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Exciton tunnelling in ZnCdSe quantum well/CdSe quantum dots

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Abstract

Exciton tunnelling through a ZnSe barrier layer of various thicknesses is investigated in a $Zn_{0.72}Cd_{0.28}Se$/CdSe coupled quantum well/quantum dots (QW/QDs) structure using photoluminescence (PL) spectra and near resonant pump-probe technique. Fast exciton tunnelling from quantum well to quantum dots is observed by transient differential transmission. The tunnelling time is 1.8, 4.4 and 39 ps for barrier thickness of 10, 15 and 20 nm, respectively.

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Carrier and exciton tunnelling in semiconductor heterostructures is important in understanding basic quantum mechanics and it is still an active area in the application of tunnelling devices [1–3]. Wide-gap II–VI semiconductors become promising materials for the investigation of tunnelling devices, in which electron and hole tunnel rapidly as an exciton due to their large exciton binding energies [4]. Much work has been concentrated on tunnelling processes in asymmetric double quantum wells (ADQWs) [5,6]. Thereinto, exciton tunnelling from a narrow well (NW) to a wide well (WW) in ADQWs structures of ZnCdSe/ZnSe [7], ZnCdTe/ZnTe [8] and semimagnetic semiconductors doped by Mn^{2+} [9] have been widely studied. However, in ADQWs structures the excitonic optical absorption in a WW occurs inevitably when an adjacent NW is excited. The accumulation of excitons in WW impeded the exciton tunnel from NW to WW, which makes a trouble for the applications of ultrafast-exciton devices. According to the properties of QDs, such as small absorption cross-section, discrete energy levels and strong peaked density of states, we introduced QDs into a coupled structure. We substituted QDs for WW and designed a coupled quantum well/quantum dots (QW/QDs) structure. We expect that a fast tunnelling process from QW to QDs can be realized in this kind of QW/QDs structure.

In this letter, we investigated exciton tunnelling in a coupled $Zn_{0.72}Cd_{0.28}Se$ QW/ZnSe/CdSe QDs structure with different barrier thicknesses. Selectively-excited photoluminescence (PL) and near-resonant pump-probe measurements were performed, and a faster exciton tunnelling process from QW to QDs was observed.

The ZnCdSe QW/ZnSe/CdSe QDs samples were grown by molecular-beam epitaxy (MBE). Fig. 1 shows a schematic structure of the QW/QDs studied in the measurements. A 1-μm-thick ZnSe buffer layer, followed by 10 periods of QW/QDs structure and a 120 nm ZnSe capping layer were epitaxy successively on an (100)-oriented GaAs substrate. The coupled QW/QDs structure consists of a self-assembled CdSe QDs layer (3 monolayers), a ZnSe barrier layer, a 5-nm-thick $Zn_{0.72}Cd_{0.28}Se$ QW layer and a ZnSe separation layer with the same thickness as ZnSe barrier layer. An atomic force microscopy (AFM) image of the uncapped surface of CdSe QDs layer shows that the average height, base diameter and density of dots are 10, 10 nm and 10 dots per μm^2, respectively [10]. The QW/QDs side was epoxied down on a sapphire disk and

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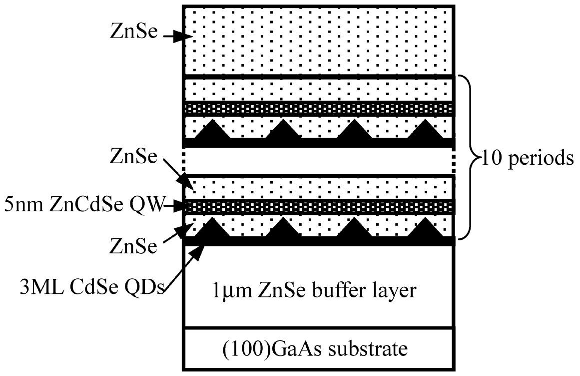

Fig. 1. Schematic of the ZnCdSe QW/ZnSe/CdSe QDs structure.

the GaAs substrate was then selectively etched for absorption and pump-probe measurements.

Three ZnCdSe QW/CdSe QDs samples with ZnSe barrier width (L_b) 10, 15, 20 nm were used in our experiments, labeled as sample A, B and C, respectively. The absorption spectra of the samples were shown in Fig. 2. The peak at 2.351 eV is attributed to the exciton absorption in ZnCdSe QW. We did not observe any obvious absorption peak originated from CdSe QDs. It indicates that optical absorption of QDs could be avoided in QW/QDs structures. Fig. 3 shows the PL spectra for the three samples at RT. The excitation source is the 514.5 nm line of an Ar^+ laser, which is close to the absorption peak of ZnCdSe QW. The emission peaks around 2.316 eV correspond to the exciton recombination in ZnCdSe QW [10]. A strong and narrow line at 2.345 ev is the 2LO phonon Raman scattering line from ZnSe interface. The emission band with dominant peak

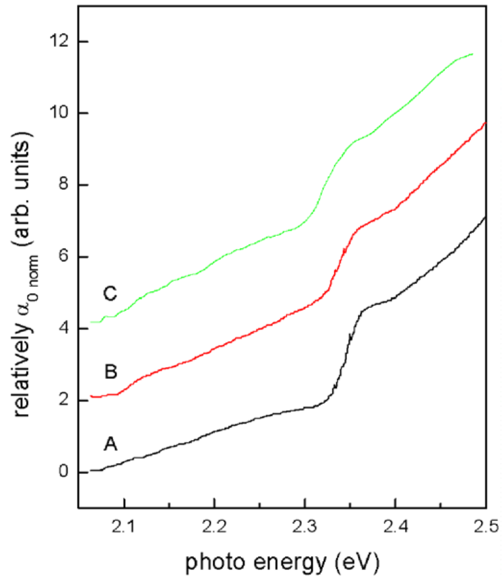

Fig. 2. Absorption spectra of ZnCdSe QW/ZnSe/CdSe QDs coupled structure with different barrier thicknesses of 10 nm (A), 15 nm (B) and 20 nm(C).

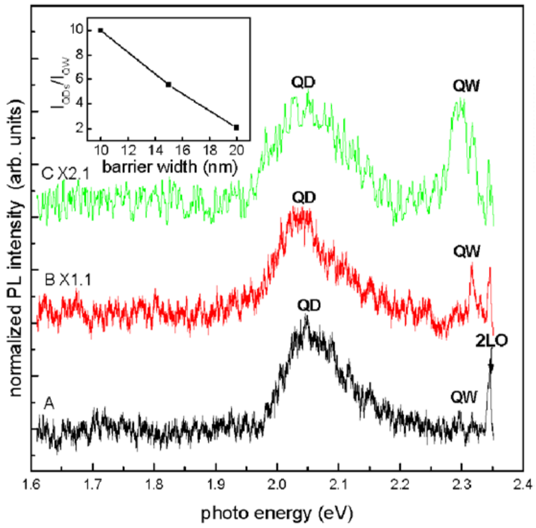

Fig. 3. The normalized PL spectra of sample A, B and C. The inset shows the ratio of the integrated intensity I_{QDs}/I_{QW} as a function of the barrier width L_b.

at 2.05 ev originates from the exciton recombination in CdSe QDs. The energy broadening of CdSe QDs exciton emission peak is attributed to the random size and spatial distribution of CdSe QDs. We normalized the three plots about the intensity of exciton emission in CdSe QDs. Although we did not observe the absorption from CdSe QDs (as in Fig. 2), a strong exciton emission of CdSe QDs was observed. The inset of Fig. 3 shows the integrated intensity ratio of the exciton emission in CdSe QDs to that in ZnCdSe QW, I_{QDs}/I_{QW}, as a function of L_b. It is seen that I_{QDs}/I_{QW} increases significantly with decreasing L_b. This relative decrease of the QW emission directly indicates excitons tunneling out of the ZnCdSe QW into the CdSe QDs.

In order to prove further the tunnelling rate of exciton from ZnCdSe QW to CdSe QDs, the RT pump-probe measurement was performed. Pump and probe pulses were generated by an optical parametric amplifier (OPA) pumped by a Ti:Sapphire regenerative amplifier. The pulse's energy and duration from OPA were 5 μJ and 130 femtosecond (fs), respectively. The wavelength was tuned to 537 nm, overlapping spectrally with the low-energy side of the ground-state exciton absorption peak in ZnCdSe QW. The laser beam was split into pump and probe beams with a pulse energy ratio $R > 15$. The two beams were focused onto the sample noncollinearly, whereas a chopper was set on pump beam. The transmitted probe intensity was measured using a photomultiplier, followed by a lock-in amplifier.

The transient differential transmission signals of samples A, B and C are shown in Fig. 4. In the case of low excitation, the differential transmission ΔT can be expressed as

$$\Delta T = \Delta T_0 \exp[-t/\tau_d], \qquad \tau_d = \tau + \Gamma, \tag{1}$$

where ΔT_0 is the differential transmission at $t = 0$, dependent on the energy, τ_d is the decay time of experiment

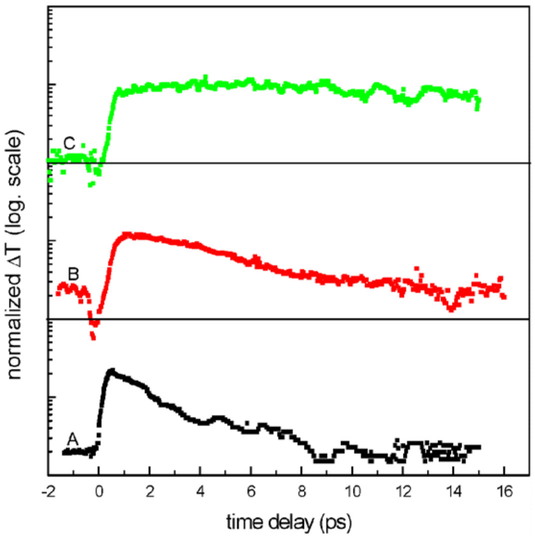

Fig. 4. Differential transmission intensity vs. delay time of pump-probe measurement for samples A, B and C. Pump light wavelength is 537 nm.

curve, τ is the exciton decay time in ZnCdSe QW, and Γ is the sum of pump and probe pulse durations (260 fs). Exciton decay in ZnCdSe QW is mainly resulted from exciton recombination and exciton tunnel to QDs of its vicinity, so the recombination rate $1/\tau$ can be written as

$$1/\tau = 1/\tau_t + 1/\tau_{re}, \quad (2)$$

where τ_t is exciton-tunnelling time from ZnCdSe QW to CdSe QDs, and τ_{re} is the intrinsic recombination time. In general, the influence of the barrier thickness on the exciton recombine rate $1/\tau_{re}$ is slight [8]. So, τ_{re} can be considered to be identical with the single QW decay time, which is in the 100-ps range for a similar design as ours [7]. By fitting the experimental data in Fig. 4 with Eq. (1), we got τ_d and τ, and calculated τ_t by Eq. (2). The results were listed in Table 1.

Table 1 shows that the exciton decay time is strongly dependent on the barrier thickness, which provides a direct evidence of exciton tunnelling behavior from ZnCdSe QW to CdSe QDs. The tunnelling rate increases rapidly with decreasing the barrier thickness. When barrier thickness decreases to 10 nm, the tunnelling time from ZnSeCd QW to CdSe QDs reaches to 1.8 ps. It indicates that the fast excitonic tunnelling rate is realized in the coupled QW/QDs structure. In addition, in the QW/QDs structure QDs penetrate into ZnSe barrier layer because the shape of self-assembled dots is conical or pyramidal. Thus, the exciton tunnelling is determined by the ZnSe thickness between ZnCdSe QW and the tip of CdSe QDs. The thickness for the ZnSe barrier is much smaller than ZnSe layer thickness. Furthermore, a slow decay involved in the fast tunnelling process was also observed in Fig. 4. We propose that exciton tunnelling from ZnCdSe QW to CdSe wetting layer through ZnSe separation layer can also exist in

Table 1
Barrier width L_b, fitted decay time τ_d, exciton decay time τ in ZnSeCd QW and the resulting tunnelling time τ_t

Sample	L_b (nm)	τ_d (ps)	τ (ps)	τ_t (ps)
A	10	2.1	1.8	1.8
B	15	4.5	4.2	4.4
C	20	28	28	39

this type of structure. Therefore, we attribute the slow decay process to this type of tunnelling process.

Based on our measurements and analysis, the introduction of QDs gives rise to two advantages for tunnelling devices: first, in QW/QDs structure, we can selectively excite ZnCdSe QW. Thus, the absorption effect of CdSe QDs due to the small optical absorption cross-section of QDs can be avoided. Second, the fast exciton tunnelling is easily carried out in experiments for our QW/QDs structure. So, the QW/QDs coupling structure is promising for ultrafast exciton tunnelling device.

In summary, we investigated the exciton tunnelling process in a coupled ZnCdSe QW/ZnSe/CdSe QDs structure utilizing PL spectra and femtosecond pulse pump-probe spectroscopy. The realization of rapid exciton tunnelling from QW to QDs provides a new idea for the investigation of novel ultrafast exciton tunnelling devices based on QW/QDs coupling structures.

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