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Multiphonon relaxation in ZnSe thin films and ZnSe/ZnCdSe superlattice

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Abstract. The photoluminescence (PL) and resonant Raman scattering (RRS) spectra of thin films and multiple quantum well (MQW) structures were investigated at room temperature. The light scattering spectra of MQW consist of intensive band of the quantum wells luminescence and additional narrow bands. The energy shift of these additional bands was equal to the multiple value of longitudinal oscillation phonon of the strained ZnSe barriers. The intensity of these bands is resonance amplified at approaching the excitation radiation frequency to MQW luminescence bands frequency. We suppose, that this process consists of several stages: absorption of stimulating light by quantum wells; exchange of energy between hot electrons and barriers; relaxation of system including longitudinal phonon generations in barriers.

Introduction

The structures with single and multiple quantum wells are objects of intensive experimental and theoretical investigations. The interest for II–VI low dimensional structures is caused by possibility of creation of effective light-emitting devices with spontaneous or coherent radiation completely overlapping visible region of spectrum. In this work the optical properties of thin ZnSe films and MQW ZnSe/ZnCdSe structures were researched by methods of a PL and RRS.

1. Experimental procedure

All investigated samples were grown by molecular-beam epitaxy (MBE) on GaAs (100) substrates. Homogeneous ZnSe films were of various width: from 100 Å up to 9000 Å. The MQW structures configuration included a ZnSe buffer layer with thickness 1 μm and periodical sequence of $Cd_xZn_{1-x}Se$ quantum wells separated by ZnSe potential barriers. The total quantum-size region thickness equals $\approx 1.5\mu$m for all structures. The thickness of the ZnSe cap layer on the surface of structures is 300 Å. The additional information on a quantum well composition was received by measuring Auger-spectrums of $Cd_xZn_{1-x}Se$ films of the greater thickness in the analytical chamber of MBE unit and from energy standing of the low-temperature photoluminescence lines of the MQW structures. All used methods gave the coinciding results. The main structure parameters are given in the Table 1. The PL and RRS spectra are registered at room temperature on a U-1000 spectrometer in

Table 1. List of structure parameters.

MQW sample number	Widths of quantum wells (Å)	Molar concent. of cadmium, x	Width of ZnSe potential barriers (Å)	Quantity of quantum wells	RS band frequensy (cm^{-1})
178	66	0.4	100	100	246
182	80	0.5	500	30	248
183	55	0.27	1000	15	250
184	61	0.3	5000	4	250

geometry of "backscattering". RRS and PL spectrums were excited by 4416 Å, 4880 Å and 5145.3 Å laser lines for all samples. The resolution was 1–5 cm^{-1}.

2. Results and discussion

In Fig. 1 the RRS spectrums both films and MQW structures at 4416 Å laser line excitation are shown. The energy of quantum of this line is higher than energy of a ZnSe forbidden region. The spectrums consist of the narrow bands and the wide bands of a ZnSe edge luminescence. On the diagram the spectrums were shifted upwards in accordance with increasing of film width or MQW barrier width. It is easy to see that the spectrums have common features. The changes in a spectrum (varying distribution of band intensity) occur in accordance with increasing of ZnSe layer width. The narrow bands with shifted on the multiple value frequencies are observed in RRS spectrums of polar semiconductors. This shift is featured by expression $n\omega_{\text{LO}}$, where ω_{LO} is energy of longitudinal oscillation of a semiconductor lattice, $n \sim 1, 2, 3 \ldots$ [1]. The smoothly varying distribution of these band intensity in RRS spectrums ZnSe films at change of film thickness from 1 μm up to 0.1 μm was observed in [2]. Results of experiments was explain on the basis of cascade model. In this model the electrons, excited highly in a conduction band, can relax, letting out one behind other LO-phonons.

Apparently in our case there is a same process of an excited electron relaxation including of ZnSe LO-phonons both in ZnSe films and in MQW barriers. The basic change of a spectrum (see Fig. 1) occurs at thickness 100–2000 Å. In paper [2] basic changes of a spectrum occurred at thickness 1000–10000 Å. It is possible to explain by higher quality of ZnSe films structure (in [2] the films obtained by cathode sputtering were explored).

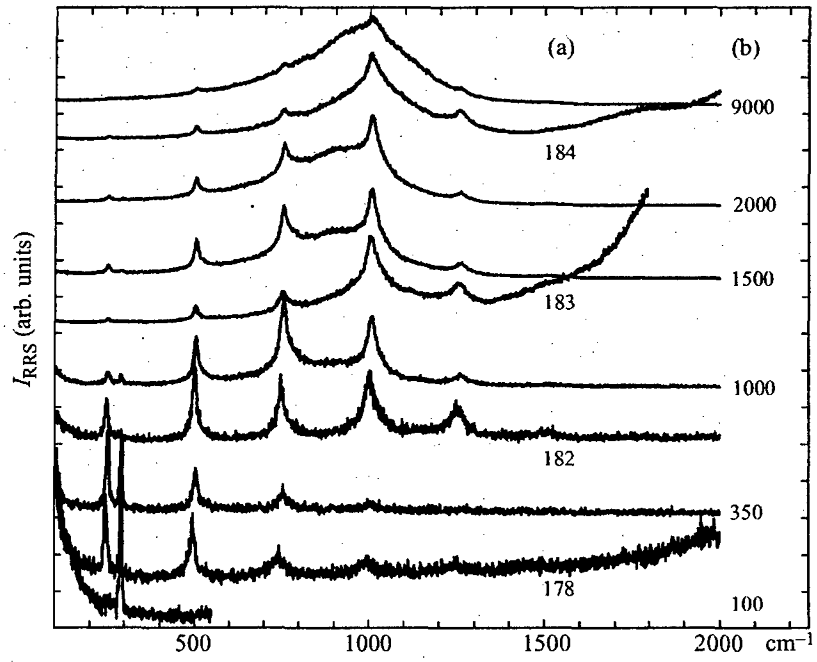

Fig. 1. Resonant Raman scattering spectra of thin films and MQW structures at room temperature. (a) MQW sample number. (b) ZnSe film width (Å).

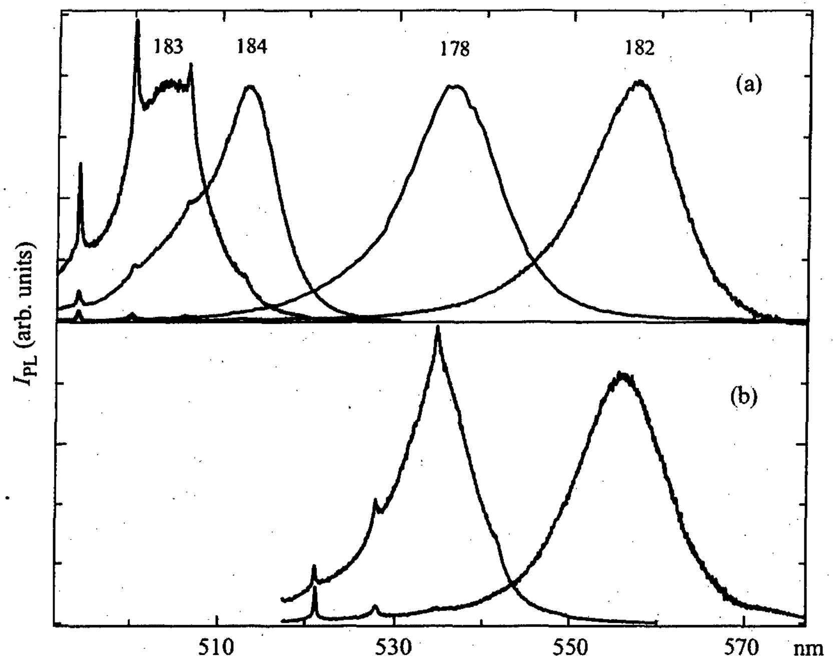

Fig. 2. PL spectra of various ZnSe/ZnCdSe MQW structures at excitation by various lines of the Ar laser: (a) 4880 Å and (b) 5145.3 Å.

PL spectra for MQW samples at excitation by 4880 and 5145.3 Å laser lines are shown in Fig. 2. The energies of quantum of these lines have an intermediate value between energy of a ZnSe-barrier forbidden region and energies of electron-hole pairs recombination in quantum wells. Each of samples has a luminescence band with frequency position determined by properties of quantum wells. Since the quantum well widths in different structures was varied in small limit, the frequency position of a luminescence band is determined mainly by concentration of Cd in the quantum wells. There is a set of narrow bands in a spectrum of each sample alongside with QW luminescence band. The frequency position of these narrow bands are shifted from excitation light energy on the value multiple energies 246–251 cm^{-1} for various samples (Table 1). The intensity of these additional bands is being resonant increased at approaching of excitation light energy to luminescent bands energy of the quantum wells.

In our samples the ZnSe buffer layer and barriers of the MQW structures are transparent for 4880 and 5145.3 Å exciting lines and, therefore, LO-phonon repetitions can not be excited in ZnSe. The availability of LO-phonon repetitions and absence of TO-phonons in spectrums testify that these spectra are obtained in RRS conditions. It would seem, these phonon repetitions could be related to solid solutions $Cd_xZn_{1-x}Se$ oscillations of a well. Nevertheless these bands in our sample spectrums can be connected with ZnSe because of their range frequencies. Really, the frequency dependence of longitudinal phonon in a solid solution $Cd_xZn_{1-x}Se$ on a cadmium concentration x is known [[illegible]]. When the value x in limits 0.27–0.5 changes (what was used in this work at manufacture of MQW) the frequency of a LO-mode in solid solutions $Cd_xZn_{1-x}Se$ must change in limits 245–240 cm^{-1} accordingly [[illegible]]. In this work the band frequencies change from 251 up to 246 cm^{-1}. The estimation of a cadmium contents in quantum wells on the basis of the measured LO-modes frequencies contradicts results of definition of the quantum wells parameters by all listed above methods *in situ* and *ex situ*. For example, the cadmium molar concentrations for samples N183 and N182, was calculated on the PL band frequency positions (Fig. 2), differs on 0.23, but concentrations calculated on the RS band frequency positions (Table 1) gives the differs in cadmium concentrations about 0.02 [[illegible]]. In addition, for each sample, energy shift of additional bands does not depend on a wavelength of exciting light. Therefore it is more reasonable to assign these bands to a material of a barrier. The small shifts of these band frequencies can be explained by internal strains in structure caused by mismatch of crystalline lattice periods of ZnSe barriers and $Cd_xZn_{1-x}Se$ quantum wells [[illegible]].

3. Conclusion

At a photoexcitation of MQW structures with exited light quantum energy higher than energy of a ZnSe forbidden region, a cascade relaxation of hot electrons in barriers including of longitudinal phonons of the ZnSe barrier occurs.

If the exited light quantum energy have an intermediate value between energy of a ZnSe-barrier forbidden region and energies of electron-hole pairs recombination in quantum wells, this process occurs in some stages:
(a) the light quantum is absorbing by a quantum well. This process is especially effective if the energy of exciting light is closer to the luminescence line energy of the quantum wells;
(b) there is an exchange of energy between hot electrons and barriers because QW thickness is close to Bohr radius of excitons in QWs;
(c) there is a relaxation of energy of an electron through sequential interaction to longitudinal oscillations of the barrier material.

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