Resonant Raman Scattering and Photoluminescence in Zn_{1-x}Cd_xSe-ZnSe Superlattice

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The Raman scattering and photoluminescence from a $Zn_{0.76}Cd_{0.24}Se$ -ZnSe superlattice at room temperature and 77 K have been studied. Two kinds of LO phonon confined modes were observed in detail for the first time, they were ascribed to those of ZnSe barrier layers and $Zn_{0.76}Cd_{0.24}Se$ well layers in the superlattice, respectively. Four orders of LO_w multi-phonon modes for well layers, including Stokes and anti-Stokes, and five orders of LO_b multi-phonon modes for barrier layers were seen clearly at room temperature. At 77 K every order of LO phonon modes shifts a little toward lower frequency. Three photoluminescence peaks located at 2.478 eV (19990 cm⁻¹), 2.554 eV (20600 cm⁻¹) and 2.387 eV (19250 cm⁻¹) were observed at room temperature.

KEYWORDS: Zn_{1-x}Cd_xSe-ZnSe superlattice, resonant Raman scattering, photoluminescence

1. Introduction

ZnSe based II-VI compound single- or multiple-quantum-well and superlattice (SL) structures, $\rm Zn_{1-x}Cd_xSe-ZnSe$, have recently attracted much attention because of their potential application. Their electronic and optical properties have been widely studied, especially for excitonic properties $^{1-3)}$. The resonant Raman scattering of single-quantum-well in $\rm Zn_{1-x}Cd_xSe-ZnSe$ was also investigated $^{4)}$

In this paper we have studied in detail the resonant or nearly resonant Raman scattering of $\rm Zn_{1-x}Cd_xSe\text{-}ZnSe$ at room temperature (RT) and 77 K. Raman scattering by two kinds of ZnSe-like LO multi-phonons respectively confined in ZnSe barrier layers and $\rm Zn_{0.76}Cd_{0.24}Se$ well layers of the SL are presented for the first time. The photoluminescence (PL) spectra from the SL structure at 77 K and RT are also presented.

2. Samples and Experiments

The SL used in this study were grown by MOCVD on (100) GaAs substrate, which had a buffer layer of 0.7 μ m ZnSe to diminish misfit of the lattices. Dimethylzinc, dimethylcadmium and H₂Se are used as the source materials. The sample is composed of 100 periods of alternating Zn_{0.76}Cd_{0.24}Se and ZnSe layer of 4 nm and 6 nm in thickness respectively. The ZnSe and ZnCdSe layers are grown at 593 K.

The Raman spectra with back scattering geometry and PL measurements were carried out by using Spex-1403 Raman spectrometer. The sample was excited by 514.5 nm, 488.0 nm, 476.5 nm and 457.9 nm lines of Spectra-physics 165 argon ion laser in either case for Raman scattering or for PL at RT and 77 K, respectively.

3. Photoluminescence Spectra

The spectra in Fig. 1 (a), (b) and (c) were measured at RT using 488.0 nm, 476.5 nm and 457.9 nm lines as an excitation light, the sharp peaks superimposed on the PL wide peaks are the Raman peaks. A stronger PL peak is located at 2.478 eV ($19990~\rm cm^{-1}$) and two weaker PL peaks are at 2.387 eV ($19250~\rm cm^{-1}$) and 2.554 eV ($20600~\rm cm^{-1}$) respectively.

Fig. 2 shows the PL spectra obtained under excitation with the 476.5 nm and 457.9 nm lines of an argon laser at 77K. From Figs. 1 and 2, it is observed that the emission peaks situated in 2.478 eV and 2.387 eV at RT shifted to

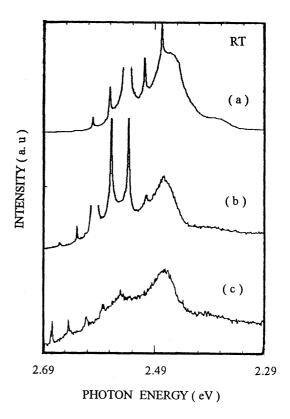


Fig. 1 PL spectra of Zn $_{0.76}$ Cd $_{0.24}$ Se-ZnSe SL excited by (a) 488 nm, (b) 476.5 nm and (c) 457.9 nm laser lines at RT. The sharp peaks superimposed on the PL spectra are the Raman lines.

2.551 eV (20580 cm⁻¹) and 2.497 eV (20140 cm⁻¹) at 77 K, respectively, the high-energy peak of 2.554 eV (20600 cm⁻¹) at RT could not be observed at 77 K by excitation with 476.5 nm and 457.9 nm lines. Using 488.0 nm line excitation, the peak at 2.551 eV disappeared and only the 2.497 eV peak remained at 77 K. According to ref. 1 , we attribute the PL main peak situated at 2.478 eV to the excitonic transition $E_{\rm 1hh}$ from the n=1 electron subband to n=1 heavy hole subband in $Zn_{0.76}Cd_{0.24}Se$ well layers.

4. Raman Spectra

The RT Raman spectra excited by 514.5 nm, 488.0 nm and 476.5 nm laser lines are manifested in Fig. 3 (a), (b) and (c), respectively. As shown in Fig. 3 (b) and Fig. 4,

under direct excitation with the 488.0 nm line of an argon laser the $2LO_w$ phonon scattering has maximum intensity, and further, the $2LO_w$ peak falls right on top of the PL peak at 2.478 eV. In such a case, the observed Raman $2LO_w$ scattering is exactly out-going resonant with the PL peak at 2.478 eV.

The experiment results in Fig. 3 show that the LO_{w} -phonon frequency is 249.5 cm⁻¹ (30.9 meV), a few cm⁻¹ smaller than that of LO for pure ZnSe ⁴⁾. It is known that the energy gap of barrier layer ZnSe at RT is much larger than photon energies of excitation lines above mentioned. Thus the ZnSe barrier layers are transparent for the incident light of 514.5 nm, 488.0 nm and 476.5 nm. However, the incident photon energies of 488.0 nm and 476.5 nm laser lines are larger than the band gap of $Zn_{0.76}Cd_{0.24}Se$ well layers in the SL at the same temperature. It is reasonable to consider that the LO_{w} phonon mode is ascribed to that of $Zn_{0.76}Cd_{0.24}Se$ well layers.

As shown in Fig. 4, a striking feature of the $2LO_w$ outgoing resonance is its sharpness and the large intensity of the $2LO_w$ phonon. The latter is almost an order of magnitude stronger, compared to the LO_w -phonon intensity. We believe the sharp $2LO_w$ resonance reported in this study is a double resonance involving the intermediate and out-going channels. The relevant electron-phonon scattering process is schematically shown in Fig. 4 as an inset. In the diagram the two-phonon scattering process is regarded as an iterated one-phonon interaction.

Because the energy difference between the 476.5 nm photon and PL peak at 2.554 eV is situated in the middle of $\rm LO_w$ and $\rm 2LO_w$ phonon energues, as a result Raman intensity both of $\rm LO_w$ and $\rm 2LO_w$ excited by 476.5 nm at RT is all enhanced resonantly. The situation can be seen clearly in Fig. 1 (b) and Fig. 3 (c). The resonance conditions are benefical to the observation of higher order multi-phonon Raman scattering. As shown in Fig. 3, four orders both of anti-

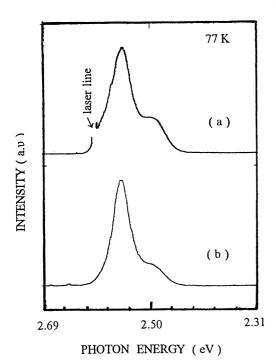


Fig. 2 $\,$ PL spectra of Zn $_{0.76}{\rm Cd}_{0.24}{\rm Se}$ -ZnSe SL excited by (a) 476.5 nm and (b) 457.9 nm laser lines at 77 K.

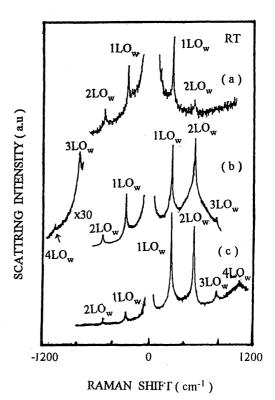


Fig. 3 Stokes and anti-Stokes Raman spectra of $Zn_{0.76}Cd_{0.24}$ Se-ZnSe SL excited by (a) 514.5 nm, (b) 488 nm and (c) 476.5 nm laser lines at RT.

Stokes and Stokes LO_w multi-phonon modes confined in the $Zn_{0.76}Cd_{0.24}Se$ well layers could be seen unambiquously.

In contrast with the case aforesaid, only the first order scattering of $\rm LO_w$ phonon could be observed at 77 K, using 476.5 nm line as an excitation light. And there is not any Raman spectra excited by 488.0 nm and 514.5 nm laser lines at 77 K.

In Fig. 5 (a) are shown Raman scattering spectra of LO_b phonon mode for the $\rm Zn_{0.76}$ $\rm Cd_{0.24}$ Se-ZnSe sample excited by 457.9 nm laser line at RT. Comparing and checking carefully the phonon frequencies in Fig. 3 and Fig. 5 (a) we find that LO_w and LO_b are two different phonon modes. As aforementioned the frequency of LO_w phonon is 249.5 cm⁻¹, however, that of LO_b phonon is 251.5 cm⁻¹, 2 cm⁻¹ larger than that of LOw, and similar trends to the higher order modes as well. The frequency 251.5 cm⁻¹ is very close to that of LO mode for pure ZnSe ⁴⁾. We believe the LO_b is a ZnSe-like LO mode, it originates probably from the barrier layers of the SL structure. In the other hand, we know that the band gap 5) of ZnSe barrier layers at RT is quite near to the incident photon energy of 457.9 nm, which satisfies nearly condition of incoming resonance, thus we believe the LOb mode is assigned to that of the ZnSe barrier layers in the SL be reasonable. In this case, five orders of LOb phonon modes are

When the experiment temperature is at 77 K, three orders Raman scattering can be observed unambiquously, using the 457.9 nm laser line excitation (see Fig. 5 (b)). In the situation, an interesting new phenominon occurs, namely the Raman peaks shift toward higher energy and make the frequency of its first order phonon mode even smaller several cm $^{-1}$ than that of LO_{w} . The seperations among corresponding Raman peaks, however, remain about the same as those of LO_{h} at room temperature. The reason

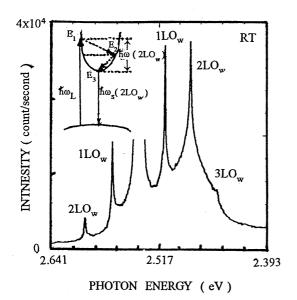


Fig. 4 PL and resonant Raman scattering spectra of $Zn_{0.76}Cd_{0.24}Se-ZnSe$ SL excited by 488 nm laser line. At the resonance, the $2LO_{\rm w}$ Raman peak falls right on top of 2.478 eV PL peak. The diagram shown as an inset illustrates the resonant Raman scattering process for the $2LO_{\rm w}$ phonon.

giving rise to this is not understood at present.

From our experiment, some features of multi-phonon Raman scattering both of $\mathrm{LO_w}$ and $\mathrm{LO_b}$ are displayed: less than 1% frequency deviation of higher order $\mathrm{LO_w}$ ($\mathrm{LO_b}$) from those of the exact $\mathrm{LO_w}$ ($\mathrm{LO_b}$) overtones and separation of the frequencies neighboring multi-phonon modes getting smaller and smaller with the order number of phonon mode increasing.

5. Summary and Acknowledgment

Multi-phonon Raman scattering of two kinds ZnSe-like LO phonon modes, LO_w and LO_b in $Zn_{0.76}Cd_{0.24}Se$ -ZnSe SL structure are studied in detail by resonance Raman scattering and up to five orders of ZnSe-like LO_b phonon modes confined in ZnSe barrier layers and four orders of

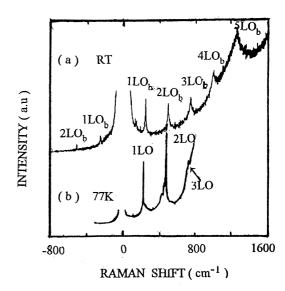


Fig. 5 Raman Spectra of Zn_{0.76}Cd_{0.24}Se-ZnSe SL excited by 457.9 nm laser line at RT and 77 K.

ZnSe-like LO_w phonon modes confined in the $Zn_{0.76}Cd_{0.24}Se$ well layers of the SL structure were observed. In addition, three PL peaks located at 2.478 eV, 2.554 eV and 2.387 eV were observed also at RT. This work is supported by Nature Science Foundation of China.

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