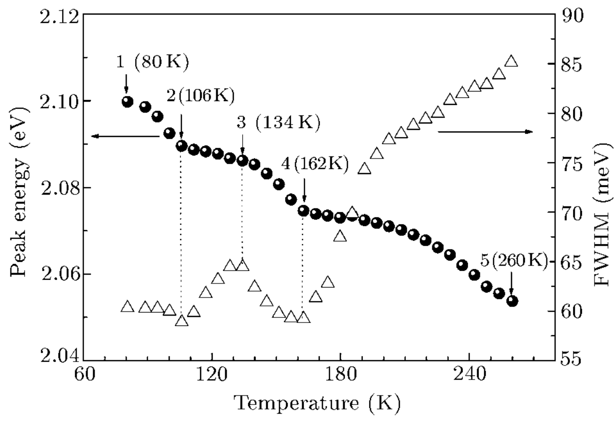

Fig. 4. Temperature dependence of the peak position (square) and FWHM (triangle) of CdSe QDs for sample *A*.

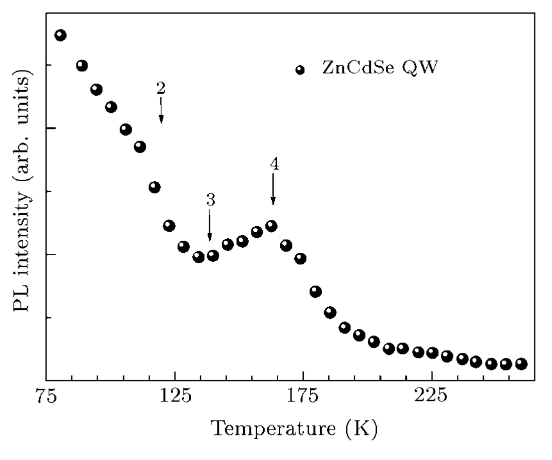

Fig. 5. Temperature dependence of PL intensity of ZnCdSe QW for sample *A*.

In summary, the temperature-dependent PL measurement reveals that the PL behaviour of CdSe QDs is strongly affected by the ZnCdSe QW via the exciton tunnelling from the QW to QDs. A strong PL of the CdSe QDs was obtained in this structure. The activation energy of the exciton dissociation in the CdSe QDs is as high as 71.4 meV. The temperature dependence of the peak energy and the FWHM of the PL band for the CdSe QDs suggest that they are related to the exciton tunnelling from the QW to QDs. The present study leads to a better understanding of the optical properties of coupling structures of CdSe QDs and a ZnCdSe QW. Based on this knowledge, further research on a new type of photoelectric device of Ⅱ-Ⅵ QDs/Ⅱ-Ⅵ QW coupling structures is undergoing.

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