Manipulating fine structure splitting in semiconductor quantum dots

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Abstract. We perform million-atom empirical pseudopotential calculations of fine structure splitting (FSS) in realistic InAsP/InP and InGaAs/GaAs QD structures. We find that in alloy In(Ga)As self-assembled QDs (001) there exist a lower bound to the tuning of FSS by applying uniaxial stress due to an anticrossing of the excitonic lines, while in pure InAs self-assembled QDs (001) the FSS goes through zero to negative values due to the crossing of latter. We further predict that semiconductor nanostructures grown along the [111] direction such as self-assembled quantum dots or nanowire quantum dots have a vanishing FSS on grounds of their symmetry and are therefore ideal candidates for the generation of entangled photon pairs.

1. Introduction
At the heart of the emerging fields of quantum teleportation [1], quantum cryptography [2] and quantum computations [3] is the generation and manipulation of entangled photon pairs. Semiconductor quantum dots (QDs) are investigated as a potential source of adequate photons. Indeed, in an idealized QD with degenerate intermediate exciton states the polarization of a photon stemming from a biexciton decay ($|XX\rangle \rightarrow |X\rangle$) is entangled with a photon stemming from the exciton recombination ($|X\rangle \rightarrow |0\rangle$) [4]. However, in real self-assembled semiconductor QDs grown along the [001] direction the existence of non-vanishing energetic difference between the intermediate exciton states, so-called fine structure splitting (FSS), destroys the polarization entanglement between the photons produced in the biexciton cascade process [5]. A schematic illustration of the biexciton cascade process generating a polarization-entangled photon pair in a QD is shown in Figure 1. When FSS is smaller than the radiative linewidth of exciton emission the two single exciton decay paths become indistinguishable and the radiative decay lose the “which path” information [6]. This is the ideal condition for the production of polarization-entangled photon pairs in a semiconductor QD. There is tremendous ongoing efforts to manipulate and reduce FSS below the radiative linewidth in order to generate polarization-entangled photon pairs using self assembled QDs grown along the [001] crystal direction. The ongoing efforts to manipulate and reduce the FSS include electric fields [7], magnetic field [8], strain [9], local annealing techniques [10], spectral filtering [6] and selection of QDs with low FSS [11]. An alternative approach to manipulate the FSS is the use of uniaxial stress [9]. But it is still an open fundamental question whether FSS can be tuned through zero by this mean.

Here, we investigated the manipulation of FSS in realistic and atomistically described self-assembled In(Ga)As/GaAs QD (001) structures by uniaxial stress applied along different crystal directions. The nanostructures grown along [001] and [111] directions with high structural...
symmetries are also investigated for the QDs which intrinsically have zero FSSs. We predict that semiconductor nanostructures grown along the [111] direction such as self-assembled quantum dots or nanowire quantum dots have a vanishing FSS and therefore, are ideal candidates for the generation of entangled photon pairs [12].

2. Results and discussion

The QD structures are constructed with the information for shape, size and composition directly from the experimental reports [13, 14]. We relax the atomic positions of this QD structure to minimize the strain energy using the valence force field (VFF) method [15]. The single particle orbitals and energies of the QD are calculated by using the atomistic pseudopotential approach [16, 15], taking strain, band coupling, coupling between different parts of the Brillouin zone and spin-orbit coupling into account, retaining the atomistically resolved structure. The Coulomb and exchange integrals are calculated from the atomic wave functions [17] and the correlated excitonic states are calculated by the configuration interaction (CI) approach [18]. For the CI calculations we use all possible determinants constructed from the twelve lowest energy electron and twelve lowest hole states (spin included), thus accounting for correlations.

2.1. Manipulating FSS by uniaxial stress

In this work, we consider In(Ga)As/GaAs lens shaped QDs with (i) circular base (base diameter = 25.2 nm, height = 3.5 nm), (ii) elongated ellipsoidal base with long axis along the [110] direction [long (short) axis = 26.5 nm (23.9 nm), height = 3.5 nm] and truncated cone shaped QDs (base diameter = 24 nm, top diameter =18 nm) with heights 3.5 nm and 7 nm. We take different alloy compositions with constant 60% and 80% In content across the QD volume and a graded In composition profile according to the “model 5” reported by Mlinar et al. [19]. In this latter model, the In concentration profile was obtained by a combination of cross-section
STM, photoluminescence and theory. The In concentration varies linearly between the base center of the dot (55% In) the base edge (20% In), the top center (100% In) and the top edge (80% In). We investigate the effect of uniaxial stress along the [110], [110] and [100] crystallographic directions. The uniaxial stress is created by changing the lattice parameters along the crystallographic direction of the applied stress. The stress values are calculated by using the relation, $S = Y[(a_0 - a)/a_0]$ where $Y$ is the Young’s modulus, $a_0$ and $a$ are the equilibrium and distorted lattice parameters, respectively. The Young’s modulus of zinc-blende cubic crystals is anisotropic. We take $Y=85.3$ GPa and 121.3 GPa [20, 21] as the Young’s Modulus of GaAs along the [100] and [110] crystallographic directions, respectively. For each applied stress the atomic positions within the simulation cell, as well as the simulation cell along the [001] growth direction, are relaxed to the minimum strain energy using the VFF method. The unaxial stress applied here is much lower than the breaking limit of 1.0 GPa [9].

### Table 1

| Structure          | Material          | Uniaxial Stress [Crystal Direction] (MPa) | FSS (μeV) |
|--------------------|-------------------|----------------------------------------|-----------|
| Circular Base      |                   |                                        |           |
| Lens-shaped        | In$_{0.6}$Ga$_{0.4}$As | 21.3 [100]                             | 2.86      |
| Lens-shaped        | In$_{0.6}$Ga$_{0.4}$As | -60.7 [110]                             | 2.74      |
| Lens-shaped        | In$_{0.6}$Ga$_{0.4}$As | 60.7 [110]                              | 2.74      |
| Lens-shaped        | In$_{0.6}$Ga$_{0.2}$As | 30.3 [110]                              | 5.25      |
| Lens-shaped        | Graded composition| -91.0 [110]                             | 5.60      |
| Truncated-Cone     | Graded composition| -91.0 [110]                             | 3.58      |
| Long Truncated-Cone| Graded composition| -151.6 [110]                            | 2.15      |
| Elongated Base     |                   |                                        |           |
| Lens-shaped        | In$_{0.6}$Ga$_{0.4}$As | 151.6 [110]                            | 2.98      |

Uniaxial stress on InAs/GaAs QDs give rise to a red-shift of excitonic energy, while uniaxial stress on alloy QDs give rise to a blue-shift of excitonic energies. These results are in agreement with experimental results [9]. The detailed understanding of these shifts in excitonic energies by uniaxial stress will be presented in the subsequent work. Whereas QD’s excitonic transition energy can be tuned substantially, the oscillator strength of transitions remain almost constant over the range of applied stresses [see Figure 2], in contrast to the use of electric fields. This property of the system is useful if the QD’s emission wavelength has to be tuned into resonance with a constant wavelength (light or cavity mode) in order to exploit the Purcell effect, and possibly use the QD as an efficient single photon source [22]. We find that in realistic alloy In(Ga)As self-assembled QDs (001) there exist a lower bound to the tuning of FSS by applying uniaxial stress due to anticrossing of excitonic lines, while in pure InAs self-assembled QDs (001) the FSS goes through zero to negative values due to crossing of excitonic lines [23]. The lower bound of FSS in alloy QDs under uniaxial stress along different crystal directions is given Table 1. We notice that for different alloy structures and stresses the lower bound to the tuning of FSS is about 2 μeV. The structural symmetry of pure InAs QDs [23] allow crossings of excitonic lines in pure QD structures under stress along the [110] or [110] direction.
2.2. Manipulating FSS by growing high symmetric QD structures

On the basis of symmetry analysis using group theoretical arguments we predicted that QD structures with $D_{2d}$ and $C_{3v}$ symmetries intrinsically have zero FSSs. We have confirmed this by empirical pseudopotential calculations on realistic QD structures [12]. The FSS in [111] and [001] grown InAs$_x$P$_{1-x}$/InP QDs with different structural symmetries are given in Table 2. Whereas QDs with $D_{2d}$ symmetry are difficult to grow, the QDs with $C_{3v}$ symmetry in nanowire heterostructures [111] can be easily grown [24]. Thus, we predict that nanowire QDs are the ideal candidates for the generation of entangled photon pairs [12]. We further investigated how robust the results are against structural deformations. The simple in-plane elongation of heterostructure nanowire QDs reduces the $C_{3v}$ symmetry to $C_s$ and leads to nonzero FSSs.

Table 2. FSSs in InAsP/InP QDs with different shape, composition and structural symmetry.

| QD material     | Shape       | Size (d, h) nm | FSS (µeV) |
|-----------------|-------------|---------------|-----------|
| InAs [111]      | disk ($C_{3v}$) | 33.0, 8.0     | 0.2       |
| InAs [111]      | hex. ($C_{3v}$) | 35.0, 8.0     | 0.0       |
| InAs$_{0.25}$P$_{0.75}$ [111] | disk ($C_{3v}$) | 33.0, 8.0     | 0.0       |
| InAs [111]      | disk ($C_{3v}$) | 25.2, 3.5     | 0.1       |
| InAs [111]      | lens ($C_{3v}$) | 25.2, 3.5     | 0.1       |
| InAs [001]      | disk ($D_{2d}$) | 25.2, 3.5     | 12.9      |
| InAs [001]      | disk ($C_{2v}$) | 25.2, 3.5     | 2.8       |
| InAs [001]      | elongated disk ($C_s$) | 10%, 3.5 | 5.4       |
|                 |             | 15%, 3.5      | 8.2       |

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