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Excitonic Properties of ZnSe-ZnCdSe Multiple Quantum Wells under High Excitation Density

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ZnSe-$Zn_{0.78}Cd_{0.22}Se$ MQWs are grown on GaAs(100)substrate by MOCVD. The photoluminescence (PL) under pulsed N_2 laser excitation is investigated in detail. The bands observed in PL spectra at 77K are attributed to the n=1,2 heavy-hole exciton transitions, respectively. In high-density excitation the excitonic recombination is mainly on the exciton interaction, involves the exciton-electron (Ex-e) and exciton-exciton (Ex-Ex) interactions. The stimulated emission due to Ex-Ex scattering is observed at 77K in ZnSe-ZnCdSe MQWs.

KEYWORDS: ZnSe-ZnCdSe multiple quantum wells, exciton-exciton scattering, exciton-electron scattering, photoluminescence, stimulated emission

1. Introduction

The wide-gap Ⅱ-Ⅵ semiconductors in which the luminescent spectra are dominated by excitonic transitions are potentially useful for light emitting diodes (LEDs) and injection lasers. In particular, the corresponding enhancement of exciton-binding energy in ZnSe-based multiple quantum wells (MQWs) and strained-layer superlattices (SLSs) make it possible to apply room temperature excitonic effects for short wavelength devices. Recently, diode laser operation at shorter wavelengthes has been achieved in MQWs structures of ZnSe-ZnCdSe. [1] In order to understand of the gain mechanism of these lasers, the investigation of excitonic properties have attracted much attention. [2,3] The origin of the stimulated emission is studied under resonant excitation and a phenomenological model of optical gain due to inhomogeneously broadened exciton resonance is proposed. [3] In this paper, we reported the properties of excitonic emission at 77K under high excitation conditions and observed the stimulated emission originating from homogeneous broadening of the exciton resonance due to the exciton-exciton interactions.

2. Experimental

ZnSe-$Zn_{0.78}Cd_{0.22}Se$ MQWs were grown on (100) GaAs substrate by atmospheric pressure metalorganic chemical vapour deposition (MOCVD). Dimethylzinc (DMZn), dimethylcadmium (DMCd) and 10% H_2Se together with H_2 gas were used as the source materials. A ZnSe buffer layer was intentionally inserted to avoid lattice mismatch between substrate and MQWs. The MQWs sample was composed of a hundred $Zn_{0.78}Cd_{0.22}Se$ wells of thicknesses 8nm and ZnSe barrier layer of thicknesses 10 nm. A 100 nm thick cap layer of ZnSe was grown to complete a wave guide structure. The MQWs were grown at 320℃.

The 337.1nm line of a model UV-24 pulsed N_2 laser was used as excitation source. The laser with 10 ns duration had a peak power $J_0 = 4MW/cm^2$. The photoluminescence (PL) spectra and time-resolved spectra (TRS) were detected using a Spex 1404 double gratings spectrometer with a RCA C-31034 cooled photomultiplier.

3. Results and discussion

Figure 1 shows the PL spectra of the ZnSe-$Zn_{0.78}Cd_{0.22}Se$ MQWs sample excited by the 337.1 nm line of a N_2 laser at 77K. The PL spectrum consists of the three bands which are located at 445 nm

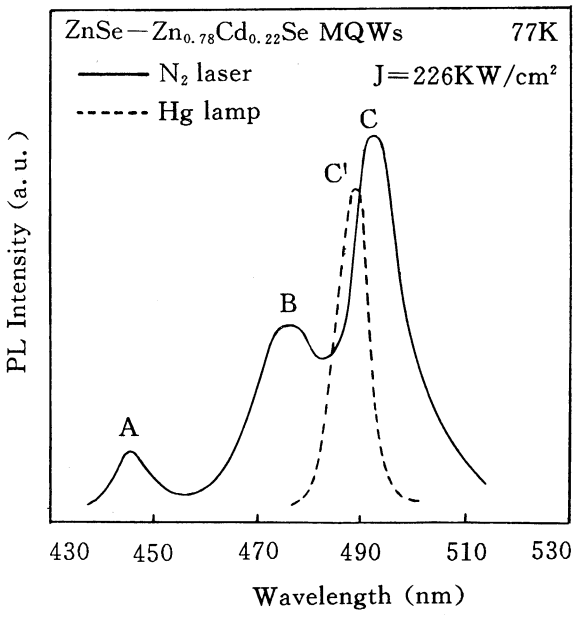

Fig. 1 PL spectra of ZnSe(10nm)-$Zn_{0.78}Cd_{0.22}Se$(8nm) MQWs at 77K excited by the 337.1nm line of a N_2 laser (solid line) and the 365nm line of a Hg lamp (dashed line).

(band A), 475 nm (band B) and 491 nm (band C), respectively. In Fig. 1 we also gave the PL spectra of the same sample excited by the 365 nm line of a 500W Hg lamp at 77K. It is noticed that only one band C' is observed at 488. 8 nm, and its linewidth

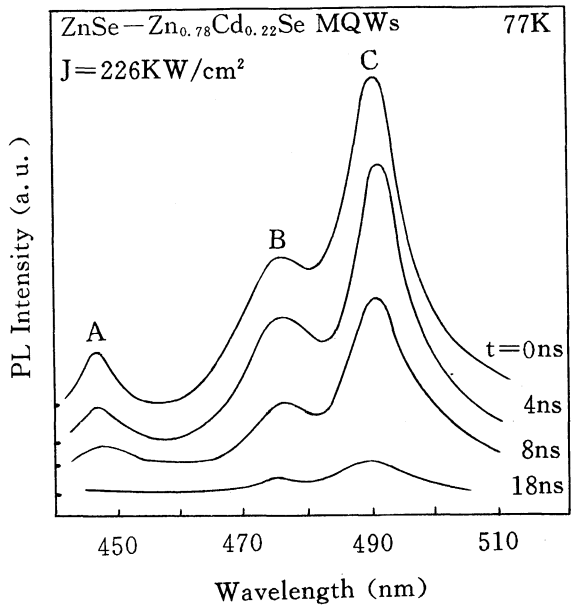

Fig. 2 Time-resolved spectra of ZnSe(10nm)-$Zn_{0.78}Cd_{0.22}Se$(8nm) MQWs at 77K.

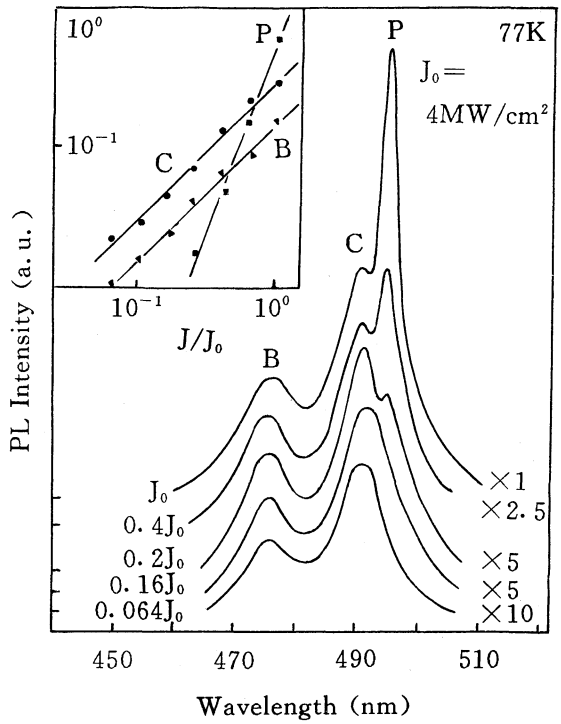

Fig. 3 PL spectra of ZnSe(10nm)-$Zn_{0.78}Cd_{0.22}Se$(8nm) MQWs under different excitation at 77K ($J_0=4MW/cm^2$), insert shows PL intensity I as a function excitation density J.

becomes narrow. The TRS of the sample at 77K by N_2 laser excitation is shown in Fig. 2. From Fig. 2 we found that these three bands have faster decay rate, and no energy shifts of the band are observed with time increasing. Above experimental results excluded the possibility of these bands coming from the donor-accptor pair process. Comparing to the energy position of the band A, we considered that the band A is due to free excitonemission of ZnSe cap layer on top of the MQWs. In order to investigate the origin of the bands B and C, we calculated the energy band structure of the ZnSe-$Zn_{0.78}$ $Cd_{0.22}$ Se MQWs by using the method which is similar to the previous work of Lozykowski.[2] The theoretical calculation results indicated that the energy positions of the emission bands due to $n=1,2$ heavy-hole excitons are located at 2. 542 eV (487. 4 nm) and 2. 615eV (473. 8 nm), respectively. We noticed that the peak position of the bands C' and B in Fig. 1 are closeto the above theoretical calculation values, but the spectral position of the band C is below calculation value about $n=1$ heavy-hole exciton. Figure 3 shows the PL spectra of the sample at different excitation densities (J) by the 337. 1 nm line of a N_2 laser at 77K, the insert gives the J dependence of the PL intensity (I). From Fig. 3 we found that I as a fumction J can be expressed as a linear relation (I~J) for the bands B and C. With J increasing the position of the band C shifts to the low energy and one of the band B is no change. At $J=0.2\ J_0$, a new emission band labeled P is observed at about 494 nm and its linewidth is rather narrow. The band P increases rapidly with J increasing and varies as $J^{2.8}$. We considered that the band P appearing at higher excitation shows the nature of stimulated emission. Ding et al.[3] has proposed a model of inhomogeneously broadened exciton resonance in providing gain in the ZnSe-ZnCdSe MQWs, when the exciton density is sufficiently low. However, our experimental observation can not be interpreted using this models. For instance, with J increasing the band C shifts to low energy side and a new band P is observed in low-energy tail of the band C. In our excitation condition, because the tatal density of excitons can reach enough high, the many-body Coulomb interactions should be occured. Above some features about the bands C and P are similar with our earlier work[5,6] on ZnSe epilayer, in which it was attributed to the exciton - electron scattering (Ex-e) and exciton-exciton scattering(Ex-Ex) processes. According to the result of Saito et al. ,[7] the energy separation of the Ex-e scattering process from the free exciton emission can be expressed as :

$$\triangle E=\frac{KT}{2}\frac{m_e+m_h}{m_e} \qquad (1)$$

where m_e, m_h are the electron and hole masses; T is the lattice temperature. The peak energy difference of the band C at $J>0$. $2J_0$ in Fig. 3 with n=1 heavy-hole exciton emission by theoretical calculation is of the order of 13 meV. This value is in good agreement with the calculation one using the Eq. (1). In our earlier work,[5,6] Ex-Ex scattering process is described as follows:

$$Exciton(k) + Exciton(k') \rightarrow h\upsilon + electron\text{-}hole\ pair \quad (2)$$

$$h\upsilon = E_g - 2E_b + E_k + E_{k'} - h^2k_i^2/2\mu \quad (3)$$

where $h\upsilon$ is the energy of the emitting photon, E_b is the exciton-binding energy, E_k *and* $E_{k'}$ are the kinetic energies of the exciton, E_g is the band gap and μ is given by $\mu^{-1} = m_e^{-1} + m_h^{-1}$. From Eq. (3), the peak position of the band due to Ex-Ex collision is about the binding energy below the free exciton emission. From this consideration, Ex-Ex scattering emission should be located at low-energy side of the band C due to Ex-e scattering. By above result and analysis, we may obtain conclusions as follow: (1) By Hg lamp excitation the luminescence dominantly come from free exciton emission; (2) Under N_2 laser excitation Ex-e and Ex-Ex scattering process exist at same time. In lower excitation Ex-e scattering gives a major contribution. For high excitation Ex-Ex scattering becomes even more important; (3) In our excitation condition, the number of the exciton coming from the high energy levels is less, the band B dominantly originates from n=2 heavy-hole exciton emission.

Much work[3,4] has been done on the exciton gain and stimulated emission in ZnSe-ZnCdSe MQWs. Taking into account the random MQW thickness variations and the alloy compositional fluctuations, the inhomogeneously broadened excitons should play a principal role in providing gain in the ZnSe-ZnCdSe MQWs. In strong excitation, we considered that the exciton-exciton, exciton-electron processes providing gain can be obtained. Figure 4 shows lasing emission spectra of ZnSe (10 nm)- $Zn_{0.78}Cd_{0.22}Se$ (8nm) MQWs with 500μm cavity length(l) at 77K excited by the 337. 1 nm line of a N_2 laser, the schematic structure is shown in the insert of Fig. 4. A rich longitudinal mode spectrum is observed. The central wavelength is 495 nm and the threshold at 77K is about 100KW/cm^2.

In summary, we have reported the excitonic emission of ZnSe-$Zn_{0.78}Cd_{0.22}$ Se MQWs under high density excitation. The origin of emission bands observed in PL spectra at 77K is attributed to the free exciton recombination between n=1,2 electron subbands and n=1,2 heavy-hole subbands. In higher excitation condition, the luminescence dominantly is due to the exciton interactions. We observed the stimulated emission due to Ex-Ex scattering at 77K in ZnSe-ZnCdSe MQWs.

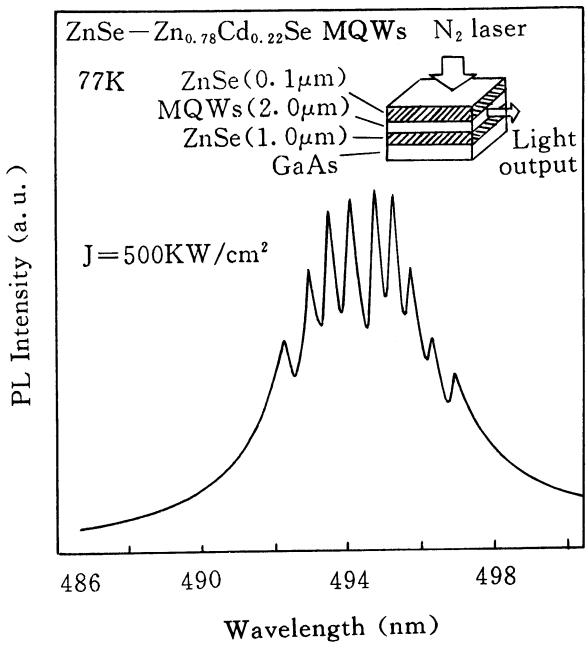

Fig. 4 Lasing emission spectra of ZnSe(10nm)-$Zn_{0.78}Cd_{0.22}Se$(8nm) MQWs at 77K for 500μm, insert shows the schematic structure.

Acknowledgements

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