Electronic and optical properties of quantum dots

T. K. Johal

National Nanotechnology Laboratory INFM– Unita di Lecce , Dip. Ingegneria, Dell'Innovazione,73100 Via Per Arnesano, Lecce

National Nanotechnology Laboratory of INFM

Outline of lectures

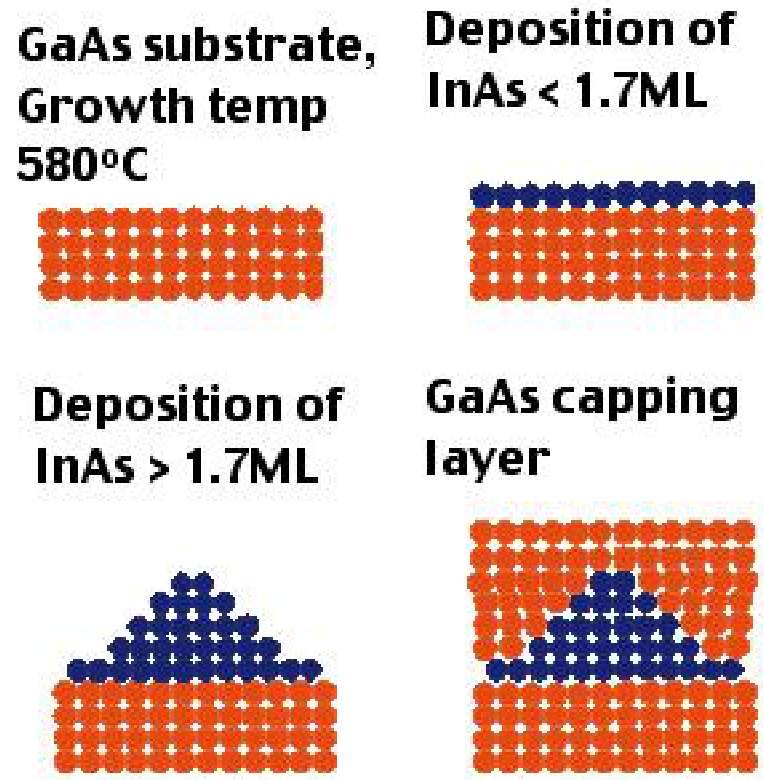
- Basics of quantum dot
- Structural properties
- Ensemble QD properties
- Theoretical methods and predictions
  - Envelope function approach
  - **k.p** method
  - Pseudopotential method
- The experimental reality
  - Single QD tunneling spectroscopy
  - Single QD optical properties
  - Single QD tunneling current induced optical properties

Semiconductor quantum dot

- Narrow band gap material nanostructure embedded in wide band material
- dimensions are comparable to the effective Bohr radius of the host semiconductor material
- Energy and charge quantization
- Occupation follows shell structure
- Can be treated like an atom
- Building block for QD molecules

Self assembled, strain driven InGaAs QD growth

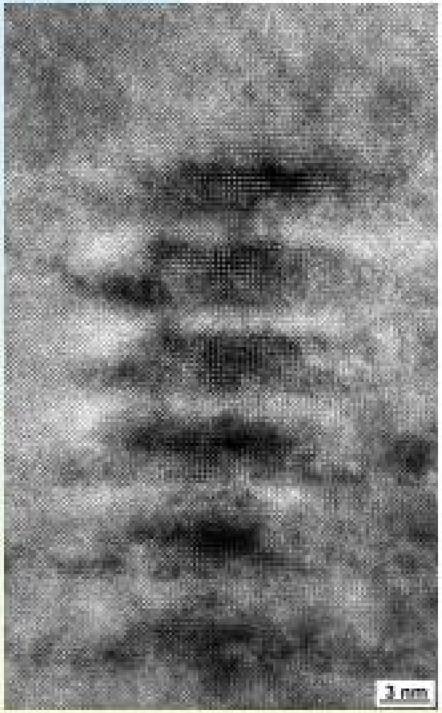
- Stranski–Krastinov, beyond critical thickness, 2D–3D transition, coherently strained islands
- Narrow size and shape distribution
- Growth process is thermodynamically driven, long range elastic interaction



National Nanotechnology Laboratory of INFM

V.A. Shchukin and D. Bimberg, Rev. Of Mod. Phys. 71 (1999) 1125

Correlated growth of stacked InGaAs quantum dots

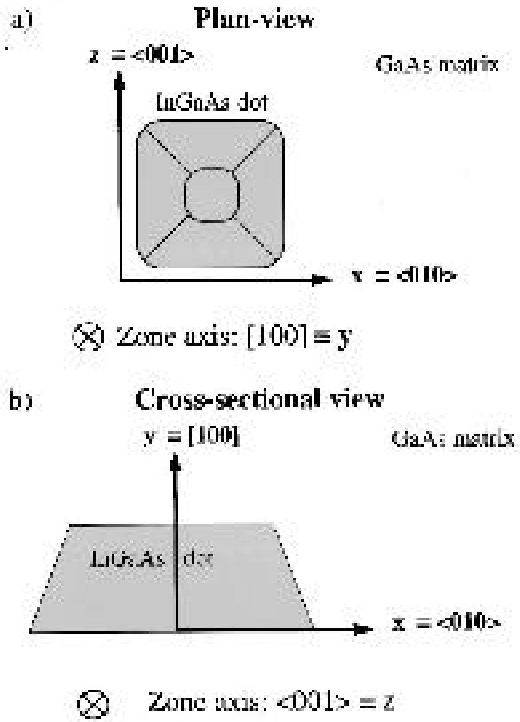


- Strain propagated growth
- High densities achievable
- Ability to stack dots for the active material of laser devices
- TEM–Phase, strain and *chemica*l contrast--zone axis along the [001]-direction

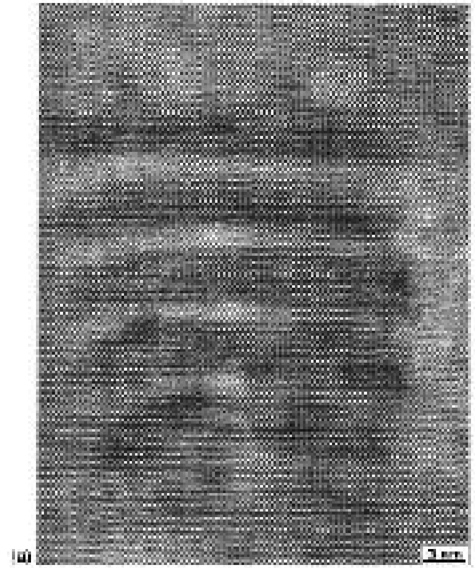
A. Passaseo, R. Rinaldi, M. Longo, S. Antonaci, A. L. Convertino, R. Cingolani, A. Taurino and M. Catalano, J. of Appl. Phys. -- . 89 (2001) 4341

National Nanotechnology Laboratory of INFM

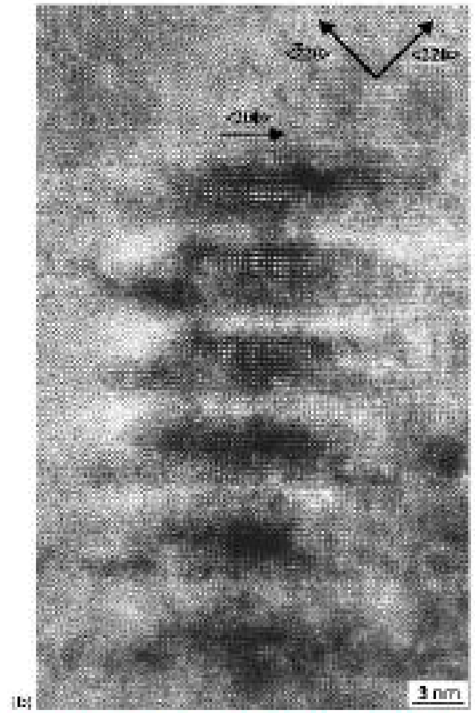
Strain field constrast Vs chemical contrast



[011] zone axis

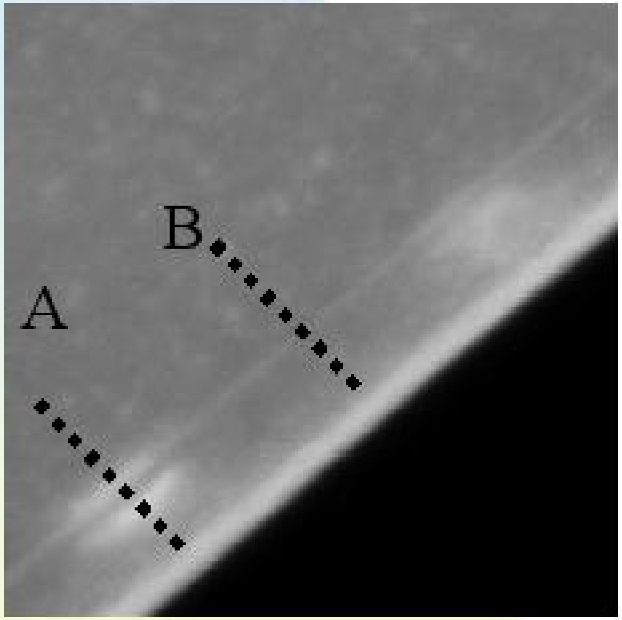


[001] zone axis

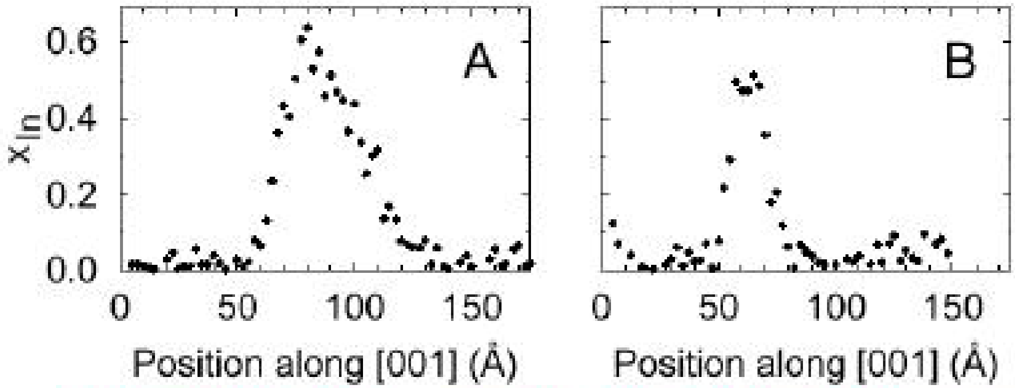


M. De Giorgi , A. Taurino ,A. Passaseo , M. Catalano ,and R. Cingolani, Phys. Rev. B, **63**, 245302

Direct measurement of compositional enrichment in InGaAs quantum dots.



- Employ electron energy loss spectroscopy, monitor In content
- In enrichment in the centre of the dot



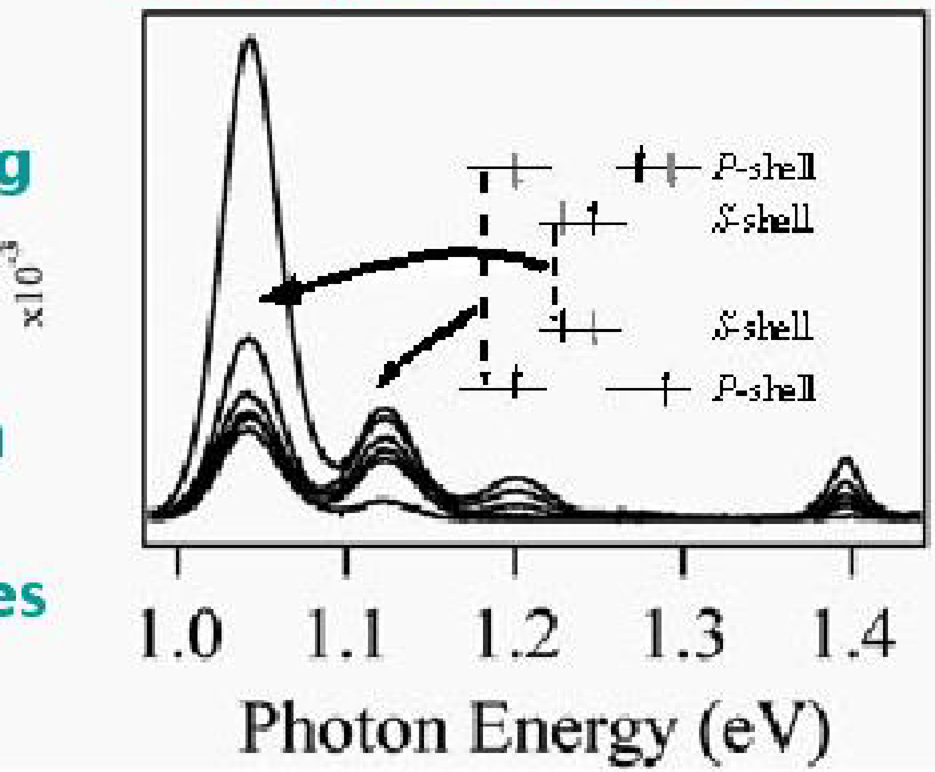
J. Shumway, A. J. Williamson, Alex Zunger, A. Passaseo , M. DeGiorgi, R. Cingolani, M. Catalano and P. Crozier Phys. Rev B, **64**, (2001) 125302

NNL National Nanotechnology Laboratory of INFM

QD ensemble properties: Photoluminescence

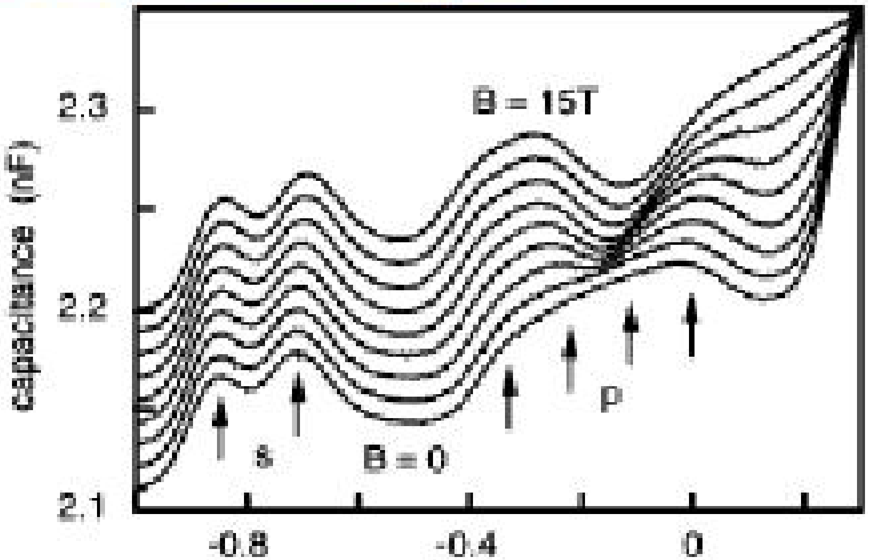
- Broad QD spectra, at least 30 meV broad
- Power dependence, evidence of state filling
- Ground state exciton energy
- With higher excitation power emission from excited excitonic states
- Intra-level spacing

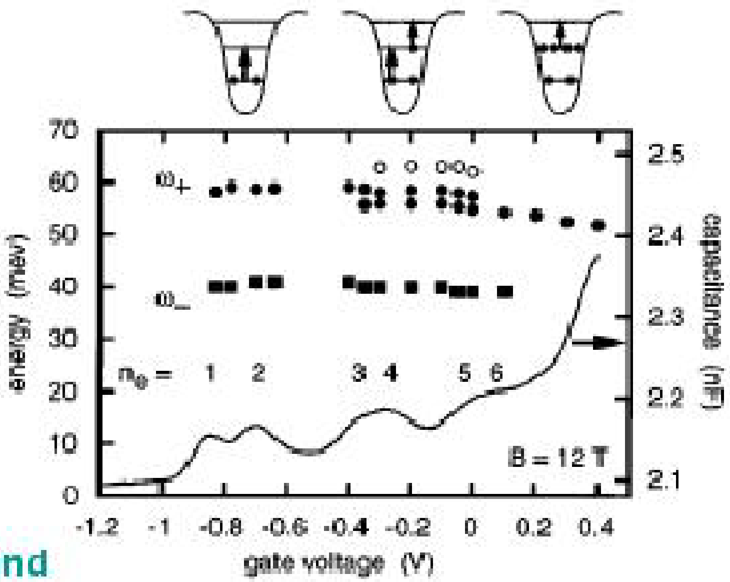
Illumination by Ar laser, 488nm line, sample temperature 30K



Single-electron charging in large-scale quantum dot arrays

- Combination of capacitance and far infrared absorption spectroscopy
- Gated QD device, electrostatically charge QD
- Measure perturbative effect of magnetic field
- Charging energy directly reflects th small overlap between the s and p state as compared to two s or two p states
- Shows the influence of the electronic shell structure on the Coulomb repulsion





M. Fricke, A. Lorke, J. P. Kotthaus, G. Medeiros-Ribeiro and P. M. Petroff, Europhys. Lett. **36** (1996) 197

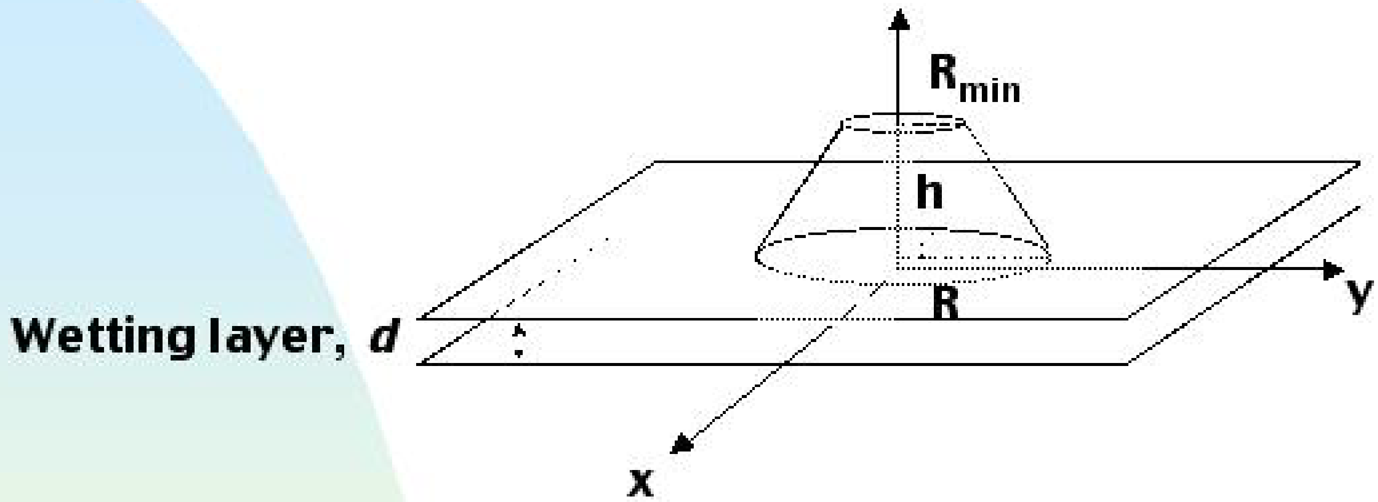
What defines a quantum dot?

- Confining potential is defined by
  - Band structure of semiconductor
  - 3-dimensional shape of QD,
  - QD size,
  - compositional gradient
    - Variations in alloy content
  - strain-field
    - Piezoelectric effects
- Energy level scheme derived from experiments
  - Inter band level spacings
  - Intraband electron and hole spacings
  - Wetting level energy position

Theoretical methods 1:

- A phenomenological approach,
- envelope function approach employing the effective mass approximation, assume parabolic bands
- Has successfully predicted ground-state eigenvalues and inter-level separations
  - Capacitance-voltage characteristics of QDs (Ph. Lelong, O. Heller and G. Bastard, Solid State Electr. **42**, 1251 (1998))
  - Far infrared magneto-absorption between conduction band states of QD (S. Hameau, Y. Guldner, O. Verzelem, R. Ferreira, G. Bastard, J. Zeman, A. Lemaitre and J.M. Gerard, Phys. Rev. Lett., **83**, 4152 (1999)

A phenomenological approach



Assume the QD shape is a truncated cone, *because of the* **cylindrical symmetry** can solve the Schrodinger equation analytically,

$$\psi_{nl}(\bar{r}) = \frac{e^{il\vartheta}}{\sqrt{2\pi}}\psi_{nl}(\rho, z)$$

National Nanotechnology Laboratory of INFM

with *l=0* for S-like levels, *l=±1* for P-like levels...

For the bound levels we consider the **separable form**

Single particle wavefunction

Envelope function

$$\psi_{nlp}(\rho, z) = F_{nl}(\rho)\varphi_{nlp}(z)$$

$$F_{nl}(\rho) = N_{nl}\rho^{|l|}P_{nl}(\rho^2)\exp\left(-\frac{\rho^2}{2\beta_{nl}^2}\right)$$

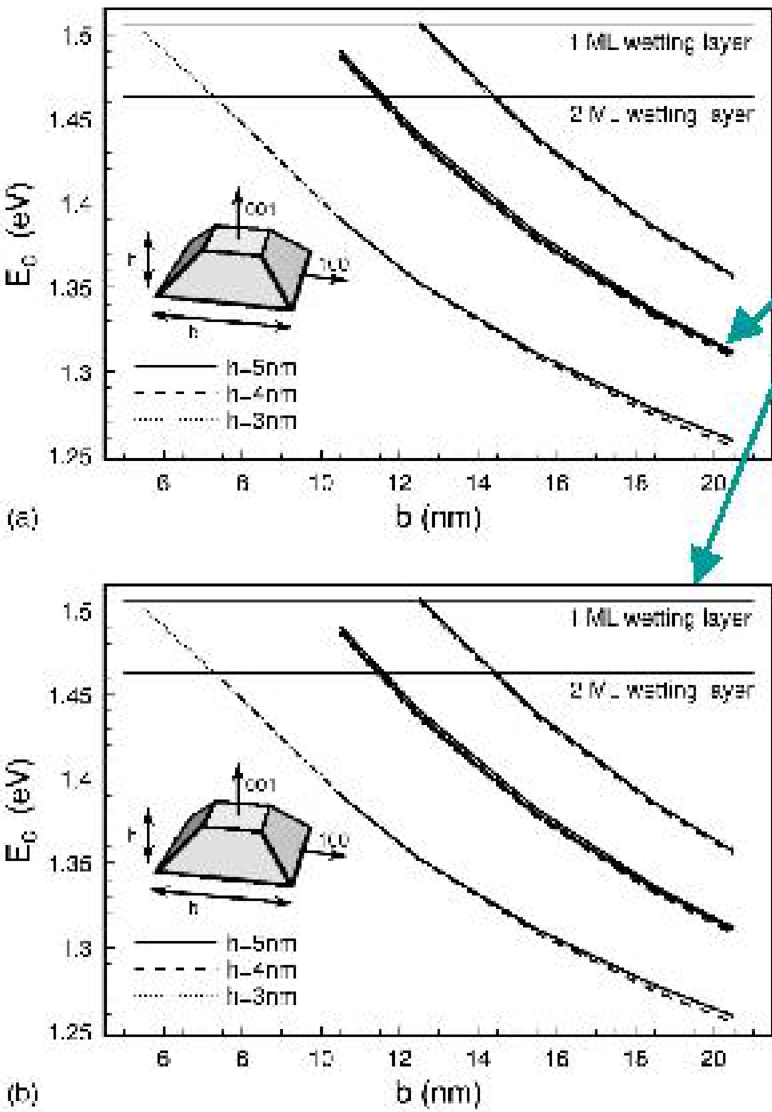
$$P_{nl}(\rho^2) = \sum_{p=0}^{n-1} a_{p,(nl)}\rho^{2p}$$

Where N_{nl} are normalization constants and $a_{0,nl}=1$, the coefficients are obtained by othornormalization.

Theoretical methods 2: *k.p* methods

- Calculate single particle energy levels of QD of arbitrary shape and composition
- Complexities of band structure included, VB mixing and CB-VB mixing
- As inputs
  - Bulk bands structure parameters
  - Bulk elastic properties
  - Size, shape and composition of QD
- Strain in, and in the vicinity of QD calculated using either valence force field model (Pryor, Phys. Rev. **B 60** (1999) 2869) or elastic continuum model (Stier *et al.* Phys. Rev. **B 59** (1999) 5688)
- Piezoelectric effects included

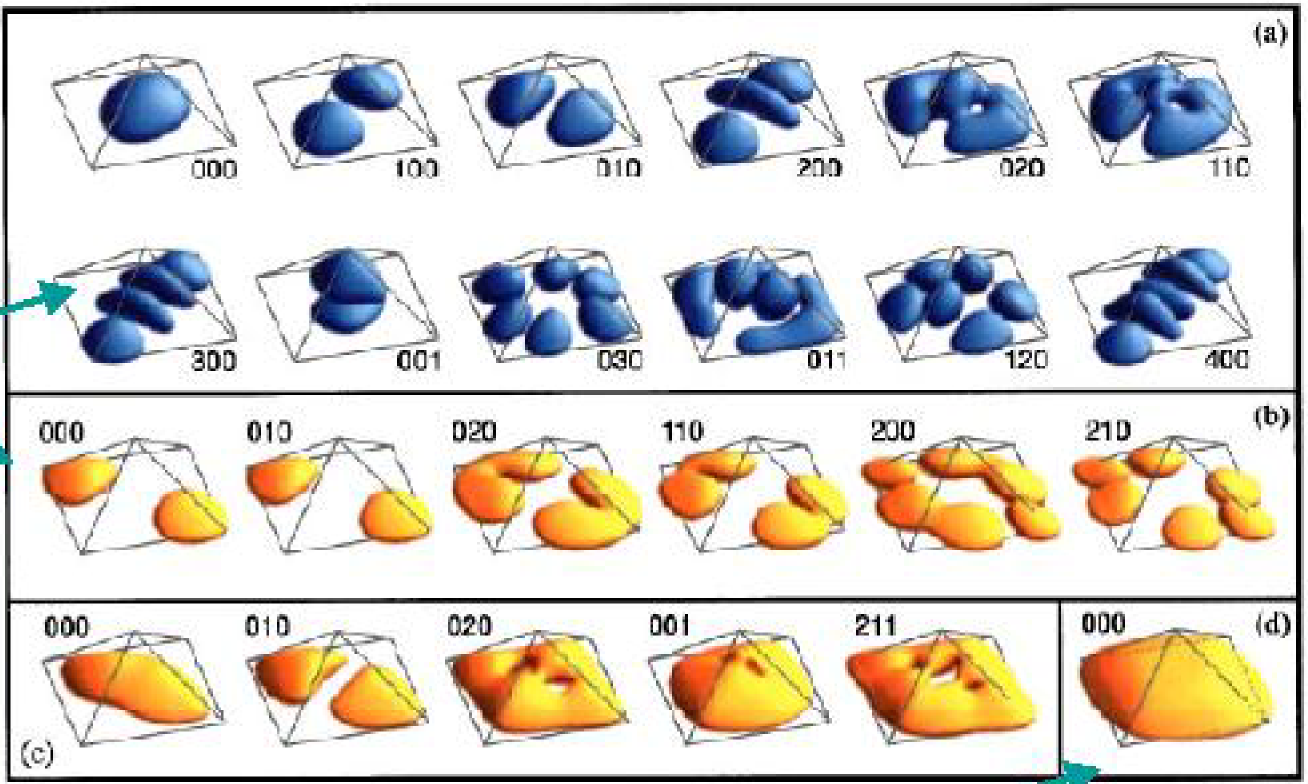
k.p methods



- Calculated electron and hole confinement energies
- Strain contribution most sensitive to the base length
- 10% error in geometry of dot caused 50 meV error in predicting exciton emission energy

k.p methods

Electron and hole states for base length=20.4nm, strain included.



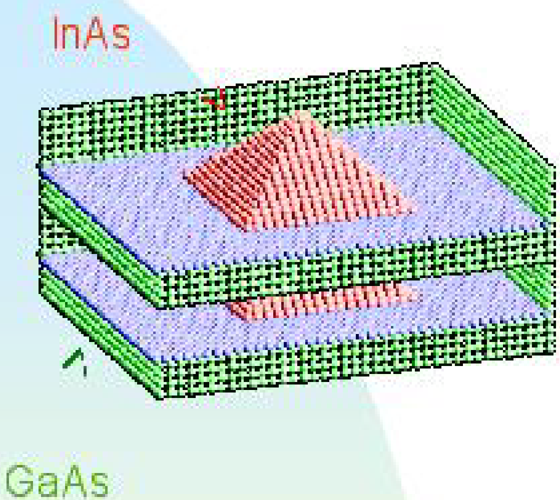
Electron and hole states for base length=13.6nm, strain included.

Hole state for base length=13.6nm, for effective mass method

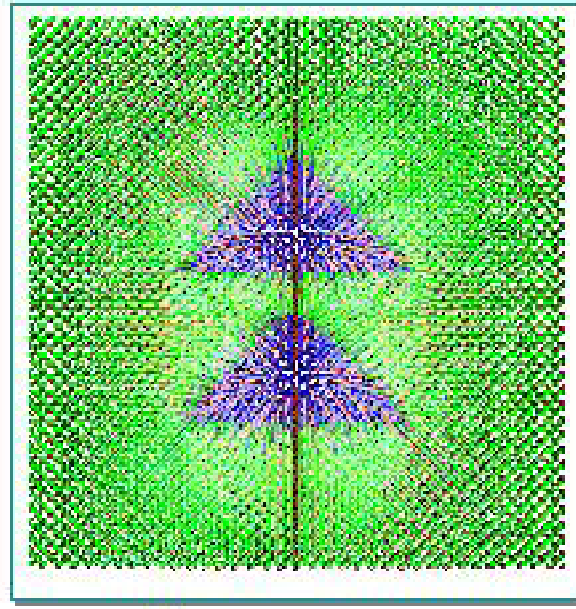
Theoretical methods 3: Pseudopotential methods

- Create smooth potentials to mimic the behavior of the valence region of real atoms
- Construct supercell of QD, WL and barrier material
- Supercell consists of ~ 30,000 atoms
- Strain in, and in the vicinity of QD calculated using valence force field model (A. J. Williamson and Alex Zunger, Phys. Rev. B 59 (1999) 1582)
- Piezoelectric effects included

Pseudopotential methods



1. Relax atomic positions using Valence Force Field (VFF)



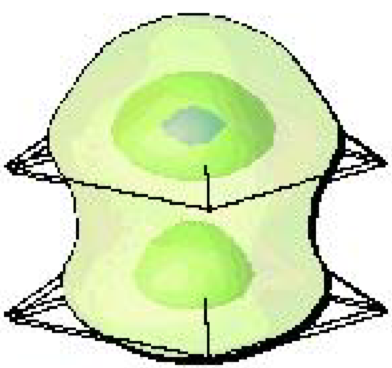
2. Construct EPM Hamiltonian

$$\hat{H} = \frac{-\hbar^2}{2m}\nabla^2 + \sum_{atoms,\alpha} v_\alpha(\mathbf{r} - \mathbf{R}_\alpha)$$

- Atomistic shape
- Atomistic surface
- 1,000,000 atoms

Calculate eigenstates using Linear Combination of Bulk Bands (LCBB) method

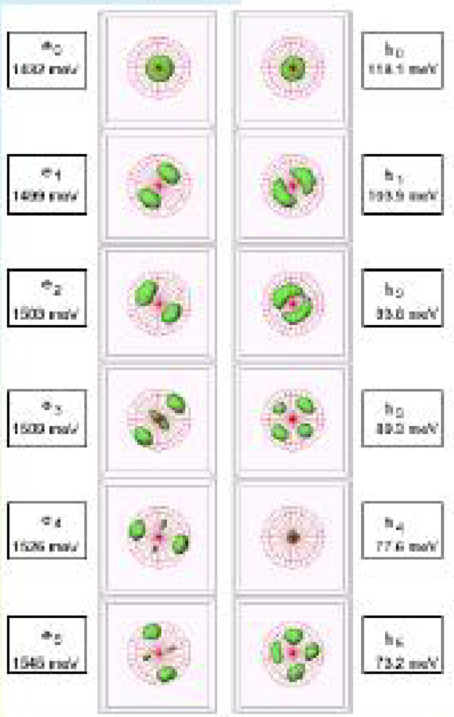
$$\psi^{dot}(\mathbf{r}) = \sum_{n}\sum_{k} c_{n k}\, u_{n k}(\mathbf{r}) e^{i\mathbf{k}.\mathbf{r}}$$

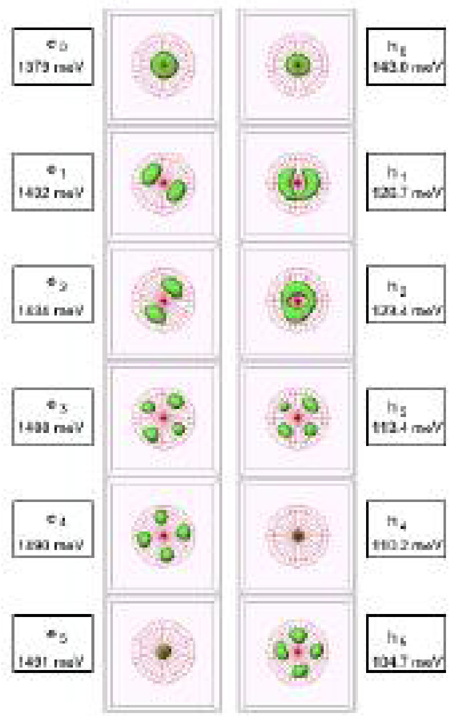


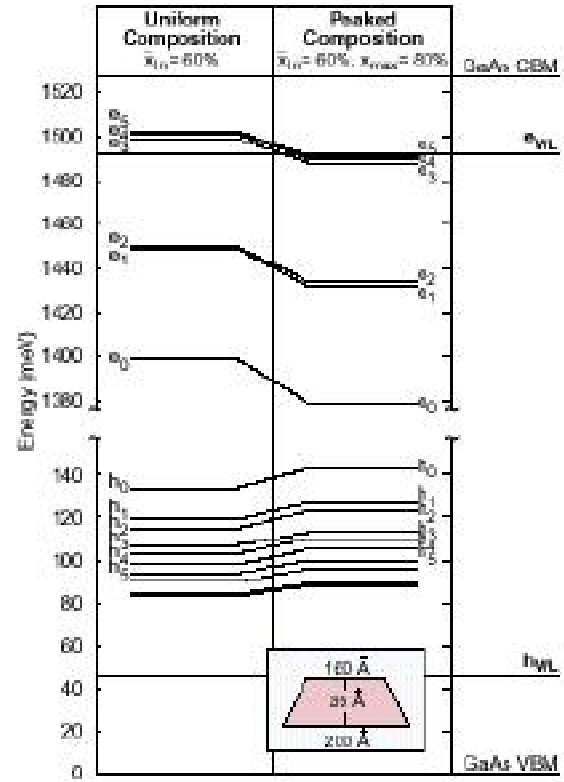
A. J. Williamson and Alex Zunger, Phys. Rev. B 59 (1999) 1582

Implications of In-enrichment

- Correctly model size, shape, compositional and strain field variations
- Compare with *ab initio* calculations



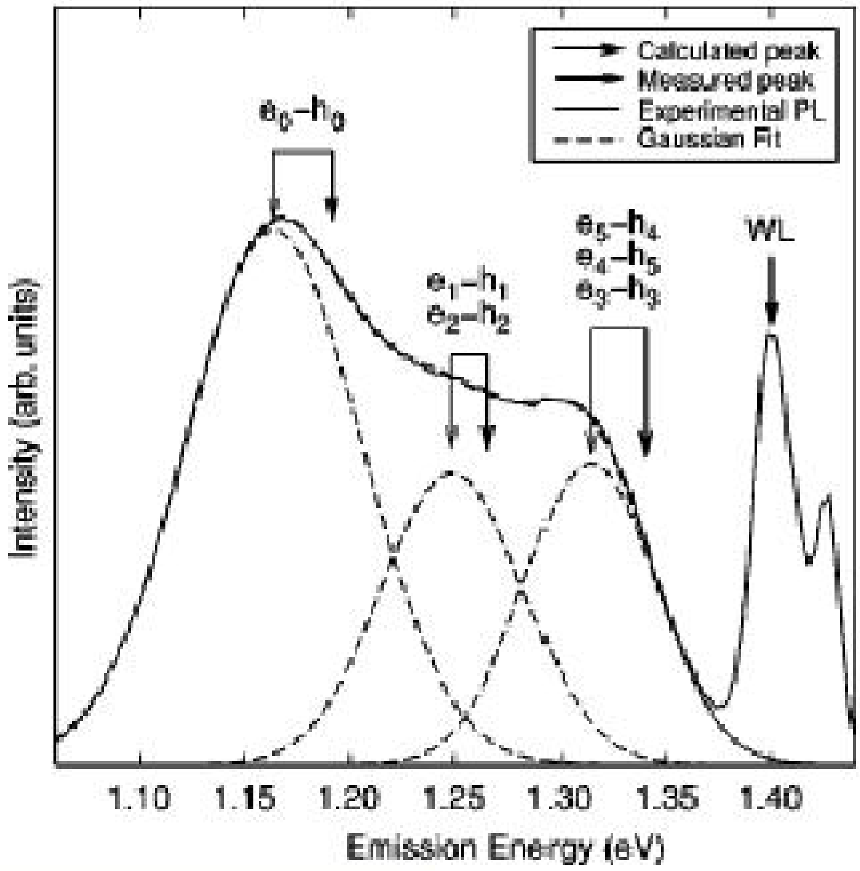




J. Shumway, A. J. Williamson, Alex Zunger, A. Passaseo, M. DeGiorgi, R. Cingolani, M. Catalano and P. Crozier, Phys Rev. B. (submitted)

NNL National Nanotechnology Laboratory of INFM

Pseudopotential methods



Comparing with ensemble PL, predicts well the ground and excited excitonic states

National Nanotechnology Laboratory of INFM

Predicted QD properties

- Discrete states, delta function like density of states: high optical efficiency
- Reduced relaxation rates: ultra narrow emission lines
- Small volume of QD: enhanced carrier interactions, correlation effects should be evident in PL spectra

Summing up

- Quantum dot – the artificial atom
  - Optical properties show that QDs obey Hund's Law
  - Calculated single particle energies agree with experimental results
- Charging properties.....
- Strong interaction with environment
- Multi-particle effects masked by inhomogenous broadening