Atomistic Simulations of Electronic Structure in Realistically-Sized Wurtzite InN/GaN Quantum Dots having Different Geometries

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ABSTRACT

Built-in/internal fields in Wurtzite quantum dots originate mainly from: (1) the fundamental crystal atomicity and the interfaces between two dissimilar materials, (2) the strain relaxation, (3) the piezoelectricity, and (4) the spontaneous polarization/pyroelectricity. In this paper, using the atomistic NEMO 3-D simulator, we study the influence of these four competing internal fields on the electronic structure of InN/GaN quantum dots having three different geometries, namely, box (rhombohedron), dome, and pyramid. It has been shown that the presence of the internal fields leads to a global shift in the conduction band one-particle energy states, anisotropy and twofold degeneracy in the P level, and formation of mixed excited bound states.

Keywords: quantum dots, strain, piezoelectricity, tight-binding, pyroelectricity, NEMO 3-D

1 INTRODUCTION

The group-III nitride material system has been the subject of intense experimental and theoretical research due mainly to their wide range of emission frequencies and potential for applications in various optoelectronic, solid-state lighting, and high-mobility electronic devices [1]. In the last decade, GaN and its related alloys especially InGaN have been viewed as the *most promising* materials for the applications of blue and green light emitting optical devices. Currently, growth of InN is carried out mainly on as-grown epitaxial GaN layers. Since the heteroepitaxy of InN on GaN involves a lattice mismatch of up to11%, a form of Stranski-Krastanov mode can be used for growing InN quantum dots (QDs) on GaN by molecular beam epitaxy (MBE) [2]. Recent studies have shown that the strain between InN and GaN can be relieved by misfit dislocations at the hetero-interface after the deposition of the first few InN bilayers and before the formation of InN islands. Relaxed InN islands with controllable size and density can be formed by changing the growth parameters (such as temperature) in either MBE or metalorganic chemical vapor deposition (MOCVD).

Theoretical calculation of the electronic structure is the first step towards the interpretation and understanding of experimental data and reliable device design. This is essentially true for nanoscale QDs where both the *atomistic* granularity of the underlying materials and the quantum mechanical nature of charge carriers play critical role in determining the overall device performance. Self-assembled quantum dots (QDs), formed spontaneously by Stranski-Krastanov growth mode, have been shown to demonstrate polarized transitions in quantized electron (hole) states and non-degeneracy in the first excited state in various spectroscopic analyses [3]. These observations suggest the existence of certain *symmetry lowering mechanisms* in these low dimensional QDs. Recent experiments with $In_xGa_{1-x}N$ thin layers have also revealed photoluminescence from QD-like zero-dimensional localization centers [4] and their dependence on the In content. The optical properties of these QDs, which to a large extent are determined by an intricate interplay between the structural and the electronic properties, have not yet been fully assessed experimentally and demand comprehensive theoretical investigations.

In this paper, we study the electronic properties of Wurtzite InN/GaN quantum dots having *three* different geometries, namely, box (rhombohedron), dome, and pyramid. The main objectives are two-fold—(1) to explore the nature and the role of crystal atomicity, strain-field, piezoelectric, and pyroelectric potentials in determining the energy spectrum and the wavefunctions, and (2) to address the shift in the energy state, the symmetry-lowering and the non-degeneracy in the first excited state, and the strong band-mixing in the overall conduction band electronic states. Efforts are made to demonstrate the importance of full 3-D atomistic material representation and the need for using *realistically-extended* substrate and cap layers (multimillion atom modeling) in the study of electronic structure of these reduced-dimensional QDs.

2 TOOLS AND MODELS

It is clear that, at nanoscale, modeling approaches based on a *continuum* representation (such as effective mass, and $k \cdot p$ [4]) are clearly invalid. On the other side, various *ab initio* atomistic materials science methods (fundamental many-electron correlated methods based on perturbation theory, quantum Monte Carlo method, or GW approach) offer intellectual appeal, but can only predict masses and bandgaps for very small systems (around 100 atoms). Thus, for quantum dot simulations, the simulation domain re-

quiring multimillion atoms prevent the use of *ab initio* methods. Empirical methods (*Pseudopotentials* [3] and *Tight Binding* [5]), which eliminate enough unnecessary details of core electrons, but are finely tuned to describe the atomistically dependent behavior of valence and conduction electrons, are attractive in realistically-sized nanodevice simulations. Tight-binding is a local basis representation, which naturally deals with finite device sizes, alloy-disorder and hetero-interfaces and it results in very sparse matrices. The requirements of storage and processor communication are therefore minimal compared to pseudopotentials and implementations perform extremely well on inexpensive Linux clusters.

This study has been performed through atomistic simulations using the Nanoelectronic Modeling tool NEMO 3-D. Description of this package can be found in Ref. [6]. Based on the atomistic valence-force field (VFF) method for the calculations of strain fields and a variety of tight-binding models (s, sp^3s^*, $sp^3d^5s^*$) optimized with genetic algorithms to match experimental and theoretical electronic structure data, NEMO 3-D enables the computation of atomistic (non-linear) strain and piezoelectric field for over 64 million atoms and of electronic structure for over 52 million atoms, corresponding to volumes of $(110\text{nm})^3$and $(101\text{nm})^3$, respectively. Excellent parallel scaling up to 8192 cores has been demonstrated. We are not aware of any other semiconductor device simulation code that can simulate such large number of atoms. NEMO 3-D includes spin in its fundamental atomistic tight binding representation. Effects of interaction with external electromagnetic fields are also included. Recent work on multi-million atom simulations demonstrates the capability of NEMO 3-D to model a large variety of relevant, realistically-sized nanoelectronic devices [6] including bulk materials, quantum dots, quantum wires, quantum wells and nanocrystals.

The overall polarization **P** in a typical Wurtzite semiconductor is given by $\mathbf{P} = \mathbf{P}_{PZ} + \mathbf{P}_{SP}$, where $\mathbf{P}_{PZ}$ is the strain-induced piezoelectric polarization and $\mathbf{P}_{SP}$ is the spontaneous polarization (pyroelectricity). The latter is dependent on the material, and has nonzero component only along the [0001]/growth direction. The piezoelectric polarization $\mathbf{P}_{PZ}$ is obtained from both the diagonal and *shear* components of the stress fields. The polarization constants used in this study are taken from Ref. [4]. Lattice mismatch parameters and strain tensors are taken from Ref. [5], and the small thermal strain contribution is neglected. The polarization induced charge density is derived by taking divergence of the polarization. To do this, we divide the simulation domain into cells by rectangular meshes. Each cell contains four cations. The polarization of each *grid* is computed by taking an average of *atomic* (cations) polarization within each cell. A finite difference approach is then used to calculate the charge density by taking divergence of the grid polarization. Finally, the induced potential is determined by the solution of the 3-D Poisson equation on an atomistic grid.

3 SIMULATION RESULTS

Figure 1 shows the simulated quantum dots having box, dome, and pyramid geometries. The InN QDs grown in the [0001] direction and embedded in a GaN substrate used in this study have diameter/base length, d~10.1 nm and height, h~5.6 nm, and are positioned on an InN wetting layer of one atomic-layer thickness. The simulation of strain is carried out in the large computational box, while the electronic structure computation is restricted to the smaller inner domain. All the strain simulations fix the atom positions on the bottom plane to the GaN lattice constant, assume periodic boundary conditions in the lateral dimensions, and open boundary conditions on the top surface. The inner electronic box assumes a closed boundary condition with passivated dangling bonds.

Figure 2 shows the first four conduction band wavefunctions for each of the quantum dots *without* any strain relaxation. Both the InN dot and the GaN barrier assume the lattice positions of perfect Wurtzite GaN. From the P level wavefunctions, it is clear that the fundamental crystal atomicity and the interfaces (between the dot material InN and the barrier material GaN) lower the geometric shape symmetry even in the absence of strain. Therefore, the interface plane cannot be treated as a reflection plane and creates a short-range interfacial potential. However, it is important to note that the magnitude of the split/non-degeneracy (defined as $\Delta P = E_{010}–E_{100}$) in the P level is largest in case of a dome (~45.294 meV) and minimum in a box (~1.476 meV). Also, the anisotropy in the P level assumes different *orientations*—for box and pyramid dots, the first P state is oriented along the [010] direction and the second along [100] direction, while the converse occurs in a dome.

Next, we introduce atomistic strain relaxation in our calculations using the VFF method with the Keating Potential. In this approach, the total elastic energy of the sample is computed as a sum of bond-stretching and bond-bending contributions from each atom. The equilibrium atomic positions are found by minimizing the total elastic energy of the system. However, piezoelectricity is neglected in this step. The total elastic energy in the VFF approach has only one global minimum, and its functional form in atomic coordinates is quartic. The conjugate gradient minimization algorithm in this case is well-behaved and stable. Strain modifies the effective confinement volume in the device, distorts the atomic bonds in length and angles, and hence modulates the confined states. From our calculations, atomistic strain was found to be long-ranged and penetrates deep (~20 nm) into both the substrate and the cap layers stressing the need for using *realistically-extended* substrate and cap layers (multimillion atom modeling) in the study of electronic structure of these reduced-dimensional QDs. *Figure 3* shows the wavefunction distributions for the first 4 (four) conduction band electronic states in a 2-D projection. Noticeable is the pronounced optical anisotropy and non-degeneracy in the P level. It is also important to note

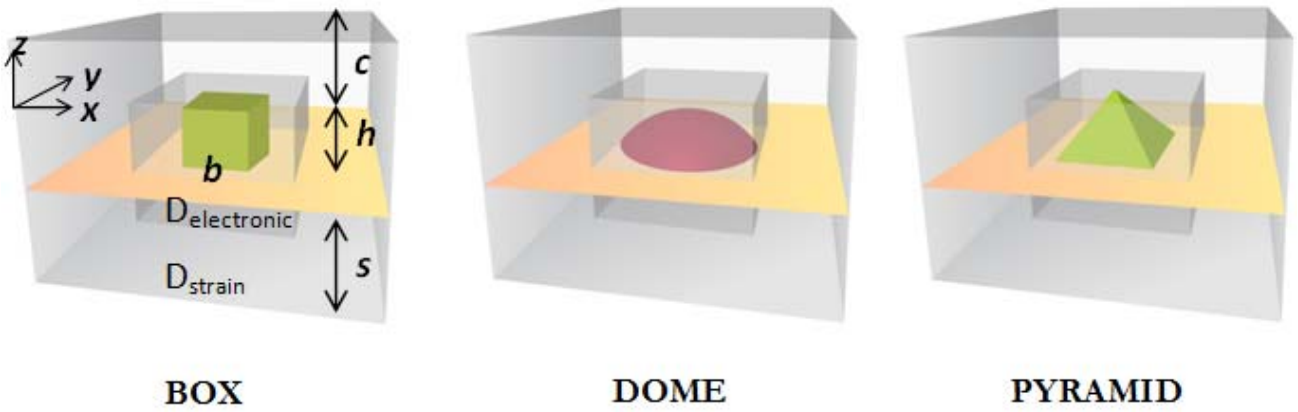

Figure 1. Simulated InN/GaN quantum dots on a thin (one atomic layer) InN wetting layer. Two major computational domains are also shown. D_{elec}: central smaller domain for electronic structure (quantum) calculation, and D_{strain}: outer domain for strain calculation. In the figure: *s* is the substrate height, *c* is the cap layer thickness, *h* is the dot height, and *d* is the dot diameter/base length as appropriate.

that strain introduces uniform orientational pressure (adds negative potential) in all three quantum dots. *Figure 3* also reveals the split/non-degeneracy in the P level associated with each of the quantum dots. This value is found to be largest in a pyramid and minimum in a dome.

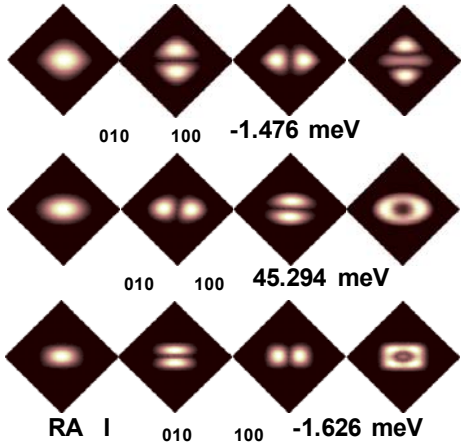

Figure 2. First four conduction band wavefunctions due to fundamental crystal and interfacial symmetry. Noticeable are the split (non-degeneracy) and the anisotropy in the P level.

In pseudomorphically grown heterostructures, the presence of non-zero atomistic stress tensors results in a deformation in the crystal lattice and leads to a combination of piezoelectric and pyroelectric field, which has been incorporated in the Hamiltonian as an external potential (within a non-selfconsistent approximation). The resulting potential distributions along the [0001] (growth) direction are shown in *Figure 4* for all three quantum dots. One can see that the potential (both the piezoelectric and pyroelectric), in accordance with the dot volume, has the largest

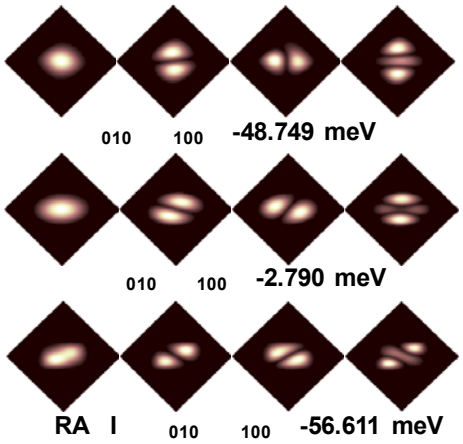

Figure 3. First 4 (four) electronic wavefunctions and split in the P levels including strain relaxation. Number of atoms simulated: 1.78 millions (strain domain), 0.8 million (electronic domain).

magnitude in a box, and is minimum in a pyramid. The pyroelectric potential is significantly larger (~5 times) than the piezoelectric counterpart and tends to oppose the latter. *Figure 5* shows the first 4 (four) conduction band wavefunctions for all three quantum dots including the strain relaxation and the piezoelectric potential. The piezoelectric potential introduces a global shift in the energy spectrum and *opposes* the strain induced field (without any significant modifications in the wavefunction orientations) in the box and pyramid dots. *Figure 6* shows the first 4 (four) conduction band wavefunctions for all three quantum dots including *all four types* of internal fields, namely, interface, strain, piezoelectricity, and pyroelectricity in the calculations. The pyroelectric potential was found to be large enough to create band mixing and strong wavefunction anisotropy in the conduction band energy landscape.

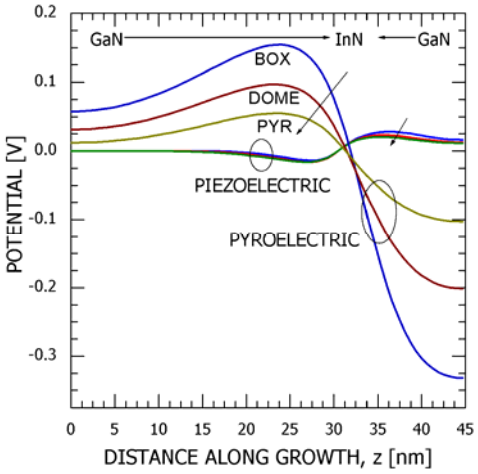

Figure 4. Induced piezoelectric potential along the *z* (growth) direction. Note the varying spread/penetration of the potential in the surrounding material matrix.

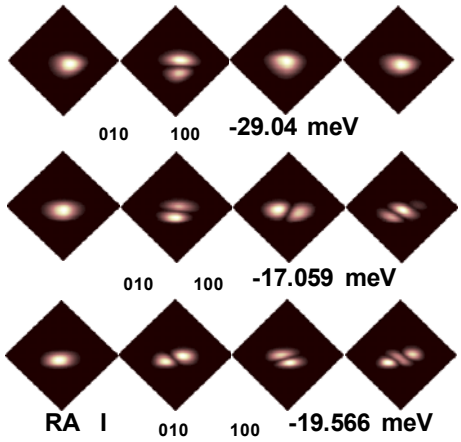

Figure 5. First 4 (four) electronic wavefunctions and split in the P levels including interfacial effects, strain, and piezoelectricity.

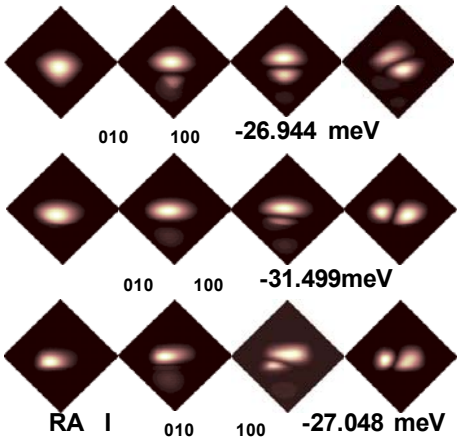

Figure 6. First 4 (four) electronic wavefunctions and split in the P levels in all three quantum dots including interfacial effects, strain, piezo- and pyro- electricity.

4 CONCLUSION

Atomistic simulations using the NEMO 3-D tool have been performed to study the influence of internal fields in Wurtzite InN/GaN quantum dots with 3 (three) different geometries, namely, box, dome, and pyramid, all having a diameter/base length of 10.1 nm and a height of 5.6 nm. atomistic strain was found to be long-ranged and penetrates ~20 nm into both the substrate and the cap layers stressing the need for using *realistically-extended* substrate and cap layers containing at least 3 million atoms in the study of electronic structure of these reduced-dimensional QDs. As opposed to the atomic/interfacial symmetry, strain is found to have a general tendency to orient the electronic wavefunctions along the [010] direction and further lower the symmetry of the system under study. The induced piezoelectric and pyroelectric potentials are significantly large (tens of meV) and anisotropic in the QD planes. Both the piezoelectricity and the pyroelectricity introduce a global shift and a band mixing in the energy spectrum, and are found to be strong enough to flip the optical polarization in certain quantum dots.

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REFERENCES

[1] FA Ponce and DP Bour, "Nitride-based Semiconductors for Blue and Green Light-emitting devices", *Nature*, 386, 351–9, 1997.

[2] W. Ke, C. Fu, C. Chen, L. Lee, C. Ku, W. Chou, W.-H Chang, M. Lee, W. Chen, and W. Lin, "Photoluminescence properties of self-assembled InN dots embedded in GaN grown by metal organic vapor phase epitaxy", *Applied Physics Letters*, 88, 19, 191913–15, 2006.

[3] Gabriel Bester and Alex Zunger, "Cylindrically shaped zinc-blende semiconductor quantum dots do not have cylindrical symmetry: Atomistic symmetry, atomic relaxation, and piezoelectric effects", *Phys. Rev. B*, 71, 045318, 2005.

[4] Momme Winkelnkemper, Andrei Schliwa, and Dieter Bimberg, "Interrelation of structural and electronic properties in $In_xGa_{1-x}N$/GaN quantum dots using an eight-band *k•p* model", *Phys. Rev. B*, 74, 155322, 2006.

[5] J. M. Jancu, F. Bassani, F. Della Sala, R. Scholz, "Transferable tight-binding parametrization for the group-III nitrides", *Appl. Phys. Lett.*, 81, 2002.

[6] Shaikh Ahmed, Neerav Kharche, Rajib Rahman, Muhammad Usman, Sunhee Lee, Hoon Ryu, Hansang Bae, Steve Clark, Benjamin Haley, Maxim Naumov, Faisal Saied, Marek Korkusinski, Rick Kennel, Michael Mclennan, Timothy B. Boykin, and Gerhard Klimeck, "Multimillion Atom Simulations with NEMO 3-D", *In Meyers, Robert (Ed.) Encyclopedia of Complexity and Systems Science*, 6, 5745–5783. Springer New York 2009.