Thermally stimulated excitation transfer in an asymmetric system of quantum wells sepa-rated by thick barriers

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The photoluminescence (PL) and reflection spectra of the heterostructure $CdTe/Cd_{0.65}Mg_{0.35}Te$ were studied in the temperature range $T = 5-300$ K. The heterostructure contains four CdTe quantum wells (QWs) with a thickness of 10.2, 5.1, 2.6 and 1.3 nm separated by $Cd_{0.65}Mg_{0.35}Te$ barriers with a thickness of 20 nm. Four emission bands corresponding to the exciton recombination in these QWs, were detected under the above-barrier excitation. It was found that the energy transfer between neighboring QWs show an activation character. The coupling of the electron states of two neighboring QWs decreases with the increasing of their thickness. An estimation of the exciton state coupling between the neighboring QWs was carried out. It is concluded that energy transfer occurs through the Förster dipole-dipole interaction or through the real or virtual phonon states.

Keywords: II-VI nanostructures, energy transfer, exciton luminescence.

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1. Introduction

Transfer of electromagnetic excitation and carriers between the elements of semiconductor heterostructure characterizes to a large extent its properties and possibilities of its application. Such scope of issues was studied in structures containing the ensembles of quantum dots (QD), quantum wells (QW) of different thickness, in hybrid structures QW — planar massive of QD and in other similar objects [1–7]. Coupling between the heterostructure elements is ensured by tunneling of the carriers and excitons, and via the dipole-dipole interaction. Efficiency of the transfer can be significantly increased under resonance conditions, when the energy levels of the heterostructure elements are close to each other or even coincide. It is worth to be noticed that resonance is realized easily in systems with QDs having significant size dispersion, or systems with QWs more fitting is necessary. Transfer in multilayer heterostructures with QWs is determined by coupling of electron states of neighboring QWs, which depends on QW thickness, and height and thickness of the barrier [1]. It is of interest to study a tunneling efficiency of carriers and excitons at temperature increasing due to extension of their energy spectrum. In our paper this issue was studied based on temperature behavior of the luminescence of heterostructure with CdTe QW. The possibilities of optical spectroscopy were used to characterize the coupling/isolation o QWs separated by thick barriers.

2. Heterostructure design and experimental details

The heterostructure $CdTe/Cd_{0.65}Mg_{0.35}Te$ under study (Figure 1) has the following structure: GaAs substrate ⟨100⟩, 4 micron CdTe, 50 nm $Cd_{0.7}Mg_{0.3}Te$, four QWs CdTe QW1–QW4 with thicknesses, respectively, of 32, 16, 8, 4 monolayers (1 monolayer — 0.32 nm), separated by barriers $Cd_{0.65}Mg_{0.35}Te$ 20 nm thick, cap layer is 20 nm thick. QWs were grown by atomic-layer deposition method (ALD), barriers — standard molecular-beam epitaxy method (MBE).

The photoluminescence (PL) spectra of the heterostructure $CdTe/Cd_{0.65}Mg_{0.35}Te$ were studied in temperature range 5–300 K. The sample was inserted in the closed-cycle optical cryostat, for the above barrier excitation of PL the semiconductor laser with photon energy 3.06 eV was used. To exclude sample heating the excitation Intensity did not exceed 30 W/cm^2, which corresponds to maximum

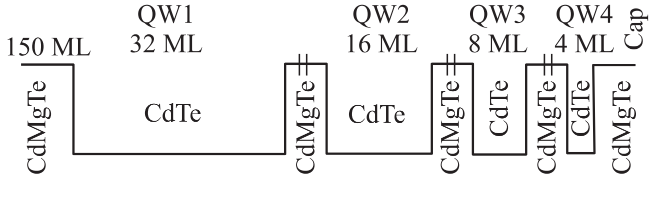

Figure 1. Scheme of heterostructure $CdTe/Cd_{0.65}Mg_{0.35}Te$.

concentration of photo-carriers in QW $< 10^{10}\,\mathrm{cm}^{-2}$. To obtain the reflection spectra the glow lamp was used.

3. Photoluminescence and reflection spectra

At low temperatures four bands are observed in the PL spectrum, which correspond to exciton recombination in QWs (QW1–QW4), and localized exciton recombination in the barrier (Figure 2). The characteristics of low temperature optical spectra of the sample are shown in the Table 1. E_{abs} — energies of exciton transitions in the reflection spectrum, corresponding to nearest levels of size quantization for each QW (see Figure 2), E_{PL} — energies of QW PL band maximums. Thus, the difference $(E_{\mathrm{abs}} - E_{\mathrm{PL}})$ is Stokes shift between the energies in absorption and emission spectra is the measure of the band tail for localized exciton states. The obtained experiment results are discussed in therms of exciton approximation, as monotonous decrease in value $(E_{\mathrm{abs}} - E_{\mathrm{PL}})$ with the increase of QW thickness is an indication of the exciton nature of the localized states which form PL spectra. Value $D = E_{\mathrm{CdMgTe}} - E_{\mathrm{PL}}$, where $E_{\mathrm{CdMgTe}} = 2.307\,\mathrm{eV}$, evaluates the energy of exciton injection from QW ground state into the barrier layer.

4. Temperature dependence of photoluminescence intensity and its analysis

Let's consider the temperature dependences of integral intensities of QWs QW1–QW4. Paper [8] shows that

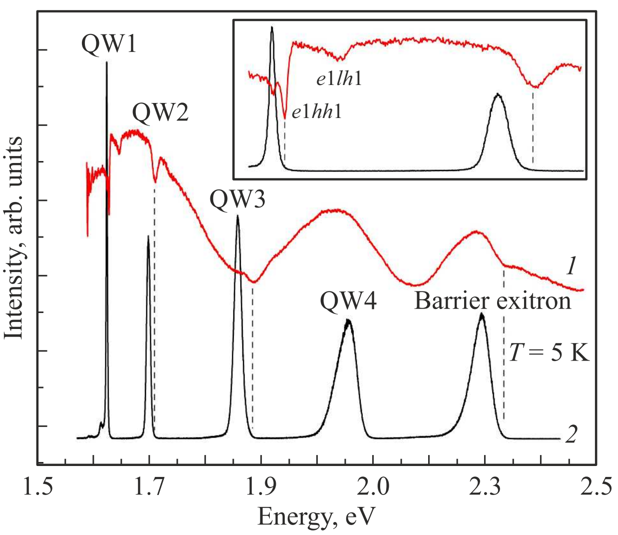

Figure 2. Reflection (*1*) and PL (*2*) spectra of heterostructure $CdTe/Cd_{0.65}Mg_{0.35}Te$. The insert shows the reflection and PL spectra at QW1 and QW2 transitions in extended scale. Peculiarities in the reflection spectrum indicated by the dashed lines correspond to exciton resonances in QWs and barrier.

Table 1. Characteristics of low temperature optical spectra of the sample

QW	E_{PL} (eV)	E_{abs} (eV)	$E_{\mathrm{abs}} - E_{\mathrm{PL}}$ (MeV)	D (MeV)
QW4	2.048	2.100	52	259
QW3	1.845	1.871	26	462
QW2	1.693	1.705	12	614
QW1	1.622	1.625	3	685

in case of the isolated QWs the temperature dependence of integral intensity $I(T)$ can be well approximated by the function consisting of two exponents with noticeably different energies E_1 and E_2:

$$I(T) = I_0/\{1 + a\exp(-E_1/k_{\mathrm{B}}T) + b\exp(-E_2/k_{\mathrm{B}}T)\}. \quad (1)$$

In this case the parameters a, b, E_1 and E_2 describe the excitation transfer to two groups of centers of nonradiative recombination which results in PL thermal quenching. For two coupling QWs with different thickness the significant redistribution of PL intensity is observed in a certain temperature range, which is due to excitation transfer from narrow QW to neighboring thick QW. If the total emission intensity from pair of neighboring QWs $I_{\mathrm{PL}}(T)$ can be described by expression (1), the temperature dependence of emission intensity from thin QW taking into account the excitation transfer to thick QW is described by introduction into the equation (1) of the additional term $c^1\exp(-E_t^1/kT)$ [8]:

$$I(T) = I_0/\{1 + a\exp(-E_1/k_{\mathrm{B}}T) + b\exp(-E_2/k_{\mathrm{B}}T) + c^1\exp(-E_t^1/k_{\mathrm{B}}T)\}. \quad (2)$$

This model assumes that after the fitting the temperature dependence of total integral intensity $I_{\mathrm{PL}}(T)$ of Pl of two neighboring QWs using the equation (1) the parameters a, b, E_1 and E_2 are fixed, and for description of the partial temperature dependence of emission of these QWs only parameters c^1 and E_t^1 stay free.

To test the proposed model of excitation transfer we considered the balance equations for three pairs of neighboring QWs QW4–QW3, QW3–QW2 and QW2–QW1. It turned out that the most closely the model proposed in [8] is applicable to describe the excitation transfer from QW4 into QW3: in temperature range $T > 100\,\mathrm{K}$ the integral intensity of PL $I_4(T)$ quickly decreases with simultaneous slowing of thermal quenching of PL $I_3(T)$ (Figure 3,*a*). Figure shows that total emission intensity from QW4 and QW3 can be described by equation (1), in this case the partial temperature dependence $I_4(T)$ is described by the equation (2) by introduction of the additional exponential term with activation energy E_t. For pair QW4–QW3 the effect of PL intensity redistribution between QW4 and QW3 becomes noticeable in range $T > 100\,\mathrm{K}$, E_t^1 for this pair is 20 meV.

The intensity redistribution with the temperature increase is also observed for the pair QW3–QW2, this also indicates

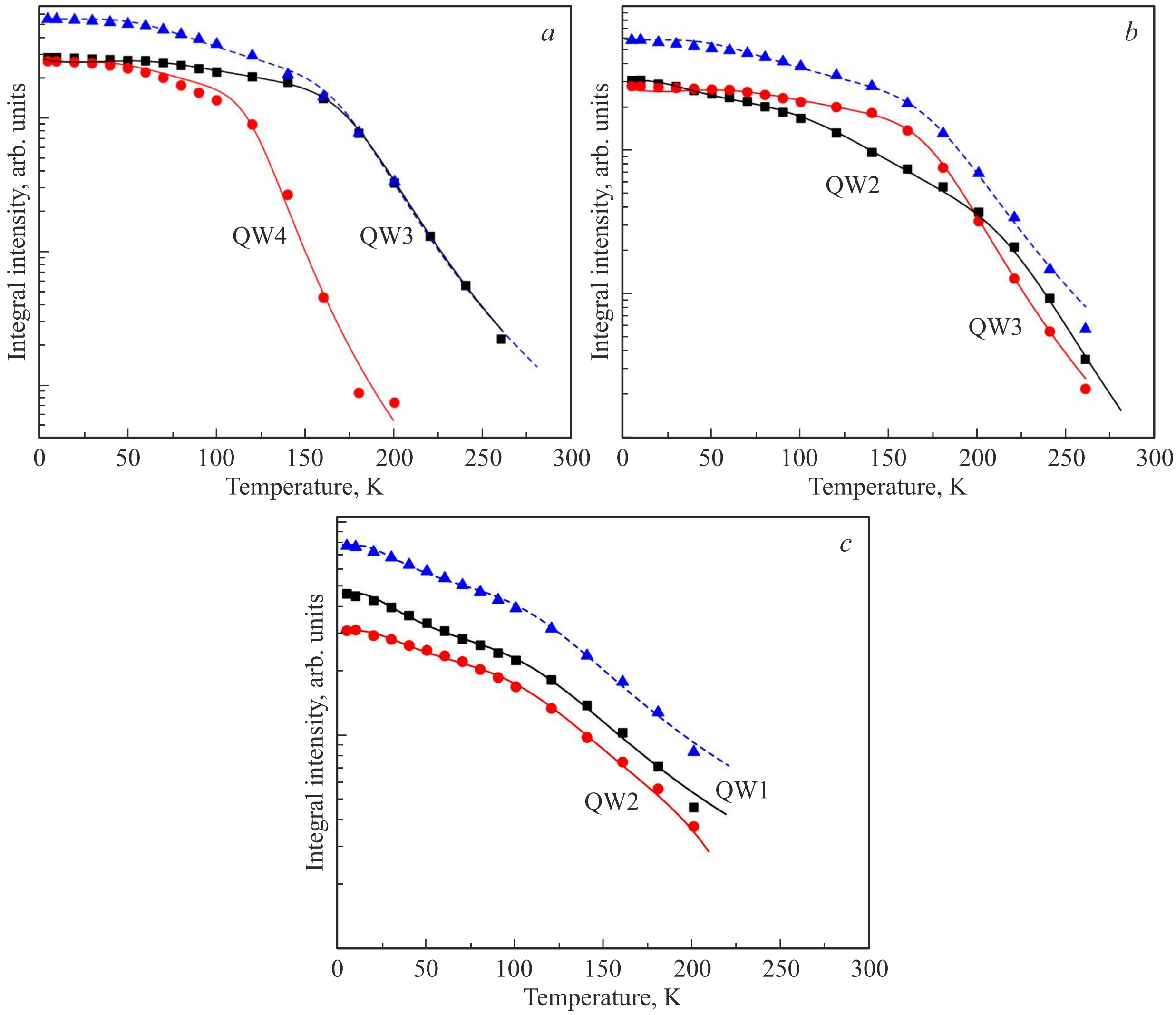

Figure 3. Integral intensities of QW luminescence vs. temperature. Markers indicate the experimental data, lines — results obtained using the formula (2) with parameters given in the Table 2. Dashed lines indicate the total integral intensity of PL of QW pair.

Table 2. Parameters of equations (1) and (2), used to describe dependences $I_1(T)-I_4(T)$

	QW4	I_{PL} = QW4 + QW3	QW3	I_{PL} = QW3 + QW2	QW2	QW1
I_0	2.63	5.44	2.80	5.9	3.07	4.6
a	8.7	8.70	13	5.12	1.5	170
E_1 (meV)	23	23	24	19	6.7	6
b	$7.45\cdot10^7$	$1.7\cdot10^6$	$3.1\cdot10^6$	$2.2\cdot10^5$	200	213
E_2 (meV)	194	202	230	180	60	60
c^1	7.46		−7.46		−0.17	
E_t^1 (meV)	20		20		4	
c^2			0.17			
E_t^2 (meV)			4			

these QWs coupling. Actually, Figure 3, *b* shows that in temperature range 100–140 K the dependence $I_3(T)$ becomes flat, which as shown above, indicates the transfer increasing from QW4 into QW3. In temperature range > 180 K the integral intensity $I_3(T)$ quickly decreases due to increase in excitation transfer into QW2. When describing quantitatively these processes along with the excitation transfer from QW4 into QW3 we shall consider also change in QW2 population due to transfer from QW3. Contribution of both processes into the partial temperature dependence

$I_3(T)$ is described by introduction in the equation (1) similar to equation (2) of two exponential terms with activation energies E_t^1 and E_t^2, negative and positive signs of these contributions reflecting, respectively, the influx of excitons into QW3 from QW4 and outflow from QW3 into QW2. In pair QW2–QW1 (Figure 3, *c*) rates of thermal quenching of PL practically coincide, which, in our opinion, indicates the absence of excitation transfer from QW2 into QW1.

Table 2 presents the parameters of equations (1) and (2), used to describe dependences $I_1(T) - I_4(T)$ in PL spectra of QW1–QW4.

We evaluated overlapping of wave functions of exciton states in QW1–QW4 and established that this effect is so small that the tunneling processes can not make significant contribution into the energy transfer. Possible mechanisms explaining the experimentally observed excitation transfer between QWs in this case could be the Förster energy transfer caused by the dipole-dipole interaction between QWs [3], or the energy transfer through the real or virtual photons [5]. These papers show that selection of the mechanism of excitation transfer between QWs separated by thick barriers can be made based on the study of this process rate dependence on the barrier thickness.

Note that temperature dependencies of rate of exciton transfer due to dipole-dipole interaction or transfer via the real or virtual photons are described in papers [3,5] with model of simple parabolic electron and hole bands. These papers show that in such simple model the dependences have sophisticated non-monotonic nature, this is due the exciton energies redistribution in thin QW due to exciton thermalization. In particular, with temperature increase the transitions between QWs are possible for excitons with non-zero wave vector. For the detailed description of the experimental results on energy transfer in the CdTe/CdMgTe structures studied in our paper, it is necessary to calculate the transfer rate in the energy range of hundreds of meV, which in turn requires taking into account the complex structure of the valence band, knowledge of the binding energy and density of states of excitons, taking into account the non-parabolicity of the spectrum of carriers, as well as the wave functions of excitons. Such calculation is beyond this paper, but we can expect, that consideration of these factors will lead in significant differences in dependence of transfer rate on temperature as compared to papers [3,5], and explains the break in temperature dependence of PL intensity at temperatures ~ 100 K.

5. Conclusion

So, characteristics of energy transfer between neighboring QWs are experimentally determined for the heterostructure with four QWs CdTe separated by barriers $Cd_{0.65}Mg_{0.35}Te$ with thickness 20 nm. At $T = 5$ K in PL spectrum four bands of comparable intensity are observed, they relate to excitons recombination in QW. It is determined that energy transfer between the states of neighboring thin and thick QWs is enhanced with temperature increase. The efficiency of energy transfer from thin QWs to thick QWs at fixed barrier thickness decreases significantly with thickness increasing of these two QWs. We concluded that possible mechanisms of excitation transfer between QWs of the studied heterostructure can be thermally-stimulated dipole-dipole interaction or transfer via states of real or virtual photons. The mechanisms of excitation transfer between QWs separated by thick barriers can be clarified based on the study of this process intensity dependence on the barrier thickness.

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Conflict of interest

The authors declare that they have no conflict of interest.

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