Single photon emission from charged excitons in CdTe/ZnTe quantum dots

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Abstract. We report on micro-photoluminescence studies of individual self-organized CdTe/ZnTe quantum dots intended for single-photon-source applications in a visible spectral range. The quantum dots surface density below 10¹⁰ per cm² was achieved by using a thermally activated regime of molecular beam epitaxy that allowed fabrication of etched mesa-structures containing only a few emitting quantum dots. The single photon emission with the autocorrelation function g(2)(0)<0.2 was detected and identified as recombination of charged excitons in the individual quantum dot.

1. Introduction
Quantum dot (QD) heterostuctures based on wide bandgap II-VI semiconductor compounds are promising for fabrication of single-photon sources of visible light, required for quantum cryptography. CdTe/ZnTe self-organized QDs are especially suitable for such applications since they can be grown by molecular beam epitaxy (MBE) with a reasonably low surface density [1]. Further decreasing of the QD density below ~10¹⁰ cm⁻² makes it possible to use routine lithography techniques and micro-photoluminescence (µ-PL) spectroscopy for even simpler extraction of the single-QD emission. The CdTe/ZnTe QD emission spectrum typically represents a set of sharp discrete lines, which correspond to multiexciton cascade recombination, i.e. recombination of excitons, biexcitons, charged excitonic states (trions), etc. [2,3]. In this work, we investigate the emission properties of single CdTe/ZnTe QDs in heterostructures grown on InAs(100) substrates by MBE, using a previously developed version of a thermal activation technique [4], which allows fabrication of the QDs arrays with the surface density noticeably lower than ~10¹⁰ cm⁻².

2. Samples and experimental techniques
The studied CdTe/ZnTe QDs were formed by MBE within Zn(Mg)Te structures pseudomorphically grown on InAs(100) substrates, by using a so-called “thermal activation technique” which comprises deposition of a strained CdTe 2D layer, covering it with amorphous Te, and fast thermal desorption of the Te layer [1,4]. The CdTe/ZnTe QD structure includes a 5 monolayer (ML)-thick ZnTe buffer layer deposited on the (2x4)As surface of an InAs buffer layer, a 10 nm-thick Zn₀.₉Mg₀.₁Te carrier confining layer, a 30 nm-thick ZnTe barrier layer followed by a single CdTe QD sheet with a nominal thickness of ~3 MLs, and a 20 nm-thick ZnTe top barrier. The Zn(Mg)Te-based layers were grown at ~300°C.

The individual QDs were spatially localized by forming cylindrical mesa-structures of several diameters ranging between 1 µm and 200 nm on the same QD structure. The mesas were fabricated by

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dry Ar⁺-ion etching through the masks formed by an electron-beam lithography. The QDs emission was extracted and spectrally resolved in a confocal μ-PL optical scheme with a 50x micro-objective and a monochromator equipped with a liquid nitrogen cooled CCD camera and diffraction grating with 1800 grooves per mm (spectral resolution is estimated as 60 μeV). A cw laser with the emission wavelength 404 nm was used for the PL excitation. A set of calibrated neutral-density filters allowed variation of the excitation power. Photon correlation measurements were performed in a Hanbury Brown – Twiss intensity interferometer exploiting two single-photon avalanched Si photodiodes providing a time resolution better than 40 ps. The measurements were performed in the 8 - 80 K temperature range in a continuous flow He cryostat with a nanopositioning piezo-driver.

3. Results and discussion
The PL spectrum of the CdTe/ZnTe QDs consists of a wide emission band spreading between 530 and 630 nm, which is transformed into a set of discrete narrow lines with a characteristic spectral width of ~ 0.3 meV at 8 K by improving the PL spatial resolution either down to 1.5-3 μm using a standart μ-PL technique with a 50 μm pinhole located in a microscope image plane or down to 200 nm provided by formation of the mesa structures (figure 1).

![Figure 1. PL and μ-PL spectra of the CdTe/ZnTe QDs. Lines marked as PL, μ-PL, 1 μm and 200 nm represent, correspondingly, a typical PL spectrum (spatial resolution 200-500 μm), a μ-PL spectrum measured within a 1.5-3 μm area, and μ-PL spectra taken from the mesas with either 1 μm or 200 nm in diameter.](image1)

At 8 K, most of the mesas with the diameter 200 nm display the spectra represented by a set of lines with fixed spectral distances between them (like shown in figure 2). Moreover, a noticeable part of the mesas show absolutely identical shape of spectra with equal intensity relations between the lines measured at the same excitation power density. Such sets include up to 8 spectrally resolved lines (the lines number depends on the excitation power density), which correspond to excitons (X), positively
and negatively charged excitons (CX and CX’), biexcitons (XX) and other charged exciton and biexciton complexes (marked as A, B, C, D lines in figure 2). Excitation power dependencies allowed us to identify the line with the highest emission energy as an exciton state, because only it emerges in the spectrum at the lowest excitation power (figure 3). CX and CX’ lines showing smaller intensity rising with excitation power as compared to XX and A-D lines are identified as charged exciton states (trions). In terms of a multiexciton model, probability of formation for biexcitons and other complexes including more than three carriers (electrons and holes) should be lower at low excitation power density, comparing to the formation probability for excitons and trions. As one can see in figure 3, XX and A-D lines are visible only at the highest excitation powers. In addition to the identification by excitation power dependencies, the QDs spectra represent linear polarization of X and XX emission lines with the fine structure splitting (FSS) up to 240 µeV, caused by anisotropic exchange splitting. FSS of the X and XX lines exhibit opposite signs of the spectral shift for two linear polarizations in each QD. Superposition of these factors allowed us to conclude that the XX line corresponds to a biexciton state.

**Figure 3.** Excitation power dependencies for the peaks of the single-QD emission. Peaks labels correspond to labels in figure 2. The excitation laser emission was focused into a 20-50 µm² area.

We assume that in the studied CdTe/ZnTe structures the formation probability for the positively charged exciton is higher than that for the negatively charged one. This is in complete agreement with the fact that ZnTe grown by MBE has usually p-type conductivity [5]. In such conditions, a typical CdTe/ZnTe QD spectrum exhibits the CX line with noticeably higher intensity as compared to all other lines. This line is spectrally separated from the X line by 9 meV on one side and by 7 meV from the XX line on other side. The CX’ line intensity is lower than that of the CX one by two orders of magnitude. As a result, only the X, CX, and XX lines are well resolved up to the liquid nitrogen temperature (figure 2).
Figure 4. The second-order correlation function for the CX charged-exciton peak. The points represent experimental data while the solid line shows fitting by equation (1).

The measured second-order correlation function of the CX state emission demonstrates an antibunching dip with a minimal value $g^{(2)}(0)$ as low as $\sim 0.2$ that is a clear signature of the non-classical single-photon nature of the emission (figure 4). To obtain this function, the measured histogram of coincidences was normalized by count rates of the detectors, width of the time bins, and the integration time [6]. The solid line represents fitting by the expression

$$g^{(2)}(t) = 1 - A \cdot e^{-t/t_a} + B \cdot e^{-t/t_b},$$  

where $t$ is the delay time. The estimated values for $t_a$ and $t_b$ are $0.8 \pm 0.1$ ns and 2-5 ns, correspondingly.

4. Conclusions

We have demonstrated that CdTe/ZnTe QDs with extremely low surface density grown by MBE with thermal activation are suitable for single-photon applications in a visible spectral range. Etched mesa-structures with a 200 nm lateral size allow easy separation of the emission of the single QDs with the value of $g^{(2)}(0)$ below 0.2.
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