Fine structure of light-hole excitons in nanowire quantum dots

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Quantum dots with light-hole ground states could find numerous applications including faster quantum bit operations or coherent conversion of photons into electron spins. Typically, however, holes confined in epitaxial quantum dots are of heavy-hole character. I show, by use of atomistic tight-binding theory, that the hole ground state undergoes a transition from heavy holelike to light holelike with increasing height of a nanowire InAs/InP quantum dot. The fine structure of the light-hole exciton consists of a dark ground state and three bright states. Two of the bright states are quasidegenerate and are in-plane polarized, whereas, the third energetically higher bright state is polarized in the perpendicular out-of-plane direction. The light-hole exciton fine structure is robust against alloying.

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Semiconductor nanostructures have attracted great interest for applications in quantum information,[1,2] computing,[3] and cryptography.[4] Epitaxial[5] quantum dots (QDs) typically are relatively flat nanostructures with heights much smaller than their lateral dimensions. The low-aspect ratio and presence of inherent strain in lattice-mismatched systems,[6] such as InAs/GaAs or InAs/InP self-assembled QDs, results in a quasi-two-dimensional confinement[7] and the dominant heavy-hole ($J_z = \pm\frac{3}{2}$) character of the QD ground hole state.[8,9] For a typical self-assembled QD, there are two dark excitonic states corresponding to parallel alignments of electron and hole spins, split by the electron-hole exchange interaction from two energetically higher bright excitonic states of antiparallel spin alignment.[10] Both bright excitonic lines are predominately in-plane polarized, whereas, the out-of-plane component has negligible oscillator strength. For realistic QDs, the bright and dark excitonic states are further split by the "anisotropic electron-hole exchange" interaction.[10] In particular, the energetic difference (typically, 10–100 μeV) between the two bright exciton states is known as the fine-structure splitting (FSS) and has recently gained tremendous attention due to its importance for the QD-based entangled photon generation.[4,11–17] The characteristic exciton fine structure of two dark states and two bright in-plane polarized states is observed for a vast family of epitaxial QD systems including, e.g., self-assembled InAs/GaAs (Ref. 18) and InAs/InP (Ref. 19) QDs or GaAs/AlAs (Ref. 20) QDs grown by a droplet epitaxy. These various and diverse nanostructures share one common feature: a ground hole state of predominantly heavy-hole character.

Nanowire QDs are expected to have similar ground hole state character and excitonic spectra.[21] Moreover, nanowire QDs also have small FSSs[22] due to their high symmetry and have been proposed as natural candidates for efficient entangled photon generation.[22] Additionally, the vapor-liquid-solid (VLS) growth[23] mechanism of nanowire QDs[24] allows for accurate positioning,[25,26] single QD measurements,[27,28] and efficient tailoring of quantum dot dimensions,[25] composition, and even crystal phase.[29,30] The VLS approach allows, in principle, for the growth of QDs of any desired height without the limits inherent to a strain-driven process (Stransky-Krastanov[5] growth). In this paper, I demonstrate using an atomistic tight-binding approach that the ground hole state undergoes a transition from heavy hole to light hole with increasing quantum dot height with a pronounced effect on the exciton fine structure. So far, light-hole ground-state excitons have been found experimentally only once in pyramidal self-assembled QDs.[31] The investigated system revealed broad emission spectra obscuring details of excitonic fine structure. Light-hole states have also been reported theoretically for nontapered (uncapped) nanowire heterostructures.[32] However, uncapped QDs have much stronger lateral confinement due to the effect of surrounding vacuum and very different strain distributions, similar to how free-standing InAs QDs differ from self-assembled QDs.[33] In particular, nontapered systems can relax strains by expanding outward, thereby distorting the surface of the nanowire, a mechanism not possible in high optical quality nanowire QDs capped due to radial growth.[34] More importantly, Ref. 32 reported single-particle spectra, whereas, the current paper focuses on excitonic many-body spectra. This paper is a theoretical calculation of light-hole exciton fine structure for realistic tapered nanowire QDs. Although very interesting from a basic research point of view, using a light hole instead of a heavy hole for the excitonic ground state could be advantageous for quantum information applications including faster quantum bit operations[35] or coherent conversion of photons into electron spins.[36] Recent progress in the reduction in InAs/InP nanowire QDs linewidths[30,37] should enable experimental studies of light-hole excitons in nanowire QDs in the near future.

I consider InAs disk-type nanowire QDs embedded into the cylindrical InP nanowire. The nanowire diameter is 72 nm, corresponding to typical diameters of InP nanowires.[25,30,38] Unless specified otherwise, results presented below refer to [001] grown nanowire. The diameter of the QD is 9.6 nm, whereas, the height of the QDs varies from 1.2 to 18 nm. The small QD diameter results in increased lateral confinement and should, in principle, lead to increased light-hole contribution.

Nanowire quantum dots involve 10^6 atoms, and their electronic properties cannot, at present, be computed using *ab initio* methods, such as, e.g., the GW-Bethe-Salpeter-equation approach.[39,40] Approximate methods, capturing atomistic structure of quantum dots and their surrounding matrices, include tight-binding[41–45] and pseudopotential approaches.[46–48]

Typically, calculations using these approximate approaches involve three steps:[49–53] (a) calculation of equilibrium position of constituent atoms, (b) calculation of quasielectron and quasihole states equivalent to the GW step, and (c) inclusion of final-state interactions by defining an effective Hamiltonian of interacting excited quasiparticles, diagonalized using the configuration-interaction method.

In this paper, I follow the above approach with relaxation of strain (calculation of atomic positions) included via atomistic valence force field theory.[54,55] With the equilibrium atomic positions known, I can attempt to calculate the single-particle electronic structure of the system. I use atomistic tight-binding theory[56] for electron and hole states with an $sp^3d^5s^*$ orbital model[56,57] and parameters set from Ref. 57. This model accounts for nearest-neighbor coupling, coupling between different parts of the Brillouin zone, and spin-orbit effects and includes strain due to lattice mismatch.

The Coulomb and exchange integrals are calculated from the tight-binding eigenfunctions[58,59] as shown in Ref. 58, and the correlated excitonic states are calculated by the configuration-interaction (exact diagonalization) approach.[50,58] The excitonic Hamiltonian does include vertex corrections in the form of electron-hole interaction, but self-energy corrections are included indirectly in the electron and hole energies fitted to the experimental transitions of the bulk material. The contribution to the self-energy correction due to the presence of image charges at the quantum dot/nanowire interface is small primarily because of cancellation between the self-energy interaction of each particle with its own image charges and the excitonic corrections: The exciton as a whole being a neutral excitation, and there is no net charging of the nanowire.[55]

Typically,[10] excitonic fine structure is calculated using a basis formed by the lowest electron and hole states only. In this paper, however, for the configuration-interaction calculations, I use all possible determinants constructed from the 12 lowest-energy electron and 18 lowest hole states (including spin), thus, accounting for correlations[22] and guaranteeing convergence of dark-bright exciton splitting within 0.02 meV.

An atomistic model is essential for describing excitonic fine-structure splitting of nanowire QDs.[17,21,22] The size of the computational domain for strain calculation, including the quantum dot and the surrounding nanowire, reaches 15×10^6 atoms, whereas, by using a multiscale[44,60] approach, the number of atoms for the single-particle calculation is reduced to 0.5×10^6 atoms. Results presented below are qualitatively the same for [001] and [111] grown nanowires, thus, unless specified otherwise, I present results for [001] case only.

Figure 1 shows hydrostatic and biaxial strain profiles calculated using the atomistic valence force field method for two nanowire QDs with the same diameter but substantially different heights. The magnitude of the hydrostatic strain is related to the InAs/InP lattice mismatch ($\approx$3.5%) and, thus, to a large degree, is unaffected by a QD size. On the contrary, both the magnitude and the character of the biaxial strain undergo a substantial modification with increasing QD height. For small nanowire QD height, the biaxial strain distribution resembles that of a typical self-assembled QD: There is a large positive biaxial strain in the QD area, and the biaxial strain changes sign at the QD-surrounding matrix interface.[6] In the

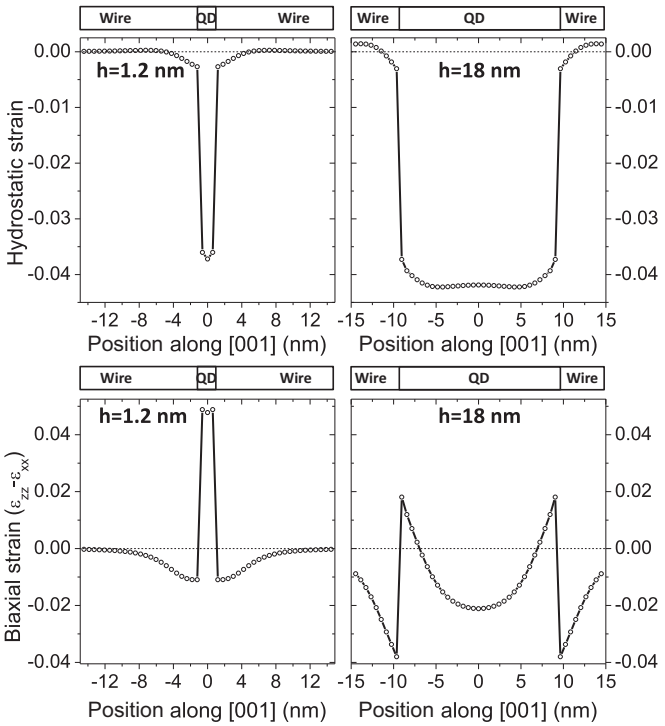

FIG. 1. Hydrostatic (upper row) and biaxial (lower row) strain profiles calculated for InAs/InP nanowire quantum dots with the same diameter ($d = 9.6$ nm) but different heights ($h = 1.2$,18 nm). The horizontal dashed line indicates zero strain.

center of the flat QD, the character of strain can be understood by utilizing a simple picture of a pseudomorphically strained layer.[61] At large QD heights, the center of the QD witnesses a significant negative biaxial strain. The strain in the center of the tall high-aspect ratio nanowire QD resembles that of a pseudomorphically strained one-dimensional cylinder[61,62] rather than a two-dimensional layer.

To illustrate the effect of strain on confining potentials, Fig. 2 shows the local band structure calculated from strained atomic positions using the Bir-Pikus model[6,63] for two QDs with the same diameter but substantially different heights. For a flat ($h = 1.2$-nm) QD system, the large positive biaxial

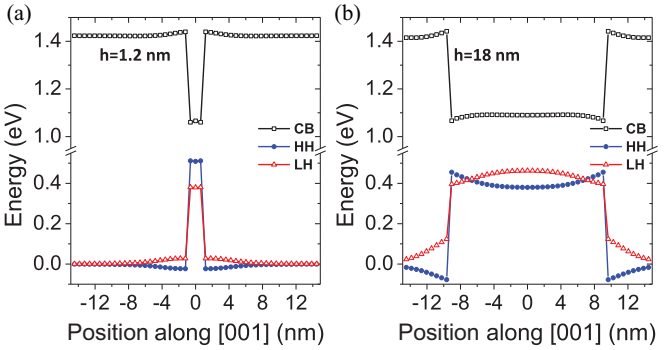

FIG. 2. (Color online) Local band structure (confining potentials) calculated using the Bir-Pikus model for InAs/InP nanowire quantum dots with the same diameter ($d = 9.6$ nm) but different heights: (a) $h = 1.2$ nm and (b) 18 nm. Squares: conduction band (CB); circles: heavy-hole band (HH); triangles: light-hole band (LH).

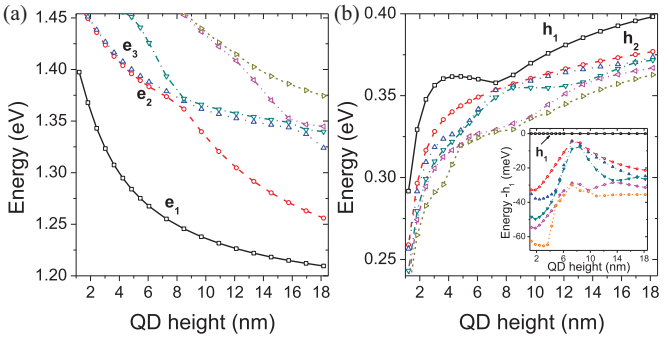

FIG. 3. (Color online) Evolution of several lowest single-particle (a) electron and (b) hole levels as a function of InAs/InP nanowire QD height. The inset shows details of the hole spectra calculated with respect to the ground hole h_1 state energy.

strain leads to large 100-meV light-hole/heavy-hole splitting [Fig. 2(a)] and, in effect, the common ground hole state dominated by heavy-hole contribution. For a tall QD system, the reversal of biaxial strain in the center of the QD leads to the reversal of the order of the light hole and heavy hole as shown in Fig. 2(b). This reversal, combined with the reduced vertical confinement, should lead to a pronounced change in the angular momenta character of the QD ground hole state. This assertion will be verified by atomistic tight-binding calculations in the following part of this paper.

Figure 3 shows the calculated single-particle energies of several: (a) lowest electron and (b) hole states as a function of InAs/InP nanowire QD height. With increasing QD height, the ground electron state undergoes a monotonous energetic redshift due to lowering of confinement. The evolution of the ground hole state energy [Fig. 3(b)] is nonmonotonous with respect to QD height with a noticeable kink at $h \approx 8$ nm. At that height, no clear shell structure in the energy spectra can be observed [inset in Fig. 3(b)]. Further increase in QD height reverses the trend and leads to a noticeable spacing of the ground hole state from the rest of the spectrum.

Figure 4 shows the probability charge densities of several lowest electron and hole states for two selected QD heights. For small QD height [Fig. 4(a)], there are only three confined electron states due to large confinement in the vertical direction. The ground electron state has s-like symmetry, and there are two nearly degenerate p-like states, each having a node at the QD center: as typical for an epitaxial QD.[7] For tall ($h = 18$-nm) high-aspect ratio QDs [Fig. 4(b)], one observes a peculiar structure of electron states, e.g., three lowest electron states have approximate in-plane s-like symmetry and an increasing number of nodal planes along the growth axis. In the vertical direction, the ground electron state is node free, the first excited electron state has one nodal plane (p_z-like symmetry), and the second excited state has two nodal planes. Higher-lying electron states have mixed nodal structures with nodes both in the vertical and in the lateral plane.

Let us now analyze wave-function symmetries of hole states. For flat QDs, the strong positive biaxial strain decouples heavy holes from light holes, leading to a single band character of hole levels. One observes a characteristic shell structure of states with well-defined nodes [Fig. 4(a)]. The ground hole state is s-like, followed by two closely spaced p-like

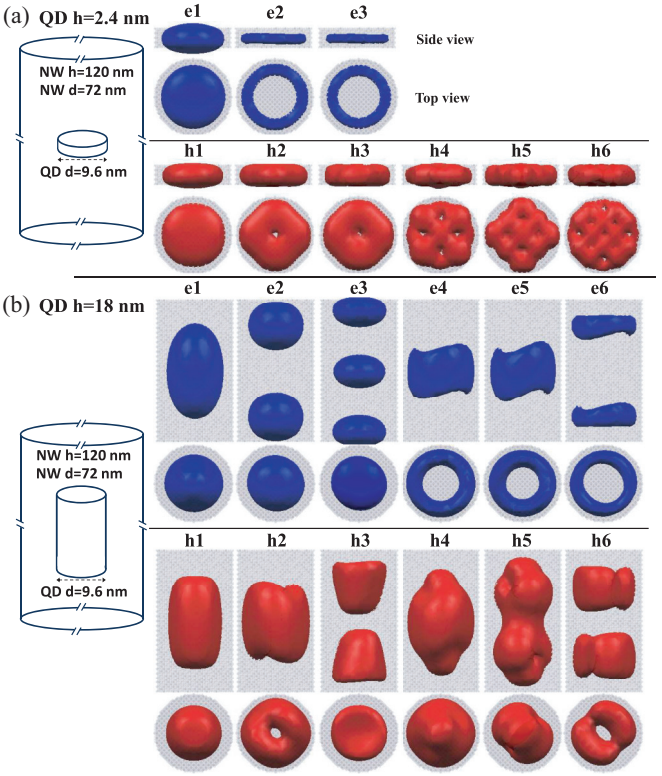

FIG. 4. (Color online) Single-particle electron and hole probability density isosurfaces for InAs/InP nanowire quantum dots with the same diameter ($d = 9.6$ nm) but different heights: (a) $h = 2.4$ nm and (b) $h = 18$ nm. On the left, site schematics of those quantum dots are shown. Gray background points correspond to quantum dot atoms (marking quantum dot shape and size), whereas, nanowire atoms are not shown for clarity.

states, then followed by three closely spaced d-like states. For $h > 14$ nm [Fig. 4(b)], the hole ground state has well-defined ellipsoidal s-like (nodeless) symmetry of the wave-function envelope and is well localized within the QD center, whereas, higher-lying hole states show complicated nodal structures.

The ground hole states of flat and tall QDs have qualitatively similar (s-like/nodeless) characters.[64] However, the microscopic (Bloch) part of the hole wave function is different for these limiting cases. Figure 5 shows that, with the increasing QD height, the ground hole state character varies from heavy

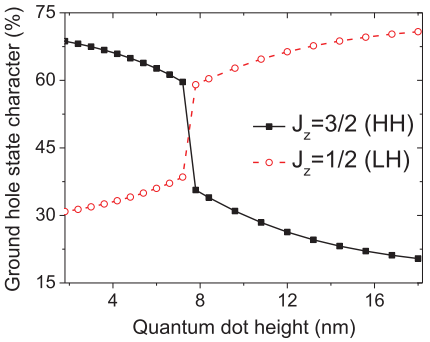

FIG. 5. (Color online) Evolution of the hole character of the ground hole state h_1 in the InAs/InP nanowire quantum dot with $d = 9.6$-nm base diameter as a function of QD height.

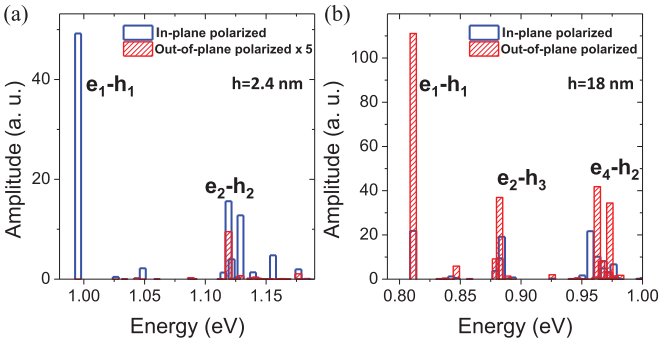

FIG. 6. (Color online) Single-particle absorption spectra calculated for light polarized in plane (blue/plain) and out of plane (red/patterned) for InAs/InP nanowire quantum dots with the same diameter ($d = 9.6$ nm) but different heights: (a) $h = 2.4$ nm and (b) $h = 18$ nm.

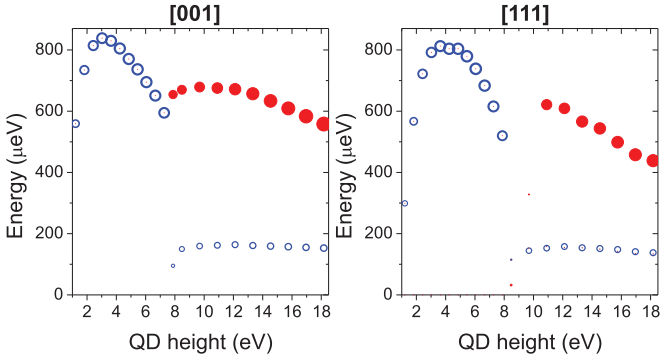

FIG. 7. (Color online) Excitonic fine structure for InAs/InP nanowire quantum dots with $d = 9.6$-nm base diameter as a function of QD height and nanowire substrate orientation ([001] and [111]). The energies are presented by setting the energy of the lowest, dark, exciton states to zero. The blue/empty circles represent the transitions polarized in the growth plane, whereas, the red/full circles represent transitions polarized along the growth direction. The size of the circles is proportional to the oscillator strengths.

holelike to light holelike. The level crossing occurs at a QD height comparable to its diameter with aspect ratio ≈ 0.8. One can conclude this section as follows: The dominant heavy-hole contribution in typical semiconductor QDs originates from: (a) strong confinement in the growth direction and (b) pronounced biaxial strain separating further heavy holes from light holes. With increasing QD height, both contributions change their characters. In particular, the confinement in growth direction is significantly reduced due to increased QD height, and, as shown earlier, the biaxial strain reverses sign and, thus, in effect, the light hole dominates the ground state. The above result should be universal for most QDs, however, nanowire QDs (thanks to the VLS approach) allow, in principle, for the growth of QDs of any desired height without the limits intrinsic to other methods. A similar crossover has been shown theoretically for uncapped nanowire heterostructures,[32] although the strain distribution and confining potential are very different from tapered (capped) nanowire QDs.

The change in the hole wave-function character has a pronounced effect on the optical spectra of the QD. Figure 6 shows the joint optical density of states (single-particle absorption spectra) calculated for nanowire QDs with different heights. For the flat system, one observes the characteristic transitions between electron and hole states of the same envelope symmetry (s-s, p-p) with a pronounced in-plane (x/y) polarization and a negligible out-of-plane (z) component. For the tall system, groups of peaks with complicated selection rules are observed. Most importantly, the dominant contribution comes from the out-of-plane (z-polarized) contribution.

Finally, Fig. 7 shows the calculated excitonic fine structure as a function of QD height. Energies are relative to the lowest exciton state, which is a dark state in all considered cases. Blue symbols show in-plane polarization, and red symbols show out-of-plane polarization. Fine-structure spectra calculated for a nanowire grown on a [111] substrate are qualitatively similar to the [001] substrate case. At small QD heights, the two bright states are in-plane polarized and degenerate due to high nanowire QD symmetry.[22] The bright-dark splitting is about 0.5 meV. Dark states are nearly degenerate (splitting $\ll 0.1$ μeV) for the [001] nanowire QDs of D_{2d} symmetry and are exactly degenerate for the [111] nanowires with C_{3v} QD symmetry.

With increasing QD dot height, the situation changes dramatically with one bright state out-of-plane polarized 500 meV above the lowest dark state and two degenerate bright and in-plane polarized states 130 μeV above the lowest dark state. These results are in qualitative agreement with the recent group-theoretical paper that predicted transitions with these polarizations.[65] It should be noted that, at the QD height comparable to QD diameter, the ground hole state has strongly light-hole/heavy-hole mixed character, and the overall oscillator strength (especially for the [111] QD) is significantly reduced, whereas, the fine structure is far from trivial.

Alloying may affect QD spectra and should be accounted for.[66] Additional calculations have been performed for the alloyed $InAs_{0.25}P_{0.75}/InP$ nanowire QDs.[37,67] Alloying significantly reduces spacings between dark and bright states and leads to the splitting of bright in-plane polarized states (typically 15–40 μeV) due to disorder and lowering of QD symmetry. Additionally, the bright out-of-plane polarized line gets a small ($< 1\%$) contribution of in-plane polarization and vice versa. Nonetheless, the characteristic structure of light-hole exciton spectra of tall nanowire QDs stays, to a large degree, unaffected by alloying.

In conclusion, the variation in the nanowire QD height can be used to engineer quantum dots and to switch the excitonic ground state from heavy hole to light hole. High-aspect ratio quantum dots have electron and hole states of different symmetries and emission spectra significantly different from typical flat epitaxial quantum dots. The light-hole exciton has one dark ground state, two in-plane polarized bright states, and an out-of-plane polarized bright state of dominant oscillator strength, consistent with group-theoretical predictions. The effect should be experimentally observed for both nonalloyed and alloyed nanowire QDs grown either on the [001] or on the [111] substrate.

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