

Emission properties of single-photon sources based on CdTe/ZnTe quantum dots

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Abstract. We report on single photon emission from single CdTe/ZnTe quantum dots (QDs) grown by thermally-activated molecular beam epitaxy providing a reduced QD lateral density below 10^{10} cm^{-2} . For micro-photoluminescence spectroscopy studies and correlation measurements, the mesa structures of 200 and 500 nm diameters are fabricated by combination of electron-beam lithography and reactive plasma etching. Autocorrelation function of photons emitted from a single QD under cw excitation demonstrates antibunching with a value of $g^{(2)}(0) \sim 0.3$ that is a signature of non-classical light.

1. Introduction

In the last decades, a lot of attention has been paid to sources of quantum light such as single photon emitters possessing non-classical photon statistics and sources of entangled pairs of single photons. These devices are key elements for the systems of quantum cryptography, information teleportation, and quantum computing. Self-organized single quantum dots (QDs) based on wide-gap II-VI compounds grown by epitaxial techniques (usually by molecular beam epitaxy – MBE), are considered as promising candidates for creation of the single photon sources, because they are less susceptible to blinking, which is the main drawback of colloidal QDs. So far, single-photon emission at elevated temperatures has been demonstrated for the structures with the epitaxial QDs made from various II-VI compounds including CdSe/Zn(S,Mg)Se [1-5] and CdTe/ZnTe [6,7]. Room temperature operation under optical [3] and electrical [4] pumping has been recently achieved. The serious obstacle hampering further progress in this field is a high lateral density inherent to the epitaxial QDs, which complicates spatial selection of a single excitonic line in a spectrum of a QD array, needed for the non-classical light observation. To distinguish the single-QD emission line, different approaches are exploited, including formation of etched mesa structures [1,2] or nanoapertures in a non-transparent mask [3,4,8], as well as insertion of a QD within a nanowire [5]. However, all these approaches require the relatively low QD density of at least 10^{10} cm^{-2} (the less the better), which can hardly be realized by conventional MBE.

As a promising approach to produce the arrays of II-VI QDs with a reduced density, the MBE technology exploiting thermal activation growth mode has been proposed [9,10]. This method implies the changing of surface energy with thermal annealing that facilitates the 2D-3D transition of a strained layer into a QD ensemble. It was first verified for the CdTe/ZnTe heterostructures [9]; then,



some modification of this technique was applied to the structures based on CdSe/ZnSe [11]. The obtained lateral densities were about $4 \cdot 10^{10} \text{ cm}^{-2}$ and $(2 - 5) \cdot 10^{10} \text{ cm}^{-2}$, respectively, for a nominal insertion thickness in the 3 - 4.5 monolayers (MLs) range. The formed objects were not canonical Stranski-Krastanov QDs but rather flat nano-islands whose average sizes in vertical/lateral dimensions are 5 nm/50 nm for CdTe/ZnTe and 2 nm /20 nm for CdSe/ZnSe. Thus, the decrease of the lateral QD density was realized on account of increasing the sizes of the nano-islands. When the island size is markedly more than 10 nm, the exciton density of states (DOS) corresponds rather to that in a quantum well that can prevent undistinguished photon generation, since single photons emission assumes recombination of excitonic states possessing DOS of distinct QDs [12]. At low temperature, nevertheless, micro-photoluminescence (μ -PL) spectra exhibited narrow emission lines [9,10], probably related to fluctuating localization potential within the extended islands, and antibunching photon behaviour was demonstrated in the CdTe/ZnTe structures grown by this method [6,7].

In this paper, we investigate CdTe/ZnTe QDs grown by MBE using the thermal activation mode with and without exposure to Te flux. This procedure is considered as a method to influence basic QD emission properties. Data of μ -PL spectroscopy allow us to estimate the QD density, which turns out to be sufficient for spatial and spectral selection of single quantum emitters. For the confirmation of single photon statistics, the correlation function $g^{(2)}(\tau)$ was measured and analyzed.

2. Experiment

We have studied emission properties of two samples (labeled further as A and B) grown by MBE on InAs (001) substrates overgrown by a 0.2- μm -thick InAs buffer. The details of the growth procedure will be published elsewhere [13]. The sequence of deposited layers includes a 5-ML-thick ZnTe interface layer, a ~ 100 nm-thick $\text{Zn}_{0.9}\text{Mg}_{0.1}\text{Te}$ barrier, a 30 nm-thick ZnTe layer followed by a 3.1 - L-thick CdTe insertion, and a 20 nm-thick top ZnTe layer. The QDs were grown in a thermal activation mode, which implies growth of the strained CdTe layer on the ZnTe surface, coverage of the growth surface with a $\sim 0.1 \mu\text{m}$ -thick amorphous Te layer at $T_s < 50^\circ\text{C}$, desorption of the tellurium layer at $\sim 220^\circ\text{C}$ during the gradual T_s increase with a ramping rate of $\sim 40^\circ\text{C}/\text{min}$, and the following gradual increase of T_s up to 300°C that was done slowly for 30-40 min either under exposure to Te flux (sample A) or without that (sample B).

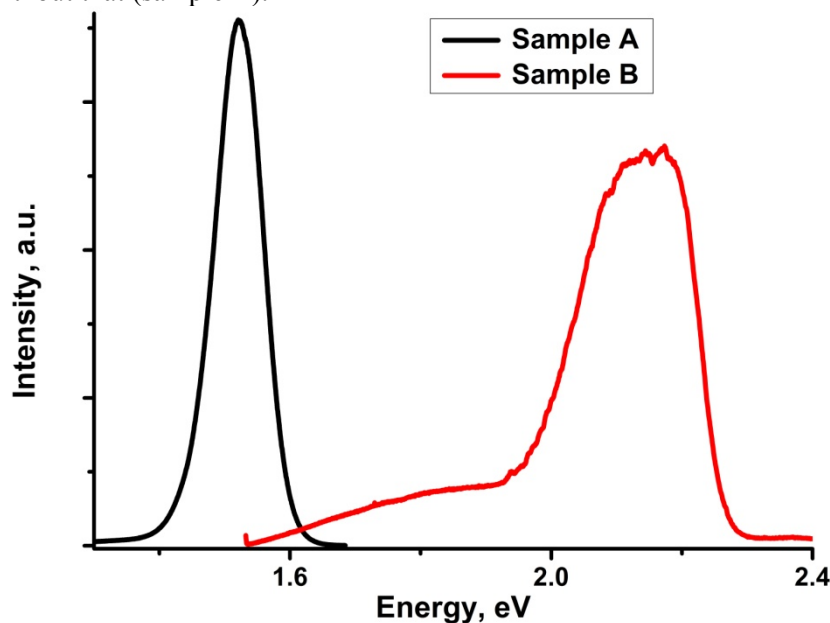


Figure 1. Normalized low temperature (77 K) spatially integrated PL spectra of structures grown either with exposure to Te flux (sample A) or without that (sample B).

The emission properties of QDs were investigated using cylindrical mesa-structures of 200 nm and 500 nm in diameter, which were fabricated by means of the combination of electron lithography and plasma etching. At first, the masks possessing the circle shape of different diameters were formed on the surface of the heterostructures with the QD layer using an e-beam-resist (MaN 2403). Then, the structures were etched by Ar plasma. Scanning electron microscopy studies have confirmed formation of cylindrical mesas with vertical sidewalls. The sample is mounted in a He-flow cryostat with an Attocube XYZ piezo driver inside, which allows us to optimize and precisely maintain the positioning of the chosen mesa with respect to a laser spot during a long time (few hours). The μ -PL measurements were carried out under optical excitation by a cw laser line (405 nm). The laser power density was 4 W/cm² before a cryostat window. Photon correlation measurements were performed in a Hanbury Brown – Twiss detection scheme exploiting two single-photon avalanche diodes (from Micro Photon Devices) possessing a full width at half maximum (FWHM) of photon timing resolution of about 40 ps.

3. Results and discussion

The cw PL spectra (integrated over the area of $\sim 200 \mu^2$) of samples A and B, measured at 77 K are shown in figure 1. The emission line in sample A is markedly red-shifted relative to that in sample B, in spite of the same amount of deposited CdTe. This shift is a signature of larger QD sizes in sample A. In sample B grown without the Te flux exposure, the mobility of Cd adatoms is higher and their certain re-evaporation can take place. As a result, the QD has smaller sizes and blue-shifted emission. Moreover, FWHM of the emission line in sample B is more than twice larger than in sample A (200 meV versus 85 meV), that indicates a stronger QD size dispersion leading to the dispersion of the emission energies of individual QDs.

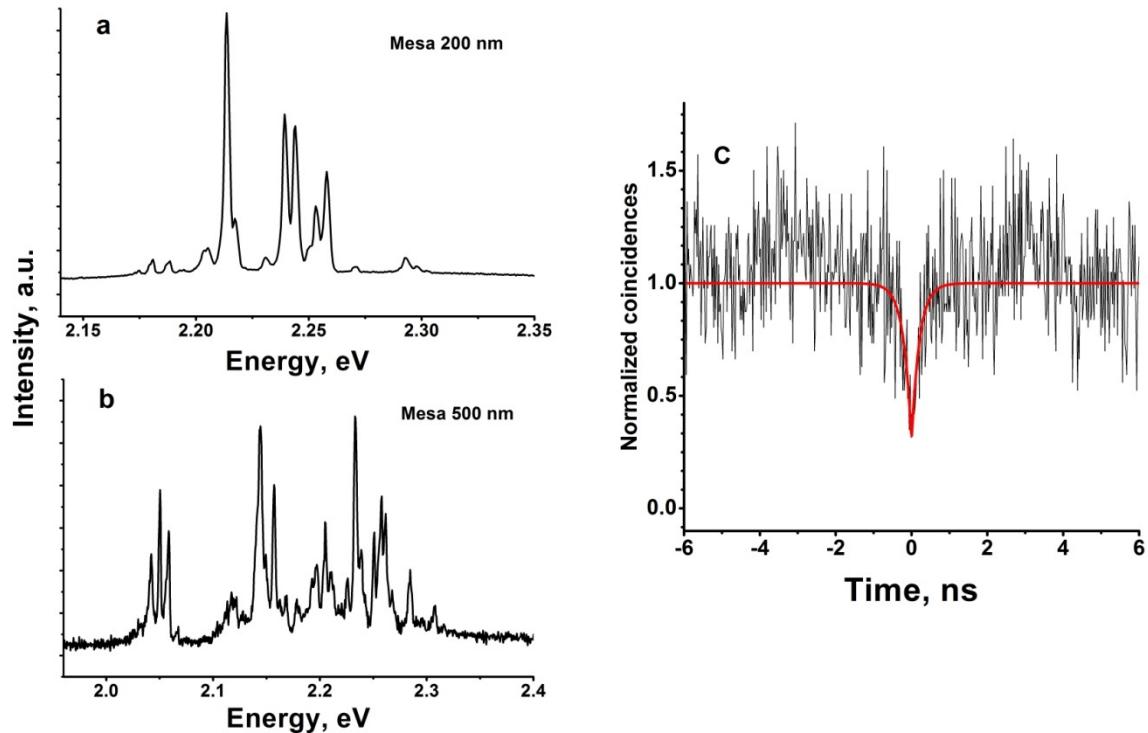


Figure 2. μ -PL spectra measured at 8 K in CdTe/ZnTe QD sample B using (a) 200-nm and (b) 500-nm mesas. (c) Normalized autocorrelation $g^{(2)}$ function of single photon emission measured in sample B at $T = 8$ K using the 200-nm mesa.

The μ -PL spectra of sample B reveal a number of relatively narrow lines, assigned to the emission of excitons and other electron-hole complexes – trions and biexcitons – from individual QDs (figure 2 (a, b)). The ballpark number of narrow excitonic lines in μ -PL spectra is estimated to be about several tens in the mesas with the 500-nm diameters. For the 200-nm mesas, the line number decreases to 2-7. We believe that the destroying of QDs adjusting to the sidewalls can contribute to such a decrease along with reducing the mesa diameter. The recorded number of lines in sample B corresponds to the QDs density as small as 10^{10} cm^{-2} . In contrast, the μ -PL spectra measured in sample A do not show pronounced narrow lines at all. This can indicate either the large density of QDs or the formation of flat extended nano-islands (quantum wells). The final conclusion needs detailed transmission microscopy studies (in progress).

Thus, sample B, grown without the Te flux exposure during the thermal annealing, demonstrates better optical characteristics from the standpoint of quantum light generation. Indeed, the distinct antibunching behavior has been recorded using the 200-nm mesas fabricated in this sample. Figure 2(c) shows the autocorrelation function $g^{(2)}(\tau)$ of a single QD, registered at $T = 8 \text{ K}$. This function is obtained by fitting the experimental data with the equation

$$f(\tau) = a - b \exp\left(-\frac{|\tau|}{c}\right) \quad (1)$$

and normalizing by the obtained value of the parameter a [14]. The fitted value $c = 210 \text{ ps}$ gives estimation of the intrinsic width of the correlation function dip, which is essentially due to the lifetime of the involved QD exciton. For the value of the correlation function at zero time, the fitting gives the value of $g^{(2)}(0) = 0.32$ that evidences the dominance of single photon emission. We underline that, with high enough power density of excitation, the decay of different states, besides excitons, such as trions and biexcitons, can contribute to this value as discussed in [5-7]. Both proper subtraction of their contribution and optimization of design, aimed to increase the separation energies, can improve this characteristic.

4. Conclusions

We demonstrate single photon emission of CdTe/ZnTe QDs grown by thermally-activated MBE. It was achieved in a sample without Te flux exposure during the thermal annealing. We assume that the mobility of Cd adatoms is higher in this case as compared with QDs affected by the Te flux that results in the formation of QDs of smaller sizes but with more dispersed size statistics. This inhomogeneous QD ensemble exhibits a wide PL band (FWHM $\sim 200 \text{ meV}$) with a maximum at 2.12 eV . Respective μ -PL spectra measured using 200-nm mesas demonstrate a countable number of narrow emission lines of single QDs corresponding to the dot density of about 10^{10} cm^{-2} . The value of the autocorrelation function $g^{(2)}(0) = 0.32$ measured in this sample is a signature of quantum light.

Acknowledgements

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