Ultra-small excitonic fine structure splitting in highly symmetric quantum dots on GaAs (001) substrate

Y. H. Huo,1,a) A. Rastelli,1,2,b) and O. G. Schmidt1

1Institute for Integrative Nanosciences, IFW Dresden, Helmholtzstrasse 20, D-01069 Dresden, Germany
2Institute of Semiconductor and Solid State Physics, Johannes Kepler University Linz, Altenbergerstrasse 69, A-4040 Linz, Austria

(Received 7 March 2013; accepted 2 April 2013; published online 16 April 2013)

We prepare symmetry-controlled GaAs/AlGaAs quantum dots (QDs) on (001) GaAs substrates by infilling GaAs into AlGaAs nanoholes. For the most symmetric QDs, we measure an average excitonic fine structure splitting (FSS) of only (3.9 ± 1.8) μeV. The FSS and polarization direction of the two bright excitonic recombination lines directly reflect the degree of the QD symmetry. Since the FSS is comparable to typical homogeneous linewidths of excitonic recombination, these strain-free GaAs/AlGaAs QDs might offer a practical platform to generate entangled photons in future quantum devices. © 2013 AIP Publishing LLC [http://dx.doi.org/10.1063/1.4802088]

The study of the fine structure of neutral excitons confined in single quantum dots (QDs) has been of great interest over the last two decades both in fundamental physics as well as for potential applications. In order to serve as central units in entangled photon-pair sources, QDs should provide a highly symmetric confinement potential, so that anisotropic exchange interaction becomes negligible. Ideally, the two bright exciton states occupying the electronic ground state should be degenerate to such a degree that the excitonic fine structure splitting (FSS) is smaller than the intrinsic recombinations of each exciton transition. However, in real QDs, the symmetry of the confinement potential is generally reduced by irregularities in QD shape, strain, chemical composition, and piezoelectricity. These introduce anisotropic in-plane electron-hole exchange interaction, which lifts the degeneracy of the bright optical active exciton pairs and split them into two orthogonally polarized components—eventually leading to a FSS too large for many envisioned applications.

There are several strategies to minimize FSS in QDs. One strategy includes post-growth treatments, such as annealing, applying magnetic or electric fields, or a combination thereof. These approaches have been successful in bringing FSS to values small enough to generate entangled photons.

From a practical point of view, it would, however, be desirable to rely on reproducible growth protocols, which provide ensembles of quantum dots with sufficiently small FSS. Stimulated by theoretical work and in order to retain high symmetry, quantum dots have been grown on high indexed surfaces. For instance, InGaAs QDs were grown on (111)B GaAs surfaces and a mean FSS of (5.6 ± 0.6) μeV averaging over 22 quantum dots was reported.

The situation on (001) surfaces is more difficult. The most widely studied QD system, i.e., In(Ga)As QDs on GaAs (001) substrates, seems not to be suited for this task because structural fluctuations tend to break the symmetry and cause an unacceptably large spread in FSS. Because intermixing effects can be suppressed, GaAs/AlGaAs QDs constitute an attractive alternative, but their geometry has not been well enough controlled yet as to provide reproducible and vanishing FSS.

In this Letter, we fabricate GaAs/AlGaAs QDs with different shapes on GaAs (001) substrates. Since complicated intermixing effects are absent, we can directly correlate the FSS to the shape symmetry of the QDs. For the most symmetric QDs, we obtain an average FSS of only (3.9 ± 1.8) μeV (20 measured QDs). Assuming a recombination lifetime of about 0.2 ns, this value is comparable to the Fourier-limited excitonic linewidth of single QDs (~3.2 μeV).

The samples were grown by solid state molecular beam epitaxy (MBE). Figures 1(a), 1(c), and 1(e) are atomic force microscopy (AFM) images of Al0.4Ga0.6As surfaces of samples A, B, and C decorated with nanoholes of different symmetries. Sample A was fabricated by etching holes into the GaAs surface at 515 °C using Ga/AlGaAs repeats with a total of 11.4 ML of excess Ga similar to previously developed methods in Refs. 29 and 30. The etched holes were then overgrown with a 7 nm-thick Al0.4Ga0.6As layer serving as a bottom barrier for the QDs. Sample B and C were fabricated by etching holes directly into an Al0.4Ga0.6As surface at 600 °C using Al droplets with a total of 0.5 ML of excess Al deposited at growth rates of 0.05 ML/s and 0.5 ML/s, respectively, similar to Refs. 31 and 32. QDs are formed by diffusing 2 nm GaAs into the nanohole template (by 2 min annealing after deposition) and by capping the filled holes with AlGaAs material as top barrier. Because of weak intermixing between GaAs and AlGaAs, the GaAs QDs assume the shape of the predefined holes. For sample C, a flat surface is recovered after the annealing step following GaAs filling, while for sample A and B, holes with a depth of about 9 nm and 2 nm are still observed. Figures 1(b), 1(d), and 1(f) provide line-scans of different holes along the [110] and [1-10] crystal directions. Going from left to right, the holes gradually evolve into structures of in-plane circular symmetry. The height of all QDs in this study is roughly the same (7–8 nm). The lateral aspect ratios between the hole (and therefore the QD) extensions along the [110] and [1-10] directions are about 1.13 (60 nm/53 nm), 1.05 (50.5 nm/48 nm), and 1.02 (48 nm/47 nm) for

a)y.huo@ifw-dresden.de
b)armando.rastelli@jku.at

© 2013 AIP Publishing LLC
samples A, B, and C, respectively. (This is measured by measuring the widths at a level of 7.5 nm above the bottom of the nanohole). The QD densities of these samples are 0.4 μm⁻², 0.04 μm⁻², and 0.3 μm⁻², respectively, small enough to record the PL spectra of single QDs with a standard micro-photoluminescence (μ-PL) setup.

In order to study the FSS and its relation to the QD shape symmetry, we carried out μ-PL measurements for all three samples. The samples were cooled to 7 K in a liquid helium flow cryostat. PL was excited by a 532 nm laser line. A microscope objective with 0.42 numerical aperture was used to focus the laser light and collect the PL signal. The PL signal was diffracted by a double 1800-grating spectrometer and detected by a liquid-nitrogen-cooled Si CCD detector with a spectral resolution of 20 μeV. The linear polarization of the emitted light was analyzed by a rotatable achromatic λ/2 plate retarder followed by a fixed polarizer in the optical path. For polarization measurements, samples were aligned in such a way that a polarization angle of 0° corresponds to the [110] crystal direction within an error of about 5°.

Figure 2 shows representative color-coded spectra of the neutral exciton emission from single QDs in the three different samples as the λ/2 waveplate rotates continuously from 0° to 180° in 2° steps (the corresponding linear polarization axis rotates by 360°). Figures 2(a), 2(c), and 2(e) are spectra collected from samples A, B, and C, respectively. In order to obtain values for FSS, we fit the different excitonic lines with Lorentzian curves for all polarization angles. The peak energies derived from this fitting procedure are shown in Figs. 2(b), 2(d), and 2(f), respectively. Similar to Ref. 27, we obtain the extremes of the peak energies and, therefore, the FSS and polarization angles by a sine wave fitting when the FSS is small (Figs. 2(c) and 2(e)) and by square wave fitting (Fig. 2(a)) when FSS is large enough to spectrally resolve the two components as two linearly polarized peaks. The fitting values for the full width at half maximum (FWHM) of the peaks of samples A, B, and C are 23 μeV, 25 μeV and 22 μeV, respectively. These values are very close to the spectral resolution of our spectrometer.

From Figs. 2(a) and 2(b), we derive that the neutral exciton spectrum of the QD in sample A is composed of two linearly polarized excitonic peaks separated by a FSS of ~47 μeV with a low energy component polarized along the [110] crystal direction, which is the elongation direction of the QD. With improved symmetry of the QD shape in sample B, the FSS decreases to ~10 μeV (Fig. 2(d)), the pattern becomes wavy (Fig. 2(c)), and the two components of the spectra can no longer be clearly separated. Figure 2(e) presents the spectrum of one QD in sample C, which has further improved symmetry. Here, the FSS further decreases to ~3.5 μeV. The measured correlation between QD FSS and QD shape is qualitatively consistent with the expected increase of the anisotropic exchange interaction with increasing dot elongation. The fact that QDs in sample C have the smallest FSS is nevertheless remarkable, since these QDs are also the narrowest and the exchange interaction is expected to increase with increasing electron-hole overlap (decreasing QD size).

To generalize our observations, we measured the FSS and polarization angle of 20 QDs for each sample. Figures 3(a), 3(c), and 3(e) shows the statistical distribution of the FSS values for samples A, B, and C, and Figs. 3(b), 3(d), and 3(f) are the corresponding polarization angles of the high energy excitonic peak. Different from Ref. 13, which determined the FSS by measuring spectra at two orthogonally oriented polarization directions, we measure spectra by varying the polarization angle between 0° and 360° and define the FSS as the absolute value of the energy difference between the two bright excitonic extremes. This is important as the polarization direction of the excitonic dipoles is not known a priori and may show substantial fluctuations. In this case, projection on two fixed polarization angles leads to...
unreliable measurements of the FSS.\textsuperscript{34} The polarization angle is given by defining the [110] crystal direction as $0^\circ$ and measuring the angular position of the high energy peak with respect to it. As the shape symmetry improves, the average values of FSS monotonically decrease from 49 $\mu$eV to 9.4 $\mu$eV (with standard deviations of 5.8 $\mu$eV, 2.8 $\mu$eV, and 1.8 $\mu$eV), respectively. The polarization directions of the emitted light also change significantly. For QDs in sample A, the high energy component of the bright exciton is firmly aligned along the [1\-10] crystal direction ($90^\circ$ in Fig. 3(b)), which is consistent with the QD shape anisotropy. For QDs in sample B, the high energy peak is still mainly found along the [1\-10] direction, but the angle distribution is broader than in sample A (Fig. 3(d)). Interestingly, for the QDs in sample C, the polarization angles appear to be random (Fig. 3(f)). We attribute the residual small FSS and the random polarization direction to the combination of small shape fluctuations and possibly atomic-scale alloy fluctuations in the barrier regions around the nanoholes, rather than the symmetry properties of the (001) surface.\textsuperscript{15,35-37} In fact, the latter would still favor bright exciton emission polarized along the [1\10] and [1\-10] directions. Figure 3 also shows that neither the FSS nor the polarization direction change significantly with the exciton recombination energy. A very small FSS in Fig. 3(e) over more than 10 meV spectral range is achieved. The QDs also have very small FWHMs, which are $(32 \pm 1) \mu$eV, $(30 \pm 6) \mu$eV, and $(23 \pm 3) \mu$eV for samples A, B, and C, respectively, close to the spectral resolution of our setup.

In summary, we have grown strain-free GaAs/AlGaAs QDs of different shape symmetries on (001) GaAs substrates. The QD morphologies are defined by the initially etched holes, which we have carefully characterized by AFM. Polarization-dependent PL measurements were carried out and analyzed with respect to FSS and polarization angle. A clear correlation between the QD in-plane shape and the excitonic FSS is revealed. We achieve a FSS of $(3.9 \pm 1.8) \mu$eV with random polarization angles for the most symmetric QDs. Since the excitonic linewidth is also very small (mostly resolution limited), these quantum dots might serve as practical sources for entangled photon generation.

This work was financially supported by the BMBF project QuaHL-Rep (Contracts Nos. 01BQ1032 and 01BQ1034) and the European Union Seventh Framework Programme (FP7/2007-2013) under Grant Agreement No. 601126 (HANAS). We are grateful to E. Zallo, R. Engelhard, and D. J. Thurmer for MBE assistance and S. Kumar and R. Trota for assistance with optical measurements.

34 See supplementary material at http://dx.doi.org/10.1063/1.4802088 for a detailed description of the fine structure splitting measurement.