Spectroscopic evidence of tunnel coupling between CdTe quantum wells in the CdTe/ZnTe heterostructures

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Abstract. The photoluminescence (PL) spectra of CdTe/ZnTe double quantum wells (QWs) are studied on a series of samples containing two CdTe layers with nominal thicknesses of 2 and 4 monolayers (ML) in the ZnTe matrix. The QWs were grown in atomic-layer epitaxy and separated by ZnTe spacers with the thicknesses d_{sp} = 40–160 ML. The dependences of the relative intensity of shallow QW_1 and deep QW_2 PL bands (I_1 and I_2, respectively) on the pump intensity (J) for excitation by lasers with different radiation wavelengths are investigated. It is found that in the sample with d_{sp} = 40 ML, the ratio $Y(J)=I_1/I_2$ depends on J and the shape of the $Y(J)$ dependency changes with a variation of the excitation wavelength. In the samples with d_{sp} > 70 ML $Y(J)$ also changes with the excitation intensity J, but the shape of this dependence is the same for various excitation wavelengths. It is concluded that the energy relaxation in these samples is influenced not only by the tunneling of charge carriers from QW_1 to QW_2, but also by the carrier relaxation to nonradiative centers which recombination rates are different for shallow and deep QWs.

1. Introduction

Investigation of the tunneling of charge carriers in semiconductor heterostructures is of great importance for a detailed study of quantum mechanical processes and for applications in devices using the tunnel effect. Of particular interest are the structures based on wide-gap II–VI compounds, in particular, Zn and Cd tellurides. A characteristic feature of heterostructures which contain several layers of narrow gap material (CdTe) embedded into a wide gap matrix (ZnTe) is the formation of a quantum well (QW) from a spatially inhomogeneous solid solution of ZnCdTe. The nanoscale regions with a large (compared to average) CdTe content arise in these QWs, which have the properties of quantum dots in many aspects. The properties of such objects depend at large extent on growth technologies.

This work is devoted to the study of charge and energy transfer processes in a system of double QWs formed by the atomic layer deposition of two and four CdTe monolayers in ZnTe matrix.

2. Samples and experimental details

CdTe/ZnTe heterostructures consisting of two CdTe QWs (DQW) separated by ZnTe barriers of different thicknesses were grown on semi-isolated GaAs (001) substrates by molecular beam epitaxy (MBE). ZnTe buffer layer of these structures with a thickness of about 2.5 micrometers was deposited in the standard mode of MBE. The subsequent growth of two CdTe wells with nominal thicknesses of 2 and 4 monolayers was performed by the atomic-layer epitaxy mode. The structures were covered with a cap layer comprising 100 nm ZnTe and 50 nm ZnMgTe grown by the standard MBE mode. In total, five samples of DQW structures with ZnTe barrier thicknesses of 40, 70, 100, 130, and 160 monolayers (ML) were grown and studied. In addition to the DQW structures two reference samples with single CdTe QW of nominal thickness of 2 and 4 ML were grown under the same conditions.

To excite photoluminescence the He-Cd laser with 442 nm wavelength and the semiconductor lasers with wavelengths in the range of 507–405 nm were used. The samples were placed in a closed-cycle He cryostat, and low-temperature PL spectra were recorded using a double monochromator and Hamamatsu R928 photoelectron multiplier.

The excitation power density in our experiments did not exceed 10 W/cm^2, which is obviously less than one e-h pair per CdTe quantum dot [1, 2].

3. Results and discussion

Figure 1 shows the low-temperature (T = 5 K) PL spectra of samples with different barrier thicknesses d_{sp} obtained at maximum pump level J_{exc} = 10 W/cm^2. The PL spectra which include two bands are normalized to the intensity of the low energy PL band. Emission band in the spectral range (2.30±0.01) eV with full width at half maximum (FWHM) about 10–12 meV is the result of exciton recombination in the shallow (narrow) QW_1, while the emission band in the range (2.17±0.02) eV with FWHM 23–25 meV corresponds to exciton recombination in the deep (wide) QW_2. As can be seen in Figure 1, PL bands from QW_1 and QW_2 in samples with the barrier thicknesses d_{sp} > 70 ML have comparable intensities I_1 and I_2, which may indicate that these QWs are isolated. In the sample with a thinner barrier (d_{sp} = 40ML) the intensity I_1 of QW_1 band (thick solid line in Figure 1) is noticeably weaker as compared with that of QW_2 band which apparently indicates a tunnel-coupling between QW_1 and QW_2. As shown in [3] and [4], the tunneling between QW_1 and QW_2 layers should manifest itself in the dependence of relative intensities I_1 and I_2 bands on the excitation level, as well as in the change in this dependence for different excitation wavelengths.

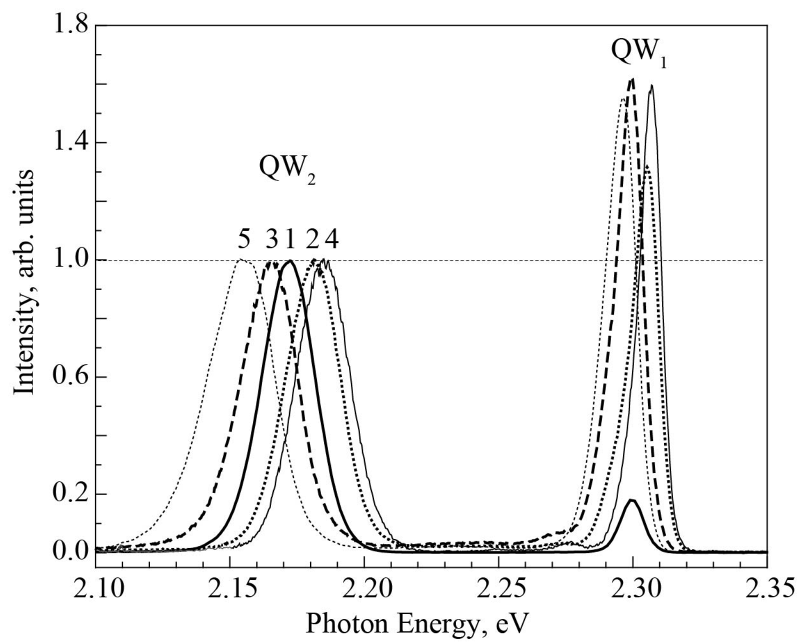

Figure 1. Low-temperature (T = 5 K) PL spectra of CdTe/ZnTe double QW structures with different ZnTe barrier thicknesses d_{sp} at above-barrier excitation in the region of 2.807 eV. Symbols 1–5 correspond to the samples with d_{sp} = 40, 70, 100, 130 and 160 ML, respectively. The spectra are normalized to the peak intensity of QW_2 PL band.

Figure 2 shows the dependences of the ratio $Y = I_1/I_2$ of the integral intensities of I_1 and I_2 emission bands on the pump intensity J in PL spectrum of the sample with barrier thickness of d_{sp} = 40 ML under above barrier excitation with 442, 480 and 507 nm wavelengths. Such variation of the $Y(J)$ dependence with a change in the excitation wavelength should be expected if the barrier is tunnel-transparent for single carriers, but the probability of a tunneling of excitons between QW_1 and QW_2 layers is lower than the probability of radiative recombination of excitons [3, 4]. Indeed, in the limit of strong excitation, when the concentration of photoexcited free electrons and holes is high enough, exciton state in the quantum dot is formed mainly as a result of the independent capture of electrons and holes. Under these conditions, the intensities of the I_1 and I_2 bands are governed only by the pump intensity, that is the I_1/I_2 ratio does not depend on the excitation wavelength. At low excitation intensity the probability of tunneling of a single carrier (electron) from the shallow QW_1 to the deep QW_2 exceeds the probability of exciton formation in QW_1. Under these conditions the PL intensity from QW_1 is determined by the recombination of hot excitons that successfully complete the energy relaxation as a whole. In this case, the value of $Y = I_1/I_2$ depends on the initial kinetic energy of hot excitons, i.e. on the excitation wavelength [4].

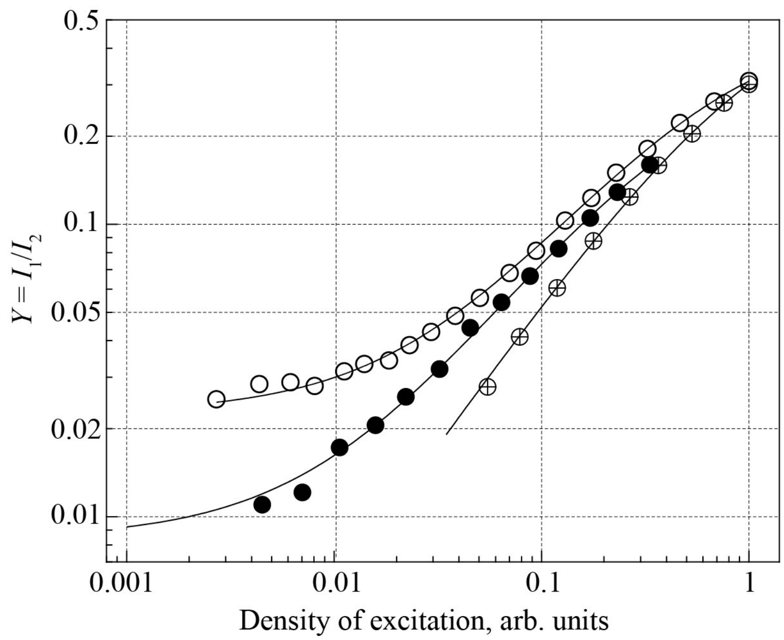

Figure 2. Dependence of the relative PL band intensities of the shallow QW_1 and deep QW_2 on the excitation intensity for the sample with a barrier thickness of d_{sp} = 40 ML when excited with wavelengths 442, 480 and 507 nm (crossed, full and open circles, respectively). Solid lines are guides to the eye.

We have found that, in contrast to the behavior of $Y(J)$ in the sample with 40 ML barrier, in the samples with a barrier thickness of 70 ML or more, the $Y(J)$ ratio does not depend on the excitation wavelength (Figure 3). At first glance, this result can be used as a more direct evidence of the tunneling independence of the QW_1 and QW_2 layers. Indeed, in the ideal case of a 100% quantum yield, the ratio I_1/I_2 should depend neither on the intensity nor on the excitation wavelength. However, as can be seen from Figure 3, the value of Y also increases in the PL spectra of samples with thick barriers with increasing excitation intensity. One can suppose that the process of energy relaxation in the DQW systems under consideration is affected not only by the tunneling of charge carriers from shallow QW to a deep QW, but also by the carrier relaxation on the centers of non-radiative recombination. It can be assumed [5] that the process of self-organization leads to the quantum dot formation in the QW plane. At the boundaries which separate the QW and quantum dots and/or quantum dots and the barrier, a whole spectrum of states appear, which act as the centers of nonradiative recombination. In this case, the experimentally observed dependence of Y on the pump intensity J corresponds to gradual saturation of the centers of non-radiative recombination, the saturation rate of these centers being different for shallow and deep QWs. It seems natural that the saturation effect is most pronounced for a shallow QW, since it has fewer centers of both radiative and non-radiative types than a deep QW.

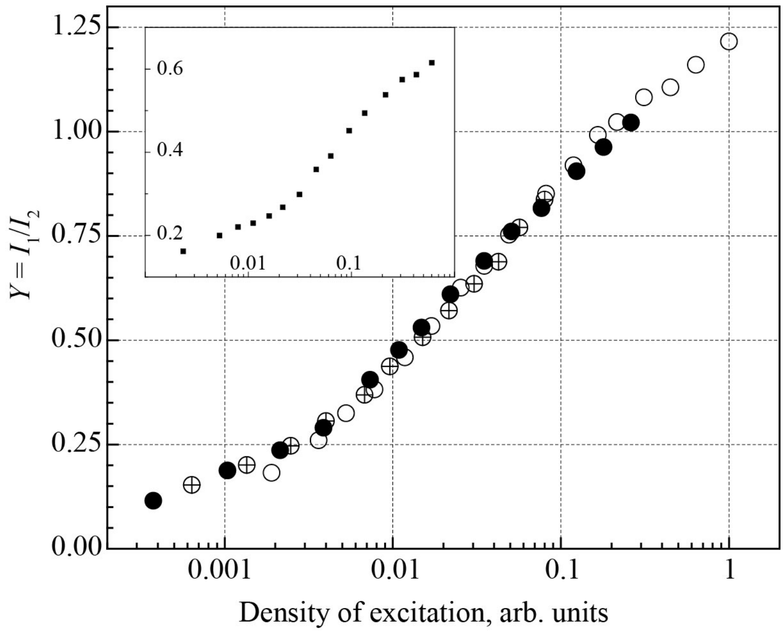

Figure 3. Dependence of the relative PL band intensities of the shallow and deep QWs on the intensity of excitation for the sample with barrier thickness of d_{sp} = 70 ML by lasers with wavelengths 442, 480 and 507 nm (crossed, full and open circles, respectively). The inset shows this dependence for ratio I_{2ML}/I_{4ML} of the PL band intensities on the excitation level in reference CdTe/ZnTe samples with the single QWs with nominal thicknesses of 2 and 4 ML with wavelengths 442 nm.

One more factor that determines the difference in the saturation rates of the centers of nonradiative recombination in the vicinity of shallow and deep QWs is the different degree of localization of their electron wave functions of the ground state of these QWs.

The influence of nonradiative recombination centers should also be manifested in the dependences of the PL band intensities on the excitation levels in the samples containing single QW. We have studied these dependences in the reference samples with single CdTe QWs 2ML and 4ML grown in the same conditions as epy DQW samples. The dependence of the ratio of the integral intensities of their PL bands of the samples on the excitation levels is shown in the inset to Figure 3. As can be seen in Figure 3, the dependences $Y(J)$ in the PL spectra of DQW separated by thick barriers, and the dependences of the ratio $I_{2ML}/I_{4ML}(J)$ in the PL spectra of reference samples are qualitatively the same.

Thus, we have shown that a comprehensive study of the dependence of the intensity of the PL bands on the pump intensity under the above-barrier excitation with different wavelengths allows to establish the presence or absence of tunnel coupling in an asymmetric double QW system, as well as to elucidate the effect of the centers of nonradiative recombination on this dependence.

Acknowledgements

Authors thank G.V. Budkin for useful discussions. This research was supported within the State Assignments from the Ministry of Science and Higher Education of the Russian Federation to the Ioffe Institute (0040-2019-0006), St. Petersburg State University for the research grant ID: 75746688. The research in Poland was partially supported by the National Science Centre through Grants No. 2017/25/B/ST3/02966 and 2018/30/M/ST3/00276.

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