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LOW-DIMENSIONAL SYSTEMS

# Influence of the ZnTe Barrier Width on Photoluminescence Spectra of CdTe/ZnTe Superlattices with Layers of Quantum Dots

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**Abstract**—The photoluminescence and Raman spectra of CdTe/ZnTe superlattices with layers of CdTe quantum dots have been measured in the temperature range of 5–85 K. It has been found that, in the photoluminescence spectra, there are three emission bands, which are attributed to the luminescence of two-dimensional wetting layers, direct excitons, and spatially indirect excitons in quantum dots, respectively. The influence of the width of the ZnTe barrier layer in the range of 2–50 ML on the energy and width of the emission lines of quantum dots and wetting layer has been analyzed. The temperature dependences of the photoluminescence intensity of direct and indirect excitons have been investigated and the corresponding activation energies of temperature quenching of luminescence have been determined.

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## 1. INTRODUCTION

Epitaxially grown self-assembled quantum dots (QDs) belong to the most promising low-dimensional structures due to the three-dimensional size quantization of their electron spectrum and excellent optical properties. In a number of studies, it was shown that, in multilayer structures under appropriate growth conditions, it is possible to obtain either vertical or lateral ordering of self-assembled quantum dots, or simultaneously both types of correlations. These phenomena were observed in nanostructures based on semiconductors III–V [1], IV–VI [2], and II–VI [3, 4]. In particular, in II–VI structures CdSe/ZnSe, there are two models of correlations depending on the thickness of the ZnSe spacer layer between the adjacent planes of CdSe [3]. In one case, islands in the overlying layers are formed directly above the islands located in the lower layers, whereas in the other case, they are located between the islands of the lower layer. A different picture is observed in CdTe/ZnTe superlattices with layers of CdTe quantum dots [4, 5]: when the thickness of the ZnTe spacer is more than 25 monolayers (ML), the positions of islands located in the adjacent layers of quantum dots are not correlated. When the thickness of the spacer is less than 20 ML, quantum dots the overlying layer nucleate between the islands located in the underlying layer. In particular, quantum dots in this structure tend to be aligned along the axis inclined by an angle of 40° with respect to the growth direction. The inclination angle is determined by the fact that the minima of the elastic energy in barrier layers appear in the [110] direction [2, 6]. A transmission electron microscopy (TEM) image of the cross section of such structure with a spacer thickness of 15 ML is presented in [4].

The use of quantum dots in high-speed optical devices requires the understanding of the relaxation of charge carriers. Investigation of the relaxation involving optical phonons is particularly important, because it is the most rapid relaxation in semiconductors. In the II–VI semiconductor structures, the electron–phonon interaction is very strong in contrast to the III–V structures. However, there are a very limited number of studies on the relaxation of hot carriers or excitons over a wide energy range, including the ZnTe matrix, the nearest environment of the quantum dots, and the wetting layer. In [7], these phenomena were investigated using the example of a single layer of CdTe quantum dots in the ZnTe matrix. Of particular interest is the interaction of pairs of quantum dots in the adjacent layers, which manifests itself in the appearance of an additional low-energy photoluminescence band.

In this work, we investigated the photoluminescence (PL) of wetting layers, direct excitons, and spatially indirect excitons in quantum dots in the CdTe/ZnTe superlattice with a change in the width of the ZnTe barrier layer from 2 to 50 ML (1 ML ZnTe = 3.05 Å). The measurements were performed in the temperature range of 5–85 K. The Raman method was used to investigate the relaxation of hot excitons when the laser excitation energies were greater and less than the ZnTe band gap: 2.409, 2.33, 2.18, and 1.91 eV.

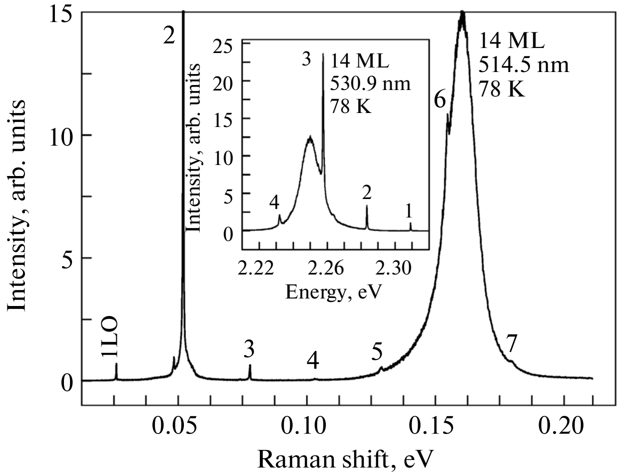


**Fig. 1.** Raman spectra of the CdTe/ZnTe superlattice with a barrier width of 14 ML upon excitation by an $Ar^+/Kr^+$ laser with wavelengths $\lambda = 514.5$ and 530.9 nm at 78 K.

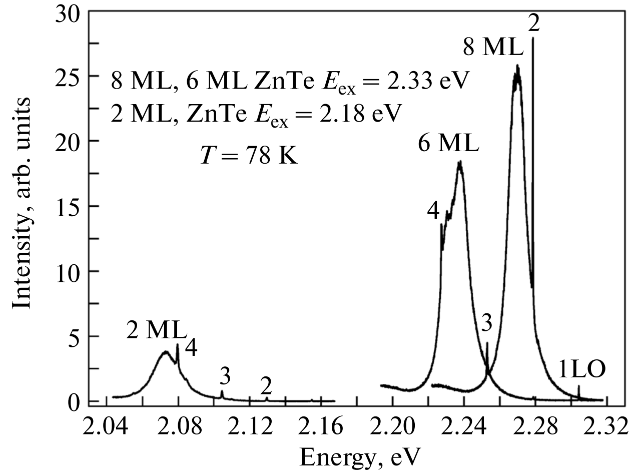


**Fig. 2.** Raman spectra of the CdTe/ZnTe superlattice with ZnTe barrier widths of 2, 6, and 8 ML upon excitation by an $Ar^+/Kr^+$ laser with energies of 2.33 and 2.18 eV at 78 K.

## 2. SAMPLES AND EXPERIMENTAL TECHNIQUE

Raman spectra of CdTe/ZnTe structures with layers of quantum dots upon excitation by an $Ar^+/Kr^+$ laser with wavelengths of 488, 514.5, 568.1, 530.9, and 647 nm were recorded on a U-1000 spectrometer equipped with a microscope in the backscattering geometry with a resolution of 1.5 $cm^{-1}$. The measurements were performed at a temperature of 78 K. The laser radiation power was 10 mW. The size of the spot on the sample was approximately 30 μm.

Photoluminescence (PL) spectra were recorded on a SpectraPro2500i spectrograph equipped with a cooled multichannel CCD detector (Spec-10). The spectral resolution was 0.05 meV. Photoluminescence was excited by an argon laser with a wavelength of 514.5 nm. The laser radiation power was varied from 0.05 μW to 9.3 mW with the spot size of approximately 200 μm. The measurements were performed in the temperature range of 5–85 K.

We studied seven 200-period structures containing layers of self-assembled CdTe/ZnTe quantum dots grown by molecular beam epitaxy on GaAs(100) substrates with a CdTe(3 μm)/ZnTe(1.7 μm) buffer layer [4]. The nominal thickness of the CdTe layers was 3 ML, and the width of the ZnTe barrier layers was varied in the range of 2–50 ML.

The examination of the cross sections of the samples on a high-resolution transmission electron microscope (TEM) revealed that quantum dots had the form of 2- to 3-nm disks with a diameter of 10–20 nm. In this case, islands in the overlying layer were located between the islands in the underlying layer [4].

## 3. RESULTS OF THE EXPERIMENT

### *3.1. Raman Spectra of a Wetting Layer in CdTe/ZnTe Superlattices with CdTe Quantum Dots at 78 K*

The Raman spectrum of the sample with the ZnTe barrier width equal to 14 ML at excitation energies of 2.41 and 2.33 eV is shown in Fig. 1. It is a multiphonon spectrum with a strong resonance amplification of the 2LO line and a weaker resonance amplification of the 6LO line. The first resonance corresponds to the energy of the electronic transition $E = 2.356$ eV, which coincides with the band gap of the ZnTe matrix at 78 K. However, the series of LO peaks is not limited by the matrix ZnTe. It can be seen from this figure that the 4LO line is already very weak, but, then, we observe an increase in the intensity of the 5LO lines and, especially, the 6LO line, which is located near the maximum of the luminescence band at an energy of 2.245 eV. The Raman spectrum of this sample at an excitation energy of 2.33 eV is shown in the inset to Fig. 1. At this excitation energy, which is less than the ZnTe band gap, the resonance character of the multiphonon spectrum is determined by the electronic transition energy equal to 2.245 eV. This emission line is attributed to transitions between the levels in a quantum well formed by the CdTe wetting layer with a thickness of approximately 2 ML (1 ML = 3.24 Å). The existence of this layer is caused by the growth of self-assembled quantum dots according to the Stranski–Krastanow model [4, 8]. The fact that the relaxation of hot excitons in the CdTe quantum well occur with the emission of ZnTe LO phonons ($\omega_{LO}$ = 209 $cm^{-1}$) suggests that the wave functions of electrons in the quantum well to penetrate into the barrier.

Figure 2 shows the Raman spectra of the structures with ZnTe barrier widths of 2, 6, and 8 ML at excitation energies of 2.33 and (in the first case) 2.18 eV. All

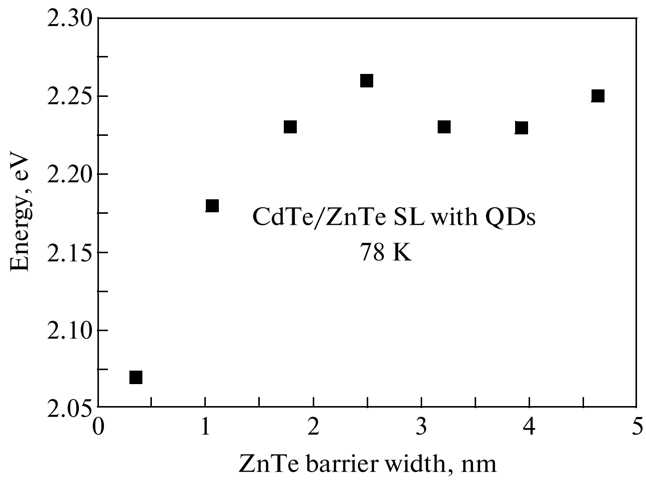


**Fig. 3.** Dependence of the energy at the PL maximum for quantum wells formed by the CdTe wetting layer on the width of ZnTe barriers in CdTe/ZnTe superlattices at 78 K.

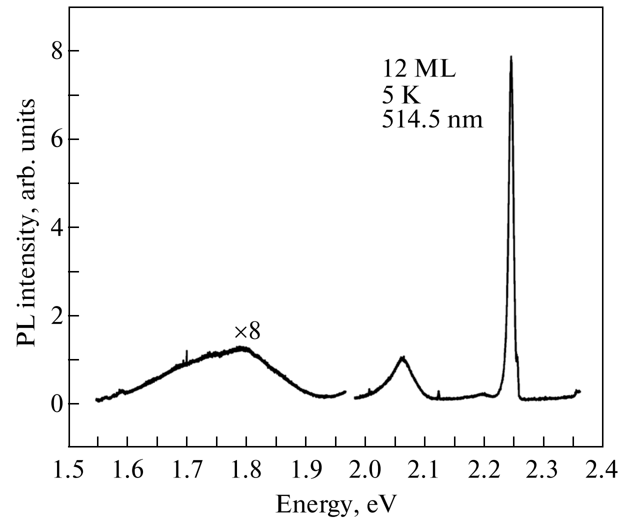


**Fig. 4.** PL spectrum of the CdTe/ZnTe superlattice with the ZnTe barrier width of 12 ML upon excitation by the laser with the wavelength $\lambda = 514.5$ nm at 5 K.

the spectra have a multiphonon resonance character [9]. It can be seen from this figure that the energy of the luminescence maximum decreases with a decrease in the width of the barrier. The dependence of this energy on the barrier width at 78 K is shown in Fig. 3. As the barrier width decreases from 14 to 2 ML, the luminescence energy decreases by 170 meV. This result agrees well with the calculation of quantum levels within the Kronig–Penney model for the two tunnel-coupled CdTe/ZnTe quantum wells with a width of 2 ML (6.8 Å), which is equal to the thickness of the wetting layer. From the calculation, it follows that the energy of the lower level decreases by approximately 150 meV with a decrease in the barrier width from 14 to 2 ML, which is in satisfactory agreement with the experimental results. We measured the dependences of the PL intensities ($I$) of the wetting layer and quantum dots on the excitation power ($P_{ex}$) in the samples with barrier widths of 12 and 4 ML at 5 K. In both samples, these dependences are close to linear with the exponent $k = 1.04–1.10$ in the expression $I = (P_{ex})^k$.

### *3.2. Photoluminescence Spectra of Quantum Dots at Low Temperatures*

In the PL spectrum of the CdTe/ZnTe structure with a barrier width of 12 ML, there are three bands with maxima at energies of 2.25 eV (band width 8 meV), 2.06 eV (band width 40 meV), and 1.79 eV (band width 270 meV) (Fig. 4). The first band is attributed to the emission of the quantum wells formed by CdTe wetting layers, whereas the second and third bands are assigned to the luminescence of quantum dots. According to the interpretation proposed in [6], the second band is determined by the recombination of excitons in isolated, not interacting quantum dots, when an electron and a heavy hole coupled into excitons are localized in a single quantum dot (direct exciton). The third, low-energy band is associated with the recombination of excitons in a vertically correlated closely spaced quantum dots. In this case, the contribution to the recombination comes from electrons and holes localized in spatially separated adjacent quantum dots (indirect exciton) [6, 10, 11]. The luminescence decay time of this band is one order of magnitude greater than the decay time of the luminescence of the direct exciton [4, 6].

The superlattice with the ZnTe barrier width of greater than 25 ML is dominated by the formation of isolated quantum dots. Our investigations revealed that, in the PL spectrum of the CdTe/ZnTe structure with a barrier width of 50 ML, there is only one emission line with the maximum at an energy of 2.04 eV at 78 K and the PL band width of 40 meV. The parameters of this PL band are close to the parameters of the second band in Fig. 4.

The PL spectrum of the sample with a barrier width of 2 ML at 5 K is shown in Fig. 5. In this spectrum, there are three emission bands at energies of 1.93 eV (band width 52 meV), 2.0 eV (band width 23 meV), and 2.08 eV (band width 19 meV). The PL band with an energy of 2.08 eV is associated with the emission of the wetting layer. At $T = 78$ K, the energy of this band is equal to 2.07 eV (Fig. 2). Bands with maxima at energies of 2.0 and 1.93 eV correspond to the emission of direct and indirect excitons, respectively. In the spectrum of this sample, in contrast to the spectrum of the sample with a barrier width of 12 ML, the intensity of the emission band of indirect excitons is significantly higher than that of direct excitons. This indicates that the number of correlated quantum dots increases and exceeds the number of uncorrelated

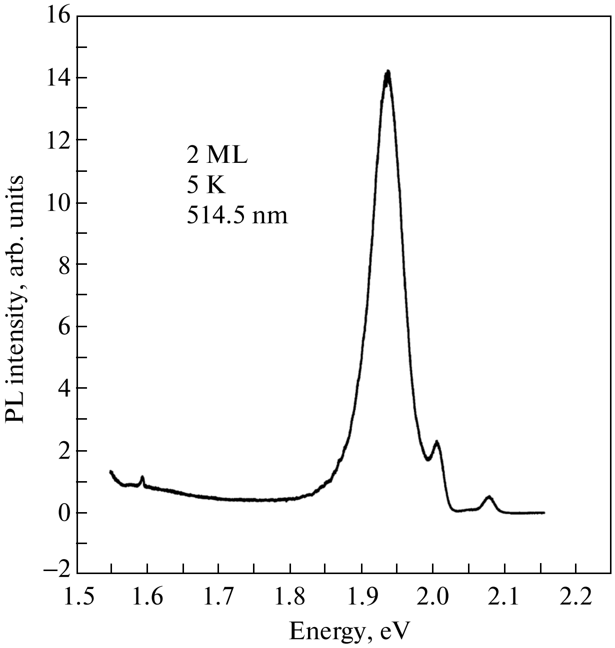


**Fig. 5.** PL spectrum of the CdTe/ZnTe superlattice with the ZnTe barrier width of 2 ML upon excitation by an $Ar^+$ laser with the wavelength $\lambda = 514.5$ nm at 5 K.

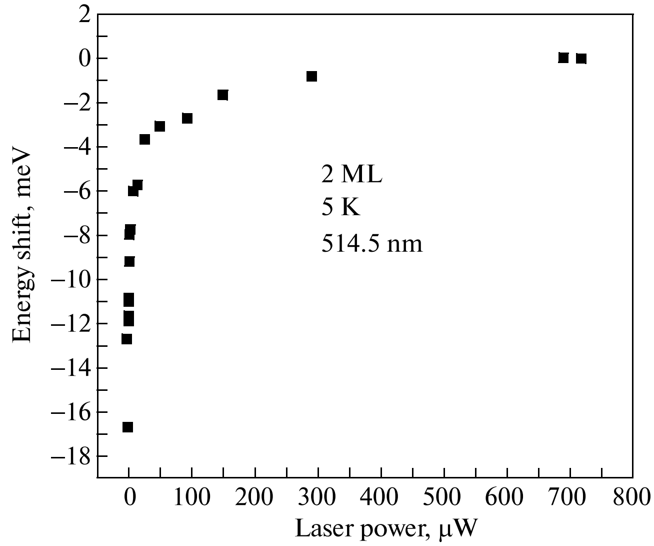


**Fig. 6.** Dependence of the energy shift of the PL maximum for indirect excitons in the CdTe/ZnTe superlattice with the ZnTe barrier width of 2 ML on the laser power at 5 K. The PL energy shift at the maximum laser power is taken as zero.

quantum dots. A decrease in the barrier width leads to a significant decrease in the PL line width of direct and indirect excitons. This is probably due to the more uniform size distribution of the quantum dots. The PL band of the direct exciton is shifted by an energy of 60 meV toward the red region with a decrease in the barrier width to 2 ML. This can be associated with the tunneling interaction of the ground states of excitons in adjacent quantum dots. The splitting of the ground state of the exciton in InAs/GaAs quantum dots is approximately 30 meV for the GaAs barrier width equal to 4 nm [10]. The PL band of the indirect exciton is shifted toward higher energies. We measured the temperature dependences of the PL intensity of direct excitons in the temperature range of 5–85 K at an excitation power of 290 μW in the samples with barrier widths of 12, 4, and 2 ML. The activation energies $\Delta E$ calculated according to the formula given in [12] are equal to 26, 22.6, and ~7 meV, respectively. The values of $\Delta E$ are determined by the activation of heavy holes through the effective barrier whose height decreases with a decrease in the ZnTe barrier width due to the decrease of the compressive strain in the CdTe layers. These results are preliminary. The relationship between the activation energy and the energy band structure requires more detailed investigation [13]. All the studied samples are characterized by a two- or three-fold increase in the PL intensity of excitons in the temperature range of 5–35 K. In our opinion, this is explained by the ionization of shallow impurities or defects in the ZnTe barrier.

An increase in the excitation power leads to a blue shift in the maximum of the PL energy of the indirect exciton in the sample with a barrier width of 2 ML, in which the PL band has the smallest width (Fig. 6). The maximum change in the PL energy is 17 meV at a laser excitation power of 720 μW. The blue shift was observed in the PL spectra of indirect excitons in the structures with two quantum wells based on III–V compounds [14, 15, 16]. The PL energy of the direct exciton does not depend on the excitation power. We failed to observe the PL of indirect excitons upon excitation with a resonance energy of 1.91 eV. The nature of the PL band with the maximum at 1.93 eV in the sample with a spacer width of 2 ML remains unclear. Possibly, it is associated with an indirect exciton of another type: an electron in the CdTe quantum dot and a light hole in the ZnTe layer. In this sample, tensile strains should arise in the ZnTe layer, which will lead to a shift of the band of light holes toward higher energies and to the formation of quantum wells for light holes. This assumption is confirmed by a decrease in the frequency of ZnTe LO phonons in the barrier by 4 $cm^{-1}$ as compared to other samples.

## 4. CONCLUSIONS

The Raman spectra of the superlattice with the ZnTe barrier width of 2–14 ML have a multiphonon (with the participation of ZnTe LO phonons) resonance character in the energy range of 2.08–2.24 eV, which is typical for processes of relaxation of excitons in a two-dimensional quantum well. Based on these data and the dependence of the luminescence energy on the barrier width, we concluded that the luminescence bands in the given energy range are attributed to the superlattice consisting of quantum wells formed by wetting layers of CdTe. It was found that the PL energy of direct excitons decreases by 60 meV in CdTe quantum dots with a decrease in the width of the barrier layer to 2 ML, which indicates tunneling interaction of 1*s* exciton states. The thermal activation energies $\Delta E$ of excitons with a heavy hole at an excitation power of 290 μW have been determined in structures with barrier widths of 2, 4, and 12 ML. It was found that the emission energy of indirect excitons increases with an increase in the excitation power, whereas the emission energy of direct excitons does not exhibit this dependence.

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