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2002 Chinese Phys. Lett. 19 1152

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<u>CHIN.PHYS.LETT.</u> Vol. 19, No. 8 (2002) 1152

Optical Properties of ZnCdSe/ZnMgSe Multiple Quantum Wells Grown by Molecular Beam Epitaxy *

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(Received 24 February 2002)

We study the optical properties of ZnCdSe/ZnMgSe multiple quantum wells using photoluminescence (PL) and Raman scattering spectra. In the PL spectra, an intense emission band coming from the free exciton luminescence of the quantum wells can be observed from 80 K to 300 K. The exciton binding energy is evaluated by the dependence of PL intensity on the temperature, showing the behaviour of the better two-dimensional excitons. The result indicates that the enhancement of the confinement effect is due to containing Mg in the barrier layers. At room temperature, Raman scattering spectra are classified into confined optical modes and folded optical modes. This confirms the formation of a multilayer system with a higher crystalline quality.

PACS: 63. 22. +m, 81. 15. Hi, 78. 55. -m, 68. 65. +g

ZnCdSe/ZnSe quantum well structures have been extensively studied for realizing short wavelength light emitting diodes (LEDs) and laser diodes (LDs).^[1,2] Heterostructures containing Mg-based II-VI compounds, which are used as cladding layers to prevent oxidation or as potential barrier layers to confine carriers and photons, are demonstrated to be very important in the fabrication of LEDs and LDs. [3,4] Most previous works have focused on the crystal structure and characterization of the ZnMg(S)Se alloy layers. [5,6] For the quantum well structure, only a few works have been reported.^[7] Compared to ZnCdSe/ZnSe, the ZnCdSe/ZnMgSe system has larger band offset and less lattice mismatch between the well and barrier layers. For this reason, we fabricate ZnCdSe/ZnMgSe multiple quantum wells (MQWs) grown by molecular beam epitaxy (MBE), in which ZnMgSe layers are used as barrier layers instead of ZnSe. The optical properties of the samples are reported by the investigation of the photoluminescence (PL) and Raman spectra.

The samples were grown by MBE on (100) GaAs substrates using elemental Zn, Cd, Mg and Se as sources. The structural configuration included a ZnSe buffer layer with a thickness of 1 μ m and 75 periods of ZnCdSe quantum wells with 3–5 nm thicknesses separated by ZnMgSe potential barriers with a thickness of 10 nm. For the samples used in this study, Cd compositions of the quantum well were 0.15 and 0.2, respectively, and the Mg composition of the potential barrier was 0.3. The samples were characterized by an x-ray diffraction measurement. In addition to the $K_{\alpha 1}$ and $K_{\alpha 2}$ (400) diffraction peaks of the GaAs substrate, the

observation of some satellite peaks confirms the formation of the multilayer structure. The PL of the sample was excited by the 325 nm line of an HeCd laser. The PL spectrum is dispersed by a monochromator (LabRam) with gratings of 1800/mm, and detected by a charged-coupled device (CCD) detector coupled to a microscope. The spectra resolution of the experimental set-up was 0.7 Å. In the temperature-dependent experiment, the liquid-nitrogen cooling system was used in conjunction with the sample stage to cool a sample down to $\sim 80\,\mathrm{K}$. The temperature was controlled by the TMS94 from 80 K to room temperature, in which the temperature has an accuracy of better than 1 K. Raman spectra were recorded using the same spectrometer equipped with a CCD detector and coupled to a microscope of the Raman system. The 488 nm or the $514.5\,\mathrm{nm}$ lines of an $\mathrm{Ar^{+}}$ ion laser were used to excite the sample. The Raman spectra resolution was less than $2 \, \mathrm{cm}^{-1}$. The power of the laser on the sample was set to about 100 mW.

Figure 1 shows the typical PL spectra at 80 K for three $\rm ZnCdSe/ZnMgSe$ MQWs with different well widths or Cd compositions labelled a, b and c. In each of the MQWs, one strong emission band is obtained in the green region, and a distinct PL peak shift is observed by varying the well width or the composition. The peak positions of the bands shift to the higher-energy side with decreasing well width, suggesting that the bands observed in Fig. 1 are related to the quantum energy level of the ZnCdSe well layer. Compared to the result of Ref. [8], we consider that the bands are due to the recombination of n=1 heavyhole free exciton from the ZnCdSe well layer.

^{*} Supported by the Innovation Project of the Chinese Academy of Sciences, the 863 High Technology Research Programme in China under Grant No 2001AA31112, the National Fundamental and Applied Research Project, the National Natural Science Foundation of China under Grant Nos 69896260 and 69977019, and the Programme of Hundred Talents of the Chinese Academy of Sciences.

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Figure 2 gives the PL spectra of ZnCdSe/ZnMgSe and ZnCdSe/ZnSe MQWs with the same width and composition at room temperature. $ZnCdSe/ZnMgSe\ MQWs\ (spectrum\ a)$, we only see one excitonic emission peak at $\sim 527 \,\mathrm{nm}$ from the quantum well. It is important that, for ZnCdSe/ZnSe MQWs (spectrum b), we can observe two bands located at about 460 nm and 527 nm. The band at $\sim 527\,\mathrm{nm}$ is attributed to the excitonic emission of the ZnCdSe quantum well, [8] and the band at $\sim 460 \,\mathrm{nm}$ is considered to be the band-edge emission from the ZnSe barrier layer. [9] To discuss the confinement effect in the two systems further, the PL spectra of the above two samples were measured at different temperatures. As the temperature was increased from 80 K to room temperature, we noticed that the luminescence from the ZnSe barrier in ZnCdSe/ZnSe MQWs became increasingly important and the luminescence from the ZnMgSe barrier in the ZnCdSe/ZnMgSe MQWs was not observed, even at room temperature. Zn_{0.8}Cd_{0.2}Se/ZnSe MQWs, the band offset is less than that of ZnCdSe/ZnMgSe MQWs; the electrons and holes at higher temperature cannot be effectually confined in the well layer due to the thermal release. The release of electrons and holes from the well layer to the barrier layers results in the appearance of stronger luminescence from the ZnSe barrier layers at higher temperatures. The inset of Fig. 2 exhibits the temperature dependences of luminescence intensities from ZnCdSe wells for samples a and b. From the plots, the thermal activation energies are equal to 32 meV and 40 meV for ZnCdSe/ZnSe and ZnCdSe/ZnMgSe MQWs, respectively. In the ZnCdSe/ZnMgSe system, the excitons are caused dominantly by the confinement effect of the quantum well. For the ZnCdSe/ZnSe system, the dominant contribution to the excitons is brought about by the Coulomb interaction between the electrons and holes due to the lower potential barrier. The spread of the electrons and holes into the ZnSe barrier partially restores a three-dimensional characteristic at higher temperature. The exciton binding energy is known to be enhanced with the increasing confining dimension. The exciton binding energy can be expressed by Eq. (1) in Ref. [10]

$$E_b = \frac{4E_0}{(\alpha - 1)^2},\tag{1}$$

where E_0 is the effective Rydberg constant for the bulk exciton and α is the dynamic space fractional dimension which measures the anisotropy of the electronhole Coulomb interaction. $\alpha = 3, 2$ or 1 give, respectively, $E_b = E_0$, $4E_0$ or ∞ , corresponding to the well-known results of the integer dimension models. For excitonic binding energy in the two-dimensional system, the investigations of free and bound excitons have been reported. [10,12] In a real quantum well structure,

 α changes continuously between 3 and 2. Actually, the thermal activation energies from the inset of Fig. 2 reflect the exciton binding energy. In $E_0=21$ meV from the exciton binding energy of ZnSe bulk material, $^{[11]}$ the calculation by Eq. (1) gives $\alpha=2.62$ and 2.45 for $E_b=32$ meV and 40 meV, respectively. The barrier layer containing Mg increases the band offset between the well layer and the barrier layer. The electrons and holes are effectively confined in the deeper potential well. The stronger confinement effect leads to the enhancement of the exciton band energy. The result indicates that excitons of the ZnCdSe/ZnMgSe system show two-dimensional behaviour better than those of ZnCdSe/ZnSe system.

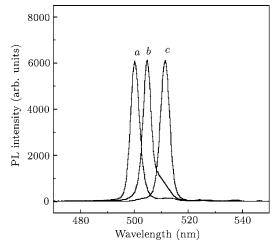


Fig. 1. PL spectra of ZnCdSe/ZnMgSe MQWs at $80\,\mathrm{K}$ excited by an HeCd laser operated at $325\,\mathrm{nm}$.

- (a) $Zn_{0.85}Cd_{0.15}Se/Zn_{0.7}Mg_{0.3}Se$ (3nm/10nm),
- (b) $Zn_{0.8}Cd_{0.2}Se/Zn_{0.7}Mg_{0.3}Se$ (3 nm/10 nm),
- (c) $Zn_{0.8}Cd_{0.2}Se/Zn_{0.7}Mg_{0.3}Se$ (5 nm/10 nm).

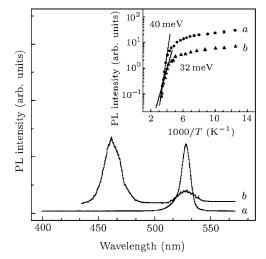


Fig. 2. PL spectra of ZnCdSe/ZnMgSe MQWs at room temperature excited by an HeCd laser operated at $325\,\mathrm{nm}$. The inset shows the temperature dependences of PL intensities of the bands at $527\,\mathrm{nm}$. (a) $\mathrm{Zn_{0.8}Cd_{0.2}Se/Zn_{0.7}Mg_{0.3}Se}$ ($3\,\mathrm{nm}/10\,\mathrm{nm}$), (b) $\mathrm{Zn_{0.8}Cd_{0.2}Se/ZnSe}$ ($3\,\mathrm{nm}/10\,\mathrm{nm}$).

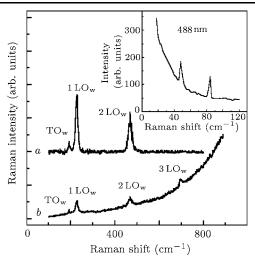


Fig. 3. Raman spectra of $\rm Zn_{0.8}Cd_{0.2}Se/Zn_{0.7}Mg_{0.3}Se$ MQWs $(3\,\rm nm/10\,\rm nm)$ at room temperature excited by (a) 488 nm or (b) 514.5 nm lines from an Ar⁺ laser. The inset gives the low-frequency region Raman spectrum at room temperature.

Typical Raman spectra of the Zn_{0.8}Cd_{0.2}Se/Zn_{0.7} $Mg_{0.3}$ Se MQWs by 488 nm line (a) or 514.5 nm line (b) excitation at room temperature are shown in Fig. 3. It is known that the energy gap of the barrier layer ZnMgSe at room temperature is much larger than the photon energies of the excitation lines mentioned above. Thus, the ZnMgSe barrier layers are transparent for the incident light of 488 nm and 514.5 nm. However, the incident photon energies of 488 nm and 514.5 nm laser lines are larger than the bandgap of $Zn_{0.8}Cd_{0.2}Se$ well layers. It is considered that the LO_w phonon mode shown in Fig. 3 is from Zn_{0.8}Cd_{0.2}Se well layers. The LO_w phonon frequency is $\sim 231 \, \mathrm{cm}^{-1}$ $(\sim 29 \text{ meV})$, this energy is smaller than that of longitudinal optical (LO) for pure ZnSe (31 meV) and larger than LO for pure CdSe (27 meV). [13] Therefore, the LO_w phonon is ascribed to be the LO phonon mode confined in Zn_{0.8}Cd_{0.2}Se well layers. For the same reason, a weaker peak at $192 \,\mathrm{cm}^{-1}$ labelled $\mathrm{TO_w}$ phonon is attributed to the transverse optical (TO) phonon mode from Zn_{0.8}Cd_{0.2}Se well layers. When the energy of exciting light (514.5 nm) is very close to the bandgap of the quantum wells, the third order of phonon modes can be seen clearly, as shown in the spectrum b of Fig. 3. The resonance condition is beneficial to the observation of higher-order multi-phonon modes. A study of the resonance Raman scattering at room temperature will be discussed in future work. The inset of Fig. 3 shows the Raman spectrum in the

region $10\text{--}100\,\mathrm{cm^{-1}}$ excited by the 488 nm line. It is reasonable to consider that the two peaks are from the folded longitudinal acoustic (LA) phonon modes in $\mathrm{Zn_{0.8}Cd_{0.2}Se}$ well layers. The doublet structure of the folded phonon modes [14] cannot be observed due to the broadening effect of the bands in the alloy layers. The observation of the folded optical modes indicates that the multilayer structure of the sample containing Mg in barrier layers is not destroyed.

In summary, the optical properties of ZnCdSe/ZnMgSe MQWs have been studied by PL and Raman scattering spectra. Only one excitonic emission is observed in PL spectra at room temperature, showing an enhancement of the confinement effect due to containing Mg in the potential barrier layers. Research on the exciton binding energy well indicates the two-dimensional exciton behaviour in such a structure. The Raman spectra are classified into confined optical modes and folded optical modes. These results confirm the formation of a multilayer structure with a higher crystalline quality.

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