Hindawi Publishing Corporation
International Journal of Photoenergy
Volume 2009, Article ID 358790, 4 pages
doi:10.1155/2009/358790

Research Article

Photoluminescence Spectroscopy of CdTe/ZnTe Self-Assembled Quantum Dots

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Received 26 June 2009; Revised 27 October 2009; Accepted 9 November 2009

Recommended by Mohamed Sabry Abdel-Mottaleb

We present photoluminescence (PL) measurements of two different, 3 monolayers and 12 monolayers (ml), CdTe self-assembled quantum dot (SAQD) samples. The spectra were recorded in the temperature range 20 K–300 K, with photoexcitation over the ZnTe barrier layer. PL spectra displayed two main emission bands. High-energy PL emission (E_1) is ZnTe LO like phonon-(ω_{LO} = 204.2 cm^{-1} (3 ml), ω_{LO} = 207.3 cm^{-1} (12 ml)) assisted deexcitation. Dominant low-energy band (E_2) presents the direct deexcitation to ground state of the CdTe quantum dots.

1. Introduction

Great interest has been observed recently in studying the properties of layers of semiconductor materials with a mismatch between their lattice parameters. Favourable conditions for the formation of self-assembled quantum dots are created in such layers [1]. The wide band-gap CdTe/ZnTe system is current interest because of its potential applications in short-wavelength optoelectronic devices, but the large lattice mismatch (6.4%) makes it very difficult to grow CdTe/ZnTe structure of high quality [2].

In the majority of experiments reported so far, dots with sizes larger than the exciton Bohr radius have been studied [3–5]. For instance, the typical size of II-VI CdSe QD's, where the exciton Bohr radius is equal to 3 nm, ranges from slightly larger than this value [3] to even 15 nm in diameter [4]. In these cases then, leakage of the exciton wave function into the barriers is not expected to change significantly for QD size distributions within an ensemble. Consequently, for large QDs no size dependence of the exciton-LO phonon coupling is observed.

Detailed measurements of PL, photoluminescence excitation (PLE), and resonant PL spectroscopy on similar systems are performed in literature [1, 5]. To study the carrier excitation processes in CdTe/ZnTe SAQD excitations below energy of ZnTe gap were used to identify major carrier excitation mechanisms in CdTe QD's [5].

In our earlier papers we using far-infrared spectroscopy [6], Raman spectoscopy [7], and resonant Raman spectroscopy [8] to investigate structural and phonon properties of CdTe/ZnTe SAQD. By extending this study to phtoluminescence spectroscopy we expect to obtain electronic structure and get complete picture of optical properties of these samples.

2. Samples and Characterization

Investigated samples were grown by molecular beam epitaxy on GaAs substrate. CdTe buffer layer, 4 μm thick, was deposited on the substrate. After a 0.6 μm thick ZnTe layer, 3 or 12 monolayers (ml) of CdTe were deposited to form a random distribution of quantum dots. The dot layer was covered by 0.1 μm ZnTe capping layer, which is schematic presented in Figure 1. High mismatch of lattice parameters of CdTe (6.482 Å) and ZnTe (6.104 Å) provokes gathering of CdTe molecules and forming of quantum dots. Further details of the samples growth can be found in

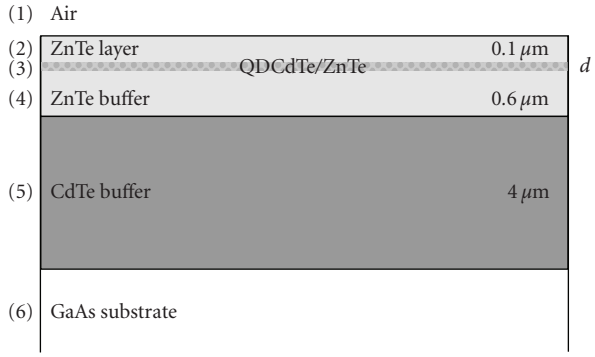

Figure 1: A schematic presentation of a six-layer structure.

[1]. It is important to note that the chosen sample set enables us to study quantum dots (QD) with different lateral sizes. Namely, CdTe QDs are very small, and, as estimated by transmission electron microscopy and magneto-photoluminescence measurements, their lateral size is of the order of 2–4 nm in diameter in sample with 3 ml. In second sample (12 ml) there are no CdTe islands; that is, there is a layer with CdTe regions surrounded by alloyed material. This work is focussed to emphasize different properties of these two cases.

Photoluminescence spectra (PL) were excited by several lines of Ar-laser line (514.5 nm, 501.7 nm, 496.5 nm, 488 nm) and Kr-laser line (647.5 nm), measured using a Jobin Yvon model U-1000 monochromator, with a conventional photo-counting system. All Ar-laser lines excite excitons over the ZnTe barrier layer.

3. Results and Discussion

The electronic band structure was characterized by a PL spectrum. PL spectra depend on energy excitation. In the case of argon laser excitations the PL spectra displayed two main emission peaks. Typical PL spectra are presented in Figures 2 and 3. Registered bands are separated and no additional analysis was needed to follow the temperature dependence of the energies corresponding to the maximum emission of these bands. Results of all measurements are presented in Figure 4.

We emphasize peaks positions for temperatures at the end of measurement interval. In the case of 3 ml; we have 2.26 eV (E_1) and 1.84 eV (E_2) at $T = 300$ K, and 2.346 eV (E_1) and 2.17 eV (E_2) at $T = 20$ K. Change in E_1 energy is about 85 meV and change in E_2 is about 330 meV. Red-shift of the high-energy peak E_1 with increasing temperature is the same order of the shift that one would expect from the temperature-induced shrinking of the ZnTe band gap. This peak is registered even at room temperature. Red-shift of the low-energy peak E_2 with increasing temperature is much larger than the shift that one would expect from the temperature-induced shrinking of the CdTe band gap. In the case of 12 mL; we have 2.307 eV (E_1) and 2.11 eV (E_2) at $T = 200$ K, and 2.337 eV (E_1) and 2.13 eV (E_2) at $T = 20$ K. Change in E_1 energy is about 30 meV and change in

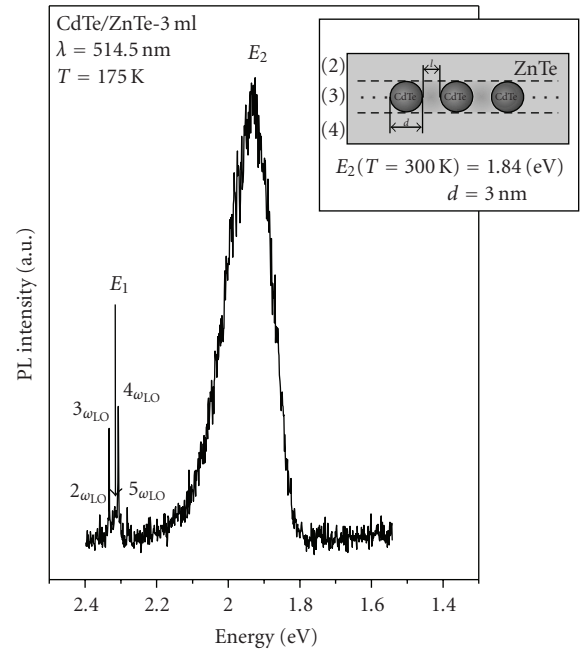

Figure 2: Typical PL spectra of sample with 3 ml of CdTe in the finishing layer; PL spectra are excited by $\lambda = 514.5$ nm Ar-laser line and recorded at 175 K. *Insert:* schematic presentation of the structure of SAQD.

E_2 is about 20 meV. Red-shift of both bands with increasing temperature is below values one would expect from the temperature-induced shrinking of the ZnTe and CdTe band gap.

Band energies vary almost linearly with temperature, as presented in Figure 4. The high-energy peak, E_1, is related to the band gap of the ZnTe barrier [9]. The low-frequency band, E_2, is related to the recombination in the CdTe quantum dots [5]. MP resonant processes are registered at temperature bellow 200 K. This process is dominant for QDs with high emission energy, that is, presumably with smaller size [5].

Energies of localized states in QDs, experimentally registered by PL emission characteristics of the QDs, can be reasonably well described by the effective-mass approximation (EMA) with parabolic energy bands. Calculation of electronic transitions is done in EMA [10]. Parameters of CdTe and ZnTe, effective masses, and conduction and valence offsets are transferred from literature [11]. If we assume spherical symmetry of a particle, that is, that single sphere of CdTe is surrounded by ZnTe, calculated value of QD diameter is 3 nm in case of 3 ml matches experimental E_2 value.

When adding a large quantity of CdTe between two layers of ZnTe, it is expected to find larger QDs. As a consequence the energy E_2 would be lower. This expectation was not fulfilled for the 12 ml sample; see Figure 4. To explain experimental results in 12 ml case, we use model core/shell type nanostructures where QD is of gradient composition. CdTe core of radius r_C is surrounded by concentric spherical

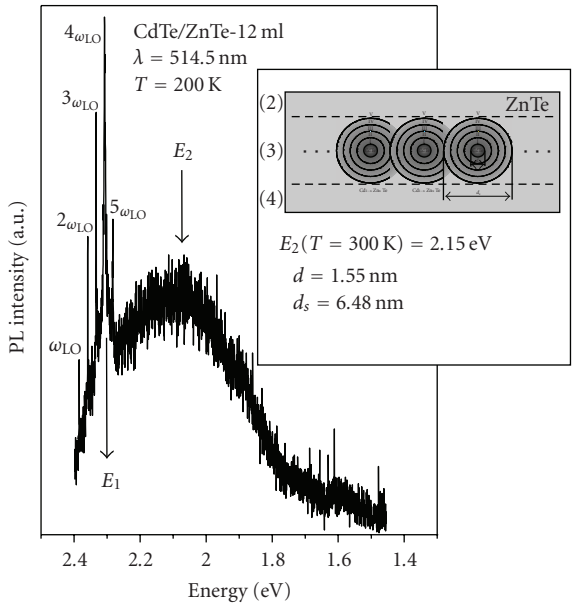

Figure 3: Typical PL spectra of sample with 12 ml of CdTe in the finishing layer; PL spectra are excited by $\lambda = 514.5$ nm Ar-laser line and recorded at 200 K. *Insert:* schematic presentation of the structure of SAQD.

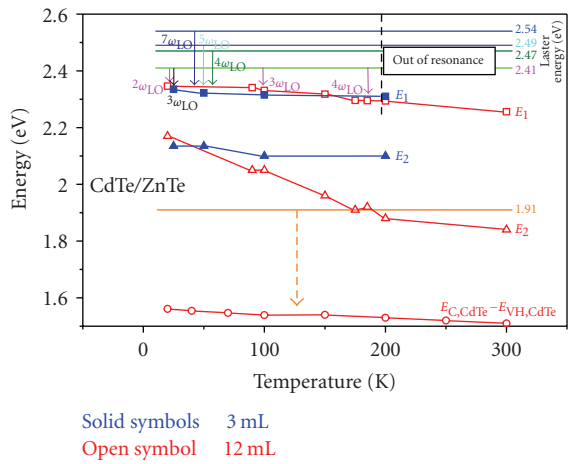

Figure 4: Temperature dependences of PL emission peaks and schematic presentation of multiphonon (MP) resonance. Results for 3 ml sample are presented as open symbols, and for 12 mL sample as solid symbols.

layers each of $Cd_xZn_{1-x}Te$ composition (x is proportional to the layer distance from the center) and finally from radius r_S by $Cd_{0.1}Zn_{0.9}Te$. We suppose that electrons and holes from QD are in spherical step-like energy well. Cd concentration in the surrounding $Cd_xZn_{1-x}Te$ gradually decreases to 10% ($x = 0.1$), step by step. We assumed that effective masses and conduction and valence offsets change proportionally to x. The existence of these intermediate layers, as a model of graded composition, influences rapidly electron, hole, and excitons spectra. For measured E_2 the diameter of the core is 1.55 nm and the diameter of outer sphere is 6.48 nm.

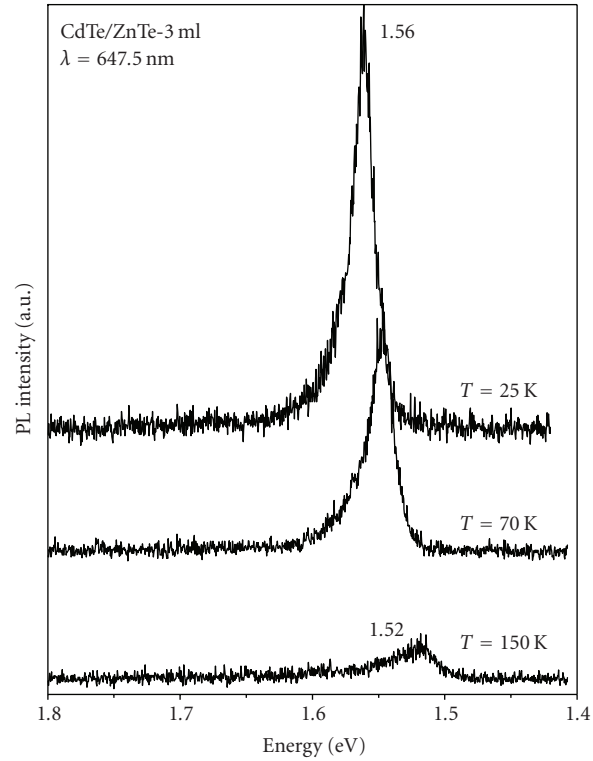

Figure 5: Typical PL spectra of sample with 3 ml of CdTe in the finishing layer; PL spectra are excited by $\lambda = 647.5$ nm Kr-laser line and recorded at different temperature.

In case of 3 ml there is red-shift of the low-energy peak (E_2) with increase of temperature, which is much larger than the shift one would expect from the temperature-induced shrinking of the CdTe band gap. In our opinion, even in the case of 3 ml, due to interdiffusion there are fluctuations of QD chemical composition and QD size distribution. So, we deal with the ensemble of QDs of different depths of the corresponding potentials. Thermally induced redistribution of carriers within the dot ensemble causes observed energy shift of photoluminescence energy. Opposed to that in the 12 ml CdTe sample all transitions are preserved due to relatively homogeneous thickness of the layer.

When the sample is excited by $\lambda = 647.5$ nm Kr-laser line, PL takes place only in the buffer layer of CdTe which is clearly seen in Figures 4 and 5. The temperature dependences of PL emissions peaks and their intensities are identical to the dependences registered for the CdTe bulk crystal.

In principle, registered MP resonant process (see Figures 2 and 3.) can be described by the following equation: $E_{\text{in}} - K\hbar\omega = E$; E_{in} and E are the incident photon energy and electron transition energy, respectively [9], $\hbar\omega$ is the phonon energy, and K is the MP order.

MP emission is not registered at $T = 300$ K. MP emission processes are registered at temperature below $T = 200$ K (for 2.41eV laser energy, $K = 4$). The equation means that when the energy of the scattered photon approached the energy E_1, the Raman line becomes strongly enhanced. This is schematically presented by arrows on the top in Figure 4.

4. Conclusion

Structural and optical properties of CdTe/ZnTe self-assembled quantum dots (SAQDs) growth by molecular beam epitaxy are investigated in this paper. Photoluminescence spectra consist of two main emission peaks: high-energy band connected to barrier band gap and low-energy band concerned to CdTe QD electron-hole recombination. Two mechanisms of relaxation are present in this system: relaxation directly to the CdTe quantum dot exciton ground state and optical phonon-assisted deexcitation. It seems that CdTe quantum dots when embeded in ZnTe barrier layers are very efficient carrier receivers. CdTe/$Cd_xZn_{1-x}Te$ heterostructures inhomogenity modelled by gradual composition influence drastically basic transition energy. The present observations can help improve understanding of the optical and the microstructural properties in the CdTe/ZnTe self-assembled quantum dots. The registered multiphonon emission processes depend on temperature. When the energy of the scattered photon approached the energy E_1, the corresponding pon line becomes strongly enhanced.

Acknowledgments

This work was supported under the Agreement of Scientific Collaboration between the Polish Academy of Science and the Serbian Academy of Sciences and Arts. The work in Serbia was supported by the Serbian Ministry of Science (Project 141028).

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