10,03

On the difference in the binding energy of 1s and 2s excitons in CdTe/(Cd,Mg)Te

© A.V. Kudinov

Ioffe Institute,
St. Petersburg, Russia

E-mail: koudinov@orient.ioffe.ru

Received May 20, 2024
Revised May 20, 2024
Accepted May 22, 2024

Photoluminescence and reflection spectra in CdTe/(Cd,Mg)Te quantum wells with a width of 60 Å with varying degrees of doping have been experimentally studied. It was found that, in accordance with previous calculations, an exciton state is observed approximately 17 MeV above the basic level of heavy exciton (1s $1e1hh$). By all indications, this condition should be associated with the exciton 2s $1e1hh$. In addition, another signal is observed higher in energy, which most likely belongs to the exciton with a light hole 1s $1e1lh$.

Keywords: photoluminescence, reflection, spectroscopy, exciton.

DOI: 10.61011/PSS.2024.06.58713.128

Quasi-two-dimensional structures such as quantum wells and superlattices based on the CdTe/(Cd,Mg)Te system have long become model objects in the physics of semiconductor nanostructures. The technology for their cultivation has been developing for about 30 years and has climbed to very great heights [1–4]. CdTe/(Cd,Mg)Te heterostructures are not much inferior to the GaAs/(Ga,Al)As family in terms of crystallographic quality. The main advantages of the CdTe/(Cd,Mg)Te system over GaAs/(Ga,Al)As are associated with spin properties. Firstly, the lower natural abundance of nuclei with spin results in a weakening of spin interactions between the lattice and band carriers and, in particular, weakens one of the channels of spin relaxation of localized electrons. Secondly, the CdTe/(Cd,Mg)Te system forms an ideal „quantum constructor" for nanostructures based on the diluted magnetic semiconductor (Cd,Mn)Te (which contains Mn^{2+} ion spins „embedded" in the crystal lattice) [5]. All of the listed materials have a zinc blende structure and belong to the $A^{II}B^{VI}$ materials, so that the magnetic ions Mn^{2+} isovalently occupy the cationic positions of the crystal lattice, without forming impurity levels in the band gap [6]. Magnetic (Cd,Mn)Te, non-magnetic (Cd,Mg)Te, and even their solid solution (Cd,Mg,Mn)Te, which provides an important opportunity to design the potential profile of heterostructures regardless of the content magnetic component in layers can be used as a wide-band material.

„Thin spectrum" of Raman scattering, consisting of five narrow lines was discovered in a recent study [7] in a (001)-CdTe/(Cd,Mg)Te quantum well with a width of 18 monolayers (60 Å) in the region of the ground exciton state 1s $1e1hh$. The width of the fine spectrum components was approximately 0.1 meV, and its total width — was approximately 1 meV with an exciton resonance width (at half maximum) of approximately 3–4 meV. There was a fine spectrum upon monochromatic excitation above the exciton energy 1s $1e1hh$ by approximately the energy of the longitudinal optical (LO) phonon. It was shown that the fine optical scattering spectrum is formed by a three-resonance denominator process involving the 2s exciton state. This process, resulting in narrow acoustic satellites LO $\pm$ LA, LO $\pm$ TA of the central LO scattering peak, will be called exciton biphonon resonance (EBR).

The mutual arrangement of levels 1s and 2s of quasi-two-dimensional excitons associated with the first levels of dimensional quantization of an electron and a heavy hole is of key importance for the EBR mechanism proposed in [7]: 1s $1e1hh$ and 2s $1e1hh$. The bond energies of both excitons were calculated in Ref. [7] using the variational method, and the basic theory of EBR was built based on the resulting difference in the calculated values. The objective of this study was to verify the calculated results by direct spectroscopic measurements. The photoluminescence (PL) and reflection spectra were experimentally studied for a series of samples with quantum wells, which allowed determining the energy positions of the main exciton resonances.

The samples for the study were grown at the Institute of Physics of the Russian Academy of Sciences on GaAs substrates and contained a series of isolated (001)-CdTe/(Cd,Mg)Te quantum wells of various widths, including quantum wells with a width of 18 monolayers. It was on samples of this series that the optical spectrum of EBR was discovered. The wells were selectively doped with rhenium in various doses. Doping results in a decrease of the quantum efficiency of PL from the doped well while maintaining other, unrelated optical properties [7].

PL was excited by the green line of an Nd-YAG laser. The reflectance spectra were measured in white light at normal incidence.

The PL spectra of all CdTe/(Cd,Mg)Te samples contain the highest energy line (over 2 eV). This line is identified as the edge luminescence of the barrier layer. The position of the maximum of this line varies slightly from sample to sample: from 2.085 to 2.119 eV. Using the known dependence of the band gap on the magnesium content x in the virtual crystal approximation [8]:

$$E_g = 1.606 + 1.755x \text{ eV},$$

we find that the magnesium concentration in the barrier varies from sample to sample in the range of approximately 27–29%.

In addition, the PL spectra of all samples contain lines associated with quantum wells 18 monolayers wide. A typical PL spectrum of CdTe quantum wells includes two closely spaced lines (as in Figure 1). The line lying higher in energy corresponds to the 1s $1e1hh$ exciton, and the line lying lower in energy — corresponds to a positive or negative trion [9–11]. The different nature of the exciton and trion lines is manifested, for example, in experiments with the magnetically-induced PL polarization, [12,13] optical orientation [14–16] and spin-flip Raman scattering [17], and the specific type of trion can sometimes be determined from the shape of the zero-field level crossing line [18].

It should be noted that there is a small (within 10 meV) scatter in the absolute energy position of the exciton resonance 1s $1e1hh$ from sample to sample, and with it the scatter of the entire PL spectrum of the quantum well, which does not correlate with the doping dose. Obviously, this scatter should be associated, first of all, with the scatter in the barrier potential due to different magnesium contents (see above).

A powerful signal associated with the exciton resonance 1s $1e1hh$ dominates in the reflection spectrum. It is at almost the same energy as the maximum of the exciton line in the PL spectrum (Figure 1). We are also interested in weaker signals located over the ground state. Circles highlight two spectral features reminiscent of exciton resonances in Figure 1. Due to the known problem with the shape of the resonant signal line, which depends on the resulting phase because of the interference of light reflected by many interfaces inside the sample, the position of these two features is not particularly accurately determined: the first is spaced from 1s $1e1hh$ by approximately 17–18 meV, the second at 23.5–24.5 meV.

Not all samples show weak signals from overlying exciton states equally well. If Figure 1 presented data related to an undoped quantum well, then Figure 2 shows the reflection spectrum of one of the most heavily doped samples. There is virtually no PL in it due to nonradiative recombination, but exciton resonances in reflection are clearly visible. Again we see a peak located at a smaller distance from 1s $1e1hh$ (16–17 meV) and a peak at a larger distance (21.5–22.5 meV).

We interpret the emerging picture as follows based on the analysis of literature data and the results of calculations

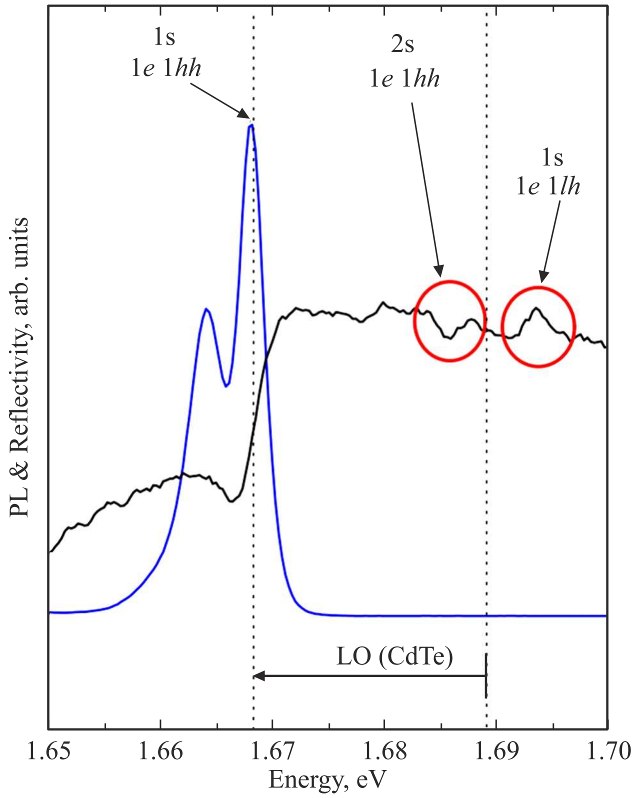

Figure 1. PL and reflection spectra of an undoped CdTe/(Cd,Mg)Te quantum well with a width of 18 monolayers (60 Å). $T = 2$ K. The main spectral features are indicated.

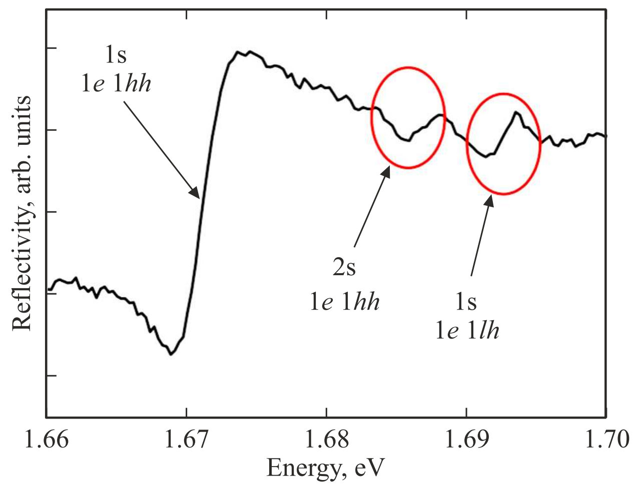

Figure 2. Reflection spectrum of a CdTe/(Cd,Mg)Te quantum well with a width of 18 monolayers (60 Å), doped with rhenium. $T = 2$ K. The main spectral features are indicated.

in Ref. [7]. The peak located at 16–18 meV over the ground exciton state 1s $1e1hh$ belongs to the 2s exciton (2s $1e1hh$), and the peak located at 21.5–24.5 meV over the main resonance — exciton with a light hole (1s $1e1lh$).

It should be noted that the idea that the critical for the EBR model developed in [7] is that 2s level of a heavy exciton is closer to the ground state (1s) than the LO phonon energy at the band center (20.88 meV), but, however, not too far. These energies were calculated separately in [7] by standard variational calculation, and their difference was 17 meV. It should be said that the theory based on these calculated values, practically without adjustable parameters, gave a satisfactory description of all the main features of the optical spectrum of the EBR. Now we see that the experiment also reveals an exciton resonance, located with good accuracy exactly 17 meV over the ground state.

As for the higher-lying resonance, its location does not allow it to be associated with the 2s exciton, not only because the energy 22−23 meV is not suitable for the EBR mechanism (> 20.88 meV), but also because it noticeably exceeds even the total 1s bond energy of heavy exciton (20.45 meV according to [7]). Over the past years, a certain consensus has developed in the literature regarding the typical values of the bond energy in CdTe/(Cd,Mg)Te quantum wells, and calculations by various authors usually gave, depending on the well width, values in the range 12−18 meV [19,20]. On the contrary, it is known (and can be easily estimated) that the light exciton 1s $1e1lh$ is approximately in this region in energy, but its energy position is not so strictly tied to the position of the heavy exciton 1s $1e1hh$, since these two exciton states are formed under different continua of single-particle states. In particular, the potential profile for the quantization of heavy and light holes is different due to such a poorly controlled, but undoubtedly present factor as the biaxial strain of the layers.

Thus, the reflection spectra for CdTe/(Cd,Mg)Te quantum wells with a width of 18 monolayers actually exhibit an exciton state in the expected position approximately 17 meV over the ground level. By all indications, this state should be associated with the 2s $1e1hh$ exciton, which confirms the assumptions made in [7] regarding the mechanism that forms the optical spectrum of the EBR. In addition, higher in energy there is another signal, which most likely belongs to an exciton with a light hole 1s $1e1lh$, and which, based on its energy position, can hardly be involved in the EBR mechanism.

Conflict of interest

The author declares that he has no conflict of interest.

References

[1] M. Czapkiewicz, V. Kolkovsky, P. Nowicki, M. Wiater, T. Wojciechowski, T. Wojtowicz, J. Wróbel. Phys. Rev. B **86**, 165415 (2012).

[2] B.A. Piot, J. Kunc, M. Potemski, D.K. Maude, C. Betthausen, A. Vogl, D. Weiss, G. Karczewski, T. Wojtowicz. Phys. Rev. B **82**, 081307 (2010).

[3] P. Olbrich, C. Zoth, P. Lutz, C. Drexler, V.V. Bel'kov, Y.V. Terent'ev, S.A. Tarasenko, A.N. Semenov, S.V. Ivanov, D.R. Yakovlev, T. Wojtowicz, U. Wurstbauer, D. Schuh, S.D. Ganichev. Phys. Rev. B **86**, 085310 (2012).

[4] I. Grigelionis, K. Nogajewski, G. Karczewski, T. Wojtowicz, M. Czapkiewicz, J. Wróbel, H. Boukari, H. Mariette, J. Łusakowski. Phys. Rev. B **91**, 075424 (2015).

[5] E. Bobko, D. Płoch, M. Wiater, T. Wojtowicz, J. Wróbel. Opto-electron. Rev. **25**, 65 (2017).

[6] Diluted Magnetic Semiconductors / Eds J. Furdyna, J. Kossut. Semiconductors and Semimetals. V. 25 / Eds R.K. Willardson, A.C. Beer. Academic, N.Y. (1988).

[7] A.V. Koudinov, E.V. Borisov, A.A. Shimko, Yu.E. Kitaev, C. Trallero-Giner, T. Wojtowicz, G. Karczewski, S.V. Goupalov. Phys. Rev. B **105**, L121301 (2022).

[8] W. Ossau, U. Zehnder, B. Kuhn-Heinrich, A. Waag, T. Litz, G. Landwehr, R. Hellmann, E.O. Göbel. Superlattices Microstruct. **16**, 5 (1994).

[9] K. Kheng, R.T. Cox, M.Y. d'Aubigné, F. Bassani, K. Saminadayar, S. Tatarenko. Phys. Rev. Lett. **71**, 1752 (1993).

[10] C.R.L.P.N. Jeukens, P.C.M. Christianen, J.C. Maan, D.R. Yakovlev, W. Ossau, V.P. Kochereshko, T. Wojtowicz, G. Karczewski, J. Kossut. Phys. Rev. B **66**, 235318 (2002).

[11] D. Andronikov, V. Kochereshko, A. Platonov, T. Barrick, S.A. Crooker, G. Karczewski. Phys. Rev. B **72**, 165339 (2005).

[12] A.V. Koudinov, N.S. Averkiev, Yu.G. Kusrayev, B.R. Namozov, B.P. Zakharchenya, D. Wolverson, J.J. Davies, T. Wojtowicz, G. Karczewski, J. Kossut. Phys. Rev. B **74**, 195338 (2006)

[13] V.F. Aguekian, D.E. Ashenford, B. Lunn, A.V. Koudinov, Yu.G. Kusrayev, B.P. Zakharchenya. Phys. Status Solidi B **195**, 647 (1996).

[14] G.V. Astakhov, A.V. Koudinov, K.V. Kavokin, I.S. Gagis, Yu.G. Kusrayev, W. Ossau, L.W. Molenkamp. Phys. Rev. Lett. **99**, 016601 (2007).

[15] Yu.G. Kusrayev, A.V. Koudinov, B.P. Zakharchenya, W.E. Hagston, D.E. Ashenford, B. Lunn. Solid State Commun. **95**, 149 (1995).

[16] A.V. Kudinov, Yu.G. Kusraev, I.A. Merkulov, K.V. Kavokin, I.G. Aksyanov, B.P. Zakharchenya. FTT **45**, 1297 (2003). (in Russian).

[17] A.V. Koudinov, Yu.G. Kusrayev, D. Wolverson, L.C. Smith, J.J. Davies, G. Karczewski, T. Wojtowicz. Phys. Rev. B **79**, 241310(R) (2009).

[18] S.V. Andreev, B.R. Namozov, A.V. Koudinov, Yu.G. Kusrayev, J.K. Furdyna. Phys. Rev. B **80**, 113301 (2009).

[19] S.R. Jackson, J.E. Nicholls, W.E. Hagston, T.J. Gregory, P. Harrison, B. Lunn, D.E. Ashenford. Superlattices Microstruct. **12**, 447 (1992).

[20] F.J. Teran, Y. Chen, M. Potemski, T. Wojtowicz, G. Karczewski. Phys. Rev. B **73**, 115336 (2006).

Translated by A.Akhtyamov