Lower Bound for the Excitonic Fine Structure Splitting in Self-Assembled Quantum Dots

Ranber Singh and Gabriel Bester

Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Germany

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The excitonic fine structure splitting describes the splitting of the bright excitons as a consequence of the atomistic symmetry of the lattice and the electron-hole exchange interaction. Efforts are underway to eliminate this natural splitting by external constraints in order to use quantum dots in quantum optics. We show by million atom empirical pseudopotential calculations that for realistic structures a lower bound for this splitting exists. We underpin our numerical calculations by an insightful symmetry analysis.

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Because of various potential applications in quantum teleportation [1], quantum cryptography [2], and quantum computations [3], entangled photons have been the subject of recent experimental and theoretical activities. The biexciton cascade process in a semiconductor quantum dot (QD) has been proposed as a source of polarization-entangled photon pairs [4]. The polarization-entangled photons are produced in the cascade, biexciton ($|XX\rangle$) $\rightarrow$ exciton($|X\rangle$) $\rightarrow$ ground state($|0\rangle$), where the polarization of the photon pair is determined by the spin of the intermediate exciton state. In an idealized QD with degenerate intermediate exciton states the polarization of the first photon (stemming from $|XX\rangle \rightarrow |X\rangle$) is entangled with the second photon stemming from ($|X\rangle \rightarrow |0\rangle$) [4]. Thus, ideally, such polarization entanglement proposals require the intermediate exciton state to be degenerate. However, for real self-assembled semiconductor QDs grown along the [001] direction, the intermediate exciton state is split as a consequence of the atomistic anisotropy in the zinc-blende structure, spin-orbit interactions, and the electron-hole exchange interaction [5–8]. The energetic difference between the two bright exciton states, $E_{[1\bar{1}0]}$-$E_{[110]}$, is known as fine structure splitting (FSS). The FSS in self-assembled In(Ga)As/GaAs QDs is typically quite large ($\approx$10 μeV) as compared to the radiative linewidth ($\approx$1 μeV) [9–11] and the exciton decay paths become distinguishable (the ''which path'' information [12] is available), which is detrimental for the production of polarization-entangled photon pairs. Recently, we have shown that [111] grown QDs have a vanishing FSS and represent ideal candidates for the generation of entangled photon pairs [13]. However, there is a tremendous ongoing effort to manipulate and reduce the FSS below the radiative linewidth to generate polarization-entangled photon pairs using the ''standard'' self-assembled QDs grown along the [001] direction. The ongoing efforts to manipulate and reduce FSS include electric fields [14], magnetic field [15], strain [16], local annealing techniques [17], spectral filtering [12], and the selection of QDs with low FSS [18]. The in-plane electric field reduces FSS but the requirements of large fields leads to a reduction in the optical intensity due to field induced reduction in electron-hole overlap [14]. The in-plane magnetic field has also been successfully used to tune the FSS [15]. However, this complicates the experimental setup. The postgrowth annealing is also shown to reduce FSS quite significantly [17]. This process, however, shifts the photon energy quite significantly towards higher values close to the energy of wetting layer emissions [17]. An alternative approach to manipulate the FSS is to apply uniaxial stress [16]. It has been shown that FSS can be tuned substantially by this procedure [16,19]. Seidl *et al.* [16] successfully reduced the FSS from 30 μeV to 15 μeV within the range of their applied stress. They argued that a larger stress or starting with a preannealed QD with FSS of about 10 μeV could tune FSS through zero. But it is still an open fundamental question whether this can be achieved in principle.

In this Letter we report that the FSS in In(Ga)As alloy QDs can be tuned to a certain degree by the application of uniaxial stresses along the [100], [110], and $[1\bar{1}0]$ directions, but cannot be reduced below a certain value. The effect is shown to result from an anticrossing driven by the atomistic nature of the studied structures and their underlying symmetries. We quantify the magnitude of these effects by large-scale empirical pseudopotential calculations for the correlated excitons.

We consider In(Ga)As/GaAs lens shaped QDs with circular base (base diameter = 25.2 nm, height = 3.5 nm), 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 of model 5 as discussed by Mlinar *et al.* [20]. We investigate the effect of uniaxial stress along the $[1\bar{1}0]$, [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, and 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 [21,22] 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 valence force field method [23,24]. The single-particle orbitals and energies of the QD are calculated by using the atomistic pseudopotential approach [23,25,26], 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 as shown in Ref. [7] and the correlated excitonic states are calculated by the configuration interaction approach [27]. For the configuration interaction calculations we use all possible determinants constructed from the 12 lowest energy electron and 12 lowest hole states (spin included), thus accounting for correlations.

We now address the stress dependence of the lowest exciton transition (not its FSS). We find that the uniaxial stress gives rise to a redshift of the excitonic energy in InAs/GaAs QDs, while it leads to a blueshift for alloy In(Ga)As/GaAs QDs. This rather surprising result can be traced back (among the several effects acting on the exciton trend with stress [28]) to the differences in the depth of the confinement potential for electrons in InAs and InGaAs. This causes the increase with applied stress of the single-particle electron states to be more pronounced in alloy In(Ga)As than in InAs QDs, while hole energies increase at the same rate. Under external stress along [110] the energies of bright excitonic states of cylindrically symmetric lens shaped InAs ($In_{0.6}Ga_{0.4}As$) QD decrease (increase) linearly with a slope of about 6.9 μeV/MPa (8.9 μeV/MPa), which is in reasonable agreement with experimental results [16,19]. For the elongated lens shaped $In_{0.6}Ga_{0.4}As$/GaAs QD we find a slope of 8.8 μeV/MPa which is also in good agreement with the experimental results of (11.6 $\pm$ 1.8) μeV/MPa [16]. These results confirm the accuracy of our atomistic empirical pseudopotential approach for the present task. For our range of applied stresses (going beyond the present state-of-the art experiments [16,19]) these slopes represent exciton shifts in the range of 3–5 meV. The oscillator strength remains constant over the range of applied stresses, 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 [29].

Before we present the numerical results of FSS under stress, we analyze the structures and their deformations on grounds of symmetry arguments. In Fig. 1 we show four

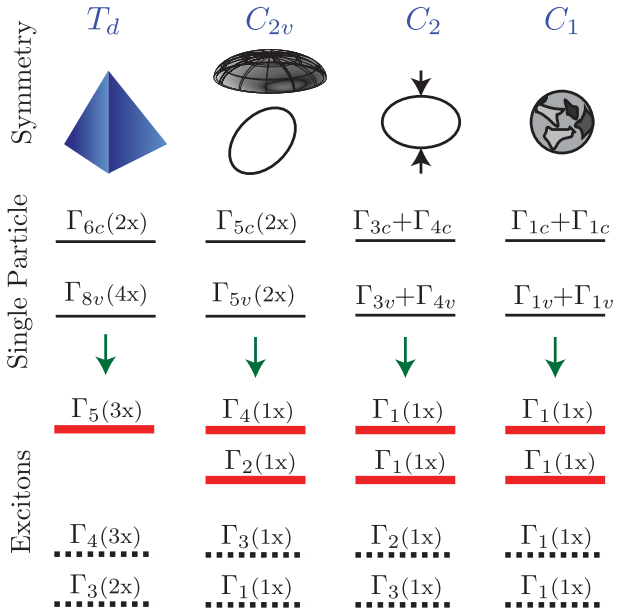

FIG. 1 (color online). Symmetry analysis for four different point groups. The single-particle levels for the highest occupied and the lowest unoccupied states are shown as black lines. The ensuing exciton states with their corresponding irreducible representation are shown as thick (dashed) lines for the bright (dark) states. The pictogram for the C_1 point group illustrates a random alloy structure.

symmetries with the corresponding single-particle highest occupied states (using the letter v) and lowest unoccupied states (with letter c). In the lower part of Fig. 1 the exciton states are created from the direct product of the single-particle states taking qualitatively electron-hole exchange into account. The bright (dark) states are shown as thick (dashed) lines. The symmetry T_d is the symmetry of bulk zinc-blende, or a sphere made of zinc-blende material. From the conduction band Γ_{6c} and the heavy- and light-hole bands Γ_{8v}, the ensuing exciton is split into a bright triplet Γ_5 and dark triplet and doublets with Γ_4 and Γ_3 symmetry. The symmetry of a pure (as opposed to alloyed) lens shaped quantum dot is C_{2v} if the base is circular or elongated along the [110] or $[1\bar{1}0]$ direction. The two bright excitons belong to the irreducible representations Γ_4 and Γ_2. If the QD is elongated along the [100] or [010] direction, or a circular dot is stressed along one of these directions, the symmetry is reduced to C_2. In this case the single-particle states belong to the Γ_3 and Γ_4 representations. The bright excitons both belong to the same irreducible representation Γ_1, while the dark states belong to $\Gamma_{2,3}$. In the case of a dot made of random alloy, e.g., $In_xGa_{1-x}As$, only the identity operation remains and the states all belong to the Γ_1 representation. Consequently all the states are in principle bright by optical selection rules. In practice, however, for typical QD structures the lowest two states are a few orders of magnitude less bright than the upper two states.

From these considerations we expect in a pure lens-shape quantum dot with circular base, or a base elongated along the high symmetry axis [110] or $[1\bar{1}0]$ (C_{2v} symmetry), the two bright states to have the ability to cross, since they belong to different irreducible representations. In the case of a deformation along the [100] or [010] direction (C_2 symmetry) the two bright states, belonging to the Γ_1 representation, should undergo an anticrossing (or "avoided crossing"). This is also true for the case of alloyed quantum dots with C_1 symmetry. The key question determining the ability to tune the FSS is about the magnitude of the interaction leading to the anticrossing. While QDs probably never have perfect C_{2v} symmetry in nature, how good is the assumption of such a symmetry? Recall that close to the entire voluminous QD literature assumes structures with C_{2v} (or higher, sometimes even parabolic 2D potentials with a continuous $C_{\infty v}$ symmetry) symmetry, so the question is legitimate.

To put a quantitative answer to the question raised by the symmetry consideration, we calculate the FSS numerically. In Fig. 2 we plot the energy of the two exciton lines $E_{[110]}$ and $E_{[1\bar{1}0]}$ for a pure InAs QD (a) and for an alloy $In_{0.6}Ga_{0.4}As/GaAs$ QD (b) as a function of stress applied along the [110] direction. Both dots have an elliptical base, elongated along the [110] direction. The stress is shown to influence the FSS significantly. This is the consequence of the deformation of the crystal unit cell and its repercussion on the atomistic Bloch functions. The overall "macroscopic" deformation of the shape of the nanostructure is less then 0.2% at the maximum stress applied and not responsible for the effect. Here the atomistic short-range origin of the FSS is striking. We note that, as pointed out earlier [7], the FSS is mostly unrelated to the overall shape asymmetry, contrary to the persistent misconception that FSS originates from an elongation or shape asymmetry. For the pure QD in Fig. 2(a) the symmetry is C_{2v} and we see a

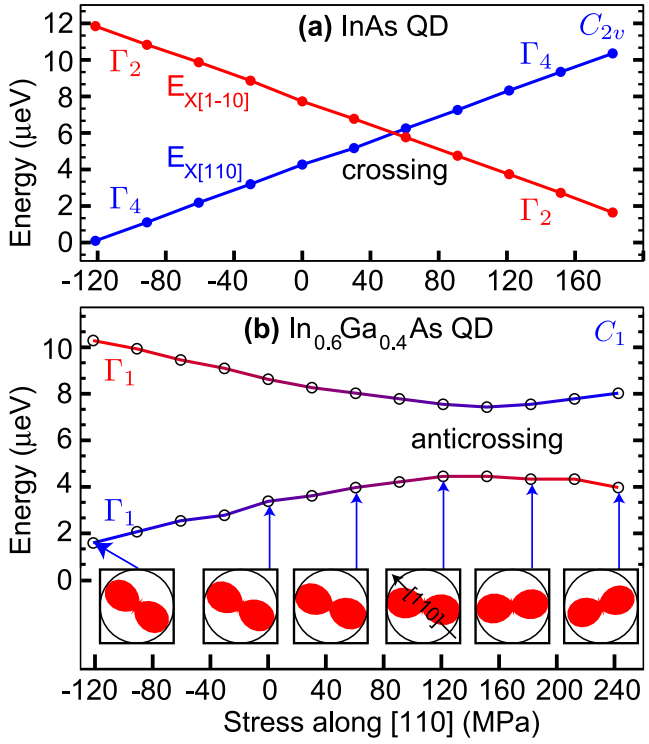

FIG. 2 (color online). Crossing and anticrossing of excitonic lines in In(Ga)As QDs elongated along the [110] direction under uniaxial stress along the [110] direction. The insets in red in Fig. 2(b) show a polar plot in the (001) plane of the oscillator strength of the lowest excitonic transition.

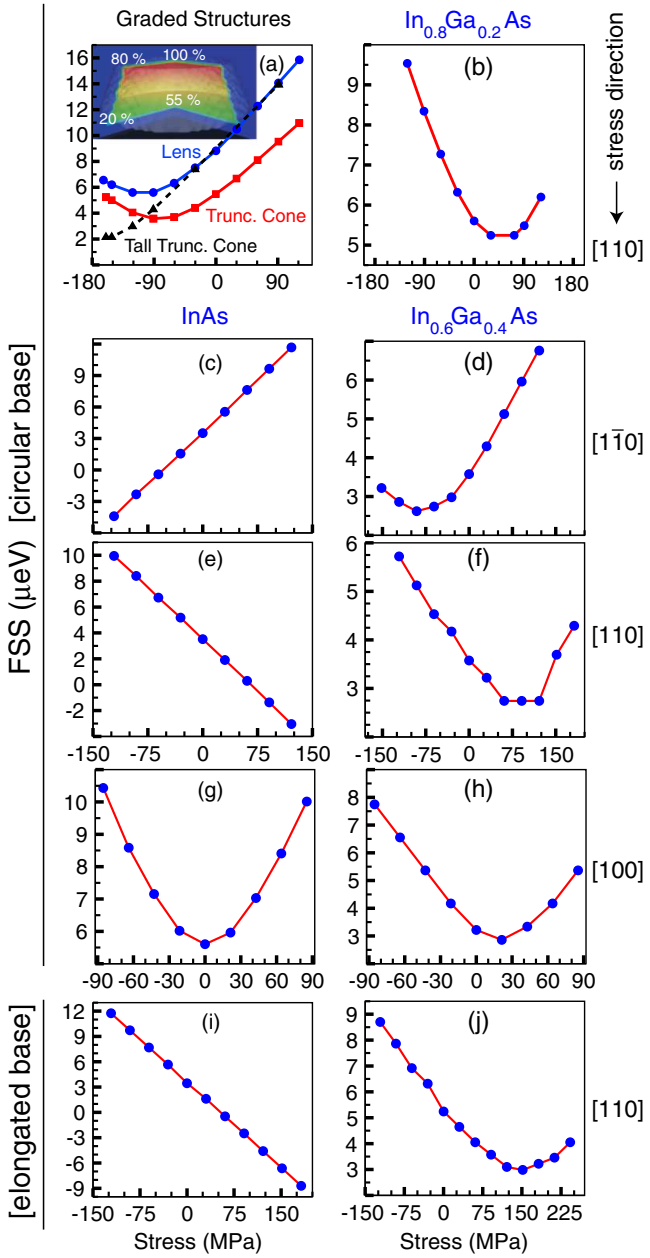

FIG. 3 (color online). Tuning of FSS in cylindrically symmetric (a–h) and elongated (i–j) InAs/GaAs and $In_{0.6}Ga_{0.4}As/GaAs$ QDs by external uniaxial stress along the $[1\bar{1}0]$ (c,d), [110] (a,b,e,f,i,j), [100] (g,h) crystallographic directions.

crossing of the Γ_2 and Γ_4 exciton states in agreement with the symmetry considerations (Fig. 1). The crossing happens at an experimentally attainable pressure of around 50 MPa. For the alloy QD in Fig. 2(b) the symmetry is reduced to C_1 and the exciton states belong to the same Γ_1 representation (Fig. 1) and anticross. As usual, the change of the state's character happens gradually along the anticrossing. In the insets to Fig. 2(b) we show a series of polar plots of the oscillator strength. For a tensile stress of -120 MPa the lowest exciton state is polarized mainly along the [110] direction and rotates to the $[1\bar{1}0]$ direction for a compressive stress of 240 MPa. The pictorial view of a pressure induced rotation of the polarization is therefore valid. The rotation of the polarization with applied stress is also relevant for experimentalists using the polarization dependence of the exciton branches to resolve the FSS from microphotoluminescence.

In Fig. 3 we show the FSSs for InAs and alloy In(Ga)As/GaAs QDs with circular base as a function of the applied stress along the [110] (a,b) $[1\bar{1}0]$ (c,d), [110] (e, f), and [100] (g,h) directions. In Fig. 3(i) and 3(j) the QDs are elongated along the [110] direction and the stress is applied along the elongation. The stresses applied along the [110] and $[1\bar{1}0]$ directions are symmetry conserving and for the pure InAs QDs it leads to crossings. This is pictured in Figs. 3(c), 3(e), and 3(i) by straight lines going through zero. In the case of the alloy dots, the most realistic cases with respect to the experimental situation, we see anticrossings as curves with negative slopes, going through a minimum and ending with positive slopes. We see that for the different structures and stresses applied on the alloy QDs, the lower bound for the FSS is around 3 μeV. For the pure InAs QD with stress along the [100] direction, the lower bound is around 5 μeV. It is worth noticing that for QDs embedded in microcavities the radiation linewidth can be larger than 1 μeV due to the Purcell effect. Under these conditions, FSS of about 2 μeV could still lead to a spectral overlap tolerating the generation of entangled photon pairs.

In summary, we showed that for alloyed self-assembled quantum dots, as usually given in experimental setups, the bright exciton states undergo an anticrossing as a function of the applied external stress. This sets up a lower bound for the FSS, in our cases of $In_{0.6}Ga_{0.4}As/GaAs$ QDs, around 3 μeV. For pure InAs quantum dots and stresses applied along the high symmetry axis [110] or $[1\bar{1}0]$, the states cross and the FSS can be reduced to zero. We give an understanding of these results based on a group theoretical analysis of the exciton manifold. As a general remark we note that the appealing idea to "restore" the atomistic symmetry by external constraints is a misconception. However, stress is shown to lead to significant variations in exciton energies, FSSs, and in the direction of the light polarization, and remains an exciting avenue of research. We suggest that the quest for a convenient and reliable source of entangled photons from [001] grown nanostructures should commence with the creation of structures of high symmetry (e.g., pure QDs) and the use of symmetry conserving external constraints.

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