Study of Alloy Induced Disorder in Quantum Dots using Tight-Binding

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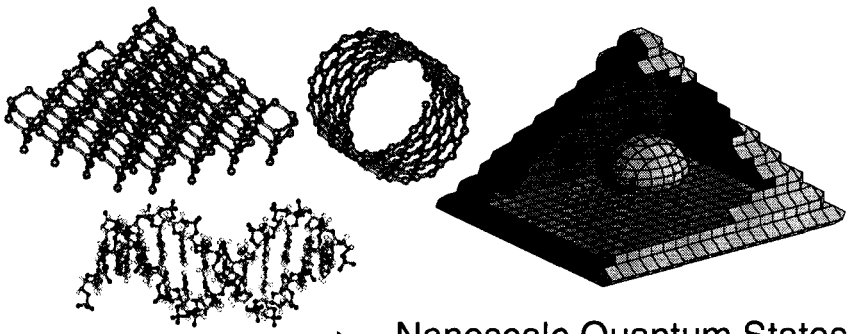
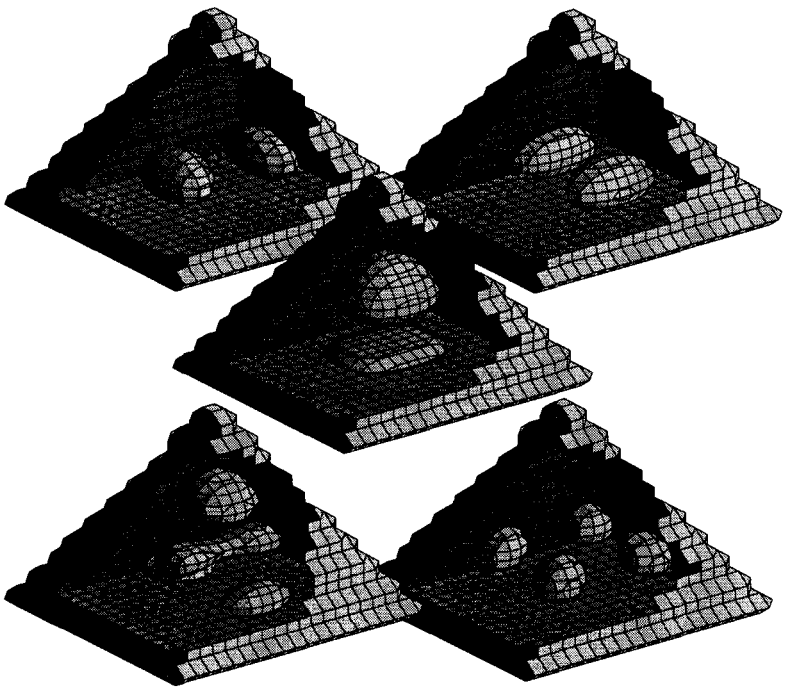
NEMO-3D: Fundamental approach



Structure → Nanoscale Quantum States (Artificial Atoms, size 20nm)



Parameterization:

- TB parameters determined from genetic algorithm to match experimentally measured band edges and masses.

Mechanical Strain:

- CG-based minimization of mechanical strain to determine atomic positions

Electronic Structure:

- Custom Lanczos eigenvalue solver

Numerical Simulation:

- Problem size is large: 100 GB is typical storage requirement for Hamiltonian
- Parallel implementation of both strain and electronic structure calculation is necessary!
- 1D data decomposition

Tight Binding Material Parameterization

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

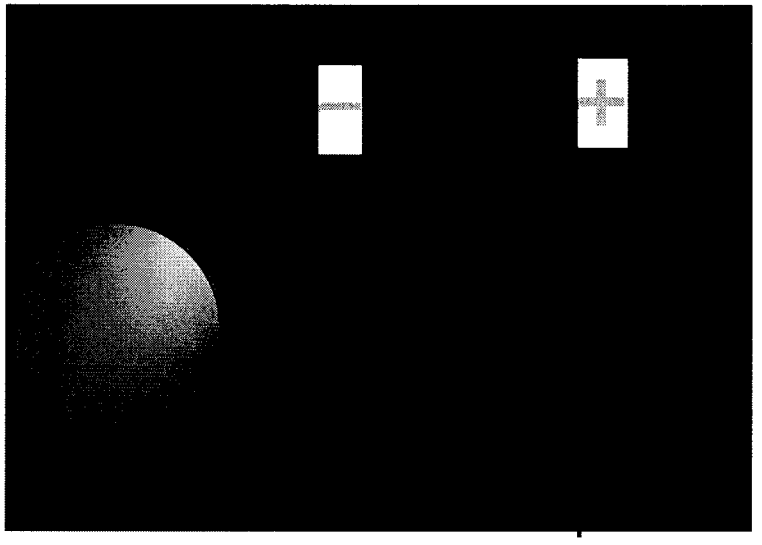
Analytical approach:

- Exact diagonalization at Γ for $sp^3d^5s^*$
- Formulas developed by Tim Boykin at UAH (subcontract) for effective masses and bandgaps from interaction energies

Numerical approach:

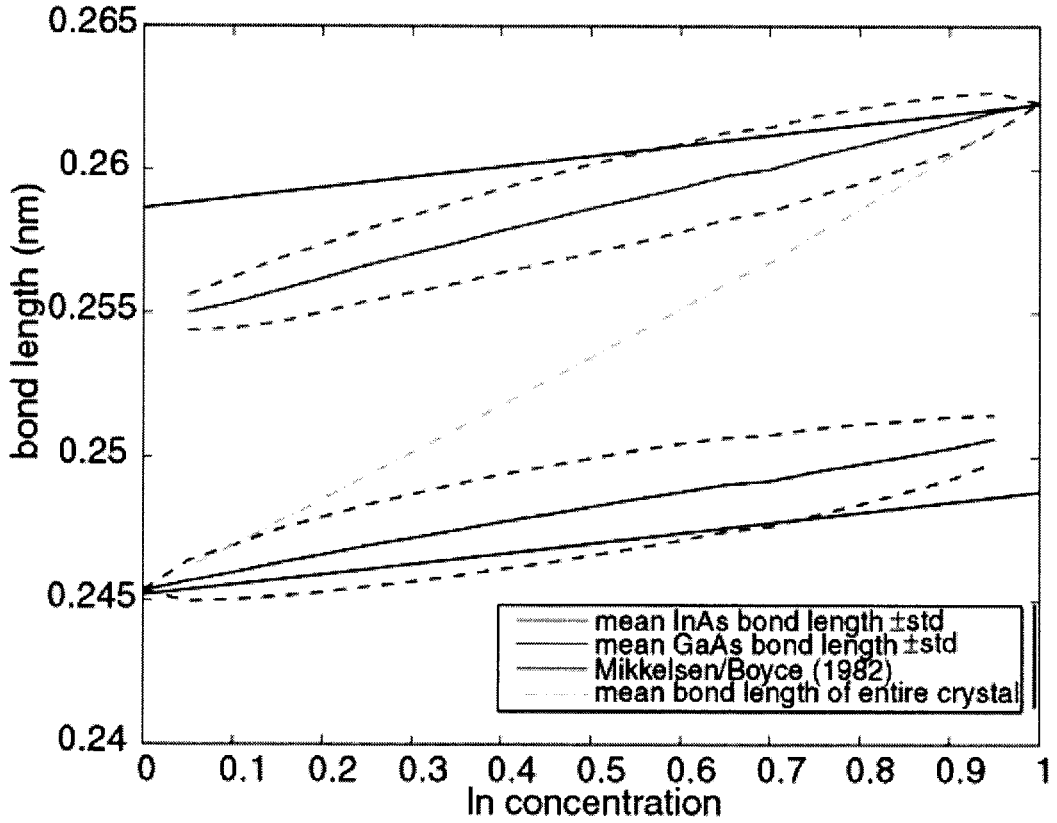
- Use a genetic algorithm to do fitting.

- 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

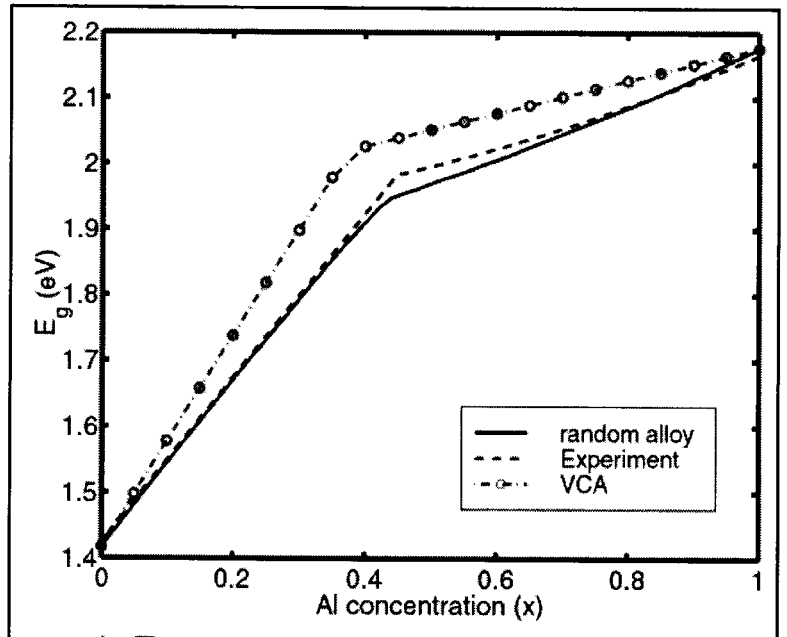


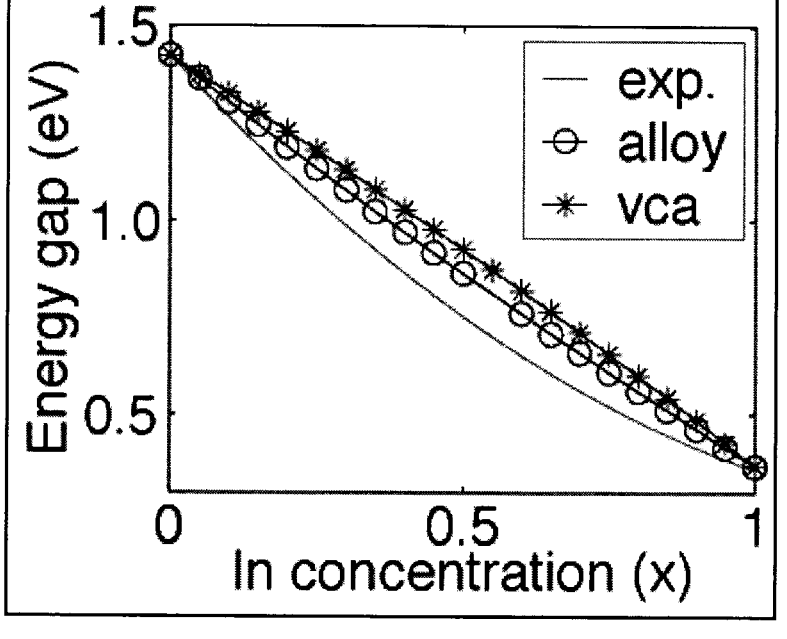
15-30 theoretical interaction energies

Minimization of mechanical strain



- **Total strain energy (computed from Keating model) is minimized through CG-based algorithm**
- **Periodic boundary conditions require relaxation of the period**
- **InGaAs bond length distribution:**
  - VCA on the bond length is incorrect
  - Locally, InAs and GaAs (mostly) maintain their bond length character





AlGaAs and InGaAs Bulk

Problem:

- VCA provides generally a linear bandgap if interpolated from the binaries GaAs, AlAs, and InAs.

Approach:

- sp3s* tight binding model
- Perform 3-D alloy simulation of the bandedges.
- Represent each individual atom in the chunk of material
- 3-D random alloy simulation matches experimental data well.

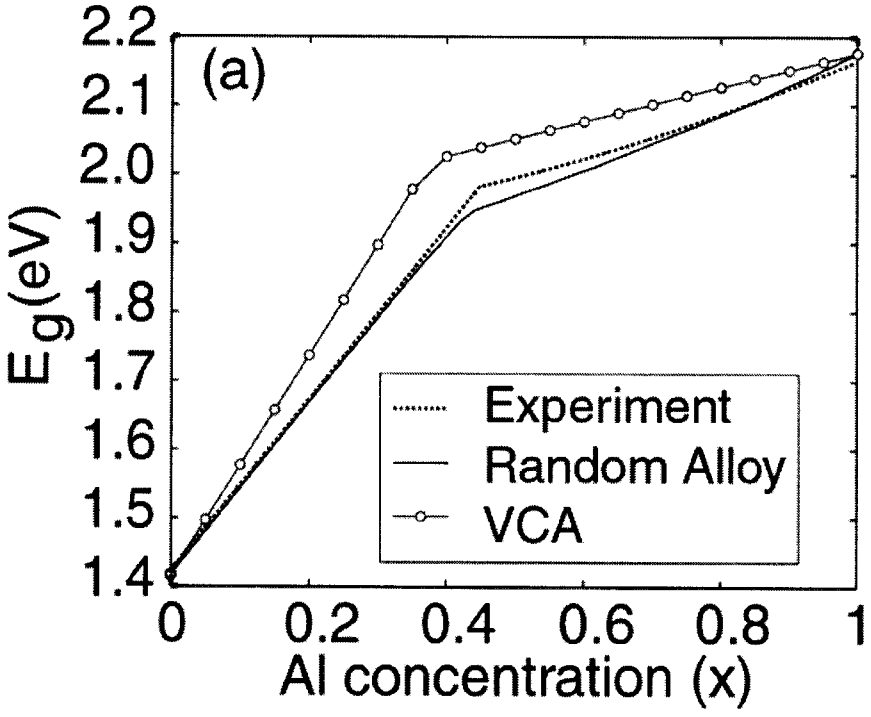
AlGaAs:

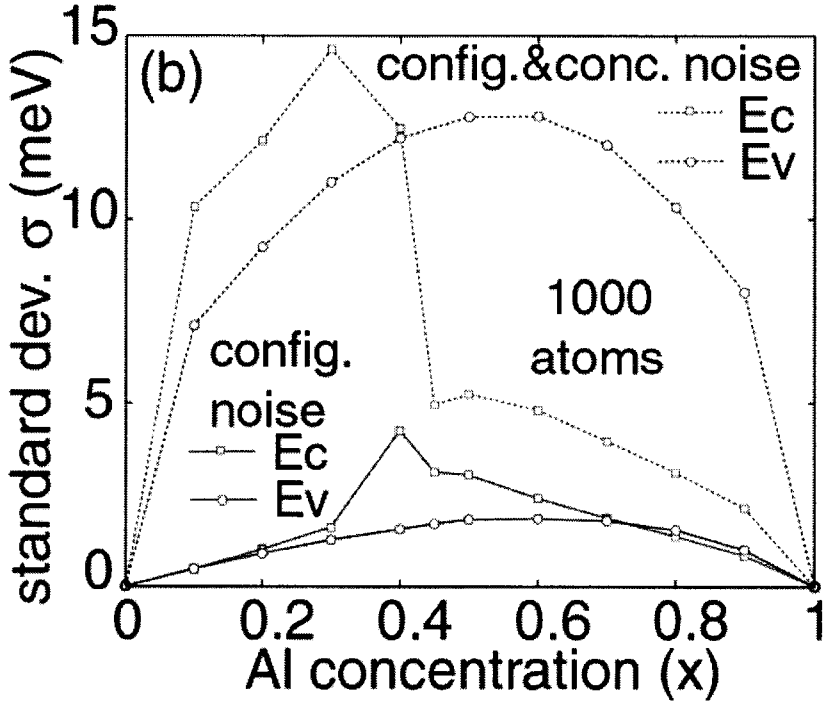
- VCA derived from pure GaAs and AlAs results in an wrong bandgap.
- 3-D simulation gives the correct bowing.

InGaAs:

- Improved bowing versus the VCA.
- Still a problem with over estimating the band gap
  -> parameterization dependent

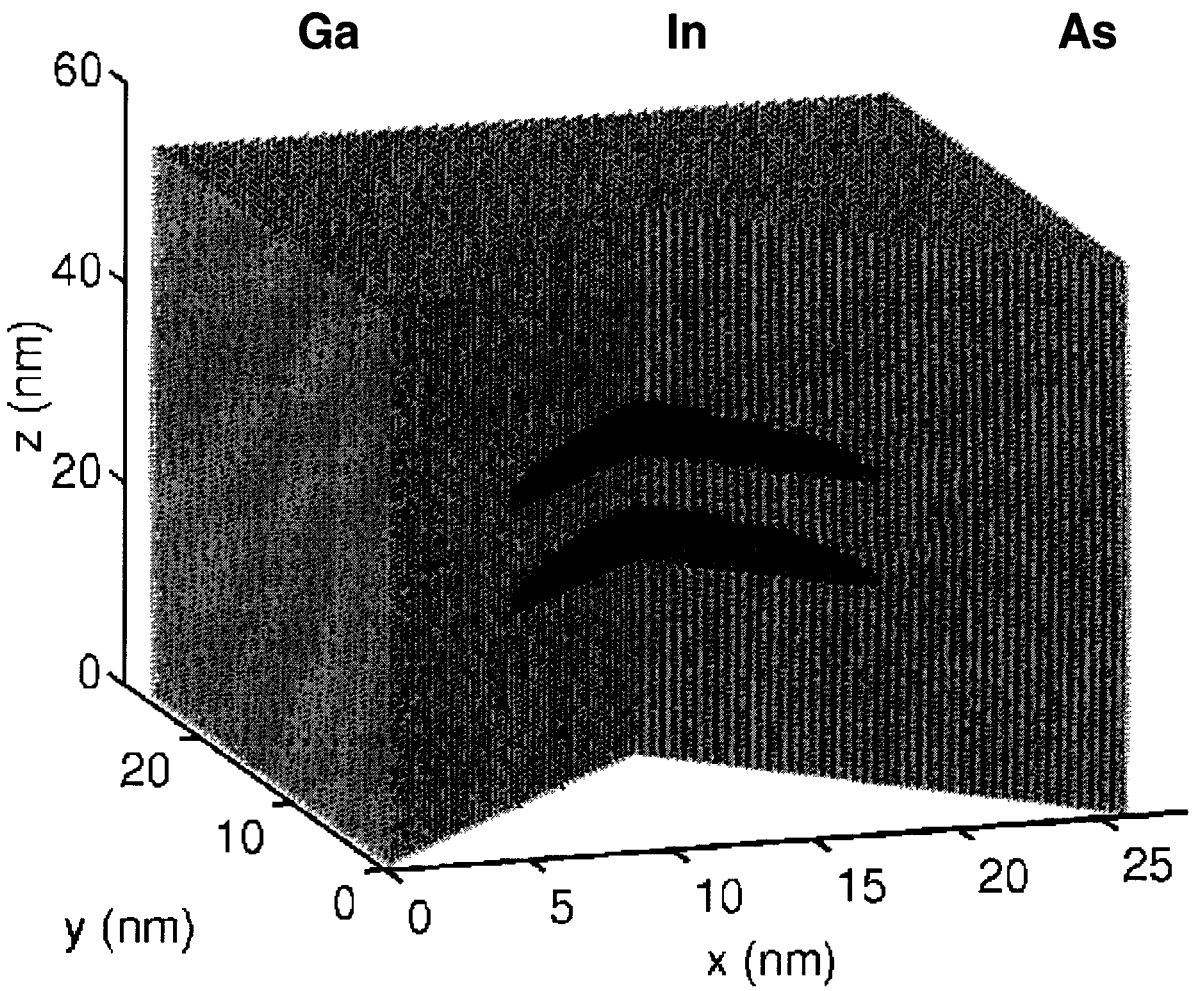
Configuration and Concentration Noise





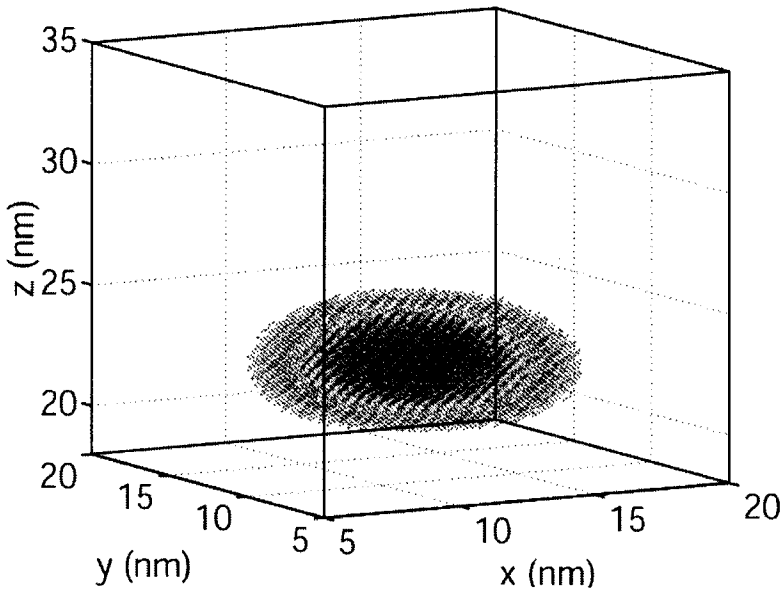
- Concentration may vary stochastically as well.
- Concentration noise is larger than configuration noise.
- For a system containing 1000 atoms, the variation is about 10-15 meV
  - Conduction band noise shows a significant feature at the Γ-X transition (Al~0.45)
  - Valence band dependence is much smoother.

Typical simulation – coupled QDs

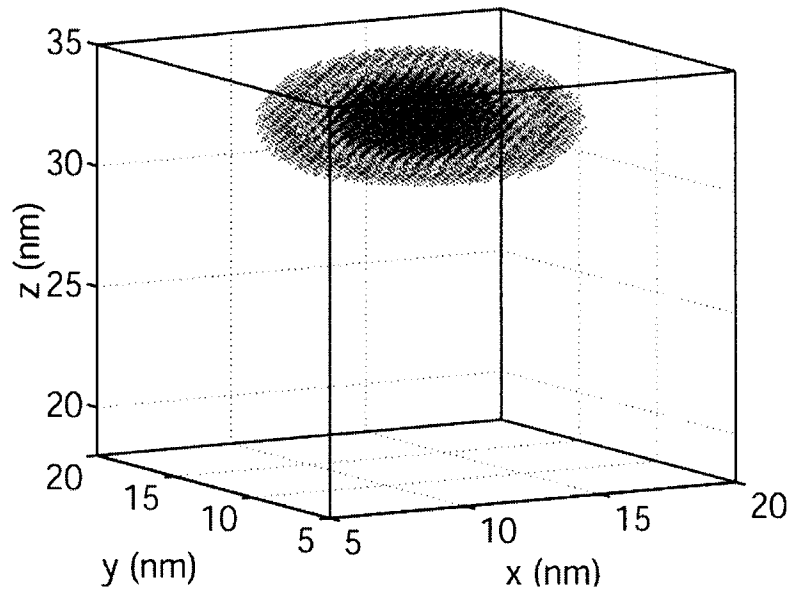


- Cutaway of simulation domain
- Coupled InAs QDs: Diameter = 18 nm; QD separation = 5 nm

Coupled Quantum Dots (Simulation without strain)



E = 0.828 eV

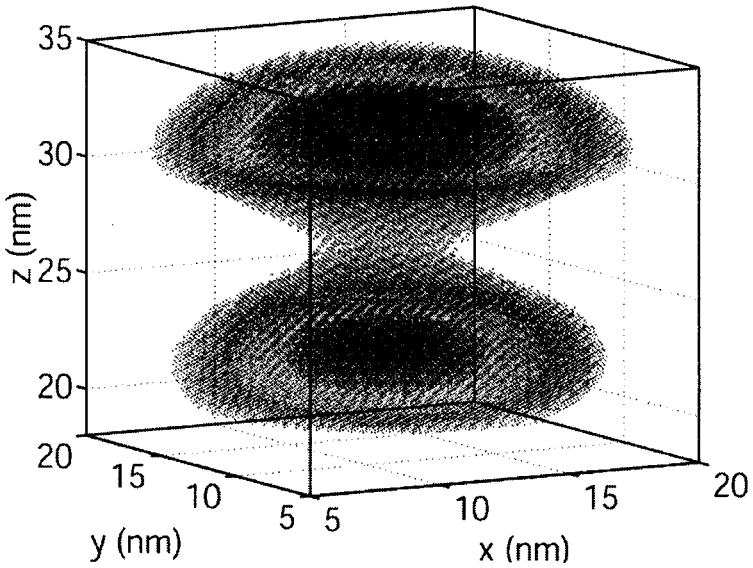


E = 0.835 eV

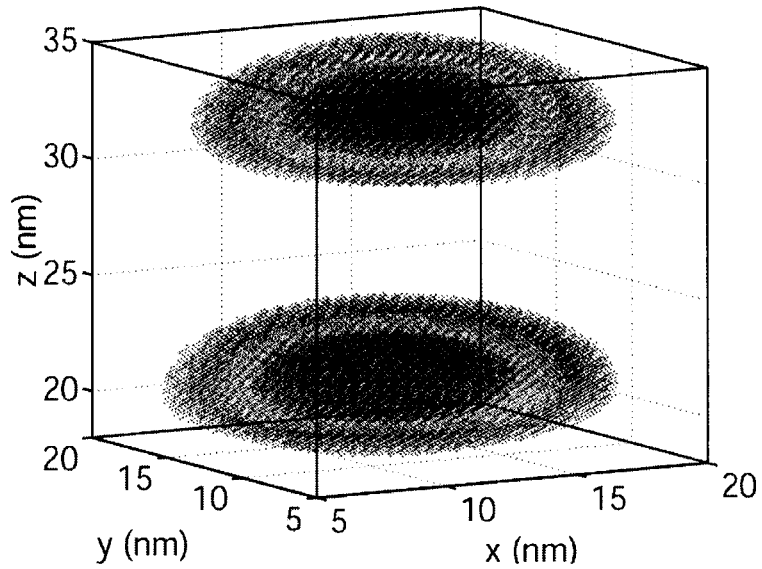
Unstrained system:

- Slight variation in the geometry of the two QDs breaks degeneracy.
- Compressive strain on the QD effectively raises E_c within QD. Without strain, potential well confining electrons is deeper and essentially decouples the two QDs.

Coupled Quantum Dots (Simulation including strain)



E = 1.364 eV

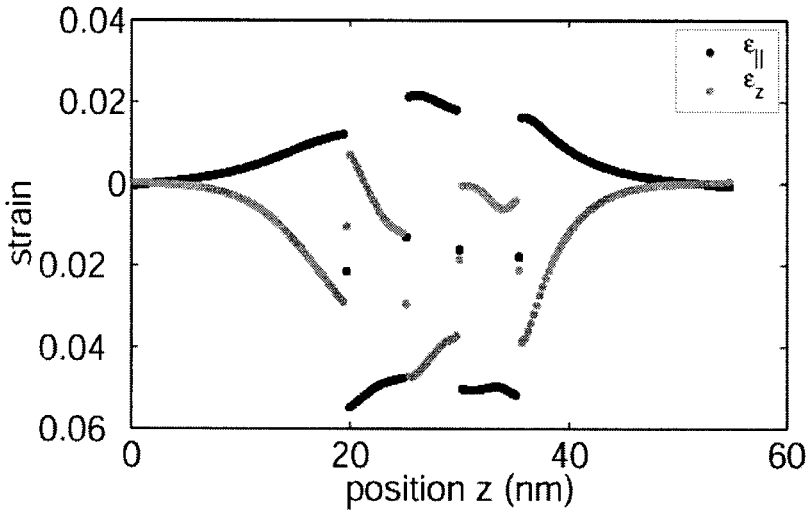


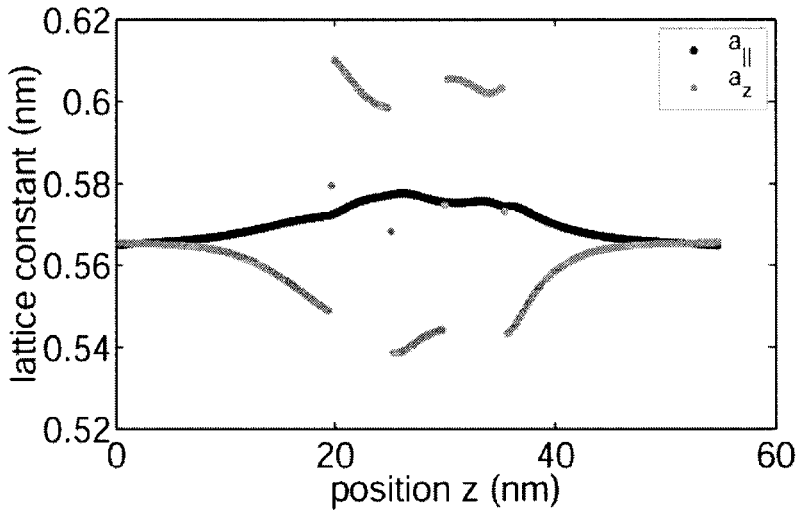
E = 1.381 eV

Strained Coupled QD:

- In absence of electron-electron interaction, ground state is bonding state; first excited state is anti-bonding state.
- Energy split (17 meV) is dependent on wave function overlap.
- Proper inclusion of strain is necessary to obtain correct eigenstates!

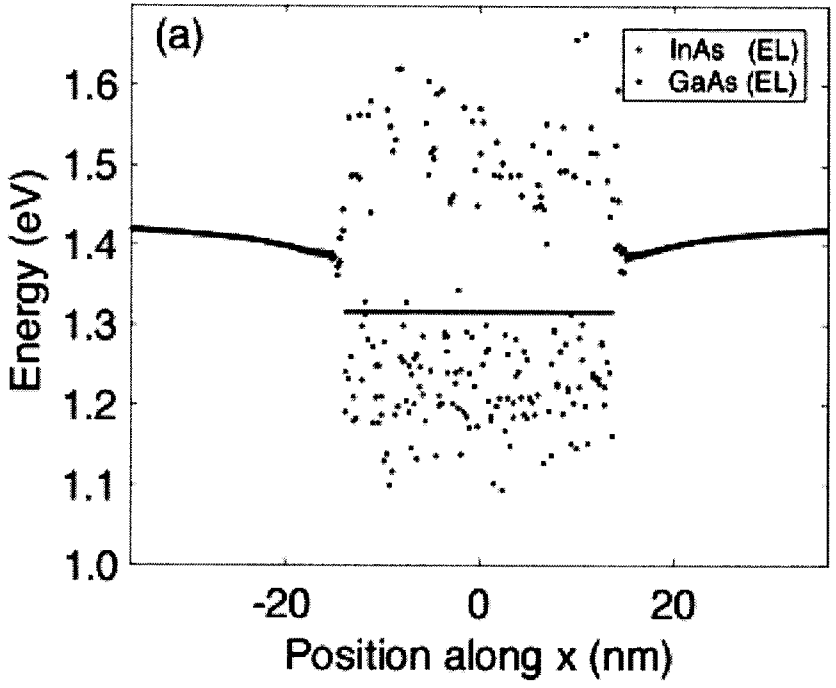
Coupled Quantum Dots – Strain Profile

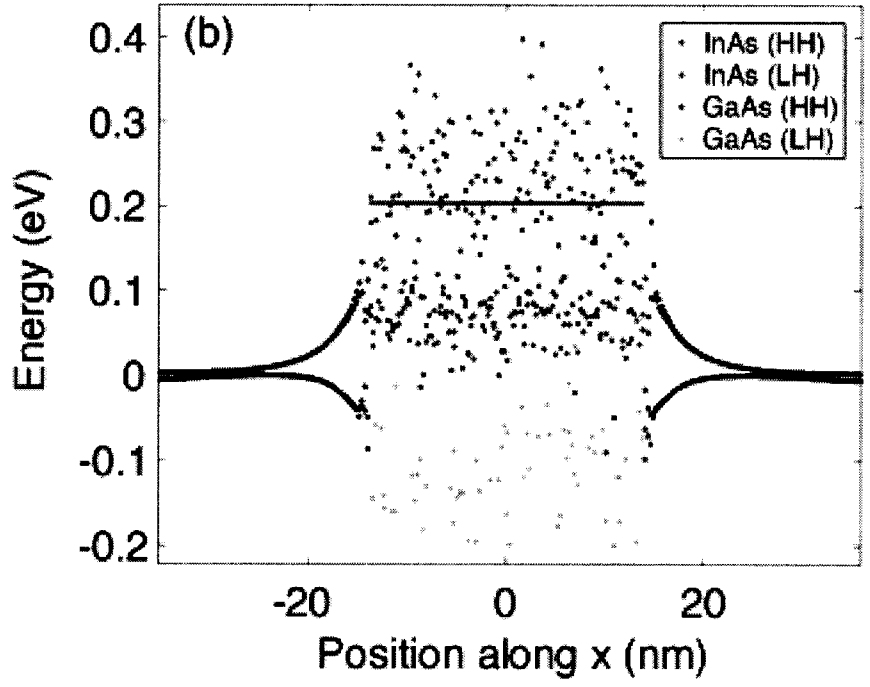




- Examine strain along major symmetry axis (z) for primitive cells centered about As atoms
- Tensile bi-axial strain outside QD ($\varepsilon_{||}>0$) due to stretching of GaAs to match InAs. Compressive strain in QD.
- Lattice constants follows same trends.

Local Bandstructure in an Alloyed QD





In-As bonds compressed in x-y

-> Ec raised from bulk value of ~0.58eV to ~1.2eV

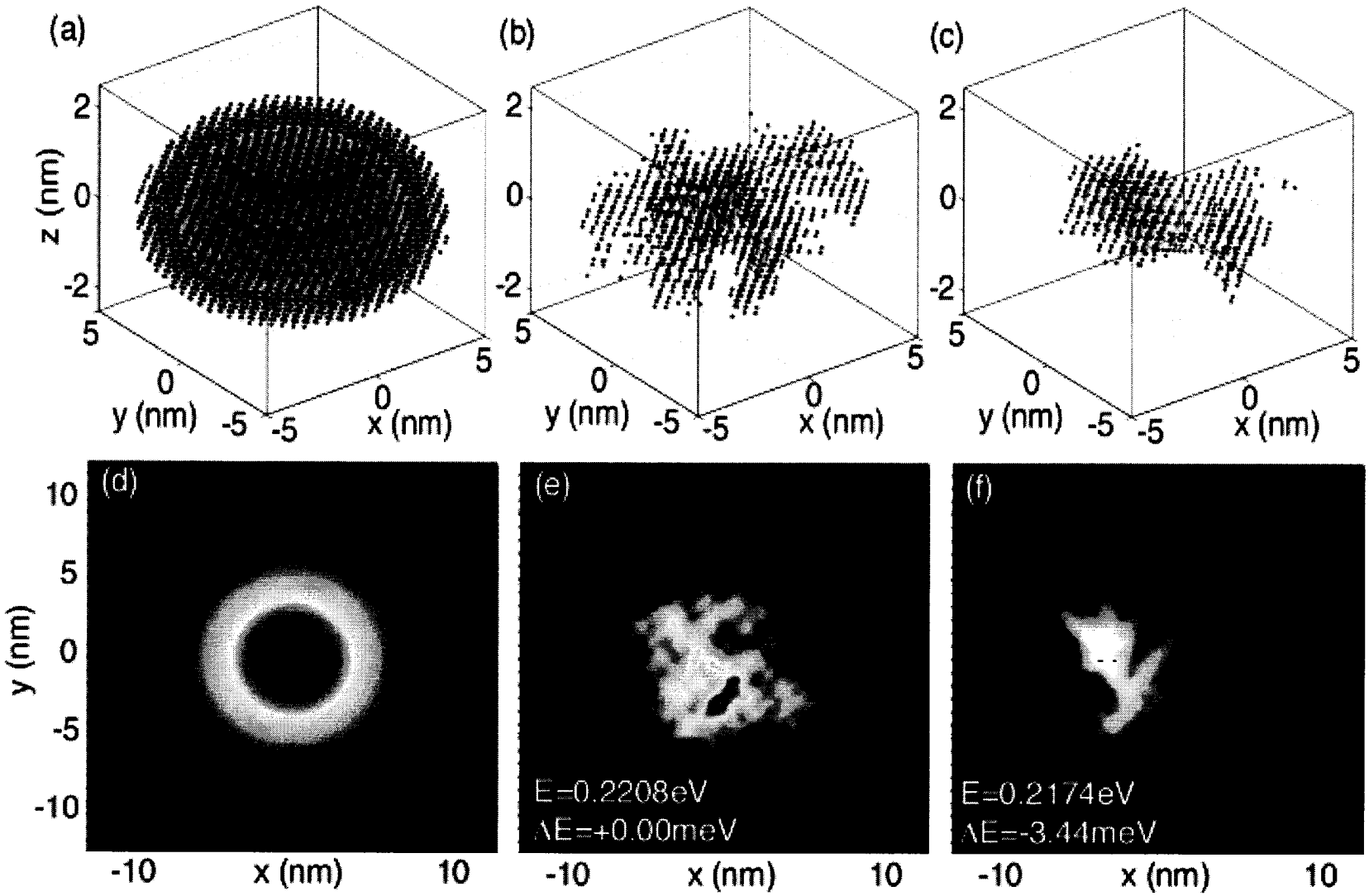
-> Ev HH raised from bulk value of ~0.22eV to ~0.3eV

Ga-As bonds compressed in x-y and stretched in z inside dot

-> Ec raised from bulk value of ~1.42eV to ~1.55eV

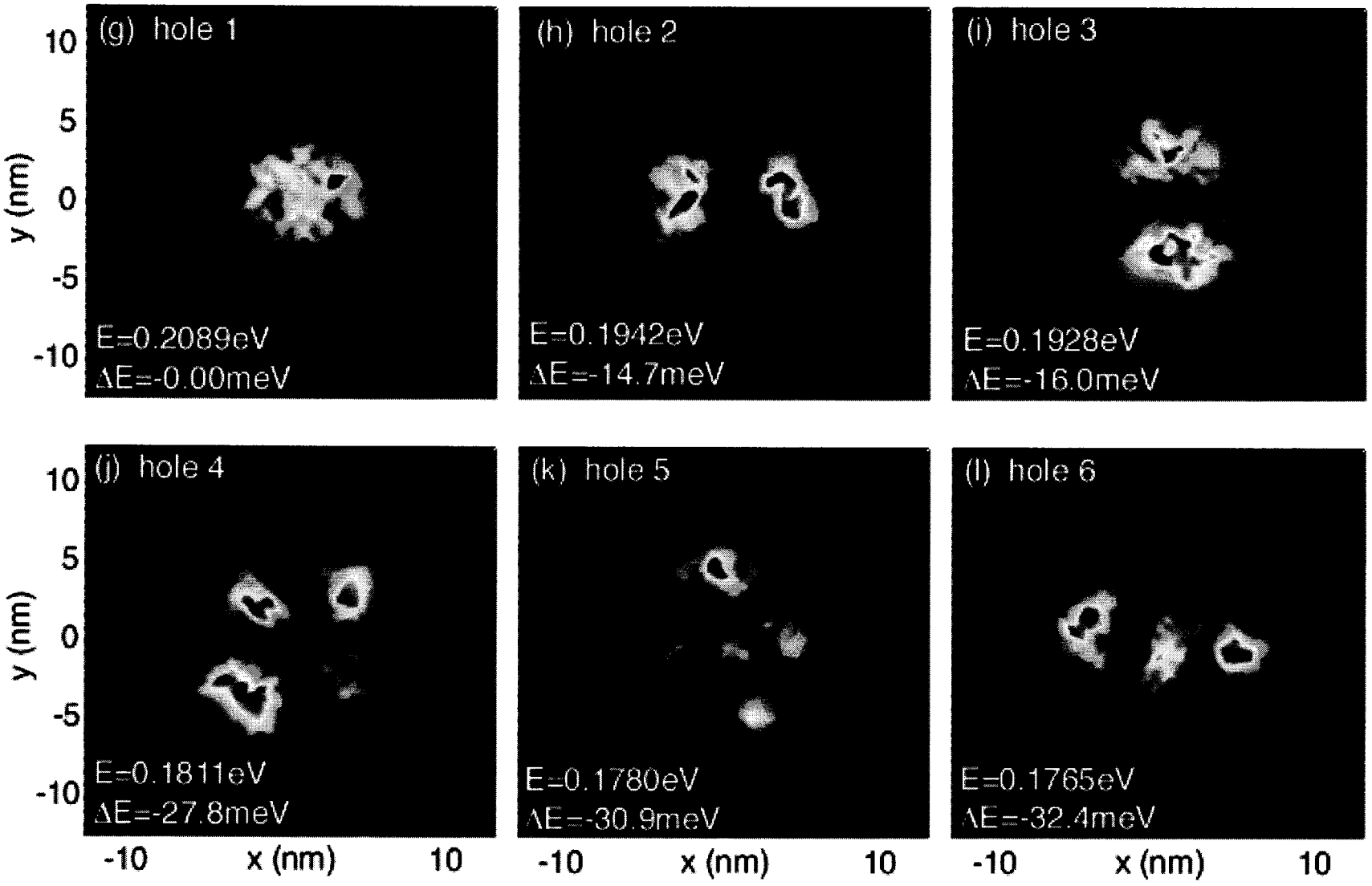
-> Ev raised from bulk value of 0eV to ~0.1eV

Spatial Irregularity in the Hole Ground State

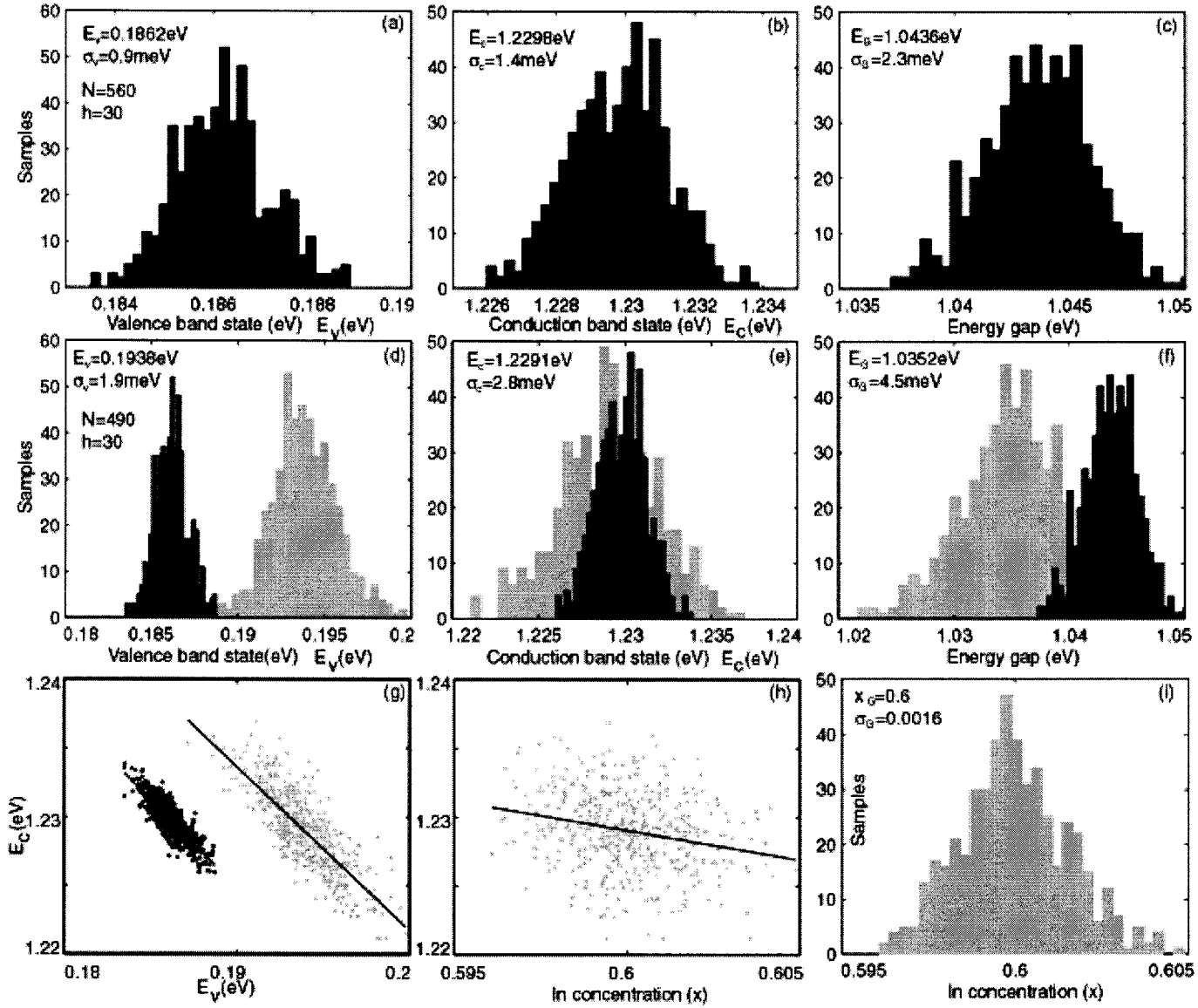


VCA / no Disorder | Disorder Sample 1 | Disorder Sample 2

Spatial Irregularity in the Hole States



>1000 Alloyed Quantum Dot Samples

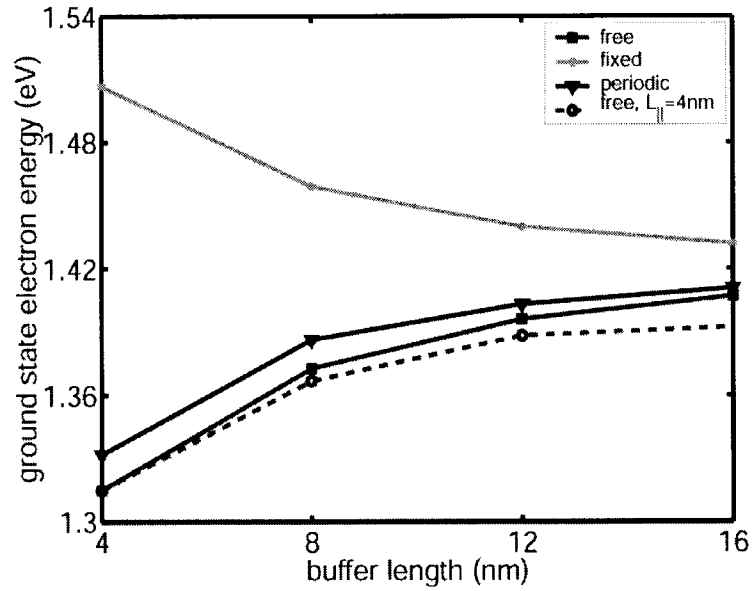


- Atomistic granularity
  σ = 2.3meV
- Cell granularity
  σ = 4.5meV
- Ec and Ev strongly correlated
- Ec and x weakly correlated

Issues with Boundary Conditions

Effect on ground state electron energy:

- System: Dome-shaped $In_{0.6}Ga_{0.4}As$ QD 15nm radius; 5.4 nm height
- ***Free** 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 ($k_{supercell}$=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.

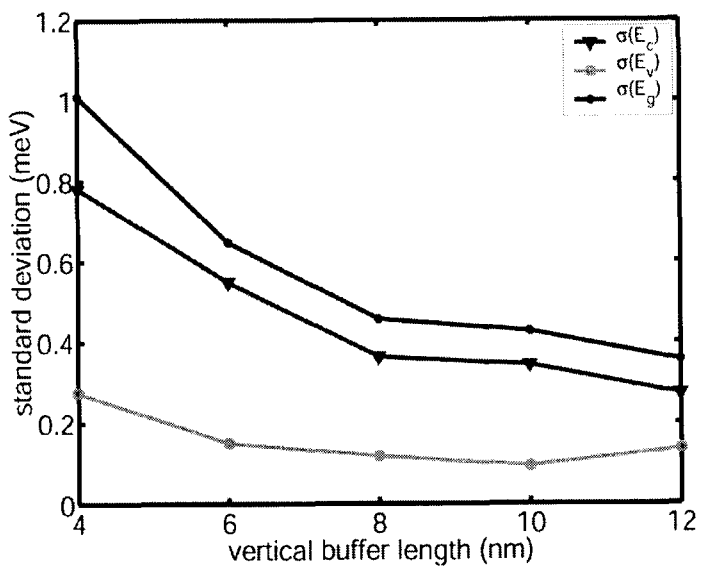
Disorder-induced Linewidth

Question: What is the contribution of alloy disorder to linewidth broadening?

- Previous PL experimental results have found large contributions due to inhomogenous broadening (~30 meV) [R. Leon et al., PRB, 60, pR8517]
- Single QD PL measurements have found narrow linewidths (~0.9 meV) [Nagamune, APL, 67, p3257]

Computation:

- Use a 'direct sampling method' (roughly 100-200 samples)
- Assume no correlation between location of In, Ga cations within the QD



Results:

- Convergence is slow but can place an upper bound of 0.35 meV, so the effect is small.
- Caveats:
  - Have not included interface interdiffusion
  - Variation is larger if there is short-range order (clustering).