Nanoelectronic 3-D (NEMO 3-D) Simulation of Multimillion Atom Quantum Dot Systems

Fabiano Oyafuso, Gerhard Klimeck, R. Chris Bowen, and Timothy B. Boykin
Jet Propulsion Laboratory, California Institute of Technology
4800 Oak Grove Dr., Pasadena, CA, 91109, USA
E-mail: fabiano.oyafuso@jpl.nasa.gov

Abstract—The convergence of electron and hole ground states of a dome-shaped In$_0.6$Ga$_0.4$As quantum dot as a function of the size of the surrounding buffer is explored within an $sp^3d^5s^*$ tight binding model. It is found that although the quantum dot encompasses only $2 \times 10^7$ atoms, proper convergence of ground state eigenenergies requires that over 10 times as many atoms need to be included in the simulation domain. It is also found that the disorder-induced broadening is very sensitive to the applied boundary conditions. Examination of local eigenenergies as functions of position shows similar convergence problems and indicates that an inaccurate resolution of the equilibrium atomic positions due to truncation of the simulation domain may be the source of the slow ground state convergence.

1. INTRODUCTION

Nonatomistic models such as $k$. $p$ are often used to characterize the electronic structures of quantum dots. However, such jellium-like models are fundamentally not well suited for the atomistic representation of nanoscale features such as interfaces and disorder [1]. We are currently developing an atomistic nanoelectronic simulation tool (NEMO-3D) to model quantum dot structures on high performance commodity clusters (Beowulfs). Modeling of realistic structures entails simulation domains encompassing many millions of atoms. Such large-scale domains result in very large eigenproblems (dimension $\geq 10^6$) which necessitate the usage of massively parallel computers. Details of the numerical implementation including performance benchmarks have been described in greater detail elsewhere [2] and are not discussed in this work.

Our simulation employs a nearest-neighbor tight-binding model ($sp^3d^5s^*$) with a 20 orbital basis, consisting of $s$, $p$, and $d$ orbitals, associated with each atomic lattice site. Since the basis set that is used is not complete in a physical sense, the parameters that enter the model do not correspond precisely to actual orbital overlaps. Thus, an analytical approach to determine these parameters is insufficient. Instead, a genetic algorithm package is used to determine a set of orbital couplings that reproduces a large number of physical observables of the bulk binary system, including bandgaps and effective masses at various symmetry points in the Brillouin zone. These orbital couplings must also depend on bond lengths to account for the shifts in atomic positions in strained systems. A power-law scaling is assumed (whose exponent is also determined with the genetic algorithm) to account for strain-induced shifts. Because the basis set used consists of orthogonalized L"owdin orbitals and not the true atomic orbitals, the diagonal (self-coupling) elements are also allowed to vary with the displacement of the nearest neighbor atoms [3]. Clearly, an accurate calculation of the electronic structure within the tight-binding model also necessitates an accurate representation of the positions of each atom. To this end, NEMO-3D uses a valence force field (VFF) model in which the total strain energy, expressed as a local (nearest-neighbor) functional of atomic positions, is minimized [4], [5].

One potential advantage of using a local orbital basis rather than a periodic plane-wave basis set as in pseudopotential calculations is the ability to study finite (non-periodic) structures. In reality such structures (e.g. quantum dots) are imbedded in a larger bulk material, and one must model a sufficiently large subset of the entire system such that the eigenspectrum is not significantly affected. Choosing appropriate boundary conditions, then, becomes a serious issue. In this work, we examine the dependence of the ground state eigenenergies as a function of various buffer sizes. We shall demonstrate that the variations of eigenenergies can depend strongly on the size of simulation domain, and that proper care must therefore be taken to ensure that a solution has “converged”.

II. SIMULATION

A. Ground state electron energy

The canonical model used for all the simulations in this work is a dome shape In$_{0.6}$Ga$_{0.4}$As quantum dot (QD) of diameter 30 nm and height 5.4 nm embedded in a finite GaAs box. The QD itself contains roughly $2 \times 10^7$ atoms. A list of the values of the tight-binding parameters necessary for the simulation is given in Table III of reference [2]. Although all the QDs are of identical size, the placement of In and Ga cations varies among simulations. We make the ansatz that no correlation in species type exists between any two atomic sites. Thus, sites on the cation sublattice are filled with a 60% (40%) probability of being In (Ga). Note that such an algorithm does not, in general, ensure that exactly 60% of the cations in the QD are In atoms. The resulting broadening of the energy spectrum, then, includes not only the configurational disorder, arising from the distribution of different cations throughout the alloy subject to the constraint of a fixed overall concentration, but also a concentrational disorder that reflects the fact that a growth process never produces nanostructures with identical concentrations each time. It has been demonstrated that for bulk (periodic) systems, the concentrational disorder dominates by an order of magnitude [6]. In the following simulations, we consider three different boundary conditions, fixed, free, and periodic, that directly impact the...
strain calculation and therefore either indirectly or directly affect the electronic computation. In the fixed case, the surface atoms are constrained to the positions they would have if the simulation domain consisted entirely of GaAs. In the free case, the entire domain is allowed to expand without any external constraints. Finally, under periodic boundary conditions, the total strain energy is minimized with respect to both position and period.

In Fig.(1), we compare the ground state electron eigenenergy for the cases of free and fixed boundary conditions as a function of buffer size in directions parallel (solid) and normal (dashed) to the base of the dome. The largest simulation employs a 20 nm buffer in the z direction, normal to the base of the dome, and encompasses nearly 3 million atoms. For variation of buffer size in the lateral (normal) direction, the buffer size in the normal (lateral) direction is fixed to 4 nm. The intent is to investigate how well such a truncated system approximates one in which the QD is embedded in an infinitely large block of GaAs. Since locally, near the center of the dot where the probability density is highest, the QD "looks" like a a quantum well and thus the eigenenergy is principally determined by confinement in the z direction. Thus, for both cases, the dependence on lateral buffer size is weak relative to the dependence on the buffer size in the z direction. In the case of free boundary conditions no external constraints are imposed, so that the strain computed for the truncated system is reduced from what it should be for an infinite buffer. The shift of the conduction band edge at \( \Gamma \), which depends linearly on the hydrostatic component, is given by

\[
\Delta E_c = \varepsilon_d^{(000)} \text{Tr}\{\varepsilon\}
\]

where \( \varepsilon_d^{(000)} < 0 \) [7]. Thus, one expects a reduction in compressive strain (an increase in Tr\{\varepsilon\}) in the QD to accompany a reduction in electron ground state energy. The situation for the fixed case, where the lattice constant on the boundary is constrained to bulk values, is inverted, since the strain effect in the QD is overestimated relative to the case of infinite buffers. Fig.(1) demonstrates that the two cases converge provided that the buffer is made sufficiently large. These results demonstrate that the simulation domain needs to extend rather far into the buffer to assure convergence.

### B. Linewidth broadening

We now consider the issue of linewidth broadening. That is, given an ensemble of ostensibly identical quantum dots, we explore the fundamental limits of linewidth broadening that arise solely as a result of variations in configurations of cations in the quantum dots and ignore any additional contributions such as size variation, strain-induced spatial perturbations on a QD due to neighboring QDs, and many-body effects. Our calculations for the case of fixed boundary conditions did not yield hole eigenenergies for small buffer sizes, so we consider only the case of free boundary conditions. We examine the electron and hole ground state eigenenergy distributions for three different buffer systems, 4nm 8nm, and 12nm. 190 samples points were obtained for the first two systems and 93 for the larger (and computationally more taxing) 12nm buffer geometry. A histogram of the distributions for the first two geometries is shown in Fig.(2), and Table (1) summarizes the statistical data for all three configurations. First we note that the electron (hole) energies are shifted up (down) with bigger buffers as found earlier. However, the electron eigenenergy broadening, given by the standard deviation, \( \sigma_E \), is two to three times larger than that of holes, \( \sigma_q \). This result differs from an earlier finding for bulk unstrained Al\(_x\)Ga\(_{1-x}\)As with periodic boundary conditions applied in which the electron broadening was found to be only slightly larger than that of holes [6]. This discrepancy might possibly be due the fact that both light and heavy holes are broadened in the bulk unstrained case, while

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**TABLE I**

<table>
<thead>
<tr>
<th>BUFFER SIZE DEPENDENCE OF EIGENENERGIES AND BROADENING</th>
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<tr>
<td>Statistical data (energies are in meV and lengths are in nm)</td>
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<tr>
<td>( L_{bf} )</td>
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<tr>
<td>( 4 )</td>
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<td>( 8 )</td>
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the confinement of a QD splits the $\Gamma_5$ degeneracy. Interestingly, the degree to which the electron and hole distributions are broadened is strongly dependent on the buffer size. Indeed, it seems likely that a significant broadening of the eigenenergy distribution is an artifact of the truncation of the simulation domain, since increasing the simulation size reduces the broadening. One possible explanation for this reduction in linewidth is that as eigenstates are pushed up closer to the bulk GaAs conduction and valence band edges, perturbations to concentration and strain produce less significant changes to the eigenvalues.

Finally, note that the sum of electron and hole standard deviations is still (roughly) equal to the standard deviation of the energy gap. This result indicates that the electron and hole eigenenergies are strongly correlated. This is not unexpected since the concentration broadening had previously been identified to be the most dominant effect in bulk unstrained systems [6]. Another way to see this correlation is to look at the dependence of the eigenenergies on the total strain energy of the system, which also depends on the alloy concentration. In the VFF model used, the total strain energy is the sum over all atoms of the local strain energy at each atom, which depends only on the positions of the neighboring atoms. Fig.(3) shows scatter plots for electron and hole ground state energies as a function of total strain for two different lateral buffer widths of 4 and 8 nm. The strain energies (not shown) seem to have a roughly Gaussian shape with a spread of approximately 25 eV (or about 1 %). Notice that an increase in strain energy reduces the band gap. The reduction in band gap is reasonable, since an increase in strain energy is likely a result of a slightly higher concentration of In atoms. The trends are the same for both buffer sizes, except that the strain energy for the system with the 4 nm buffer is reduced, since the overall system size is smaller. However, the slopes of the curves are much larger for the smaller buffer, indicating a stronger dependence of eigenenergy on strain.

C. Local bandstructure

We next examine the effect of the deformation of the primitive cells under strain on the local electron and hole band structure within the GaAs buffer. At each cation (Ga) site, we define a “local” eigenenergy obtained by computing the band minima at $\Gamma$ of a bulk solid constructed from the single primitive cell formed by the cation and its four neighboring As anions. In essence, these local eigenenergies define a spatially dependent band edge. It is important to note that the eigenenergy local to a cation depends only on the relative position of the surrounding As atoms. Thus, these local eigenvalues essentially reflect local strain conditions. Fig.(4) shows local eigenvalues of the buffer material for our canonical problem with the three boundary conditions discussed earlier and with buffer sizes of 4nm, 8nm, and 12 nm. Larger dot size corresponds to larger buffer size.
Fig. 5. Local hole eigenenergies of the GaAs buffer along a lateral axis of the QD for different boundary conditions and buffer sizes of 4nm, 8nm, and 12 nm. Larger dot size corresponds to larger buffer size.

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REFERENCES