Study of Strain Boundary Conditions and GaAs Buffer Sizes in InGaAs Quantum Dots

Fabiano Oyafuso, Gerhard Klimeck, Timothy B. Boykin*, R. Chris Bowen, and Paul von Allmen
Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA 91109
*The University of Alabama in Huntsville, Dept. of Electrical Engineering, AL 35899
email: gekco@jpl.nasa.gov website: http://hpc.jpl.nasa.gov/PEP/gekco

NEMO 3-D has been developed for the simulation of electronic structure in self-assembled InGaAs quantum dots on GaAs substrates. Typical self-assembled quantum dots in that material system contain about 0.5 to 1 million atoms. Effects of strain by the surrounding GaAs buffer modify the electronic structure inside the quantum dot significantly and a large GaAs buffer must be included in the strain and electronic structure. However, to reduce computational expenses it is desirable to keep the GaAs buffer as small as possible. The simulations presented in this paper analyze the local band structure in the system as well as the dependence of the confined hole and electron states in ensembles of alloyed InGaAs quantum dots on the total strain of the system and the GaAs buffer size.

The local strain in the 3-D system modifies bond lengths and angles for individual atoms in a unit cell. Local band structure can be computed with a periodic repetition of such a unit cell. InGaAs alloys maintain a bi-modal distribution of InAs and GaAs-like bond length resulting in a bi-modal distribution of local band edges. Figures 1a and 1b show a scatter plot of local conduction band edges along a line laterally through a 30nm diameter, 5nm high dome shaped In₀.₆Ga₀.₄As quantum dot for two different buffer sizes. The bi-modal distributions of InAs and GaAs-like band edges are evident inside the quantum dot. The GaAs-like bonds are compressed by the surrounding InAs resulting in local band edges that are higher than the surrounding GaAs. The InAs-like bonds are compressed by the overall surrounding GaAs resulting in a local band edge raised from the unstrained value. The 4nm and 12nm buffer layer simulations show significantly different scatter of local band edges in the quantum dots. 2nd degree polynomial fits in the spatial regions of the buffer and central quantum dot region are shown in Figure 1a-b) as well for InAs and GaAs-like bonds. The interface regions are left out the fits because of the distributed interface of the InGaAs dome shaped dot. The dashed lines in the quantum dots show the average local band structure including both InAs and GaAs-like bonds. Figure 1c compares the average local potentials for the 4nm and 12nm buffer size. The effects of the different buffer size are clearly evident.

We have previously analyzed the statistical distribution of electron and hole ground state energies in InGaAs quantum dots. Here we analyze the effects of the finite GaAs buffer sizes on the statistical energy distribution and their dependence on the local strain energy. The electron and hole ground states are computed in two ensembles of 200 random alloy InGaAs quantum dots for a 4nm and 8nm surrounding GaAs buffer assuming open strain boundary conditions. The ground state energies are graphed in a scatter plot against the total strain energy in the system in Figure 1a-d). Several observations can be made: 1) the absolute average energies vary with the buffer size (band gap increases with increasing buffer size), 2) the standard deviation induced by random disorder decreases with increasing buffer size, 3) the dependence on the total strain in the system is reduced for increasing buffer size. Some of these characteristics can be immediately shown to be dependent on the local band structure in the quantum dot and the surrounding buffer size. The 4nm buffer clearly is not enough to properly model the effect of the surrounding GaAs buffer.
Figure 1: (a-b) Local band edge along a line laterally through an InGaAs/GaAs quantum dot system for two GaAs buffer sizes. Bi-modal distribution of InAs and GaAs bond lengths in InGaAs leads to a bi-modal distribution of local band edge energies computed in a single primitive cell. Solid lines are 2nd order polynomial fits to the scatter. Dashed line is a fit to the overall band edge inside the quantum dot. (c) Average confining potentials from (a) and (b). Small buffer size imposes a significantly more bent potential profile inside the quantum dot.

Figure 2: (a-d) 200 statistical samples for each buffer size of electron and hole eigen energies with their corresponding in InGaAs quantum dots plotted against the total strain energies. Electron energies are decreasing with increasing strain while hole energies are increasing with increasing strain. The solid lines represent linear regression fits and are referenced to the average strain and average electron/hole energy. Increasing the buffer size from 4nm to 8nm reduces the sensitivity of the eigen energies to strain (reduced slope factor m) and reduces the statistical noise in the statistical variation in the eigen energies (reduced $\sigma_{E}$).
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Jet Propulsion Laboratory, Caltech, Pasadena CA 91109
University of Alabama in Huntsville, AL 35899

gekco@jpl.nasa.gov, 818-354-2182
http://hpc.jpl.nasa.gov/PEP/gekco
Study of Alloy Disorder in Quantum Dots through Multi-million Atom Simulations
Gerhard Klimeck, Fabian Ovadoss, Timothy B. Gray, K. Chris Leven, and Paul von Allmen

Problem:
Nanoscale device simulation requirements:  
- Cannot use bulk / jellium descriptions, need description of the material atom by atom  
  => use pseudo-potential or local orbitals  
- Consider finite extent, not infinitely periodic  
  => local orbital approach  
- Need at minimum one million atoms.  
  => need massively parallel computers  
- The design space is huge: choice of materials, compositions, doping, size, shape.  
  => need a design tool

Approach:  
- Leverage NEMO 1-D:  
  - 25 person years at TI / Raytheon  
  - 250,000 lines of code.  
- Use local orbital description for individual atoms in arbitrary crystal / bonding conf.  
  - Use s, p, and d orbitals  
  - Use genetic algorithm for fitting  
- Compute mechanical strain in the system.  
- Develop parallel algorithms to generate eigenvalues/vectors of very large matrices (N=3.2x10^8 for a 16 million atom system).  
- Develop prototype GUI for (NEMO-3D)  

Gerhard Klimeck
Applied Cluster Computing Technologies Group
Four Generations of Cluster Experience

**Hyglac (1997)**
- 16 Pentium Pros 200MHz
- 128 MB RAM per node
- 2 GB total
- 5GB Disc per node
- 80 GB total
- 100 Mb/s ethernet crossbar
- Linux, MPI
- 3.2GFlops

**Nimrod (1999)**
- 32 Pentium IIs 450MHz
- 512 MB RAM per node
- 16 GB total
- 8GB Disc per node
- 256 GB total
- 100 Mb/s ethernet crossbar
- Linux, MPI
- 14.4 GFlops

**Pluto (2001)**
- 64 Pentium IIs 800MHz
- dual CPUs
- 2 GB RAM per node
- 64 GB total
- 10 GB Disc per node
- 320 GB total
- 2 Gb/s Myricom crossbar
- Linux, MPI
- 51.2 GFlops

**NewYork (2002)**
- 66 Xserve G4 1GHz
- 1GB RAM per node
- 33 GB total
- 60 GB Disc per node
- 2 TB total
- 100 Mb/s ethernet crossbar
- MAC OS X, MPI
- 495GFlops

Gordon Bell Prize 1997

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Parallel Eigenvalue Solver on a Beowulf
(32 node, dual CPU Pentium III, 800MHz, Linux)

- Divide Simulation domain into slices.
- Communication only from one slice to the next (nearest neighbor)
- Communication overhead across the surfaces of the slices.
- Limiting operation: sparse matrix-vector multiplication
- Enable Hamiltonian storage or recomputation on the fly.

- Systems from 1/4 to 16 million atoms
- sp3s* basis, Matrix sizes up to 1.6 \(10^8\times1.6 \times 10^8\)
- Large problems do not fit on one CPU
- Recompute Hamiltonian on the fly.
- Measure time for 30 Lanczos iterations
- 1 million atoms 5000 iteration 1 CPU: ~48 hours  20 CPUs: ~3.4 hours
- Computation time linear in system size.

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Parallelization of Strain Calculation

Problem (1 million atoms):
- Serial strain computation: ~43 min.
- Serial electronic structure calculation (1000 iterations): ~ 9.6 hours
- Parallel electronic structure computation on 20 CPUs: ~41 min.

Solution:
- Parallelize strain calculation as well

Result:
- Reduce time to 2-5 minutes on a parallel machine.
- See difference between a fast 2Gbps and a 100Mbps network.
- Do not see that difference in the electronic structure calculation.
- Parallel strain computation is more communication dependent than the electronic structure calculation.

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- Quantum dot: 30nm diameter, 5nm height
- Vary GaAs buffer from 4nm to 20nm in all directions
Ground State Energy Dependence on Buffer Size

- Quantum dot: 30nm diameter 5nm height
- Vary GaAs buffer from 4nm to 20nm in all directions

- Electron and hole ground states depend strongly on the GaAs buffer size
- Dependence is weaker for the electron states in the alloyed dot.

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* Strain shows a long-range effect
* Electron confinement in x changes shape
* Electron confinement in z vertical shift
Strain Effects on Local Band Structure

- Strain shows a long-range effect
- Electron confinement in x changes shape
- Electron confinement in z vertical shift
- Hole confinement in x small modulations
- Hole confinements in z vertical shift, no convergence yet!
Decoupled Strain and Electronic Boundary Conditions

- Compute strain in 24nm buffer system (9 million atoms)
- Vary size of the electronic system buffer => Electronic states virtually unaffected

- Long-range strain effects dominate the quantum dot states.
- Hard-wall electronic boundary conditions have little effects.

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Inhomogeneous Broadening due to Alloy Disorder

Problem:
• Cations are randomly distributed in alloy.
• Does alloy disorder limit electronic structure uniformity for dot ensembles?
• Requires atomistic simulation tool.

Approach:
• Simulate a statistical ensemble of dots.
  • Identical in size and shape
  • Different only in cation ordering.

Simulation of Alloy Dot Ensemble

\[ \Gamma = 0.1 \sim 5 \text{meV} \quad E_{eh} = 1.05 \text{eV} \]

Measured \( \Gamma = 34.6 \text{ meV} \) (R. Leon, PRB, 58, R4262)

1~5meV Represents Theoretical Lower Limit

**In\text{\textsubscript{0.6}}Ga\text{\textsubscript{0.4}}As** Lens Shaped Dot
Diameter=30nm, Height=5nm, GaAs embedded
\sim 1,000,000 Atom Simulation, sp3s’ basis

In and Ga atoms are randomly distributed
Inhomogeneous Broadening?

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Range order (clustering).
- Variation is larger if there is short.
- Heterodimer.
  - Have not included interface.


Caveats:

Small upper bound of 0.35 meV, so the effect is
Convergence is slow but can place an

Results:

[\text{Nagamune, APL, 67, p3257}]
- Narrow linewidths (~0.9 meV)
  - Single QD PL measurements have found

[\text{R. Lehn et al., PRB, 60, p8517}]
- Inhomogeneous broadening (~30 meV)
  - Found large contributions due to
  - Previous PL experimental results have

Alloy disorder to linewidth broadening?

Question: What is the contribution of

Disorder-induced linewidth

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Assume no correlation between localization

100-200 samples

Use a direct sampling method (roughly)

Computation:

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Atomistic Simulation (NEMO 3-D):
- Fitting tight binding sp3s*,sp3d5s*
- General structure input
- Several million atom solutions
- Parallel eigenvalue solvers
- Strain simulations

- Strain effects are long-range.
- Studied effects of alloy disorder.

Extension of NEMO 3-D to Spintronics:
- Arbitrary magnetic fields
- Magnetic impurities
- Many-body interactions (electrons, phonons, photons)
- Open boundary conditions

- Extension of NEMO 1-D to Spintronics:
  - DC Spin transport RTD-like structures
  - Time dependent spin - transport

Quantum Dots

End of SIA Roadmap

Dopant Fluctuations in Ultra-scaled CMOS

Electron Transport in Exotic Dielectrics

(Ba,Sr)TiO_3 TiO_2
Strain Effects on Local Band Structure InGaAs

- Strain shows a long-range effect
- Electron confinement in x changes shape
- Electron confinement in z vertical shift
- Hole confinement in x small modulations
- Hole confinements in z vertical shift, no convergence yet!

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Local band structure in an Aloyed Cd
Spatial Irregularity in the Hole Ground State

VCA / no Disorder

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Spatial Irregularity in the Hole Ground State

VCA / no Disorder

Disorder Sample 1

Disorder Sample 2

E = 0.2208 eV
\Delta E = +0.00 meV

E = 0.2174 eV
\Delta E = -3.44 meV

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Spatial Irregularity in the Hole States

(g) hole 1
E=0.2089eV
ΔE=-0.00meV

(h) hole 2
E=0.1942eV
ΔE=-14.7meV

(i) hole 3
E=0.1928eV
ΔE=-16.0meV

(j) hole 4
E=0.1811eV
ΔE=-27.8meV

(k) hole 5
E=0.1780eV
ΔE=-30.9meV

(l) hole 6
E=0.1765eV
ΔE=-32.4meV
>1000 Alloyed Quantum Dot Samples

- Atomistic granularity
  \( \sigma = 2.3 \text{meV} \)

- Cell granularity
  \( \sigma = 4.5 \text{meV} \)

- Ec and Ev strongly correlated
- Ec and x weakly corelated

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Effect on ground state electron energy:

- System: Dome-shaped $\text{In}_{0.6}\text{Ga}_{0.4}\text{As}$ QD 15nm radius; 5.4 nm height

  $BC$: no constraints on QD; strain and ground state energy are underestimated

- Fixed $BC$: QD boundary pinned; strain and ground state energy are overestimated

- Periodic $BC$ ($k_{\text{supercell}}=0$): Eigenvalues lie in between free and fixed case, but results are much closer to case of free $BC$.

Conclusions:

- Overall convergence is slow.

- Varying only vertical buffer size gives a good approximation.
Inhomogeneous Broadening due to Alloy Disorder

**Problem:**
- Cations are randomly distributed in alloy.
- Does alloy disorder limit electronic structure uniformity for dot ensembles?
- Requires atomistic simulation tool.

**Approach:**
- Simulate a statistical ensemble of dots.
  - Identical in size and shape
  - Different only in cation ordering.

**Simulation of Alloy Dot Ensemble**

![Graph showing bond length vs. In concentration](image)

- $\Gamma = 0.1 \sim 5 \text{meV}$
- $E_{eh} = 1.05 \text{eV}$

- Measured $\Gamma = 34.6 \text{ meV}$ (R. Leon, PRB. 58, R4262)
- $1 \sim 5 \text{meV}$ Represents Theoretical Lower Limit

**InGaAs has bi-modal bondlength distribution:**
- InAs-like and GaAs-like bonds!

**In$_{0.6}$Ga$_{0.4}$As Lens Shaped Dot**
- Diameter=30nm, Height=5nm, GaAs embedded
- $\sim 1,000,000$ Atom Simulation, sp3s' basis

In and Ga atoms are randomly distributed

Inhomogeneous Broadening?

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**Bulk Semiconductors are described by:**
- Conduction and valence bands, bandgaps (direct, indirect), effective masses
- 10-30 physically measurable quantities

**Tight Binding Models are described by:**
- Orbital interaction energies.
- 15-30 theoretical parameters

**Analytical approach:**
- Exact diagonalization at Γ for sp³d⁵s*.
- Formulas developed by Tim Boykin at UAH for effective masses and bandgaps from interaction energies

**Numerical approach:**
- Use a genetic algorithm to do fitting.

- Match experimental data in various electron transport areas of the Brillouin zone:
  - Effective masses of electrons at Γ, X and L
  - Effective masses of holes at Γ
  - Band edges at Γ, X and L

15-30 theoretical interaction energies
Genetic Algorithm

- Genetic algorithm parameter optimization is based on:
  - Survival of good parameter sets
  - Evolution of new parameter sets
  - Persistence of diversity (ensures global exploration)

- Basic Operations:
  - Crossover – gross exploration
  - Mutation – fine tuning

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Leverage NEMO 1-D:
A User-friendly Quantum Device Design Tool

- NEMO was developed under a government contract to Texas Instruments and Raytheon from 1993-97
  - >50,000 person hours of R&D
  - 250,000 lines of code in C, FORTRAN and F90
- Based on Non-Equilibrium Green function formalism (Datta, Lake, Klimeck).
- NEMO in THE state-of-the-art heterostructure design tool.
- Used at Intel, Motorola, HP, Texas Instruments, and >10 Universities.