In(Ga)As quantum dots grown on GaAs(111) substrates for entangled photons pairs

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Abstract. Here we present the first successful growth and spectroscopic investigation of In(Ga)As/GaAs (111) quantum dots (QD). These QD were recently predicted as promising emitters of entangled photon pairs due to their intrinsic threefold degenerate symmetry resulting in zero fine structure splitting (FSS). Low spatial densities of the QDs were grown by droplet epitaxy in an MBE system. Emission of truly single photons is proved by correlation measurements. Using spatially-resolved power- and polarization-dependent experiments we discovered the characteristic excitonic luminescence fingerprints from a number of QDs. Excitonic FSS below 10 µeV, our resolution limit, is observed in agreement with our predictions.

1. Introduction
An easy-to-operate entangled-photon-pair source is required for efficient quantum key distribution in future secure data transmission [see e.g.1]. QDs have been proposed [2] and demonstrated [3, 4] as electrically-driven or optically-pumped sources of entangled photon pairs from a biexciton-exciton recombination cascade on condition that the FSS of the bright exciton state is less than the homogeneous linewidth of the transitions.

For In(Ga)As QDs on GaAs (001) even for highly symmetric QDs the lateral symmetry of the confining potential is lowered to C2v. This originates from the lateral anisotropy of the confinement potential for the charge carriers due to elongation of the QD during the growth process, significant piezoelectric effects [5] and atomistic symmetry anisotropy [6]. The post-growth attempts to reduce the FSS to zero by annealing [7], external fields [4, 8], and stress [9] have to be adjusted for every single QD, therefore making it hardly applicable.

In contrast, for (111) substrates the piezoelectric field is directed along the growth direction leading to a confining potential of symmetry C3v. Realistic calculations predicted recently zero FSS for this case [10, 11, 12].

2. Experiment
The self assembled QD samples were grown by solid-source molecular beam epitaxy using a Riber32P system on n-type doped GaAs (111) wafers. To increase the effective decomposition of the
metallorganic precursors and improve the GaAs buffer layer quality we used a miscut of 2° in the [211] direction. A 500 nm-thick layer of the GaAs was followed by a 50 nm Al$_{0.6}$Ga$_{0.4}$As diffusion barrier and then 90 nm of GaAs at 580 °C. The QDs layer was grown in a droplet epitaxy mode [13] at 510 °C. With closed As shutter nominally 1.5 ML of Ga were deposited, and then followed by nominally 2 ML of In to form In droplets on the GaAs surface, which in turn crystallized into InGaAs QDs under the As flux. The dots were then capped with 65 nm of GaAs, 50 nm of Al$_{0.4}$Ga$_{0.6}$As, followed finally by 10 nm GaAs. During the QD growth the wafer rotation was switched off, this allowed areas with low QD densities in the samples.

The samples were investigated with a microphotoluminescence set-up at 15 K with and excited with a frequency doubled Nd:YVO laser (532 nm) and a spot diameter of ~1 µm. The PL signal was dispersed by a triple monochromator and detected with a liquid-nitrogen cooled CCD array, resulting in a spectral resolution of 20 µeV. Using line shape analysis allows determine the energetic position of single line within an accuracy of 10 µeV. For the measurement of the second-order correlation function $g^{(2)}(\Delta t)$ a Hanbury-Brown-Twiss set-up (HBT) with 700 ps time- and 0.3 meV spectral resolution was used.

3. Results and discussion
Switching off the wafer rotation resulted in areas of the different QDs densities. Some of them show spectral sharp luminescence lines stemming from well separated luminescence centres, whereas we also observed areas with spectral broad luminescence. A typical spectrum of sharp emission lines, characteristic for the discrete QD energy levels is shown in Fig.1. The typical measured FWHM of the lines is up to 80 µeV, broadened by spectral diffusion. The spatial resolved investigation of the samples revealed optical active QD densities less than $10^{9}$ cm$^{-2}$ allowing single QD spectroscopy experiments without shadow masks or mesas.

True single photon emission was proven by the measurements of the second-order correlation function with a HBT. The inset of the Fig.1 shows the normalized $g^{(2)}(\Delta t)$ function of a single luminescence line with a measured value of $g^{(2)}(0)=0.3$, limited by the set-up time-resolution. The simulation with accounts for the time resolution [12] leads to true correlation function value $g^{(2)}(0)=0.1$. The cross-talk between the avalanche photo diodes of the HBT leads to the increased value of $g^{(2)}(\Delta t)$ at $\Delta t=\pm5$ns.

**Figure 1.** A typical microphotoluminescence spectrum of a sample showing single and sharp QD lines. The inset displays a second-order correlation measurement and a simulation for the measured in $g^{(2)}(\Delta t)$ (black line) [12]

Since the predicted FSS from In(Ga)As QDs grown on (111) GaAs substrates is zero we faced a challenge of investigation of the electronic origin of the emission lines. For this “novel” system we
found some recurring luminescence line patterns in power-dependent measurements and performed polarization-dependent investigations.

![Excitation power dependence of the photoluminescence. The lines from excitonic complexes are labelled A, B, C](image1)

**Figure 2.** Excitation power dependence of the photoluminescence. The lines from excitonic complexes are labelled A, B, C

For several different QDs we found a similar pattern of excitonic emission behavior. One example is shown in Fig 2. Labeled with A, B and C the lines originate from the same QD and appear simultaneously with increasing excitation power. After saturation of these three lines (at about 100 nW excitation power) they fade out and further lines appear (presumably from multiexcitonic complexes). Moreover A, B and C show almost linear intensity dependence on excitation power. Since this is a characteristic excitonic behavior, we ascribe the lines to stem from single excitonic complexes of a single QD. Line A appears at 1.306 eV, whereas B and C at 1.308 eV with a 300 μeV spectral separation. The intensity ratio is found to be 4:1:2 for A:B:C respectively. Almost the same energetic separation, intensity ratio and power dependence between the lines was observed for five other QD in the same spectral luminescence range as a typical fingerprint of the In(Ga)As QD on the (111) GaAs substrate.

![Polarization dependence of the lines A (a), B and C (b). The dotted lines show the center energy of the lines from a Gaussian fit. (c) Spectra of lines B and C for 90° and 180° polarizer position with a splitting of line B of 40 μeV.](image2)

**Figure 3.** Polarization dependence of the lines A (a), B and C (b). The dotted lines show the center energy of the lines from a Gaussian fit. (c) Spectra of lines B and C for 90° and 180° polarizer position with a splitting of line B of 40 μeV.

Fig. 3 presents the result of the polarization investigations of the excitonic lines (lines A, B and C from Fig.2). Lines A and C (Fig 3a, b) show clear polarization dependence of their intensity with no spectral position change. For an angle of 90° the intensity is 1.5 times larger, than that for 180°. Line B on the contrary shows the same intensity for all polarizer angles, but shifts in energetic position (Fig. 3b) up to 40 μeV with a periodicity of 180°. It is more visible in Fig 3c with spectra for 90° and 180° for B and C. In general, the FSS can be measured on uncharged X and a doubly charged exciton X++ [14]. Therefore lines A and C are possibly trions, whereas line B gives access to the FSS and stems either from X or X++.

For the other five QDs with the same luminescence fingerprint (like that described for Fig.2) we found the FSS of the line B ranging form 30 μeV down or below the determination limit (Fig.4 a, the dot shown on Fig. 2-3 corresponds to number 1). The polarization dependence of the lines B and C for...
the QD number 2 is shown on Fig. 4b. In contrast to the dot shown on Fig. 3 the excitonic line B exhibits an energetic shift on the order of the determination limit (10 µeV).

![Figure 4](image)

**Figure 4.** (a) FSS of excitonic line B, measured for five QDs with the same luminescence fingerprint. (b) Polarization dependence of lines B and C for QD number 2. The dotted lines show the centre energy of the lines from a Gaussian fit. Line B shows splitting below 10 µeV.

We ascribe the nonzero FSS to two effects mainly. The miscut of 2° of the (111) substrate may lead to growth of elongated QDs on the steps on the (111) surface. Besides even the growth of GaAs material exhibits high densities of dislocations. This can hinder the three fold confining potential symmetry and lead to a preferential direction of the strain distribution, similar to (001) substrates.

4. Conclusion

We report on the realization of self-assembled In(Ga)As QDs on (111) GaAs substrates via droplet epitaxy. These novel types of QDs show sharp and intense lines. Densities lower than $10^9$ cm$^{-2}$ were achieved. True single photon emission was proven by antibunching measurements and $g^2(0)<0.3$ was observed. Power-dependent measurements reveal typical excitonic luminescence fingerprint for several QDs and one of the lines gives access to the FSS. The measured FSS is in some cases below our resolution limit of 10 µeV, precondition for entangled photon pairs.

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