Single-photon sources of visible light

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Abstract. We report on emission properties of single-photon sources of visible light, based on InAs/AlₓGa₁₋ₓAs, CdTe/ZnTe, and CdSe/ZnSe quantum dot (QD) heterostructures grown by molecular beam epitaxy. For all types of QDs, the studies performed by microphotoluminescence and photon correlation spectroscopies demonstrated the single-photon nature of emission with typical values of the second-order correlation function \(g^{(2)}(0)\) less than 0.2 and allowed evaluation of sub-microsecond kinetics of blinking governed by local charging effects.

1. Introduction

The sources of single-photon emission (SPE) and entangled photon pairs, based on epitaxial semiconductor quantum dots (QDs) emitting in the visible spectral range, are promising for applications in the systems of quantum cryptography, intended for secure communications through free-space atmospheric optical connection lines or plastic fibers. Commercial devices of this type are currently absent due to both technological complexity of reproducible fabrication of the samples with the isolated single QDs and material constraints limiting possibility to distinguish emission lines of single excitons, charged excitons (trions), and biexcitons at elevated device operation temperatures. In this work, we present comparative optical studies of the SPE sources based on different QD heterostructures: InAs/AlₓGa₁₋ₓAs (x>0.35), CdTe/ZnTe, and CdSe/ZnSe, whose emission spectrum jointly covers extremely wide spectral range between 500 and 1000 nm.

2. Samples and experimental techniques

2.1. InAs/AlGaAs quantum dots

Advantages of the InAs/AlₓGa₁₋ₓAs QDs consist in possibility to achieve SPE in the red spectral range using well-developed epitaxial techniques for the growth of the required QD heterostructures [1-5]. A potential disadvantage of previously studied single InAs/AlₓGa₁₋ₓAs QDs is enhancement of the spectral width of the single-QD emission lines with an increase of the Al content in the AlₓGa₁₋ₓAs barriers [4] that is a signature of the emission blinking and spectral diffusion. Besides, the enlarged content of Al results in a significant (up to 1 meV) enhancement of the anisotropic exchange splitting of a single-QD exciton [5], that hampers generation of polarization entangled photons during a
cascade recombination of biexcitons. To the best of our knowledge, single-photon characteristics of the QDs of this type have never been studied.

To elucidate these issues we have investigated systematically emission properties of single self-organized InAs/Al<sub>x</sub>Ga<sub>1-x</sub>As (0.35<x<0.5) QDs formed by molecular beam epitaxy (MBE) on (001)GaAs substrates, using the Stranski-Krstanow growth mechanism. Two different designs were considered. In the first one (type A), the layer of InAs QDs was directly embedded in an Al<sub>x</sub>Ga<sub>1-x</sub>As matrix. In the second design (type B) a two-monolayer-thick interlayer of GaAs was deposited prior to the QDs growth. The developed MBE growth regimes allowed achievement of the QDs surface density below10<sup>10</sup> cm<sup>-2</sup>.

2.2. CdTe/ZnTe and CdSe/ZnSe quantum dots

Single QDs based on wide bandgap II-VI compounds CdTe and CdSe are of particular interest due to the possibility to obtain SPE in green, yellow, and orange spectral ranges [6-8]. Yet unsolved problem (especially for CdSe QDs) has been the reproducible growth of the structures with small enough QD surface density. For both types of heterostructures, we have developed MBE growth regimes resulting in the formation of a single sheet of QDs with the QD surface density in the range of 10<sup>10</sup>- 10<sup>11</sup> cm<sup>-2</sup>. The achieved decrease in the surface density of CdTe/ZnTe QDs relied on further development of so-called thermal activation technique, where the QD formation is induced by the reduction of the surface energy of a strained 2D CdTe layer by means of covering it with an amorphous Te layer, followed by its fast desorption [9,10]. The formation of a low-density array of CdSe/ZnSe QDs was realized using a reorganization process occurring during growth interruption and thermal activation [11].

2.3. Micro-photoluminescence and photon correlation spectroscopy

For single-QD micro-photoluminescence (µ-PL) measurements, cylindrical mesa-structures containing only a few emitting QDs were fabricated by electron-beam lithography and Ar<sup>+</sup>-ion beam etching. The samples with mesa-structures were attached to a 3D Attocube piezo-driver located within a Janis continuous flow He micro-cryostat. The QD PL was excited by a continuous wave semiconductor laser diode at 405 nm and collected by using a confocal microscope with a 50x Mitutoyo microscope objective. The emission was dispersed by a 50 cm grating monochromator and registered by a liquid-nitrogen-cooled CCD camera. Statistical characteristics of the single-QD emission were evaluated by means of photon correlation measurements performed under the continuous wave excitation with a Hanbury Brown – Twiss intensity interferometer equipped with two single-photon avalanchen Si photodiodes providing time resolution better than 40 ps. To obtain the second-order correlation function g<sup>(2)</sup>(τ), the measured histogram of coincidences was normalized by count rates of the detectors, width of the time bins, and the integration time [12].

3. Results and discussion

The type-A heterostructures with InAs QDs embedded immediately in Al<sub>x</sub>Ga<sub>1-x</sub>As (x>0.35) demonstrate a wide PL spectrum extending over the spectral range between ~1000 and ~620 nm (see Fig. 1). From the blue side, the spectrum is limited by the wavelength corresponding to the direct-indirect bandgap transition in the Al<sub>x</sub>Ga<sub>1-x</sub>As ternary alloy.No distinct PL line related to a wetting layer can typically be observed in the type-A structures. In contrast to that the type-B structures demonstrate a narrower spectrum whose maximum is located between 900 and 1000 nm, while the blue boundary of the QD emission spectrum corresponds to a distinct narrow PL peak near 700 nm, attributed to the emission of the wetting layer. The emission spectrum of CdTe QDs is adjacent to that of InAs QDs and spreads approximately between 650 and 550 nm (see Fig. 2). From the blue side, it overlaps with the PL band of CdSe QDs, extending towards 500 nm.
Figure 1. PL spectra measured at 77 K in InAs/Al$_x$Ga$_{1-x}$As QD samples with a 0.3-mm-size excitation spot. Solid and dashed lines correspond to a type-A (x=0.4) and type-B (x=0.35) samples, correspondingly.

Figure 2. Typical PL spectra measured at 77 K in CdSe/ZnSe (solid line) and CdTe/ZnTe (dashed line) QD samples with a 0.3-mm-size excitation spot.

For both types of InAs/Al$_x$Ga$_{1-x}$As QD structures, the $\mu$-PL spectra measured at 8 K in small enough mesa-structures demonstrate a set of ultra-narrow lines attributed to the emission of single individual QDs (see Fig. 3 (a)). Many of them possess the spectral width that is below resolution of the used monochromator (~50 μeV). In accordance with Ref. [5], some of these lines demonstrate a fine structure splitting of linearly polarized exciton lines, which can be as large as ~800 μeV. It was found that an increase in the Al content leads to enhancement of the average density of the QDs with the largest splitting at the expense of the QDs with the smallest one. Nevertheless, for both kinds of structures and any Al mole fraction in the range of 35-50% there always exists a pronounced number of QDs demonstrating emission lines with the anisotropic exchange splitting smaller than the set-up spectral resolution. These lines presumably correspond to the emission of either single excitons confined in relatively isotropic QDs or charged excitons (trions) and are promising for obtaining efficient SPE. The respective emission lines of individual CdTe QDs (Fig. 3 (b)) and CdSe QDs (Fig. 3 (c)) are noticeably wider: 0.2 – 0.8 meV for the former structures and 0.1 – 0.2 meV for the latter.

Figure 4 represents the second-order correlation functions obtained in representative mesa-structures containing InAs/Al$_x$Ga$_{1-x}$As QDs (Fig. 4 (a)), CdTe/ZnTe QDs (Fig. 4 (b)), and CdSe/ZnSe QDs (Fig. 4 (c)). The strongest and narrowest emission lines were selected for these measurements. The functions are fitted with the expression

$$g^{(2)}(\tau) = 1 + c_1 e^{-\tau/\tau_1} + c_2 e^{-\tau/\tau_2}.$$  

(1)

For all types of QDs, the data exhibit the antibunching dip at zero time delay with the average $g^{(2)}(0)$ as low as 0.1 – 0.2 that is a clear signature of the single-photon nature of the emission. The time width
of the dip, described by the factor $\tau_1$, corresponds to the PL lifetime being in the range 1-1.5 ns. The second exponential term is responsible for the appearance of two symmetric bunching peaks, which reflect the kinetics of blinking induced by local charging effects [13]. One can see in Fig. 4 that the emission of single CdSe/ZnSe QDs is practically free of these parasitic effects, while both InAs/Al$_x$Ga$_{1-x}$As and CdTe/ZnTe QDs suffer from certain blinking caused most probably by the QDs dynamical charging taking place on the time scale of ~10 ns.

![Image](a)

**Figure 3.** $\mu$-PL spectra measured at 8 K in representative InAs/AlGaAs (a), CdTe/ZnTe (b), and CdSe/ZnSe (c) mesa-structures.

![Image](b)

**Figure 4.** Typical second-order correlation $g^{(2)}$ functions measured at 8 K in representative InAs/AlGaAs (a), CdTe/ZnTe (b), and CdSe/ZnSe (c) mesa-structures. Solid lines represent fits of the experimental data, obtained with Eq. (1).

4. Conclusions

We have demonstrated possibility to fabricate sources of single photons, based on low-density arrays of InAs/Al$_x$Ga$_{1-x}$As ($x$$>$$0.35$), CdTe/ZnTe, and CdSe/ZnSe self-organized QDs, whose emission spectra progressively cover all visible spectral range above~500 nm. While both CdTe/ZnTe and InAs/Al$_x$Ga$_{1-x}$As QDs suffer from certain blinking taking place on the time scale of ~10 ns, all the types of QDs show the second-order correlation function at zero delay $g^{(2)}(0)$ as low as 0.1 – 0.2, that is promising for applications in effective systems of quantum cryptography.

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