Optical properties of blue-green single-photon sources based on self-assembled CdSe quantum dots

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Abstract. We report on single photon emission with a wavelength below 500 nm from single CdSe quantum dots (QDs) grown by migration-enhanced epitaxy providing a reduced QD lateral density below 10¹⁰ cm⁻². The QD photoluminescence was observed at the temperature of 8 K in 200-nm-wide mesa-structures made of CdSe QD heterostructures. The antibunching effect under cw excitation with g²(0) ~ 0.2 was demonstrated.

1. Introduction
Reliable generation of single photons on demand is essential for quantum cryptography and quantum computing [1]. Using visible single photons instead of the infrared ones is important because of the availability of efficient high-speed single-photon detectors in this wavelength range. Single self-organized quantum dots (QDs) based on wide-gap II-VI CdSe and CdTe compounds grown by epitaxial techniques (either by conventional molecular beam epitaxy, MBE [2,3], or by migration enhanced epitaxy, MEE [4,5]), are considered to be promising candidates for creation of single-photon or even entangled-photon sources of visible light because of their high quantum efficiency, fast radiative decay time and ability to operate at elevated temperatures. They are also less susceptible to blinking, which is an essential drawback of colloidal QDs. However, there are only a few reports on the single-photon emission in epitaxial QDs at room temperature, which is mainly hampered by spectral overlapping of the emission bands of different QDs due to their large density [6,7]. In particular, the conventional approaches to distinguishing the single-QD emission lines (fabrication of etched mesa-structures [8] or nanoapertures in a non-transparent mask [9]) require a relatively low QD density of at least 10¹⁰ cm⁻². Note that CdSe QDs with a wider band gap are more preferable for high-temperature operation than the CdTe ones due to the larger binding energies of excitons, biexcitons, and trions, providing a reduced overlap of the respective PL lines originating from individual QDs [10]. Moreover, the emission of CdSe QDs can be obtained in an extended spectral range including blue light (450-590 nm) [11], whereas the emission of CdTe QDs is limited to the spectral range above 540 nm.

In this paper, we apply micro-photoluminescence (µ-PL) spectroscopy to compare emission properties of individual CdSe QDs grown by either MBE or MEE techniques. The 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. The correlation function $g^{(2)}(\tau)$ was measured and analyzed in order to confirm single photon statistics.

2. Experiment

We have studied emission properties of two samples (labeled as A and B) grown on GaAs (001) substrates. The structures included the following layers sequentially from the substrate: a 10-nm-thick ZnSe buffer layer, a 100-nm-thick ZnSe/ZnSSe short-period superlattice (SL), CdSe QDs surrounded on both sides by 50-nm-thick ZnMgSSe, 1.5-nm-thick ZnSSe and ZnSe layers for better carrier confinement, and a top layer of 3-nm-thick ZnSe. CdSe QDs were fabricated by MEE in sample A, while they were grown by conventional MBE in sample B.

![Figure 1](image)

**Figure 1.** (a) Normalized low temperature (77 K) spatially integrated PL spectra of the structures fabricated using the MEE (sample A) and MBE (sample B) growth modes. (b) $\mu$-PL spectra measured at 8 K in sample A, using a 200-nm mesa.

The emission properties of QDs were investigated using cylindrical mesa-structures of 200 nm in diameter, which were fabricated using a combination of electron lithography and plasma etching. Scanning electron microscopy studies have confirmed the formation of cylindrical mesas with vertical sidewalls. The sample was mounted in a He-flow cryostat with an Attocube XYZ piezo driver inside, which allowed us to optimize and precisely maintain the positioning of the chosen mesa with respect to a laser spot during an extended period (several hours). The $\mu$-PL measurements were carried out under optical excitation by a cw laser line (405 nm). The laser power density was 4 W/cm$^2$. 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 ~200 $\mu$m$^2$) of samples A and B, measured at 77 K, are shown in figure 1(a). The emission line in sample B is markedly red-shifted relative to that in sample A, in spite of the same amount of deposited CdSe. This shift is a sign of larger QD sizes in sample B. Besides, samples A and B have more or less similar FWHM of the emission line (45 meV and 60 meV, respectively), which indicates approximately the same QD size dispersion leading to the dispersion of the emission energies of individual QDs.

The $\mu$-PL spectra of sample A, measured at 8 K, reveal a number of relatively narrow lines assigned to the emission of excitons, biexcitons, and other electron-hole complexes from individual QDs (figure 1(b)). For the 200-nm mesas, the ballpark number of narrow excitonic lines in $\mu$-PL...
spectra is estimated to be in the range of 2 - 7. The recorded number of lines in sample A corresponds to the QDs density as small as $10^{10}$ cm$^{-2}$. The obtained density of QDs is perfectly suited for creation of single-photon sources based on QD emission. In contrast, the μ-PL spectra measured in sample B do not show pronounced narrow lines at all, indicating a large density of QDs.

Figure 2. Spectrally integrated PL intensity vs temperature in sample A. The intensity is normalized to the maximum value. The inset shows the band structure of sample A.

Figure 3. Normalized autocorrelation $g^{(2)}$ function of the single-photon emission measured in sample A at $T = 8$ K using the 200-nm mesa.
The spectrally integrated PL intensity is plotted in figure 2 for the temperature range between 8 and 250 K. The integrated intensity is normalized to the maximum value. The carriers are trapped in a ZnSe/ZnSSe SL at low temperatures and, due to the lack of energy, they cannot pass through a sufficiently thick layer of ZnMgSSe (inset in figure 2). So, a rise in the PL intensity is observed with increasing temperature from 8 to 130 K. The decrease in the PL intensity, observed between 160 to 250 K, is due to carrier delocalization and migration towards centers of nonradiative recombination. In total, the integrated PL intensity drops by less than a factor of 1.5.

Sample A fabricated by the MEE growth mode demonstrates better optical characteristics with respect to quantum light generation. Indeed, the distinct antibunching behavior has been recorded using the 200-nm mesas fabricated in this sample. Figure 3 shows the autocorrelation function \( g^{(2)}(\tau) \) of a single QD, registered at \( T = 8 \) K. This function is obtained by fitting the experimental data to the equation:

\[
f(\tau) = a - b \exp\left(-\frac{\tau}{c}\right)
\]

and normalized according to the formula \( C_n(t) = c(t) / (N_1 N_2 \omega T) \), where \( c(t) \) is the raw coincidence number, \( N_{1,2} \) are single counter rates, \( \omega \) is the width of the time bin, and \( T \) is the total acquisition time [12]. The fitted value \( c \approx 510 \) ps gives an estimation of the intrinsic width of the correlation function dip, which is essentially due to the lifetime of the involved QD exciton. For the measured correlation function, the fitting gives the value of \( g^{(2)}(0) = 0.19 \pm 0.05 \) that evidences the dominance of single-photon emission.

4. Conclusions
We have presented comparative studies of self-assembled CdSe QDs grown either by MEE or MBE. Only the approach involving the MEE growth mode allowed us to obtain a low enough QD density of \( 10^{10} \) cm\(^{-2} \) and to extend the available spectral range of the single-photon emission below 500 nm. The low value of the second-order correlation function measured in this sample, as well as rather high carrier confinement, provide an opportunity to use similar CdSe QDs in various quantum systems working at high temperatures.

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