Exciton fine structure in strain-free GaAs/$Al_{0.3}Ga_{0.7}As$ quantum dots: Extrinsic effects

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We report polarization-resolved high spectral resolution photoluminescence measurements in self-assembled strain-free GaAs/$Al_{0.3}Ga_{0.7}As$ quantum dots designed and realized in order to reduce as much as possible strain and segregation, which affected previous fine-structure splitting (FSS) experiments. Photoluminescence from isolated quantum dots exhibits a linearly polarized FSS. FSS clearly shows a quantum size effect monotonically decreasing from 90 to 20 μeV by decreasing the quantum dot size (increasing emission energy). While this finding is similar to that observed in strained In(Ga)As/GaAs quantum dots, clearly it requires a different explanation, being our quantum dots not affected by strain-induced piezoelectricity. We ascribed the observed FSS to a size dependent reduction in dot shape anisotropy as evidenced by structural data analysis. Moreover the linear polarization in dots with shape close to cylindrical symmetry is not along the [110] crystallographic axis but it turns out randomly distributed, highlighting the role of extrinsic effects.

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Quantum dots (QDs) are often referred to as artificial atoms as their electronic states are quantized in three dimensions, resulting in a discrete energy-level spectrum, similar to that of an atom. They thus provide a quantum system, which can be engineered to have a wide range of desired properties, and, rather important, they can be grown within robust monolithic semiconductor devices. From the fundamental point of view these designable atoms provide a unique system to investigate carrier-carrier interactions in a precise manner. Among these, the fine-structure splitting (FSS), which is the degeneracy lift of the bright excitonic states induced by asymmetries in the QD confining potential,[1] holds an extremely high importance. The understanding and control of FSS in QDs is a relevant issue for quantum information applications[2] because triggered polarization-entangled photon pairs could be obtained by the radiative decay of biexcitons in QDs provided that FSS is tuned to zero.[3]

Since the first observation of FSS for excitons localized in three dimensions by monolayer fluctuations in GaAs/AlGaAs quantum wells,[1] FSS is a common feature of the excitonic emission from epitaxially grown QDs.[4,5] FSS shows an extremely wide range of values depending on the specific measured QD system,[6] ranging from 8 μeV in InGaAs/GaAs Stranski-Krastanow (SK) QDs (Ref. 7) up to the giant value of 1 meV in InAs/AlGaAs SK-QDs.[8]

The major contribution to the lifting of the bright exciton degeneracy in QDs comes from asymmetries in the QD confinement potential, which induce a splitting of the exciton states via the exchange interaction.[1,9,10] As a matter of fact, the confinement potential anisotropies in epitaxially grown QDs arise from several sources, such as shape,[9,10] strain,[6,11] piezoelectricity,[6,12] and crystal symmetry.[11] Several theoretical and experimental works tried to assess the role of each of the aforementioned anisotropy sources in determining the QD exciton FSS. However, the majority of the papers report about FSS in SK-QDs where a number of not well determined and interconnected QD structural parameters, such as actual shape,[12] average composition,[13] and composition profile,[14] make the quantitative comparison with theoretical prediction rather difficult. In addition, it must be mentioned that all the theoretical predictions[6,11,15] fail to justify the observed large FSS observed in SK-QDs (where FSSs larger than 100 μeV are often observed[6]) when realistic QD structural characteristics are used in the calculations. It clearly appears that for a deep understanding of the FSS it is necessary to experimentally clarify the influence of strain, piezoelectricity, geometry, and composition separately.

In this paper we present a high-resolution, polarization-dependent photoluminescence (PL) study in single epitaxial GaAs/AlGaAs strain-free QDs as a function of the QD size. We designed and realized our sample in order to reduce as much as possible FSS structural-related sources, such as strain, composition, and shape, which affected previous FSS experiments in SK-QDs. The exclusion of the strain-related piezoelectric effect, whose relative importance in determining the FSS in SK-QDs is still a matter of strong debate,[6,11,12] allows us to obtain a nice correlation between the observed FSS and structural data. Our QD PL lines exhibit FSS, which monotonically decreases from 90 to 20 μeV when decreasing the quantum dot size. The FSS decrease closely follows the reduction in $[110]/[1\bar{1}0]$ shape anisotropy that takes place in our QDs as the size is reduced. When the structural asymmetry in our QDs is close to zero, we find that the QD linear polarization axis turns out significantly distributed along random oriented directions. These findings, together with the observed cylindrical symmetry of the QD shape, highlight the role of extrinsic effects as significant contribution in determining the QD potential anisotropy.

The sample studied in this paper consists of GaAs QDs embedded in an $Al_{0.3}Ga_{0.7}As$ barrier. The QDs were grown by droplet epitaxy (DE).[16–19] Unlike SK, DE is a growth method that allows for QD self-assembling also in lattice

matched systems. The DE-QDs can be tuned in size and density independently, thus allowing for the realization of a sample with a relatively low QD density ($\approx 1 \times 10^9$ cm^{-2}) and a broad size dispersion. Particular care was taken in order to avoid modifications of the structural properties of the QDs during the capping step, which has been realized using mobility enhanced epitaxy[20] at an extremely low temperature (200 °C). After the growth, the sample was annealed either at 680 °C for one hour or at 800 °C for 8 min in As atmosphere. The QDs PL properties for the two samples are very similar. This allows for a substantial reduction in the defectivity of the material at the expense of a relatively small (around 1 nm) group III interdiffusion at the interfaces.[18,21] More details on the growth procedure and annealing effects can be found in Refs. 18 and 19.

Micro-PL experiments were performed using a ×100 microscopy objective (NA=0.7) and a single mode optical fiber acting as a confocal pinhole, leading to a lateral resolution of ~0.5 μm. The QD sample was cooled down to cryogenic temperature in a low vibration cold finger cryostat at T =10 K. With a 1 m focal length double-grating monochromator, we could reach a spectral resolution of 25 μeV in full width at half maximum (FWHM). With a fitting procedure we are able to identify a PL line shift as small as ~5 μeV. For illumination we used a continuous-wave solid-state laser emitting at a wavelength of 532 nm to excite the continuum states of the $Al_{0.3}Ga_{0.7}As$ barrier. The spot size was 5 μm. The excitation polarization was set to be linear to avoid Overhauser effects.[22,23] The linear polarization detection was controlled by rotating a half-wave plate inserted in front of a linear polarizer.

A typical PL spectrum of a single GaAs QD is shown in Fig. 1(a). At 1.5 mW, two lines labeled X and T are observed. With increasing power to 15 mW, another line labeled XX emerges on the lower-energy side. The power dependence of lines X, T, and XX is presented in Fig. 1(b); for each line the intensity increases with power until it reaches maximum and then starts to quench. A fit of the experimental data to a power law indicates a superlinear behavior for both T and XX; note that XX increases almost quadratically with respect to X. Such a spectral feature has been frequently observed in our GaAs QDs.[24] We interpret lines X, T, and XX as the neutral exciton, trion, and biexciton radiative recombinations, respectively. This attribution has been confirmed by single-photon correlation experiments[25] showing antibunching (bunching) in the cross correlation X-T (X-XX) measurement. The linewidths are found to vary between tens to hundreds of microelectronvolts reflecting spectral diffusion due to the presence of charged defects in the QD environment.

In Fig. 1(c) we report the orthogonally polarized PL spectra of two different QDs. A mirror-symmetric fine structure is evidenced for X and XX, while no splitting is observed for T, being consistent with the attribution of these lines. The measured FSS Δ defined as the maximum splitting of two orthogonally polarized lines is Δ_X=62 μeV and Δ_{XX} =60 μeV (top panel), and Δ_X=18 μeV and Δ_{XX}=20 μeV (bottom panel) with an error of ±5 μeV. As expected we find that $\Delta_X = \Delta_{XX}$ within the experimental error.

We measured the FSS magnitude for several QDs. The summary is presented in Fig. 2 where the mean value of Δ_X and Δ_{XX} is reported as a function of the X energy. Spanning an interval of ~200 μeV, the FSS Δ clearly monotonically decreases by increasing the X energy. When the X line is at low energy, the FSS is found to be approximately 90 μeV, while for smaller QDs corresponding to emission at 1.89 eV the value of FSS is as small as 18 μeV.

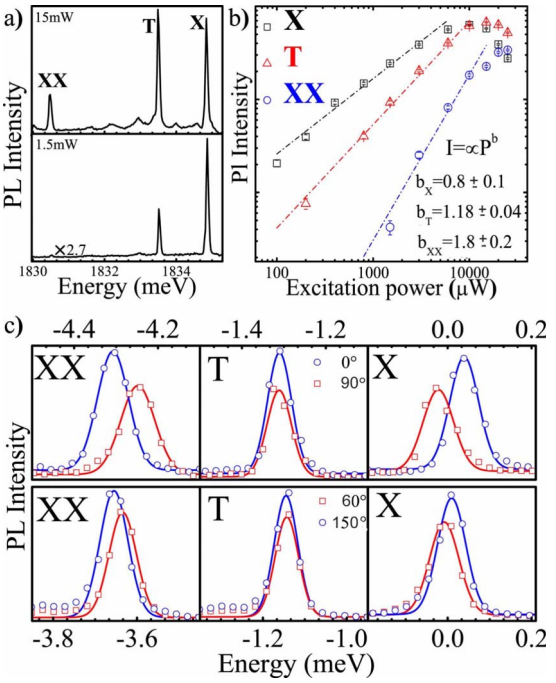

FIG. 1. (Color online) (a) PL spectrum of a single GaAs/AlGaAs QD at two excitation powers. The three lines X, T, and XX are interpreted as neutral exciton, trion, and biexciton emissions. (b) Power dependence of lines X, T, and XX. Dashed lines show fits of the data to a power law, $I \propto P^b$. (c) Polarized PL spectra of two different QDs. The top panel refers to the QD of (a). Blue circles and red squares indicate the spectra of two orthogonal polarizations of which relative angle is shown in the legend. Lines are Gaussian fits. The horizontal axis is shifted by the energy of X (1834.9 and 1875.7 meV for top and bottom panels, respectively).

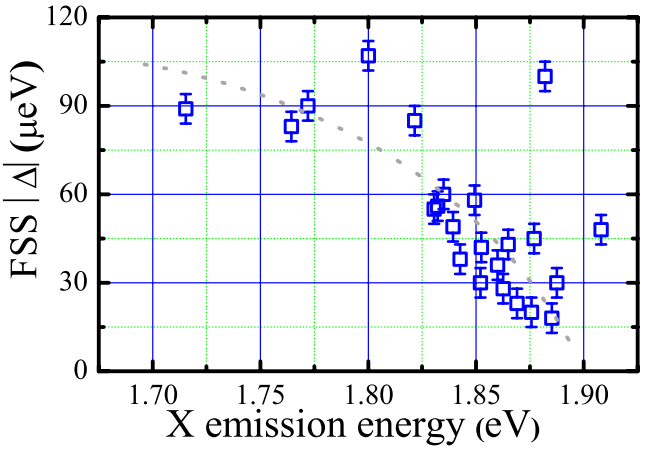

FIG. 2. (Color online) FSS of QDs emitting at different energy. The reported values are the average value of FSS's for X and XX. The dotted line is a guide to eye.

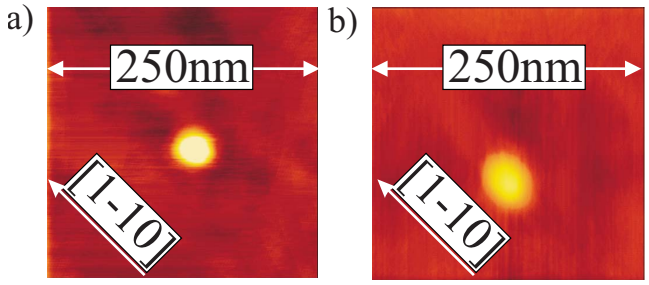

FIG. 3. (Color online) [(a) and (b)] Two-dimensional (2D) representation of the AFM analysis of a "small" QD (h=3.9 nm) and a large QD (h=11.8 nm), respectively.

Comparing the values of FSS of our QDs with those of SK In(Ga)As/GaAs QDs, we observe that both the FSS absolute values and the FSS behavior with the QD size are similar[6] despite the absence of strain-related piezoelectricity in the present case. In fact the relatively high value of FSS in SK-QDs and its reduction in size have been attributed to potential anisotropies caused by the piezoelectric field.[6] Clearly the assignment to piezoelectric effect cannot be applied to the case of strain-free GaAs/AlGaAs QDs, and then our findings raise doubts also in the attribution to piezoelectric effect for SK-QDs. As a matter of fact, among the possible origins of FSS, the effect of the piezoelectric field is the more controversial one.[11,12,26] Due to high value of the strain in the InAs/GaAs SK-QDs, linear and nonlinear terms in the piezoelectric tensor can be of comparable intensities[26] and, depending on the QD shape and composition profiles, the nonlinear terms may effectively balance out the linear ones.[12,26]

To understand the role of geometry in the QD asymmetry, we measured the size and shape of several GaAs/AlGaAs QDs on the same but uncapped sample. Typical atomic force microscope (AFM) images of the lens-shaped DE-QD, in the case of a large and a small QD, are shown in Fig. 3. Extensive AFM measurements show relevant dispersion of size and shape. This analysis points out that QDs with small height h show a small elongation in the $[1\bar{1}0]$ direction, while for large value of h, the QDs show a larger elongation in the same direction [respectively, ~8% and ~20% in the case of graphs (a) and (b) of Fig. 3]. The relative error in the diameter measurement is assumed to be 5%. The observed dependence of the DE-QD shape anisotropy with size was expected considering the kinetics of the DE-QD formation.[19] We expect that the DE-QDs retain their shape also after capping (being realized at extremely low T) and annealing (only interdiffusion at the interfaces is expected). From AFM data a simple and likely explanation of the dependence of the FSS on the QD emission energy emerges. The in-plane asymmetry augments when increasing the QD size leading to a larger splitting of the exciton recombination line along the crystallographic axis $[1\bar{1}0]$ and [110] for large QDs. It is worth noticing that even a relatively small elongation gives rise to FSS of 90 meV, thus showing that geometrical anisotropies play a much larger role than expected from theoretical predictions.[9,11,12]

In order to get further information on the origin of FSS

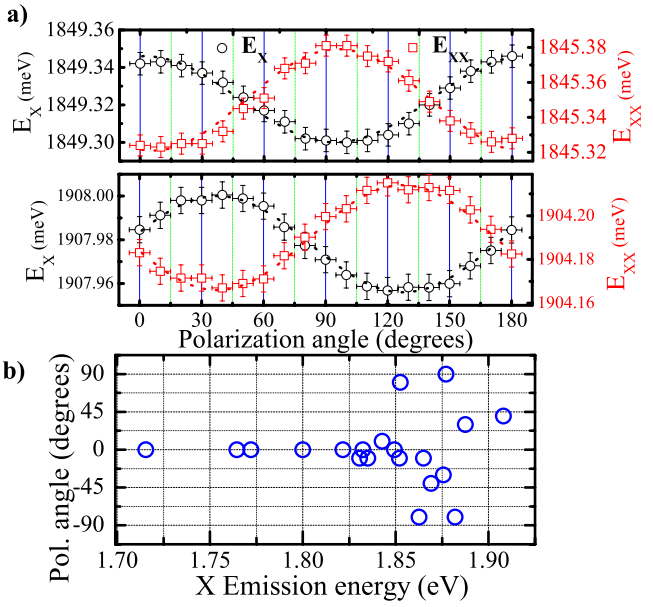

FIG. 4. (Color online) (a) In top and bottom panels circles (squares) show the energy evolution of X (XX) with polarization angle for two different QDs. Left and right vertical axes are referred to X and XX cases, respectively. The PL peak evolutions are fitted with sine functions as evidenced by the dotted lines thus determining amplitude (the FSS) and the phase shifts (the main polarization axis). (b) One polarization axis plotted versus X emission energy.

and to obtain an accurate characterization of the polarization splitting, we measured a series of spectra varying the polarization angle. Following the evolution of the centroid of PL lines with polarization angle, we obtain a clear sinusoidal behavior as shown in Fig. 4(a) for two different QDs. Fitting the experimental data to a sine function, we can measure the amplitude (the FSS shown in Fig. 2) and phase, thus determining the polarization axis of each QD. Not all the QDs show anisotropy along the crystallographic axis [110] (corresponding to the polarization angle at the 90° direction), the measured asymmetry axis of the DE-QDs. On the contrary we found that there are several QDs showing the maximum FSS at angles different from 0° and 90°. The summary of this study is shown in Fig. 4(b) where the plot of the polarization axis angle versus the X emission energy is reported. It clearly follows that large QDs show the expected anisotropy along 90° and that the stronger deviations for the expected polarization axis are mainly associated to smaller QDs. In fact for emission between 1.70 and 1.86 eV the polarization axis is close to 90°, while for higher emission energy, where the FSS is smaller, the deviations from this value become more important. Deviations of the polarization direction of the FSS from the crystallographic axes can be found in SK-QDs,[15,27,28] but this effect has not been attributed to a definite origin.

Therefore our data point out that higher symmetric QDs showing smaller FSS are influenced in their internal symmetry by a factor different from the geometrical anisotropy. This is likely due to the fact that small extrinsic QD potential symmetry reduction effects can be present in addition to intrinsic ones. We believe that a tentative explanation may rely

on the presence of charged defects in the environment nearby the QDs. Besides the fluctuating quantum confinement Stark shift, which is well known to induce spectral diffusion in the emission of the QDs, the averaged Coulomb interaction also defines an asymmetric potential for charges confined in the QDs, therefore leading to a FSS. The random relative position of the charged defects and the QDs then results in a random orientation of the observed polarization axes, thus explaining our findings. This picture agrees with recent findings on the random oscillatory dependence of the FSS on the externally applied electric field.[27] Charging/decharging, under the action of the electric field, of traps located randomly in the vicinity of the QD should cause fluctuations of the axis as well of the magnitude of the QD anisotropic potential. A theoretical discussion of this effect can be found in Ref. 29. Alternatively the FSS may be related to AlGaAs composition fluctuations nearby the QDs.

In conclusion we have reported on detailed measurements of FSS of strain-free DE-QDs. The use of GaAs/AlGaAs DE-QDs allows us to experimentally investigate the role of the geometrical anisotropy in determining the FSS getting rid of strain, piezoelectricity, and segregation processes, which make the correlation between structural data and FSS rather inconsistent. The FSS polarization axis points usually along the [110] direction. FSS values as large as 90 μeV were observed for QDs with relatively small elongation. We find a clear FSS quantum size effect related to the reduction in the QD asymmetry as the size is reduced. As the QD geometry approaches a cylindrical symmetry, the FSS does not go to zero but shows a randomly oriented polarization axis. This observation highlights the role of extrinsic effects so far neglected in the attribution of the FSS in semiconductor QDs.

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