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Fine structure splitting reduction in droplet epitaxy
GaAs quantum dots grown on (111)A surface

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Abstract. In this contribution we compare the geometrical symmetry of self–assembled GaAs quantum dots (QDs) grown by a refined droplet epitaxy (DE) method on (100) and (111)A AlGaAs substrate. By atomic force microscopy (AFM) we study the morphology of the QDs and by micro–photoluminescence spectroscopy (μPL) we compare the dependence of the fine structure splitting on QD size and shape. We show that: i) the refined DE enhances the elongation of QDs grown on (100) AlGaAs substrate increasing the fine structure splitting; ii) by growing the QDs on the (111)A AlGaAs surface it is possible to recover a good geometrical circular symmetry obtaining a small value of the fine structure splitting.

1. Introduction
The control of the anisotropic exchange Coulomb interaction between electron and hole spin in QDs (usually said fine structure splitting, FSS) is a relevant issue for quantum information applications. Single QDs in fact represent a promising building block for the realization of polarization–entangled photons [1] from the biexciton–exciton–ground state cascade (XX-X-gs) provided that the FSS is tuned to zero [2].

Generally speaking, the FSS originates from asymmetries in the QD confinement potential [3, 4] and brings to a polarization splitting in the X photoluminescence (PL) which complicates the opportunity of realizing entangled photon pairs. In Stranski Krastanov (SK) QDs, anisotropies came from several sources (shape, strain, atomistic effects and piezoelectricity) and claims of the major role of strain induced effects on the FSS have been reported [3].

Taking advantage of the DE growth method [5, 6, 7], we can realize strain–free GaAs/AlGaAs QDs in order to investigate only the effect of geometrical asymmetry. In previous studies on DE QDs grown with a conventional low–temperature method [5, 6, 7] on (100) AlGaAs surface, a reduction in the FSS was shown for small circular–symmetric nanoemitters [8]. Nevertheless, a significant drawback of the low temperature used during the growth was a large PL linewidth of the excitonic recombination from single QDs (≈100 μeV) ascribed to the fluctuating charged environment [9]. In this paper we demonstrate that a refined high–temperature DE [10] on AlGaAs (100) surface improves the QDs quality in terms of spectral linewidth [9, 10] but increases the geometrical elongation and the FSS [11] respect to the conventional low–temperature growth [8]. Moreover, the same refined DE on the AlGaAs (111)A surface allows also a reduction in the FSS.
The paper is organized as follows: in section 2 we describe the sample growth and the method of investigation, in section 3 we show the analysis of AFM measurements on uncapped samples and μPL measurements on single QDs, finally, in section 4, we summarize the results.

2. Experimental method

Two GaAs QD samples were grown on AlGaAs (100) and (111)A (respectively indicated as sample A and B) in a solid-source molecular beam epitaxy (MBE) system with a refined DE procedure [10] which is here summarized. After the growth of a AlGaAs barrier layer at 580 °C, the temperature was set to 350 °C. At this stage Ga droplet were deposited and then irradiated by As flux for the crystallization of GaAs QDs at 200 °C. The QDs density, determined by substrate temperature, was as small as $10^8$-$10^9$ QDs/cm$^2$. The GaAs QDs were annealed at 400 °C and than covered by 40 nm AlGaAs capping layer grown by standard MBE at the same 400 °C plus 20 nm thick AlGaAs layer at 580 °C. Than a 10 nm thick GaAs capping layer was grown on top by standard MBE at 580 °C and finally the samples were annealed at 800 °C.

The main difference between this refined procedure and the conventional DE is the annealing step after the QDs crystallization. The increased temperature allows to grow a high quality top barrier which is the key factor ruling the PL quality in terms of spectral linewidth [9, 10].

Low-temperature μ–PL experiments were performed by using a confocal setup in illumination–collection configuration allowing for a lateral resolution better than 1 μm. The small QDs areal density allowed collecting the PL from few nanoemitters and it was enough for accessing the single nanostructure emission after spectral filtering. The PL polarization was detected by using a half-waveplate and a polarizer. The collected PL was fed into a grating monochromator and detected by a silicon-based CCD camera allowing to detect lineshift as small as $\sim 10$ μeV and line broadening as small as $\sim 50$ μeV after a fitting procedure. For illumination we used a CW solid–state laser emitting at a wavelength of 532 nm to excite the high energy states of the AlGaAs barrier.

3. Results and discussion

Figure 1 (a) and (b) shows AFM profiles of QDs from samples A and B before capping. We note that the QD shape in sample A is elongated along [1-10] direction. We ascribe this behavior to the different adatom migration along the orthogonal in–plane directions [110] and [1-10][10]. Furthermore we observe a increase in the geometrical elongation respect to QDs grown with the conventional DE on (100) surface [10, 11, 8, 12]. We relate this effect to the increased temperature during the refined growth process which enhances the material migration along [1-10] direction [10]. Differently from A the case of sample B, despite the high temperature of the growth, doesn’t show any preferential elongation axis in the QDs shape. We ascribe this feature to the three fold rotational symmetry of the (111)A substrate.

Single QD PL of samples A has been studied in previous works and emission from neutral exciton (X) charged exciton (T) and neutral biexciton (XX) was demonstrated by FSS measurements [10]. In addition, the refined DE growth allowed for a drastic reduction in the inhomogeneous broadening (in the high excitation regime the linewidth was as small as $\sim 30$ μeV, about 4 times smaller respect to samples grown with conventional DE)[10, 9]. Moreover, in the low excitation regime, we can achieve the very small linewidth of 4 μeV as measured by scanning Fabry–Perot interferometry (not shown). This accounts for the improvement achieved thanks to the refined DE in terms of line broadening.

Typical PL spectra of QDs from sample B are shown in Fig. 3 (a) and (d) for different excitation power. The power dependence of X and XX lines is reported in Fig. 3 (b) for the QD of graph 3 (a). Power dependence of X and T of the QD shown in graph 3 (d) are displayed in graph 3 (e). On the basis of first and second order Poissonian or quasi–Poissonian dependence of the peak intensity on the excitation power we interpret these lines as neutral exciton X,
neutral biexciton XX and charged exciton T recombination[13]. This attribution is confirmed by polarization resolved PL reported in graphs 3 (c) for the QD of graph 3 (a) and in graph 3 (f) for the QD of graph 3 (d). While X and XX show the typical mirror–symmetric polarization splitting, the charged complex T is not split. The measured FSS are \(\approx 22 \ \mu eV\) for the QD in 3 (a) and \(\approx 26 \ \mu eV\) for the QD in 3 (d). We note also that the anisotropy axes of the two different QDs are noticeably different [8, 12]. In the merit of this we note that a random orientation of the polarization axes was found for all the QDs grown on (111)A surface (not shown) and in small, circular–symmetric QDs grown on (100) surface [8, 12]. The linewidth, which cannot be resolved by this experimental setup, is below 50 \(\mu eV\).

Similar \(\mu PL\) investigation was performed on several QDs from sample A and B. The results of the measured FSS are reported as a function of the X energy in Fig. 3. A comparison between the FSS in the present sample A and previous samples grown by conventional DE on (100) highlights the enhanced geometrical elongation of the QDs shape due to the different diffusion process which takes place in [100] and [1-10] directions. In fact the FSS is in average larger than the previous reported results [11, 8, 12]: the new refined growth has the relevant drawback of increasing the FSS from \(\sim 90 \ \mu eV\) up to \(\sim 140 \ \mu eV\). This points out that the annealing step, after the QDs crystallization, rules the enhanced geometrical elongation. On the contrary the FSS in sample B is as small as \(\approx 17 \ \mu eV\) (note that, due to the poor resolution of the experimental setup, this value should be considered as an upper limit).

This results account for the overall quality improvement of the DE QDs grown on (111)A surface in terms of linewidth and FSS. In fact, by keeping a high growth temperature we obtain a small PL broadening which is the signature of the improved material quality. At the same time, thanks to the choice of the (111)A plane for the droplet deposition on which any diffusion process is equivalent every 120 degrees, we can realize circular symmetric QDs.

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**Figure 1.** (a) and (b): AFM profiles of uncapped samples in orthogonal directions for a large (top panel) and a small (bottom panel) QD grown on AlGaAs (100) (sample A) and (111)A (sample B) respectively. Dashed (continuous) lines refer to scan in [1-10] ([110]) for sample A and [01-1] ([211]) for sample B.
Figure 2. (a) and (d): PL spectra of a QD grown on (111)A at different excitation power. (b): power dependence of X and XX peak intensity of the corresponding QD in (a) as a function of the incident power. (e): power dependence of X and T peak intensity of the corresponding QD in (d) as a function of the incident power. The symbols represent the experimental data. Solid lines represent first and second order Poissonian fit for X and XX (graph (b)), first order Poissonian and quasi-Poissonian fit for X and T (graph (e)). (c): energy shift of X and XX of the QD in (a) as a function of the detected polarization angle represented in a polar plot. (f): energy shift of X and T of QD in (d) as a function of the detected polarization angle represented in a polar plot. Symbols are the experimental data while solid lines are sinusoidal fit to data.

4. Conclusions

In conclusion we have studied two samples of GaAs QDs grown on AlGaAs (100) and (111)A by a refined DE technique. By PL spectroscopy we accessed the single nanoemitter discriminating between X, T and XX recombination. By polarization resolved PL analysis we investigated the FSS of several QDs allowing a comparison of the geometrical properties in different samples. The effect of the refined DE technique on QDs grown on (100) AlGaAs surface is an enhanced elongation which results in a larger FSS. On the contrary in the case of AlGaAs (111)A surface, due to the lack of preferential direction for the material diffusion, a more circular–symmetric shape of the QDs is obtained leading to a reduction in the FSS. We believe that the present achievement represent a relevant step toward the realization of solid–state sources of
polarization–entangled photon pairs

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