Emission properties of individual InAs/Al$_{0.44}$Ga$_{0.56}$As quantum dots

M V Rakhlin$^1$, K G Belyaev$^1$, G V Klimko$^1$, I S Mukhin$^{2,3}$, S V Ivanov$^1$, and A A Toropov$^1$

$^1$Ioffe Institute, Saint Petersburg 194021, Russia
$^2$St. Petersburg Academic University, St. Petersburg 194021, Russia
$^3$ITMO University, St. Petersburg 197101, Russia
maximrakhlin@mail.ru

Abstract. We report on micro-photoluminescence studies of InAs/AlGaAs single self-assembled quantum dots grown by molecular beam epitaxy. A variation of the exciton fine structure splitting induced by the anisotropic part of the electron-hole exchange interaction is systematically studied depending on the exciton emission energy and the insertion of a GaAs interlayer prior formation of the InAs QDs.

1. Introduction
In recent years, an electron-hole exchange interaction in semiconductor quantum dots (QDs) has been the subject of careful studies [1] because detailed understanding of the relevant exciton fine structure is of crucial importance for applications of the individual QDs as single photon emitters possessing non-classical photon statistics, and especially as sources of the entangled pairs of single photons. In principle, the polarization-entangled photons can be generated by recombination of a biexciton cascade in a single QD. However, in the QD structures with low enough confinement symmetry ($C_{2v}$ or lower), the anisotropic part of the electron-hole exchange interaction lifts twofold degeneracy of the exciton bright states, resulting in the exciton fine structure splitting (FSS). If the FSS is larger than the exciton homogeneous linewidth, it inhibits the emission of two entangled photons [2]. The issues of controlling both the FSS and the exciton linewidth have been relatively well understood for conventional InAs/GaAs QDs [1,2], whereas for InAs/AlGaAs single QDs which are promising non-classical light sources of visible light, this has been yet poorly investigated [3,4].

In this work, we apply micro-photoluminescence ($\mu$-PL) spectroscopy to elucidate the emission properties of individual InAs/Al$_{0.44}$Ga$_{0.56}$As QDs, grown by molecular beam epitaxy (MBE). In particular, we investigate the influence on the FSS of the presence of a thin GaAs interlayer deposited on a AlGaAs bottom barrier prior to the InAs QDs formation.

2. Experiment
We have studied emission properties of individual InAs/Al$_{0.44}$Ga$_{0.56}$As QDs either with (sample A) or without (sample B) a 2-monolayer (ML)-thick GaAs interlayer inserted between the bottom AlGaAs...
barrier and the InAs QDs. The QDs were formed by MBE on GaAs (001) substrates, using the Stranski-Krastanow growth mode. Special measures were taken to reduce the density of QDs down to around $10^{10}$ cm$^{-2}$. The emission of a limited number of individual QDs was registered by the confocal $\mu$-PL set-up in cylindrical mesa-structures of 200-800 nm in diameter, which were fabricated by electron beam lithography and Ar$^+$-ion etching. Scanning electron microscopy studies have confirmed formation of perfect 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 $\mu$-PL measurements were carried out under optical excitation by a $\textit{cw}$ laser line (405 nm). The laser power density was 4 W/cm$^2$ before a cryostat window. Photon correlation measurements were performed in a Hanbury Brown-Twiss detection scheme exploiting two single-photon avalanche photodiodes possessing the photon timing resolution of about 40 ps.

3. Results and discussion
Typical $\textit{cw}$ PL spectra (integrated over the area of $\sim$200 $\mu$m$^2$) of samples A and B, measured at 77 K, are shown in Figure 1a. The emission spectrum in sample A with a GaAs interlayer reveals two peaks attributed to the emission of QDs and a wetting layer (WL). The QD PL line is centered between 900 and 1000 nm, that is similar to the emission spectrum of most common InAs/GaAs QDs. However, the blue wing of the PL line spreads over the significant spectral range right up to the narrow PL peak from the WL, located near 1.8 eV (~690 nm). Thus, most part of the InAs/AlGaAs QDs in sample A emit in the same spectral range as InAs/GaAs QDs, possessing, however, much larger activation energy for the confined charge carriers, that is promising for higher temperature operation. Only one strongly asymmetric peak attributed to the QDs emission can be seen in the PL spectrum of sample B. The peak is markedly blue-shifted relative to the QD PL line in sample A, that is a signature of a smaller QD size in sample B, confirmed also by atomic force microscopy (not shown here), and/or a different proportion of Ga and Al atoms incorporation into the QDs due to inter-diffusion. Thus, the absence of the GaAs interlayer in sample B causes the shift of the PL spectrum towards higher energy and the lack of the WL emission. This sample reveals a wide PL spectrum extending over the energy range between $\sim$1.4 and $\sim$2 eV (890 – 620 nm), which allows one to tune the individual QDs emission wavelength over the extremely wide wavelength range from visible red to near infrared.

![Figure 1](image-url)

**Figure 1.** (a) Normalized low temperature (77 K) spatially integrated PL spectra of the structures grown either with a GaAs interlayer (sample A) or without it (sample B). (b) $\mu$-PL spectra measured at 8 K in InAs/AlGaAs QDs, using 500-nm etched mesas.
The μ-PL spectra measured at 8 K reveal a number of relatively narrow lines assigned to the emission of excitons and other electron-hole complexes – biexcitons, trions, and charged excitons [5] – in individual QDs (Figure 1b). In both samples, the ballpark number of narrow excitonic lines in a μ-PL spectrum is estimated as about several tens in the mesas with 500-nm diameters, that corresponds to the QDs density as small as $1.7 \times 10^{10} \text{cm}^{-2}$ (sample A) and $1.3 \times 10^{10} \text{cm}^{-2}$ (sample B). The obtained QDs densities are low enough and suit perfectly for creation of single-photon sources based on the emission of single QDs.

Figure 2. (a) Typical single-exciton FSS as large as 440 μeV, measured in sample A at 8 K. Solid and dashed lines represent the spectra for orthogonal linear polarizations corresponding to [110] and [1-10] crystal directions. (b) Measured FSS as a function of the exciton recombination energy for samples A and B.

It was found that different QDs in sample A, located in the same mesa-structure, display various FSS ranging from the values less than 70 μeV (set-up spectral resolution) up to extremely large FSS of the order of 500 μeV. A typical μ-PL spectrum with FSS equal to 440μeV is shown in Figure 2a. Solid and dashed curves are the spectra for orthogonal linear polarizations corresponding to [110] and [1-10] crystal directions of the QD sample. Such a large FSS is consistent with the results of Finley et al. who reported on the FSS values up to 1 meV [3]. In contrast, different QDs in sample B demonstrate approximately the same FSS equals to 100-150 μeV. This value is quite comparable with the typical exciton linewidth (~130 μeV) detected in this sample. Figure 2b displays the measured exciton FSS as a function of the exciton recombination energy for the samples with and without the GaAs interlayer. We found out that there is no pronounced dependence of FSS on the exciton recombination energy in both samples. The presence of the GaAs interlayer in sample A results in enhanced scattering of the FSS magnitude among different QDs. Nevertheless, most of the excitonic lines are not split in both samples, at least within the set up spectral resolution. On average, there are 11 unpolarized exciton lines per one line with FSS.

Figure 3 shows the autocorrelation function $g^{(2)}(\tau)$ of a single QD, obtained at T= 8 K in sample B. The experimental data on photon coincidences is fitted with the equation

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

and sequentially normalized by count rates of the detectors, width of time bins, and the integration time [6]. The autocorrelation function exhibits an antibunching dip at zero time delay with the average $g^{(2)}(0)$ as low as 0.24 that is a clear signature of the single-photon nature of the emission. The
obtained value \( c = 650 \) ps gives estimation of the intrinsic width of the antibunching dip, which is essentially due to the lifetime of the involved QD exciton.

**Figure 3.** Normalized second-order correlation function \( g^{(2)}(0) \) of single-photon emission, measured at \( T = 8 \) K in a 500-nm mesa in sample B.

**4. Conclusions**

We have represented comparative studies of self-assembled InAs/Al\(_{0.44}\)Ga\(_{0.56}\)As QDs grown either with or without a GaAs interlayer, whose emission spectrum overlaps the extremely wide wavelength range from visible red to near infrared (1.1-2 eV). The performed measurements of exciton fine structure splitting reveal that the predeposition of the 2 ML-thick GaAs interlayer leads to enhanced scattering of the FSS values ranging between 70 μeV (the set up spectral resolution) and \(~ 500 \) μeV. The measured second-order correlation function at zero delay \( g^{(2)}(0) \) as low as 0.24 is a promising value for applications in effective systems of quantum cryptography.

**Acknowledgments**

The authors gratefully acknowledge the financial support of the Russian Science Foundation (project #14-22-00107).

**References**

[1] Seguin R, Shliwa A, Rodt S, Pötschke K, Pohl U W, Bimberg D, 2005 *Phys. Rev. Let.* 95 257402
[2] Santori C, Fattal D, Pelton M, Solomon G S, Yamamoto Y, 2002 *Phys. Rev. B* 66 045308
[3] Finley J J, Mowbray D J, Skolnick M S, Ashmore A D, Baker C, Monte A F G, Hopkinson M, 2002 *Phys. Rev. B* 66 153316
[4] Grijseels S C M, van Bree J, Koenraad P M, Toropov A A, Klimko G V, Ivanov S V, Pryor C E, Silov A Yu, 2016 *J. of Lum.* 176 95
[5] Karlsson K F, Oberli D Y, Dupertuis M A, Troncale V, Byszewski M, Pelucchi E, Rudra A, Holtz P O, Kapon E, 2015 *New J. of Phys.* 17 103017
[6] Broui R, Beveratos A, Poizat J-P, Grangier P, 2000 *Opt. Lett.* 25 1294