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Temperature-dependent exciton recombination in asymmetrical ZnCdSe/ZnSe double quantum wells

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Abstract. Temperature-dependent exciton recombination in asymmetrical ZnCdSe/ZnSe double quantum wells is studied by recording photoluminescence spectra and photoluminescence decay spectra. The exciton tunnelling from the wide well to the narrow well and the thermal dissociation of excitons are two factors that influence the exciton recombination in this structure. In the narrow well, both of the two processes decrease the emission intensity, whereas, in the wide well, these two processes have contrary influences on the exciton density. The change of the emission intensity depends on which is the stronger one.

1. Introduction

Carrier recombination in semiconductor heterostructures such as quantum wells has been a field of active research for the last few years because this process is not only of basic physical interest but also of fundamental importance for optoelectronic devices. Many kinds of optoelectronic devices including laser diodes (LD) and light emitting diodes (LED) are based on this mechanism. The carrier recombination in a special quantum-well structure, namely an asymmetrical double quantum well (ADQW) structure that consists of two wells of different widths coupled by a thin barrier, shown in figure 1, is more interesting because it is modulated by carrier tunnelling which is one of the main characteristics of this structure [1, 2]. So the tunnelling rate is an important factor that influences the recombination. Previous studies [3, 4] have shown that longitudinal optical (LO) phonon emission can enhance the carrier tunnelling process efficiently when the energy separation between the lowest carrier sub-bands in the wide well (WW) and narrow well (NW) is larger than the LO phonon energy. In the wide-gap II–VI compound semiconductor ADQW, in which carriers tunnel from the WW to the NW in the form of excitons rather than independently due to the strong interaction between electrons and holes, the LO phonon emission still plays an important role in the tunnelling process [3]. Increasing the temperature would enhance the exciton tunnelling rate. On the other hand, temperature also influences the stability of excitons. The probability of thermal dissociation of excitons

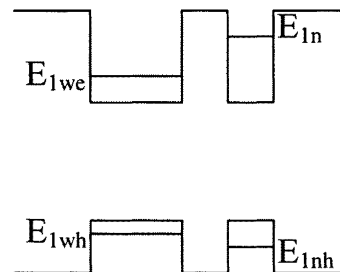

Figure 1. The band structure of an ADQW: E_{1we}, the $n = 1$ electron energy level in the WW; E_{1ne}, the $n = 1$ electron energy level in the NW; E_{1wh}, the $n = 1$ heavy-hole energy level in the WW; E_{1nh}, the $n = 1$ heavy-hole energy level in the NW; E_c the conduction band; and E_v, the valence band.

increases with temperature, which will reduce the exciton recombination. Therefore, temperature has an influence on the exciton recombination in more than one respect for the II–VI ADQW structures. Authors of most of the previous studies [1, 3, 4] have concentrated on studying the tunnelling process in ADQW at low temperatures at which the thermal dissociation of excitons can be neglected. However, it is well known that useful devices usually should work at room temperatures, even at high temperatures. To know how the exciton recombination changes with temperature is significant. In this paper, we report temperature-dependent exciton recombination in a ZnCdSe/ZnSe ADQW structure.

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2. Experimental details

The $Zn_{0.72}Cd_{0.28}Se/ZnSe$ ADQW samples studied were grown on Si-doped (100) GaAs substrates by low-pressure (LP) MOCVD at 350 °C with a reactor pressure of 76 Torr. The structure consists of a 1 μm ZnSe buffer layer followed by ten periods of $Zn_{0.72}Cd_{0.28}Se/ZnSe$ ADQW and then a 60 nm ZnSe cap layer. Each period of ZnCdSe/ZnSe ADQW includes one narrow ZnCdSe quantum well, one thin ZnSe barrier and one wide ZnCdSe quantum well, which will be denoted later as $L_n/L_b/L_w$, where L_n, L_b and L_w are the widths of the narrow well, thin barrier and wide well, respectively. Each period of the ADQW was separated by a 40 nm ZnSe barrier. Photoluminescence (PL) spectra were excited by the 457.9 nm line of a CW Ar^+ laser with output power 30 mW and signals were measured by a JY-T800 Raman spectrograph. The time-resolved set-up was based on a Ti : sapphire laser which provided a 80 MHz train of 25 ps pulses. The 400 nm line was selected to excite samples with output power 80 mW. The time-resolved signal was measured by a streak camera. The temperature of the samples was controlled by a cold-finger cryostat.

3. Results and discussion

Figure 2 shows the PL spectrum of a 5 nm/3 nm/3 nm ADQW at 98 K. The emission peaks on the high-energy and low-energy sides correspond to the $n = 1$ heavy-hole exciton recombination from the NW and WW, respectively [5]. It is obvious that the emission from the WW dominates the spectrum at this temperature. The main reason that causes the difference between the emission intensities of the NW and WW is the exciton tunnelling from the NW to the WW. Owing to the difference in $n = 1$ heavy-hole exciton energy between the two quantum wells, most of the excitons excited in the NW tunnel through the thin barrier to the WW, which induces the difference between the exciton distributions in the NW and WW. Considering the excellent sample quality and absence of unintentional doping, the influence of bound excitons on the emission can be neglected, even at low temperatures. Figure 3 shows the dependence of the integrated intensities of emission from the NW and WW on the temperature of this sample. We can see that the intensity emitted from the NW (I_{nw}) dominates the spectra at low temperature, whereas, with increasing temperature, the intensity emitted from WW (I_{ww}) becomes the dominant one. Another interesting phenomenon is that I_{ww} increases with temperature in the range 12–80 K, which is in contrast to usual experimental results and theories.

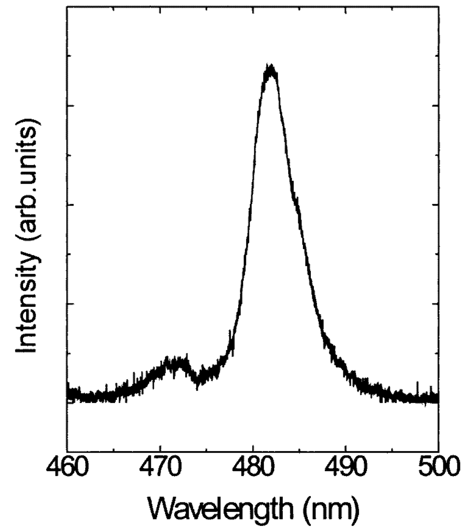

Figure 2. The PL spectrum of the 5 nm/3 nm/3 nm sample at 98 K.

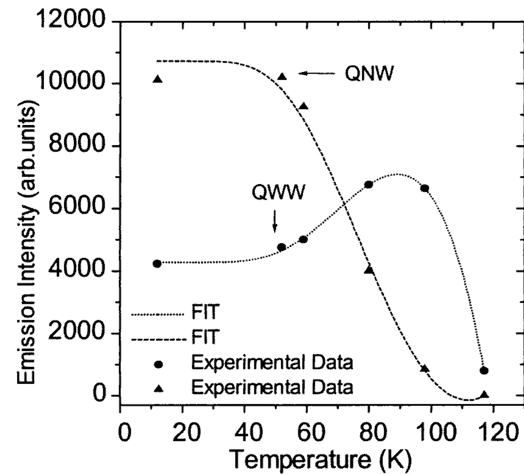

Figure 3. The temperature dependence of the emission intensity for the 5 nm/3 nm/3 nm ADQW.

Within the experimental temperature range, the energy of the excitation light (2.706 eV) is smaller than the band gap of the ZnSe barrier layer. We can exclude the possibility that the excitons excited in the barrier layer diffuse into quantum wells. Hence, the possibility that the dependence of the diffusion coefficient on temperature contributes to the change of the emission intensity in figure 3 should be excluded. These phenomena should be related to exciton tunnelling, thermal dissociation of excitons and processes whereby excitons undergo nonradiative recombination. The last term usually includes the excitonic Auger mechanism, surface recombination, recombination via defects or impurities and multi-phonon emission. For the exciton recombination in quantum wells, surface recombination can be neglected. The recombination via defects or impurities does not depend on temperature strongly [6]. The probability of multi-phonon emission is also very small [6]. Therefore the Auger mechanism seems to be the important one. In narrow-band-gap semiconductors, the Auger process really depends on temperature, but, in wide-gap materials, such as II–VI compounds, the Auger process should be related to the concentration of impurities [6]. In our experiment, the samples are not doped intentionally and the density of impurities is low ($\simeq 10^{16}$ cm^{-3}), so the Auger process can be neglected here. Considering what we discuss above, we neglect the nonradiative-recombination process in the experimental temperature range. In a normal quantum-well structure, the temperature dependence of the integrated excitonic emission intensity can be expressed as [7] $I = A/\{1+B_i \exp[-E_B/(K_B T)]\}$, which shows that the emission

intensity decreases with increasing temperature throughout the temperature range, but the situation in ADQWs is different.

In the wide gap II–VI ADQW, carriers tunnel from the NW to the WW in the form of excitons and the most efficient tunnelling mechanism has been proved to be assisted by emission of LO phonons. Haacke *et al* [8] have given the tunnelling time as

$$\tau_T \propto \hbar(4\varepsilon^2 + \Gamma_1^2)/(2\Gamma_1 E_0^2) \tag{1}$$

where ε is the difference in energy between the NW and WW states, Γ_1 is the energy-level broadening and E_0 depends slightly on the form of the electronic wavefunction. For the 5 nm/3 nm/3 nm ADQW sample, $4\varepsilon^2 \gg \Gamma_1^2$, so equation (1) can be rewritten as

$$\tau_t \propto \frac{2\hbar\varepsilon^2}{\Gamma_1 E_0^2}. \tag{2}$$

For a given sample, Γ_1 is one of the factors that dominate the tunnelling time. Here, Γ_1 can be expressed as [9]

$$\Gamma_1(T) = \Gamma_{inh} + \Gamma_A + \Gamma_{LO}\{\exp[\hbar\omega_{LO}/(k_B T)] - 1\}^{-1} \tag{3}$$

where Γ_{inh} is the linewidth originating from inhomogeneity, Γ_{LO} represents the strength of the exciton–LO-phonon coupling, Γ_A is the broadening contribution from the scattering of excitons by acoustic phonons and k_B is the Boltzmann constant. According to the approach of Lee *et al* [10], Γ_A is much less than that from the LO phonons. Γ_A can be neglected, so equation (3) can be rewritten as

$$\Gamma_1(T) = \Gamma_{inh} + \Gamma_{LO}\{\exp[\hbar\omega_{LO}/(k_B T)] - 1\}^{-1}. \tag{4}$$

In fact, equation (4) is widely used in dealing with ZnSe-based systems [11, 12]. It is well known that Γ_{LO} increases with the polarity of the material. For II–VI compounds, whose polarities are greater than those of III–V compounds, Γ_{LO} is greater and hence the tunnelling is enhanced. From equations (2)–(4), it is easy to find that increasing the temperature T can shorten τ_t. (Even though Γ_A is taken into account, it will not effect the influence of temperature on the tunnelling rate because Γ_A also increases with T.) Furthermore, we neglect the influence of the interaction between excitons and acoustic phonons on the lifetime of excitons in the later discussion.

The change of the exciton tunnelling time with temperature has also been observed in experiments. Figure 4 shows the time decay of the emission from the WW and NW at 100 K. The lifetime of excitons defined as the time from the maximum intensity of the excitonic emission to $1/e$ of the maximum value, is shorter for those in the NW than it is for those in the WW. The difference between the lifetimes implies that exciton tunnelling from the NW to the WW occurs [13]. The larger the difference, the faster the tunnelling. Figure 5 shows the temperature dependence of the difference between the exciton lifetimes in the NW and WW. We find that the difference increases with temperature, which means that the tunnelling time really decreases with increasing temperature. Considering what we discussed above, we find that the temperature T is an important factor in the exciton-tunnelling process and that τ_t decreases with increasing T.

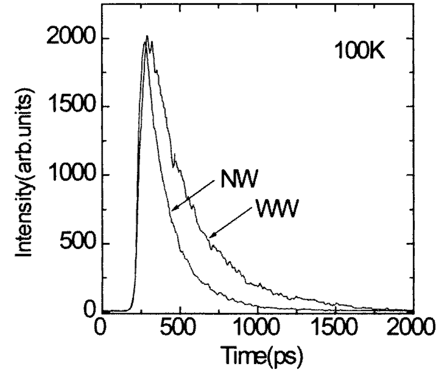

Figure 4. PL decay spectra of the 5 nm/3 nm/3 nm sample at 100 K.

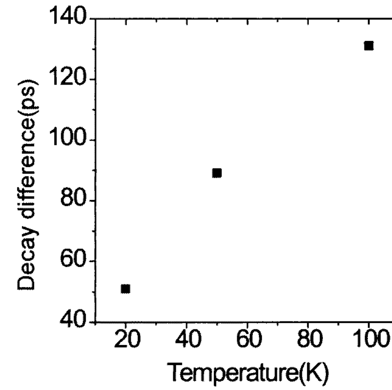

Figure 5. The temperature dependence of the difference between the exciton lifetimes in the NW and WW for the 5 nm/3 nm/3 nm sample.

Besides enhancing exciton tunnelling, increasing T also can reduce the exciton population by thermal dissociation. At any (experimental) temperature, free carriers and excitons are in thermodynamic equilibrium. Under such a condition, the relationship among the electron density n_e, hole density n_h and exciton density n_{ex} can be expressed as [14]

$$\frac{n_e n_h}{n_{ex}} = \frac{k_B T}{2\pi\hbar^2}\frac{m_e m_h}{m_{ex}}\exp[-E_B/(k_B T)] \tag{5}$$

where m_e, m_h and m_{ex} are the effective masses of the electron, hole and exciton, respectively, and $m_{ex} = m_e + m_h$. Obviously, the thermal dissociation of excitons becomes strong and n_{ex} decreases with increasing T.

Now we can interpret figure 3. The exciton population N_w in the WW, can be described by the rate equation

$$\frac{\mathrm{d}N_w}{\mathrm{d}t} = G_w + \frac{N_n}{\tau_t} - \frac{N_w}{\tau_{wth}} - \frac{N_w}{\tau_{wr}}. \tag{6}$$

G_w is the rate of generation of excitons in the WW excited by the photon. N_n is the exciton population in the NW and $1/\tau_t$ is the tunnelling rate, $1/\tau_{wth}$ and $1/\tau_{wr}$ are the rates of thermal dissociation and recombination of excitons in the WW, respectively. The rate equation for the NW can be obtained by the same method:

$$\frac{\mathrm{d}N_n}{\mathrm{d}t} = G_n - \frac{N_n}{\tau_t} - \frac{N_n}{\tau_{nth}} - \frac{N_n}{\tau_{nr}} \tag{7}$$

where G_n, $1/\tau_{nth}$ and $1/\tau_{nr}$ are the rates of generation, thermal dissociation and recombination of excitons in the NW, respectively.

For the WW, under steady-state conditions,

$$G_w + \frac{N_n}{\tau_t} - \frac{N_w}{\tau_{wth}} - \frac{N_w}{\tau_{wr}} = 0. \quad (8)$$

G_w is a constant for a certain excitation. $1/\tau_t$ and $1/\tau_{wth}$ increase with T, but they have different influences on the last term N_w/τ_{wr} which determines the emission intensity. In the low temperature range ($T < 80$ K in our experiment), due to the smaller tunnelling rate, the population of excitons tunnelling from the NW to the WW is not very large. On the other hand, $1/\tau_{wth}$ is smaller at low temperature. Under such conditions, $N_w/\tau_t - N_w/\tau_{wth}$ increases with T (the tunnelling is stronger than the thermal dissociation), which means that N_w/τ_{wr} increases with T. With increasing T, $1/\tau_t$, $1/\tau_{wth}$ and $N_w - N_n$ become large. (At higher temperature, $N_w - N_n$ becoming large is relative to N_w and N_n, but not relative to the form of $N_w - N_n$ at low temperature.) Above a certain temperature, $N_n/\tau_t - N_w/\tau_{wth}$ decreases with increasing T (the thermal dissociation is stronger than the tunnelling) and N_w/τ_{wr} decreases. This is what we see from the change of I_{ww} in figure 3.

For the NW, under steady-state conditions, equation (7) can be rewritten as

$$G_n - \frac{N_n}{\tau_t} - \frac{N_n}{\tau_{nth}} - \frac{N_n}{\tau_{nr}} = 0. \quad (9)$$

G_n is a constant here. Both N_n/τ_t and N_n/τ_{nth} increase with T, so the total effect is to reduce N_n/τ_{nr} with increasing T. Because of the different rates of change of $1/\tau_t$ and $1/\tau_{nth}$ at different T, N_n/τ_{nr} decreases with a nonlinear rate.

In the above discussion, we neglect the influence of the free-carrier tunnelling. Besides the exciton tunnelling, there is also the free-carrier tunnelling. The free-electron tunnelling can be assisted by LO phonons, which means that it is a fast process; however, the free-hole tunnelling is a very slow process because it cannot be assisted by LO phonons. (The difference between the energy levels of the holes in the WW and NW is smaller than the energy of LO phonons.) As a result, a built-in electrical field is formed, which prevents the free carriers from tunnelling. On the other hand, the formation and recombination of excitons needs both of the two kinds of carriers. Therefore, the free-carrier tunnelling does not influence our discussion.

On the basis of the exciton-tunnelling and dissociation model, the temperature dependence of the PL intensity from the WW and NW can be expressed by the equation

$$I = \frac{A}{\exp\left(\frac{E_1}{k_B T}\right) - 1} + \frac{B}{1 + C\exp\left(\frac{-E_2}{k_B T}\right)} + D. \quad (10)$$

The first and second terms on the right-hand side of equation (10) represent the contributions of the exciton tunnelling and thermal dissociation to the emission, respectively. By fitting the experimental data for the WW, we obtain $A > 0$, $E_1 = 32.5$ meV, $E_2 = 43.2$ meV and $A > 0$ makes the first term

$$\frac{A}{\exp\left(\frac{E_1}{k_B T}\right) - 1}$$

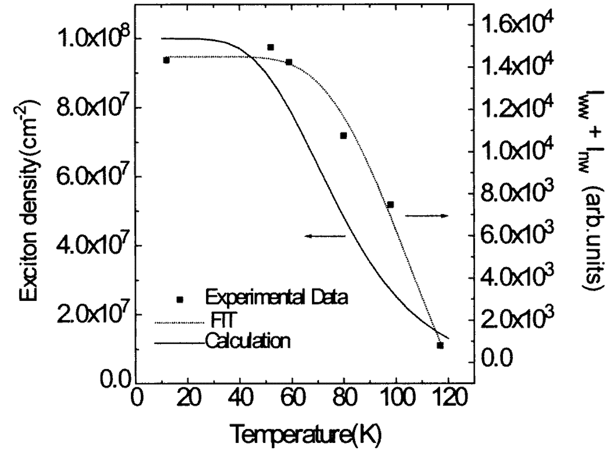

Figure 6. Temperature dependences of the total emission intensity and exciton density (points are experimental results and the full line is the calculated result) for the 5 nm/3 nm/3 nm ADQW.

positive, which means that the contribution of the exciton tunnelling to the emission from the WW is positive. This agrees with our discussion. The value of E_1 is close to the LO-phonon energy of $\hbar\omega_{LO} \approx 30$ meV and that of E_2 is close to the exciton binding energy of 40 meV for the present Cd fraction. This supports our discussion further. On fitting the experimental data for the NW, we obtain $A < 0$, $E_1 = 31.4$ meV and $E_2 = 40.4$ meV. It is easy to find that the contribution of the exciton tunnelling to the emission from the NW is negative, which implies that the exciton tunnelling can decrease the emission from the NW. The values of E_1 and E_2 also support our discussion.

Although excitons tunnel from the NW to the WW, the total exciton population in the NW and WW is influenced only by the thermal dissociation of excitons and so is the total emission intensity. (Given the strong Frohlich interaction in the polar II–VI compounds and also considering that the influence of nonradiative recombination is negligible, as discussion above, we pay attention only to the thermal dissociation of excitons here.) In figure 6, we show the dependences of the total integrated emission intensity $I_{ww} + I_{nw}$ (experimental results) and the exciton density (calculated according to equation (5)) on T. On comparing the experiment results and the calculated curve, we find that the trends of the changes are similar, which supports our discussion. (In the calculation, we assume that the exciton density excited at $T = 0$ is 10^8 cm^{-2} and that $E_B = 40$ meV [3] and neglect the difference between E_B in the NW and WW.) The deviation between the experimental results and the calculation is probably caused by our neglecting the slight difference between the binding energies in the WW and NW in the calculation. To fit the experimental data in figure 6, we should omit the exciton tunnelling term from equation (10), giving

$$I = \frac{B}{1 + C\exp\left(\frac{-E_2}{k_B T}\right)} + D. \quad (11)$$

By doing that, we obtain $E_2 = 43.3$ meV. This value is also close to the exciton-binding energy.

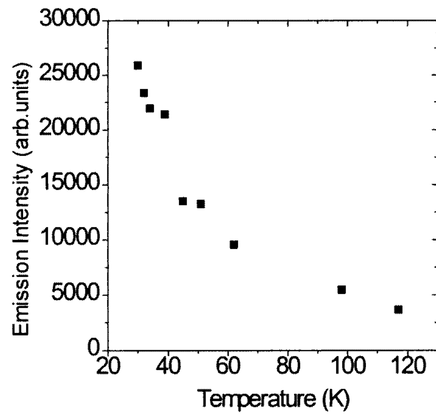

Figure 7. The temperature dependence of the emission intensity from the WW for the 5 nm/5 nm/3 nm ADQW.

It is known [8, 13, 15] that the thickness L_b of the thin barrier between the NW and WW is another dominant factor that determines the exciton tunnelling. In the limit of weak delocalization of the electron and hole wavefunctions, the exciton-tunnelling time τ_t depends exponentially on the barrier width L_b, $\tau_t \propto \exp(L_b)$. For a sample with a thin barrier, excitons are partially delocalized and tunnel from the NW to the WW rapidly due to the energy difference. However, for a sample with a thick barrier, excitons are mainly localized in the WW and NW, respectively, and the tunnelling time is long. So the thinner the barrier the faster the tunnelling. From the analysis above, we get the conclusion that the exciton tunnelling and thermal dissociation processes have contrary influences on the emission intensity I_{ww} and hence the change of I_{ww} with T depends on which is the stronger one. The exciton tunnelling is changed by L_b and then the change of I_{ww} is different with L_b. Figure 7 shows the dependence of I_{ww} on T of the 5 nm/5 nm/3 nm ADQW sample. We find that I_{ww} does not increase with T and that there is only an interval around $T = 40$ K. Considering that L_b of the 5 nm/5 nm/3 nm ADQW is larger than that of the 5 nm/3 nm/3 nm ADQW, the tunnelling rate $(1/\tau_t)$ is smaller for the former ADQW [16, 17]. For the 5 nm/5 nm/3 nm ADQW, in the competition between exciton tunnelling and thermal dissociation, the second one always occupies the dominant position; in another words, the term $N_n/\tau_t - N_w/\tau_{wth}$ equation (8) decreases with T throughout the experimental temperature range. So I_{ww} cannot increase figure 7. The interval in figure 7 probably means that N_n/τ_t in equation (8) has its maximum value at $T = 40$ K. (At this temperature, although $1/\tau_t$ is not very fast, N_n is large.)

4. Conclusion

We have studied the temperature-dependent recombination of excitons in a ZnCdSe/ZnSe ADQW by recording PL spectra and PL decay spectra. The exciton recombination both in the NW and in the WW is influenced by two factors, the exciton tunnelling and thermal dissociation processes. For the NW, the two factors have the same influence on the emission intensity, but, for the WW, the influences of the two factors are contrary. The change of the emission intensity is determined by the stronger one.

Acknowledgments

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