Multi-Million Atom Electronic Structure Calculations for Quantum Dots

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Ph.D. Research Work Summary

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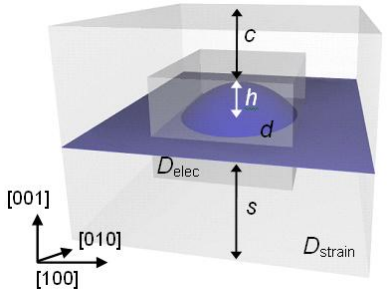
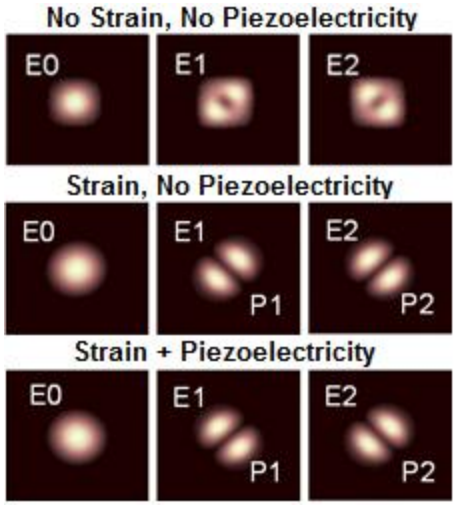
Outline

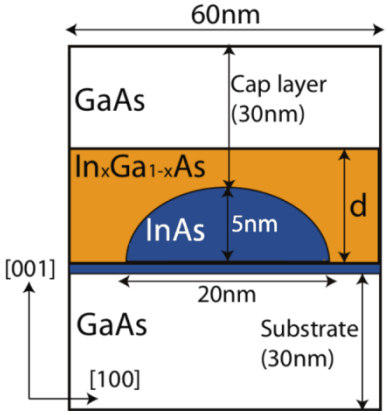
(1) Single Quantum Dot



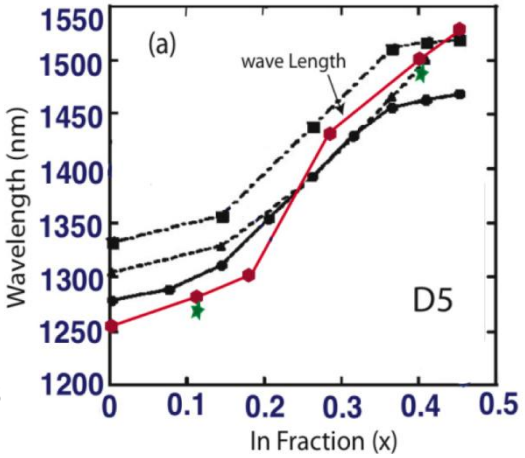


- Atomistic Interface
- Strain
- Piezoelectricity
- Optical Transitions

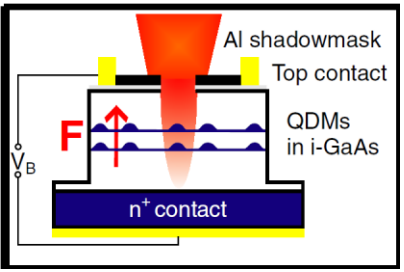
(2) Single Quantum Dot inside InGaAs-SRCL

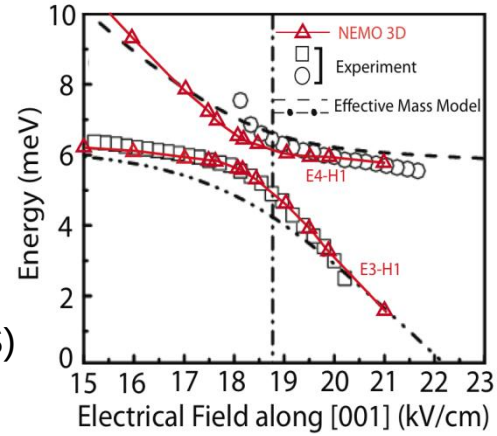


1500nm Optical Emissions



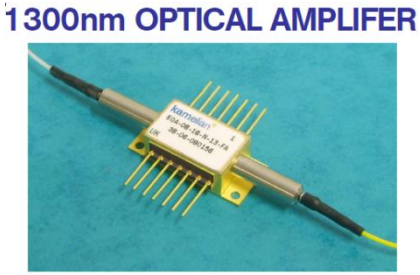
(3) Bilayer Quantum Dot Stack

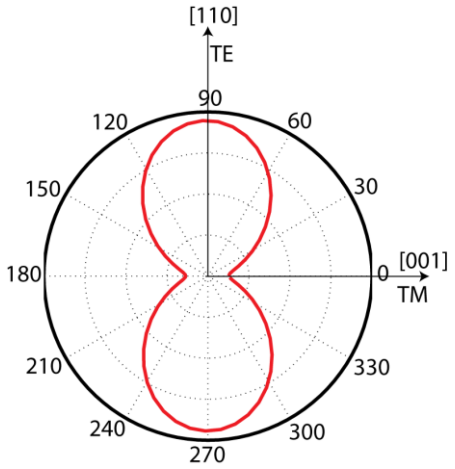




- Level Anti-crossing Spectroscopy (LACS)
- Exciton Tuning
- Piezoelectricity

(4) Polarization Resolved Optical Emissions

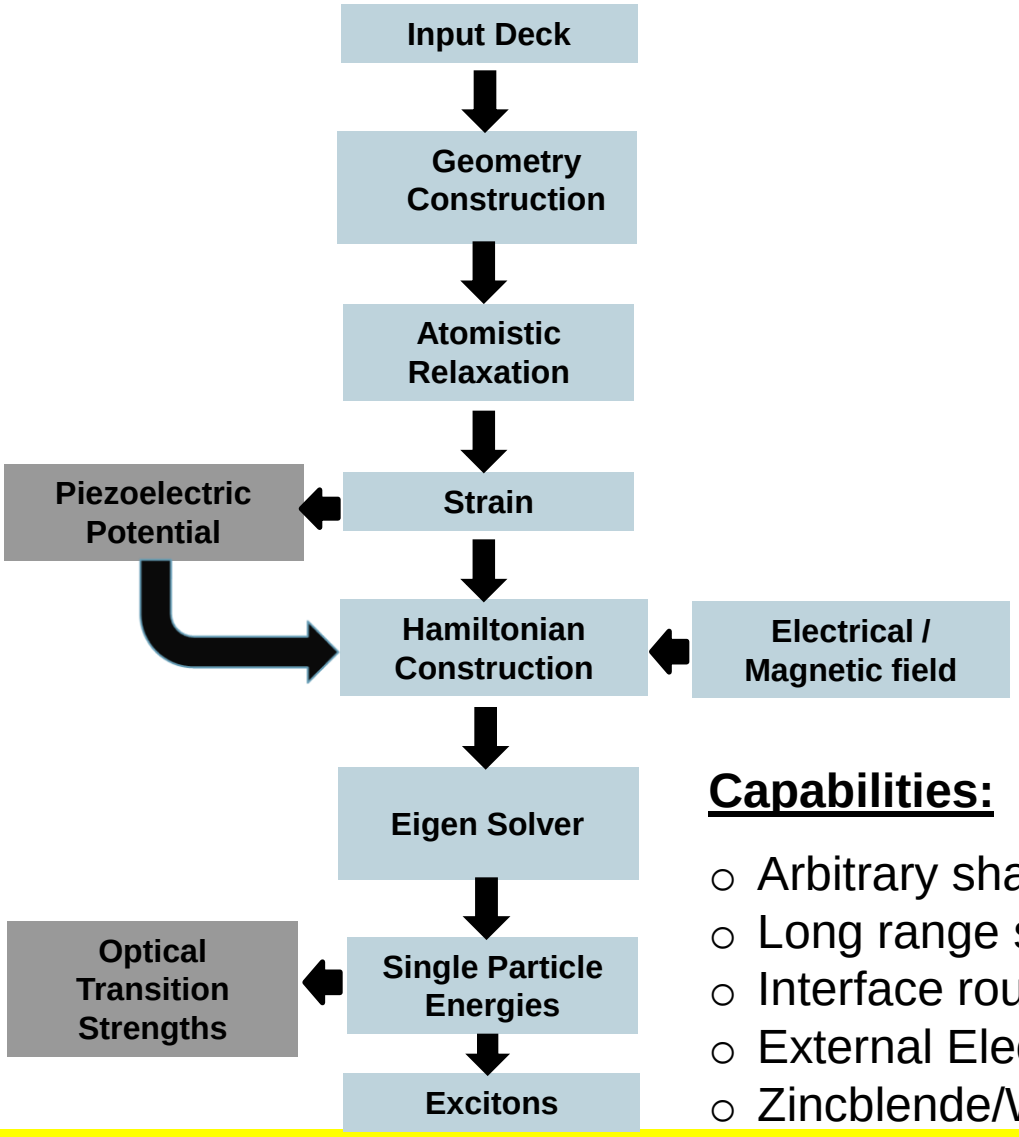




- Single vs. Bilayers of QDs
- 1300-1500nm Emissions
- Polarization-resolved Optical Transitions

NEMO 3-D – A fully atomistic simulation tool

Program Flow Chart:



Methodology:

- Experimental geometry in input deck
- Strain is calculated using Valence Force Field (VFF) Method
- Linear and Quadratic Piezoelectric potentials by solving Poisson's equation over polarization charge density
- Empirical tight binding parameters -- $sp^3d^5s^*$ band model with spin orbital coupling
- Multi-million Atom Simulations

Capabilities:

- Arbitrary shape/size of quantum dot
- Long range strain, piezoelectric fields
- Interface roughness, atomistic representation of alloy
- External Electrical/Magnetic fields
- Zincblende/Wurtzite crystals

Ref: IEEE Transactions on Electron Devices, Vol. 54, Issue 9, Sept. 2007, Page(s):2079 - 2089 (2007)

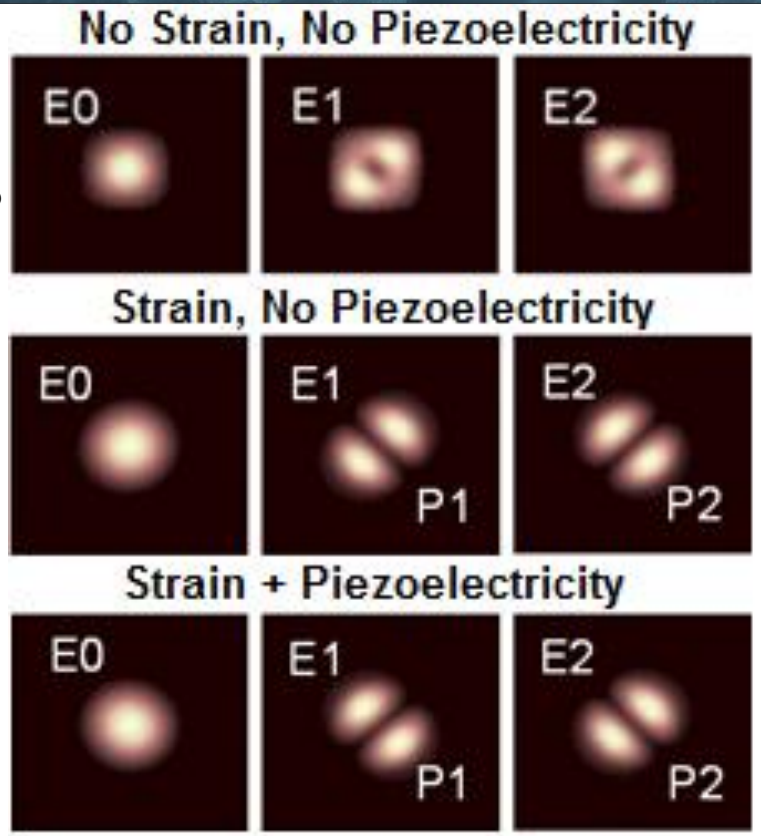
Fine Structure Splitting in QDs Crystal Symmetry, Strain, and Piezo-Electricity

Objective:

- Understand experimental data on QD spectra fine structure
- Electron P_x and P_y states are split in energy – Why?
- critical physical items

Approach:

- Implement piezo-electric charges through electrostatic potential
- Compare effects of
  - Crystal Symmetry
  - Strain
  - Piezoelectricity
- Disk shaped dots d=10nm, h=2.5nm, 10nm cap, 20nm substrate, 0.6nm wetting layer



Impact:

- Demonstrated quantitative agreement with experiment
- Atomistic approach is essential

Ref: (1) Springer Encyclopedia for Complexity, 2008
(2) proceedings of IEEE NEMs Jan 16-19, 2007, pgs. 937-942, Bangkok Thailand.

Impact:

- Crystal symmetry alone breaks symmetry of set of first excited states (weak)
- Strain breaks symmetry stronger
  => effective mass, k.p fail!
- Piezoelectric effect opposes strain
  => can flip orientation of the excited state

InAs Bilayer QD Stacks --- Strain Coupling between the QDs --- Atomic vs. Molecular Electronic Structure

Objective:

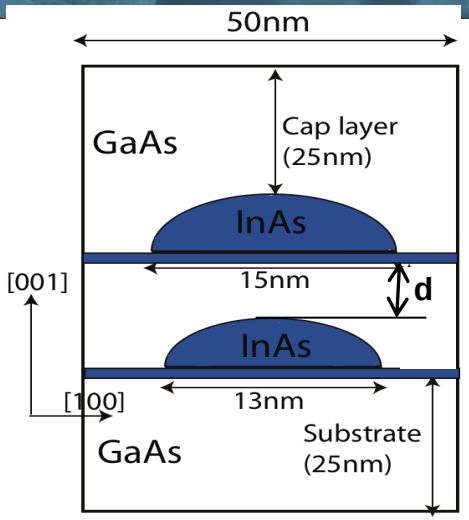
- Calculate electronic structure of a bilayer QD stack with varying dot-to-dot separation “d”
- Determine strain coupling between the QDs as a function of “d”
- Demonstrate atomic and molecular electronic states

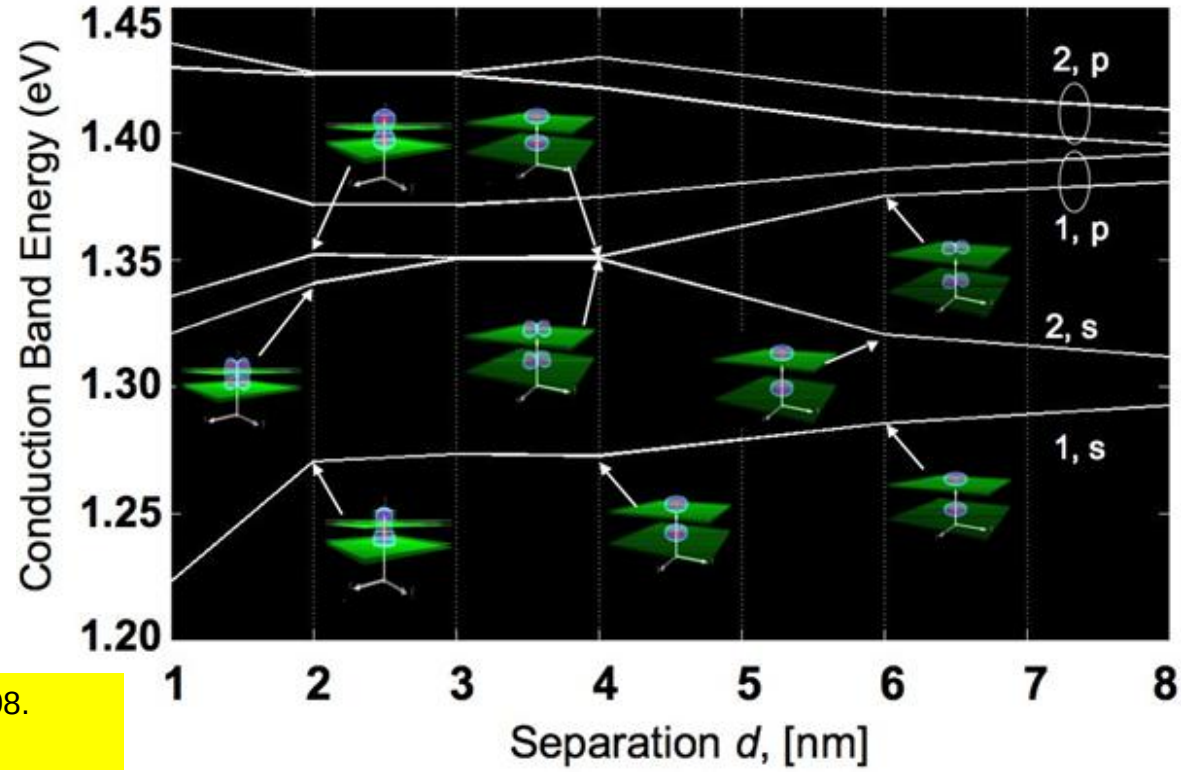
Approach:

- Bilayer QD Stack – Dome Shaped QDs – Upper QD slightly larger than lower QD
- “d” varies from 1nm to 8nm
- Strain over ~15Million atoms
- Electronic Structure ~10Million atoms

Impact:

- Long range strain couples the QDs
- Lowest electron and hole states reside in larger QD
- Small “d” → Molecular States
- Large “d” → Atomic States
- Cut off “d” ~ 6nm





Ref: 8th IEEE Conference on Nanotechnology, 2008. NANO '08. Aug. 18-21 2008, Page(s):541-544

First Electron and Hole Wave function Symmetry – Lateral Electrical Field – Polarization Dependent Optical Transitions

Objective:

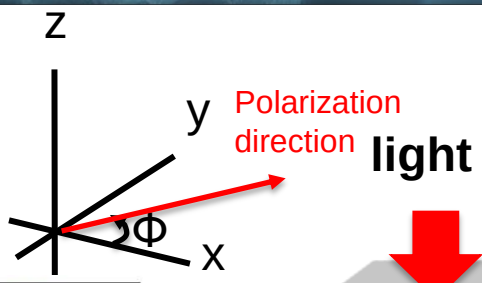
- Calculate polarization dependent inter-band optical transition rate
- Impact of in-plane electrical field on electronic structure and optical transitions

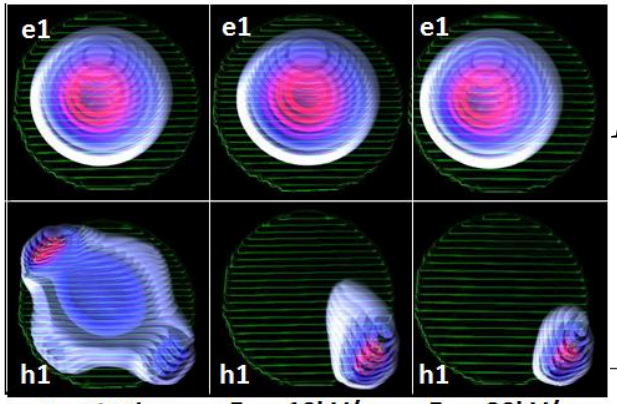
Approach:

- Optical transition strength as squared magnitude of momentum matrix element summed over spin degenerate states using Fermi's golden rule
- [100] E field from 0 to 20kV/cm

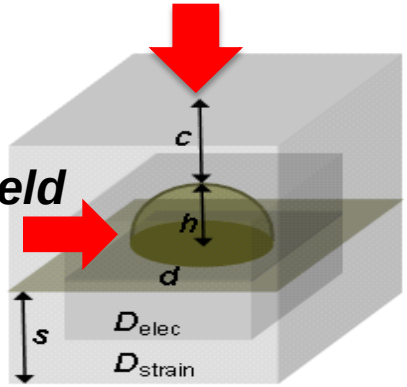
Impact:

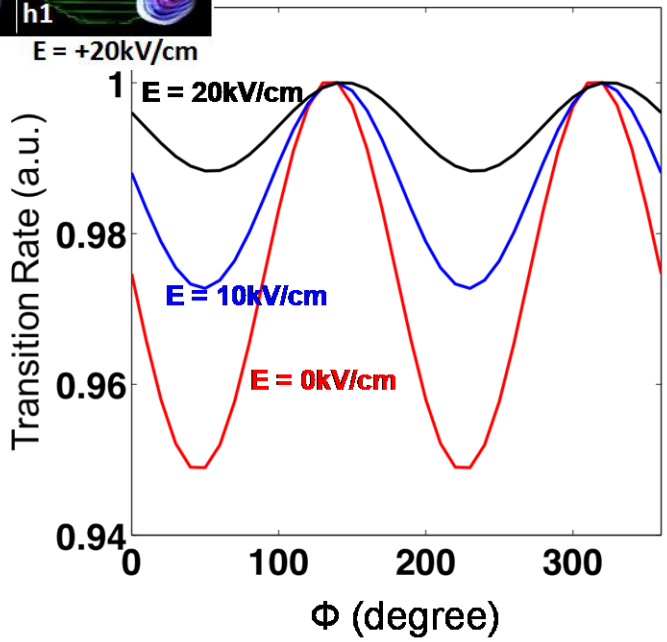
- Hole wave function is aligned along [-110] direction due to strain and piezoelectric field effects
- Sinusoidal pattern of transition rate is due to asymmetric hole wave function distribution
- Electrical field shifts the hole wave function
  → lesser electron-hole overlap
  → optical transition rate reduces





$\vec{E}$ *Field*





Ref: IEEE proceedings of the 13th International Workshop on Computational Electronics, Tsinghua University, Beijing, May 27-29 2009.

Quantitative Quantum Dot Modeling Selective Overgrowth and Wavelength Tuning

Objective:

- Optical emission at 1.5μm without GaN
- Understand experimental data on QD spectra in selective overgrowth

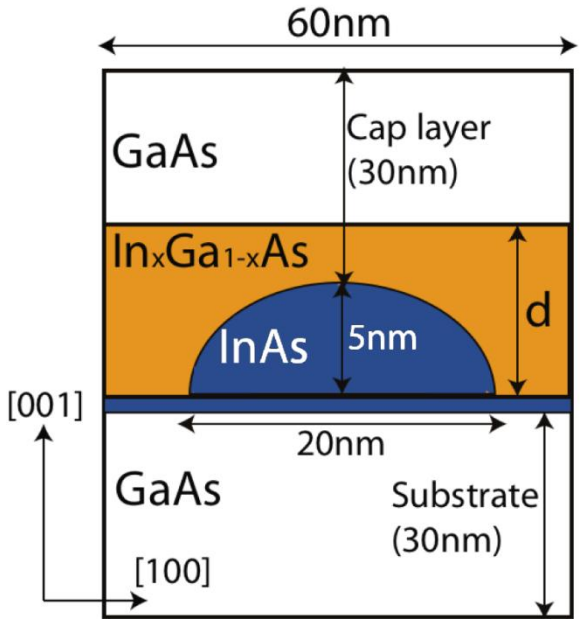
Approach:

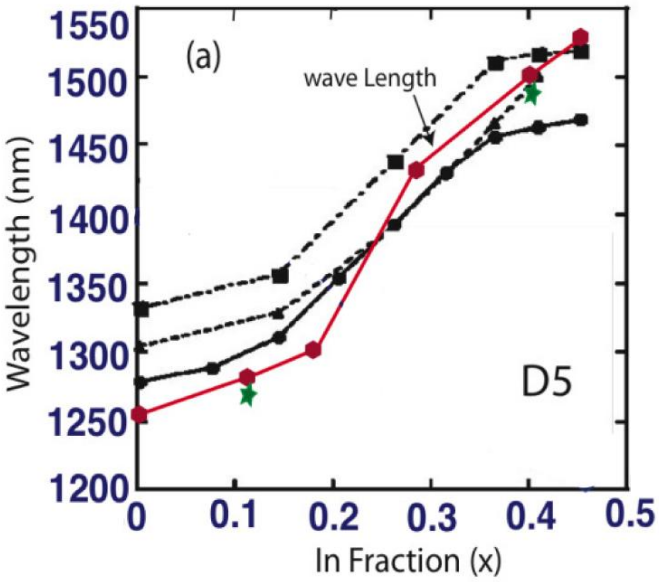
- Model large structure
  - 60nm x 60nm x 60nm
  - 9 million atoms
- No changes to the published tight binding parameters

Impact:

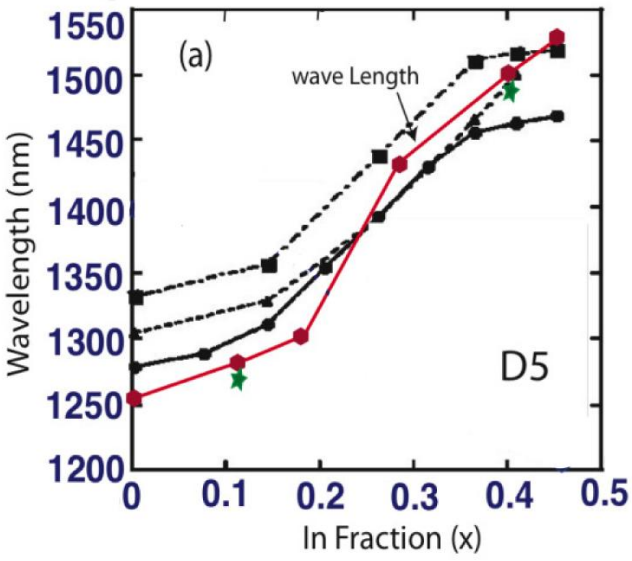
- Match experiment remarkably well
- Experimental red shift is contributed by:
  - Strain
  - change in quantum dot aspect ratio
- Quantitative model of complex system

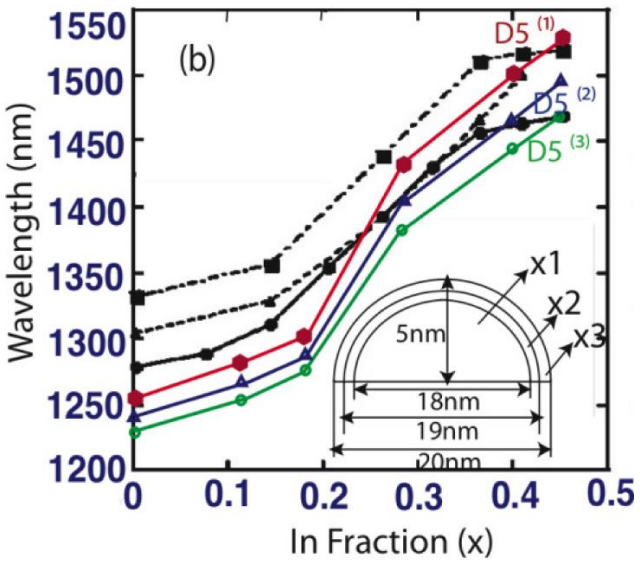
Ref: IEEE Transactions on Nanotechnology, Vol. 8, pp. 330-344 (2009)

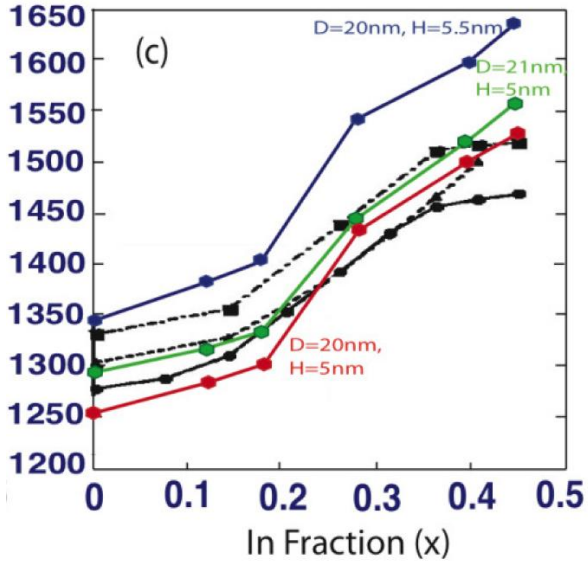




Selective Overgrowth and Wavelength Tuning Structural Sensitivity







Questions – How important are:

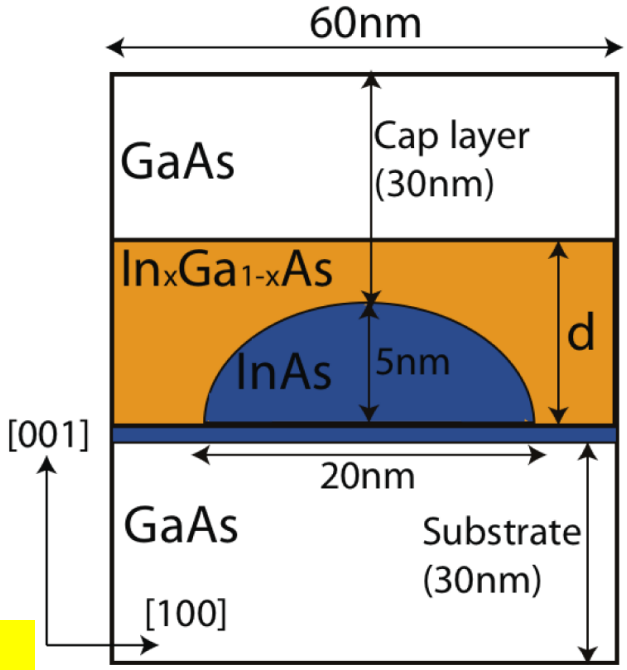
- Interface details - gradients
- Size variations

Numerical Experiment:

- Introduce interfacial gradients over 2nm (100, 90, 80%) and (100%,70%,60% InAs)
- Samples of size variation (20/5.5nm) and (21/5nm) vs. nominal (20/5nm)

Results:

- Weakly sensitive!!!



Ref: IEEE Transactions on Nanotechnology, Vol. 8, pp.330-344 (2009)

Bilayer QD Molecule
Electrical Field Tuning of Excitons

Objective

- Coupling of bilayer QD electronic structure under [001] electrical field
- Understand experimental optical strengths
  → identification of electronic states

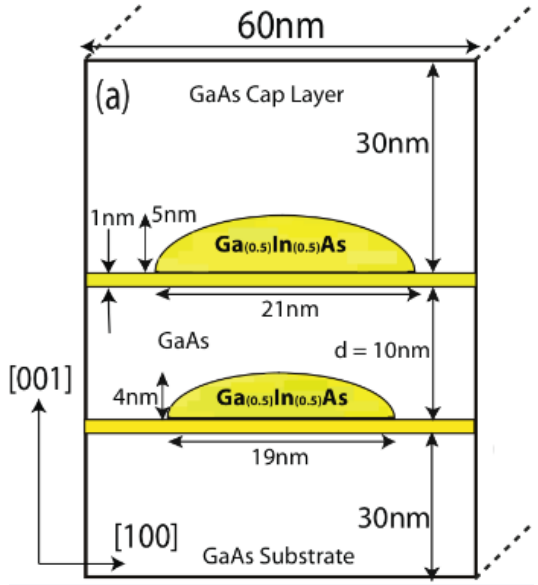
Approach

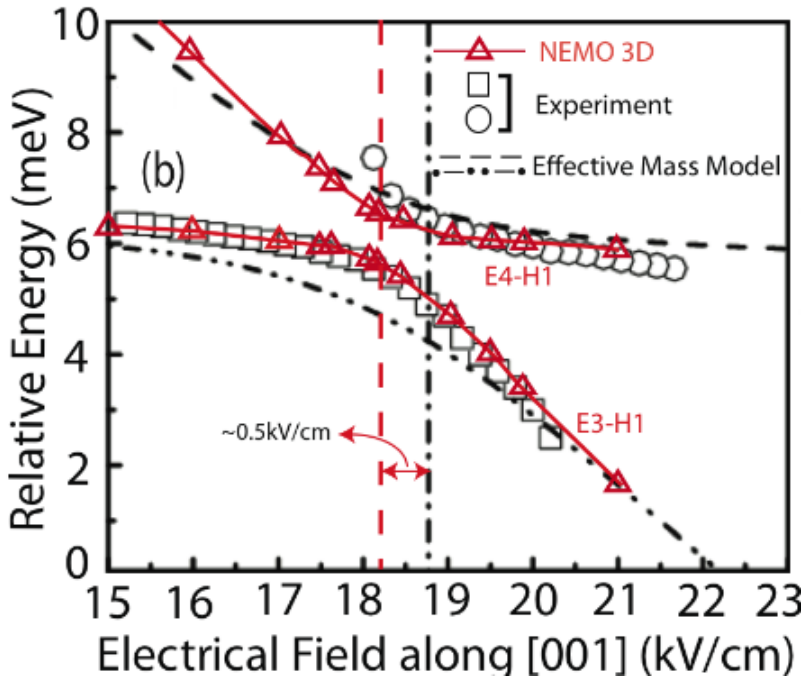
- Electronic structure calculation
  → Experimental device geometry
  → [001] Electrical field from 15 to 23kV/cm
  → Strain, Linear and Quadratic piezoelectricity
- Optical transition strengths

Impact:

- First hole energy level H1 always in upper QD
- First two electron energy levels E1, E2 are in the lower QD in the range of applied field
  → E1-H1, E2-H1 are dark excitons
- E3 and E4 exhibit inter-dot tunneling
  → E3-H1 and E4-H1 anti-cross at 18.4kV/cm
- Calculated anti-crossing quantitatively match experimental measurement

Ref: under review ACS Nano 2010
Full Paper at arXiv:????





Bilayer QD Molecule Ladder Anti-crossing Spectroscopy

Objective:

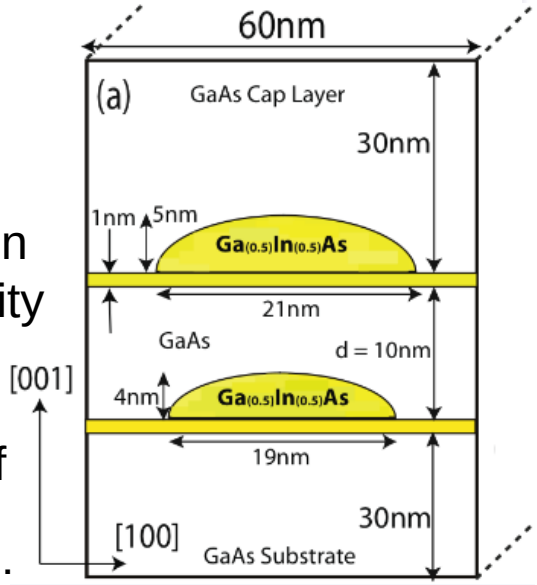
- Calculate electronic structure of bilayer QD stack as a function of [001] electrical field
- Demonstrate anti-crossings between electron and hole states with increasing field

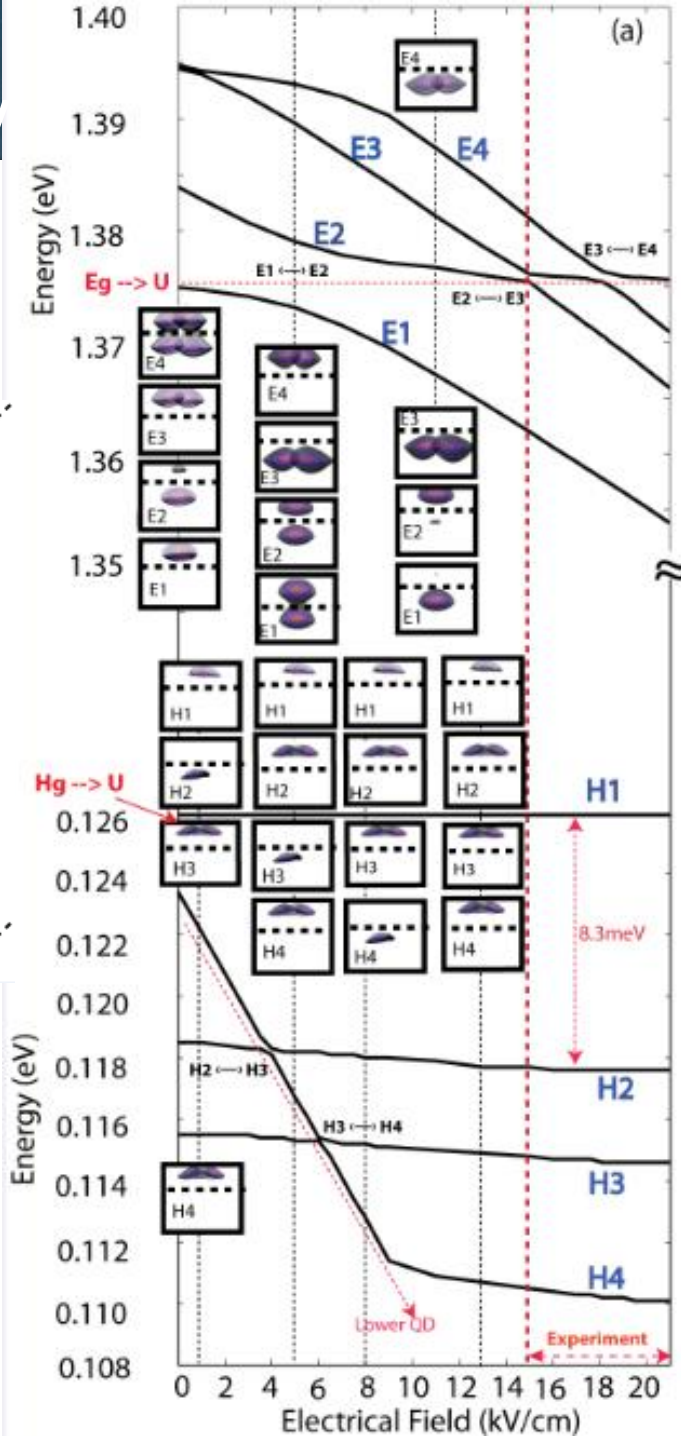
Approach:

- Experimental device geometry
- Multi-million atom electronic structure calculations with strain
- Linear+Quadratic piezoelectricity
- H1 is taken as reference

Impact:

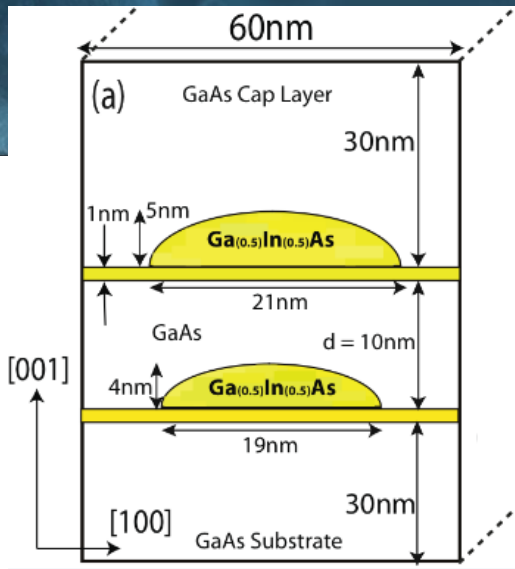
- Probing the electronic states of one QD by anti-crossings with electronic states of second QD:
  - → Symmetry of states
  - → Energy level ladder
- Determination of tunneling coupling energies
- From energy level slopes:
  - → Separation between QD centers ~ 13.3nm
  - → Intra-dot electron-hole separation ~ 1.67nm





Ref: under review ACS Nano 2010
Full Paper at arXiv: ???

Bilayer QD Molecule
Critical Importance of Piezoelectricity



Objective:

- Demonstrate field dependence of E2-H1, E3-H1, and E4-H1 excitons with and without piezoelectricity
- Calculate optical transition strengths and compare with experimental measurements

Approach:

- Calculation of excitonic spectra and optical transition strengths with and without piezoelectricity
- Linear+Quadratic Piezoelectricity

Impact:

- With Piezoelectricity

→ E2-H1 is dark

→ E3-H1↔E4-H1 Anticross

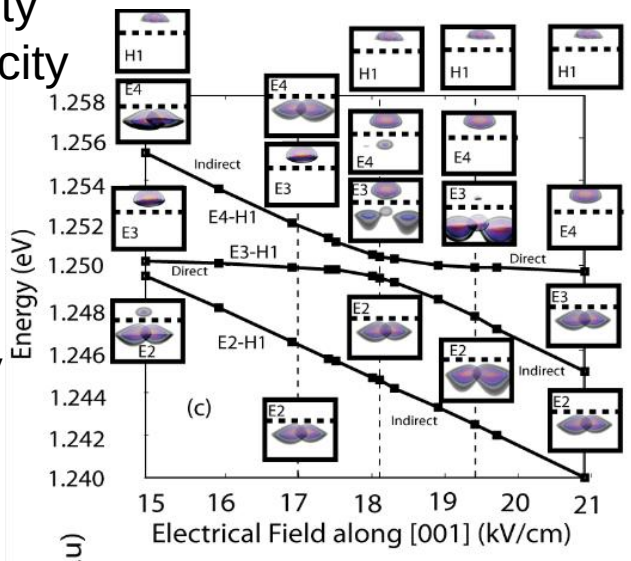
→ Calculated transition intensity match experiment

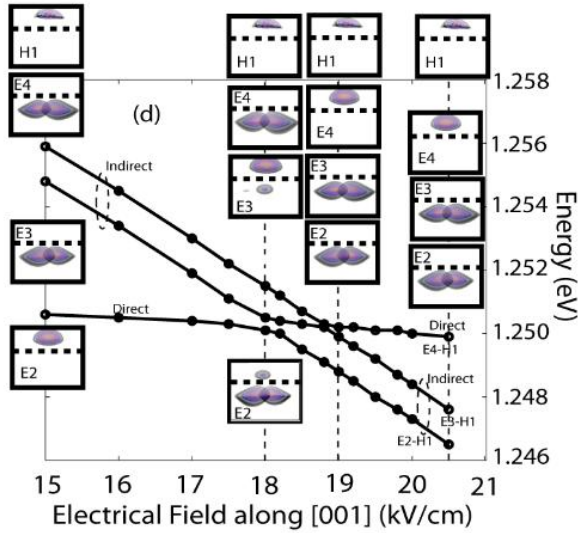
- Without Piezoelectricity

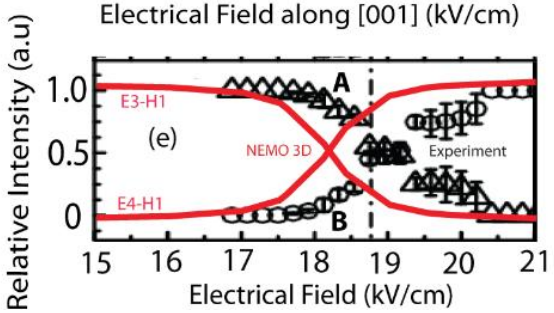
→Two Anti-crossings

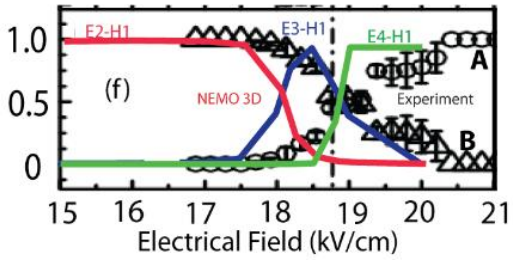
E2-H1↔E3-H1, E3-H1↔E4-H1

- Incorrect optical spectrum!
- Piezoelectricity is critical!









Ref: under review ACS Nano 2010
Full Paper at arXiv: ????

Self Assembled Single and Bilayer QDs --- Polarization dependent Optical Spectra --- TE/TM Ratio Tailoring

Objective:

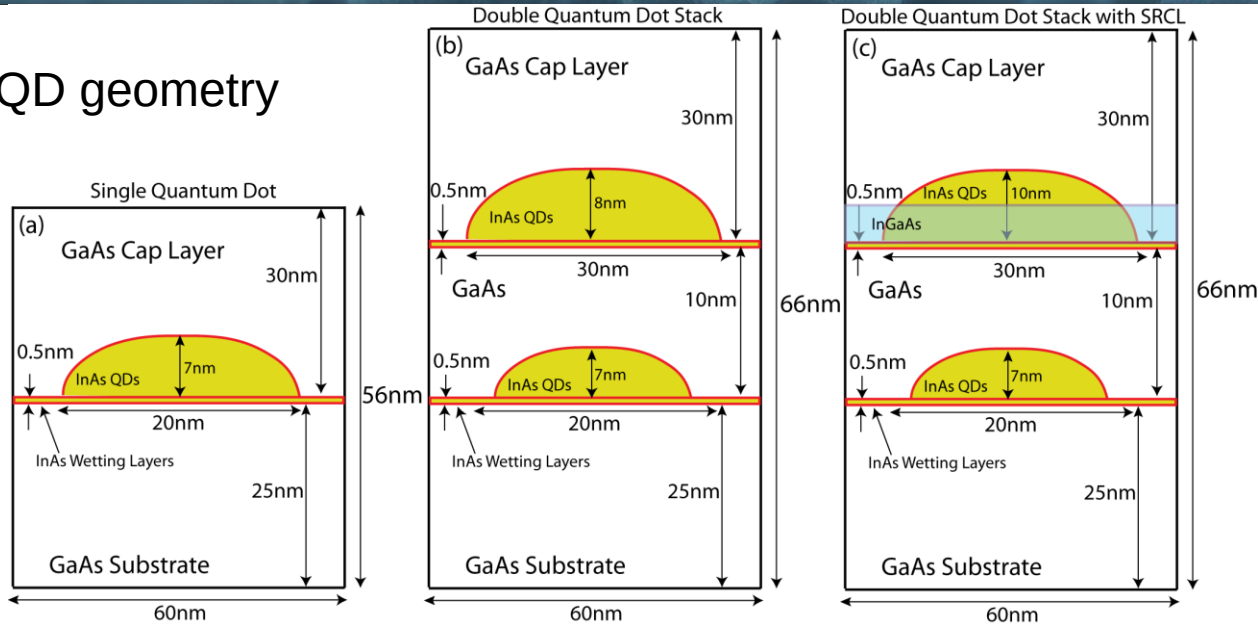
- TE/TM Ratio tailoring using QD geometry parameters

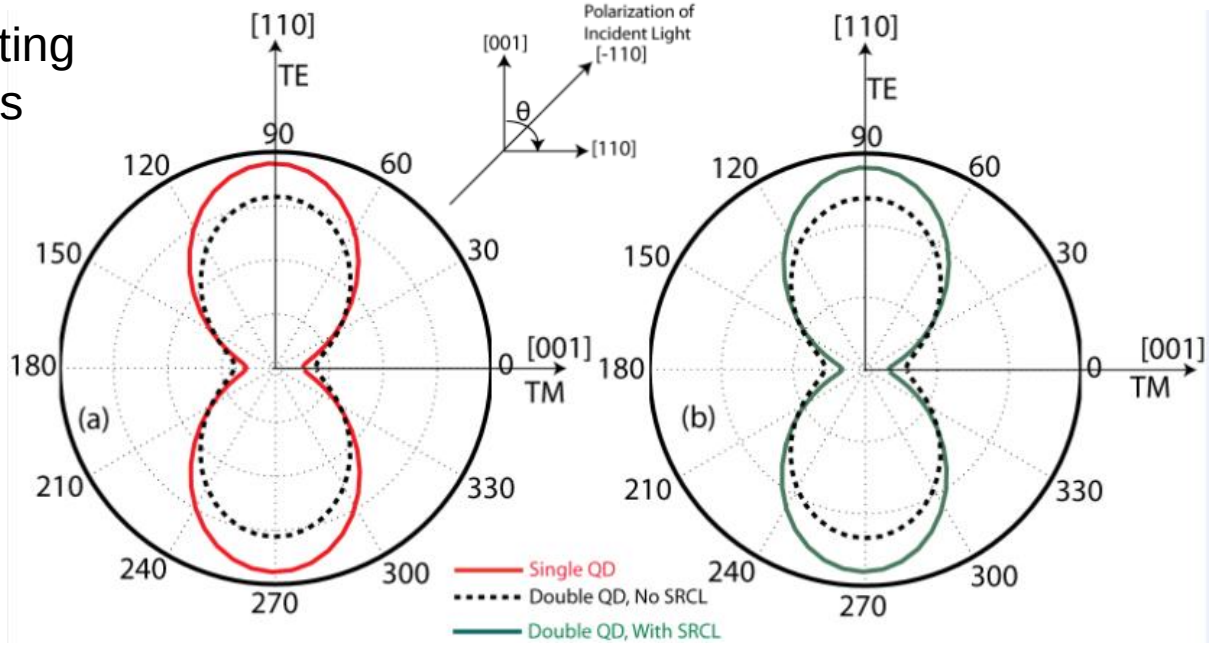
Approach:

- Calculation of TE[110] and TM[001] optical transition strengths and comparison with experiment

Impact:

- Bilayer QD exhibit lowest TE/TM ratio
- InGaAs increases HH-LH splitting and TE/TM ratio also increases
- NEMO 3-D trends match experiment





(c) TE/TM Ratio Table

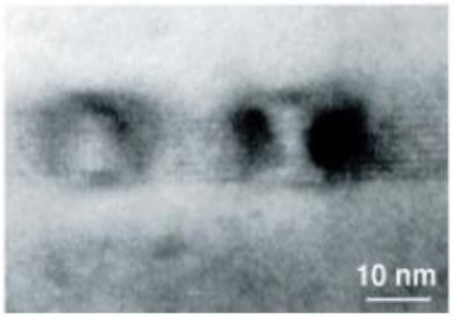
	Experiment	NEMO 3-D
Single QD No SRCL	~4.0	~7.3
Double QD No SRCL	~3.5	~4.06
Double QD With SRCL	~5.2	~8.7

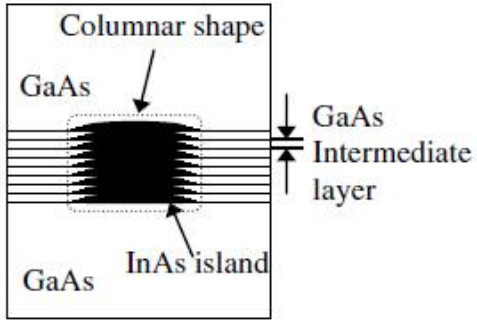
Outlook

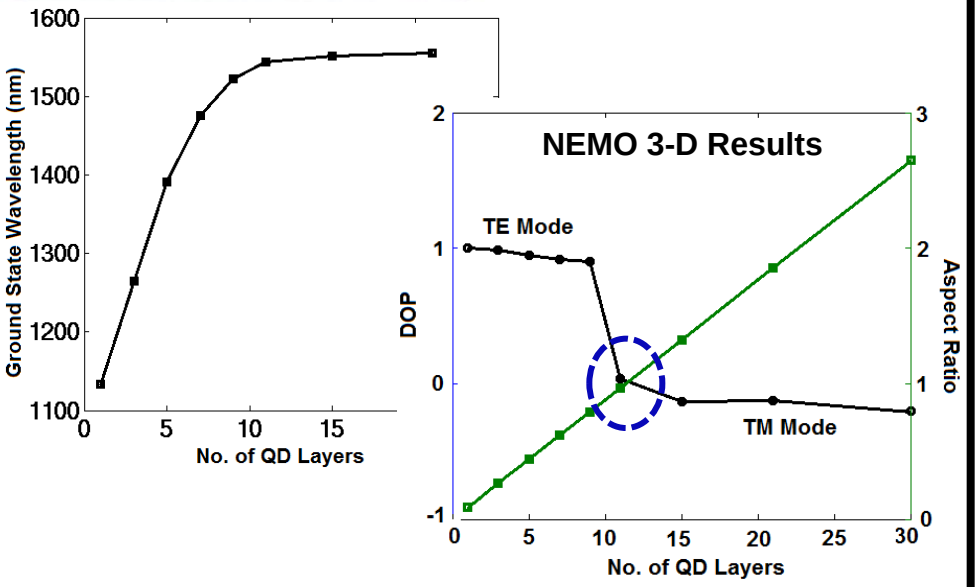
Short term projects

Columnar Quantum Dots

Ref: phys. stat. sol. (c) 0, No. 4, 1137– 1140 (2005)



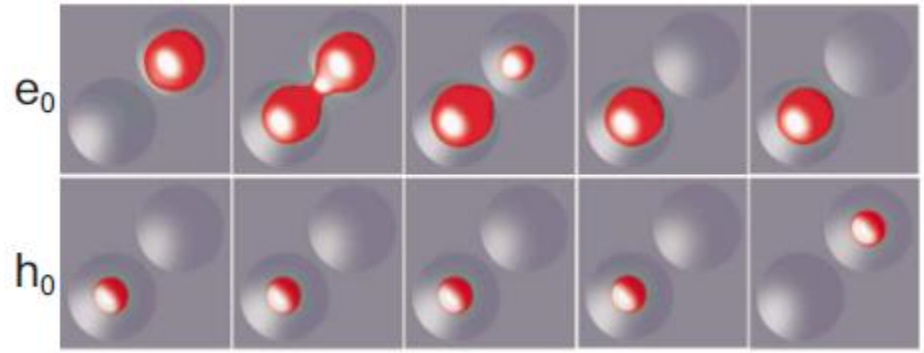


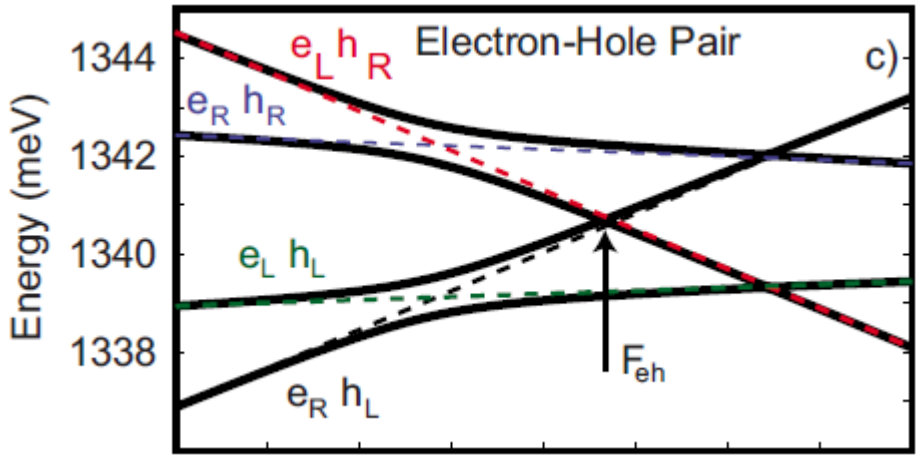


- Large Stacks of InAs QDs
- [001] confinement is relaxed
- No. of QDs ~ 9-11 → TE[110] ~ TM[001]
- Theoretical design recipe for devices

Laterally Coupled QD Molecules

Ref: PHYSICAL REVIEW B 81, 205315 (2010)



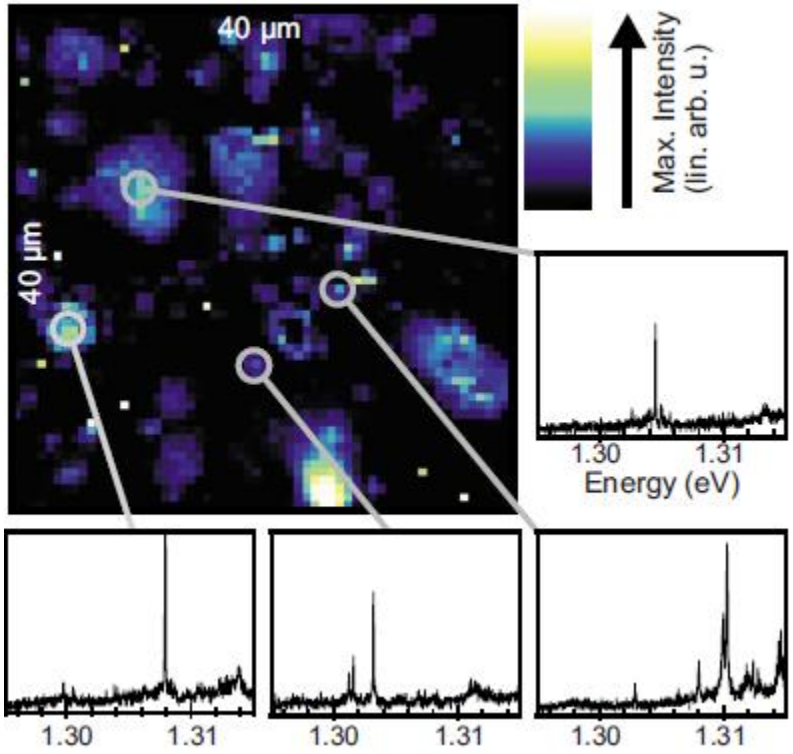


- Laterally coupled QDs for Quantum Information Science
- External electrical field determines the dot-to-dot coupling
- Little theoretical guidance available to-date

Long term projects

QDs Grown on (111) Substrates

Ref: Appl. Phys. Lett. 96, 093112 (2010)



- QDs grown on [111] substrate are potential candidates for entangled photons i.e. biexciton → exciton → 0
- Lowest symmetry is C_{3v}
- Piezoelectricity is along the growth direction and does not lower the symmetry

Growth Simulation → NEMO 3-D

Ref: Cryst. Res. Technol., 1-6 (2009), Wiley Inter Science

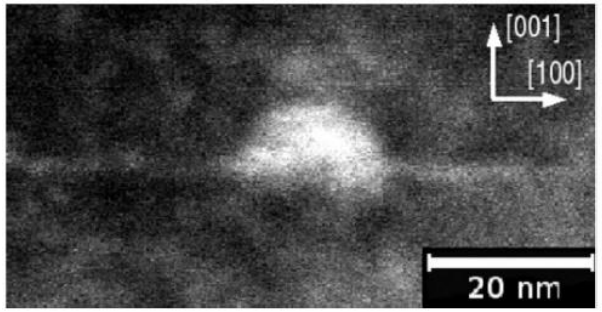


Fig. 4 HAADF STEM image of the InGaAs quantum dot under investigation. One and the same quantum dot was characterized by CELFA (see Figs. 5 and 6).

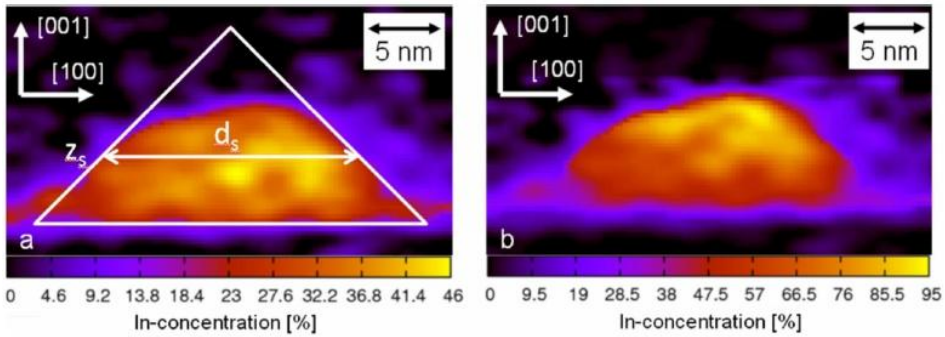


Fig. 5 Color-coded local In-concentration of a QD a) as obtained by CELFA with superimposed {101}-

- Only little is known about QDs
- Shape, Size, Composition, 'In' Segregation?
- Electronic/Optical Spectra have strong dependence on geometry of QDs
- Possibility to drive fully atomistic electronic structure calculations of NEMO 3-D by simulations of the self-assembly growth process?