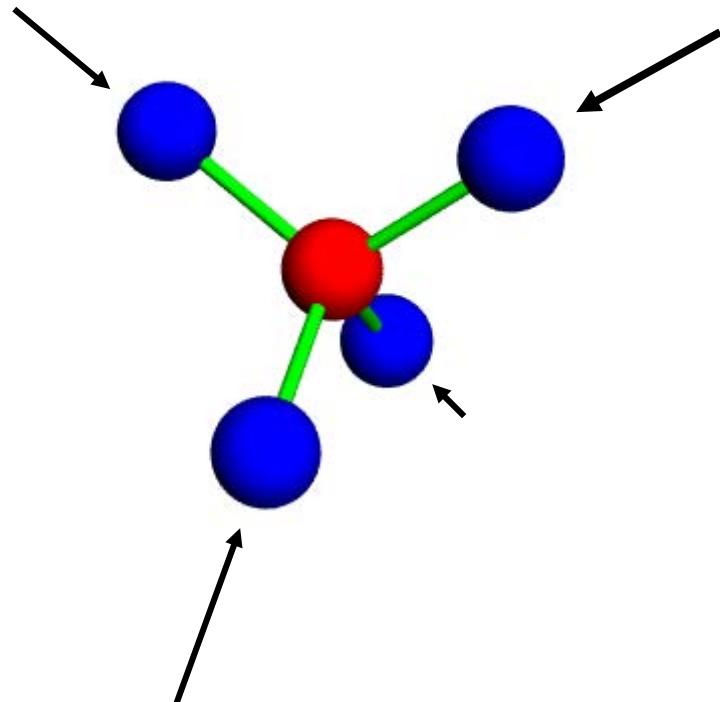


FITTING OF STRAIN

HYDROSTATIC – BIR-PIKUS MODEL



LINEAR SHIFTS OF BAND EDGES

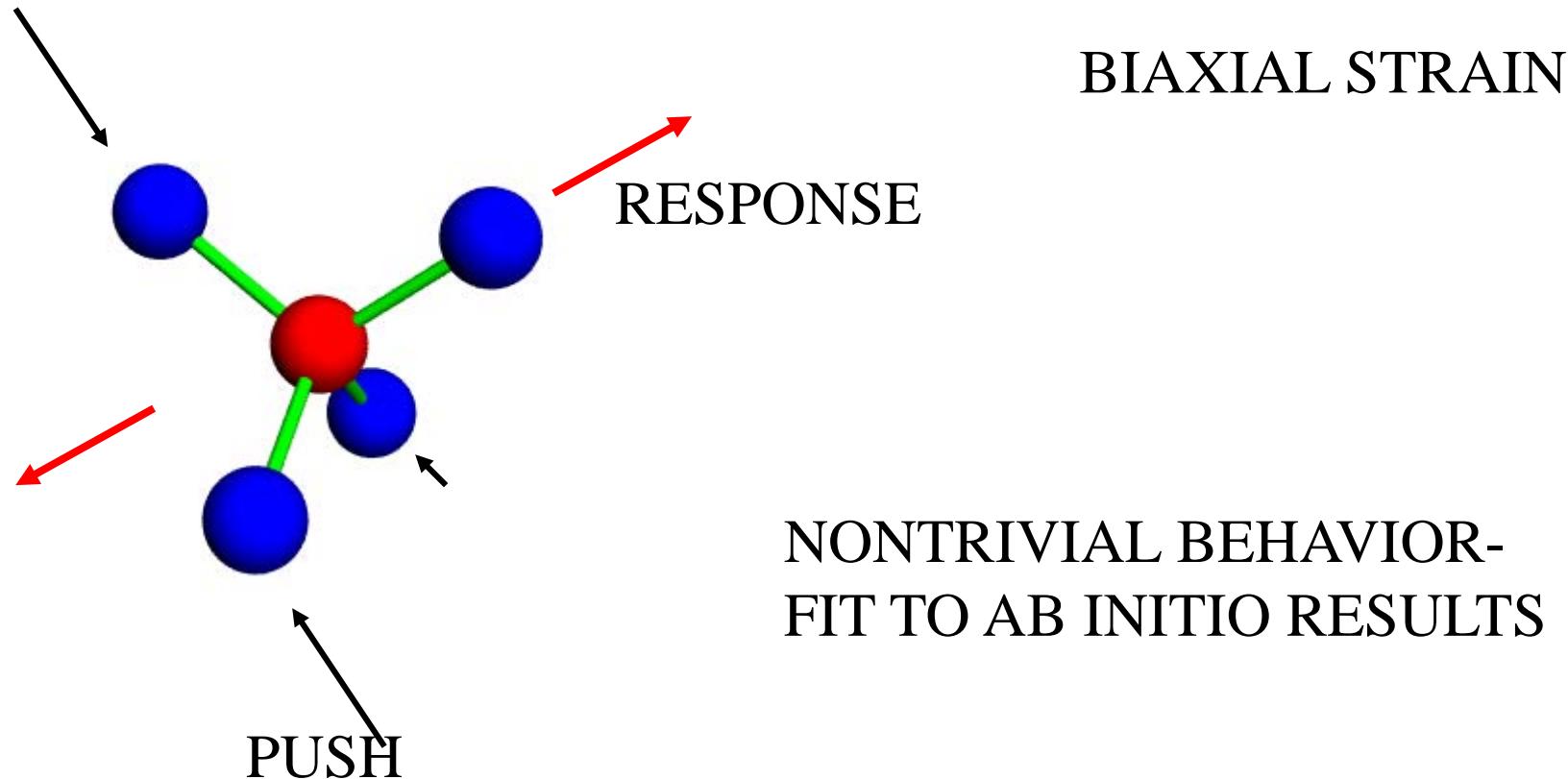
$$E_{CB} = E_{CB}^0 + a_C (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})$$

$$E_{VB,HH} = E_{VB,HH}^0 + a_V (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})$$

$$E_{SO} = E_{SO}^0 + a_V (\varepsilon_{xx} + \varepsilon_{yy} + \varepsilon_{zz})$$

a_C, a_V - DEFORMATION POTENTIALS

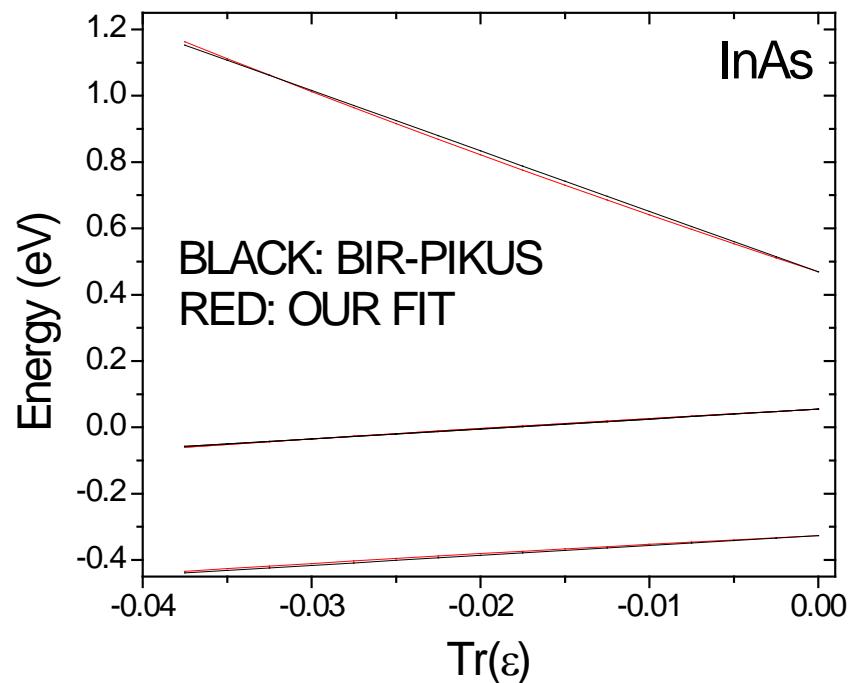
FITTING OF STRAIN



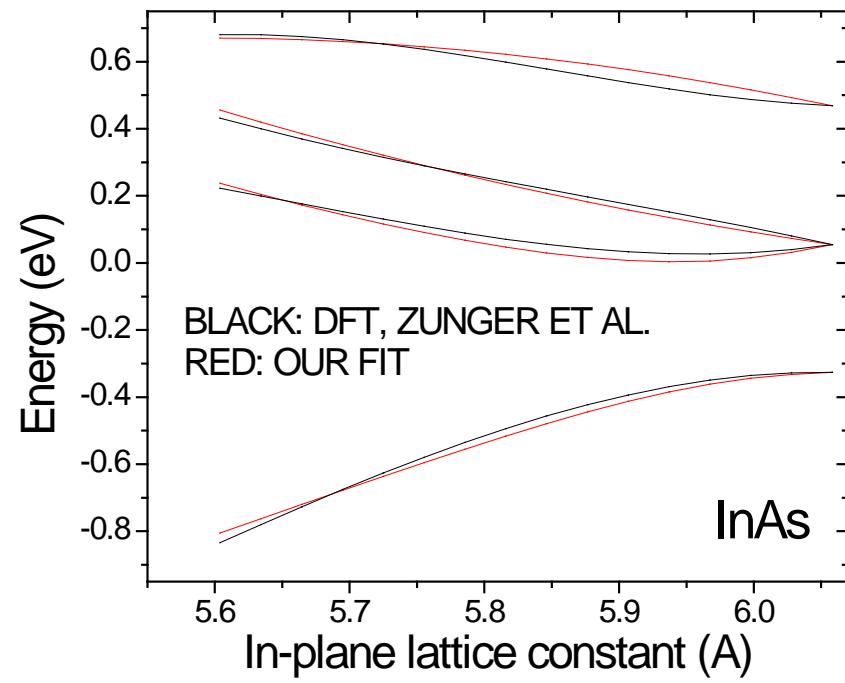
FITTING OF STRAIN PARAMETERS

BAND EDGES AS A FUNCTION OF STRAIN

Hydrostatic strain fitted to Bir-Pikus model



Biaxial strain fit to DFT (LDA) model

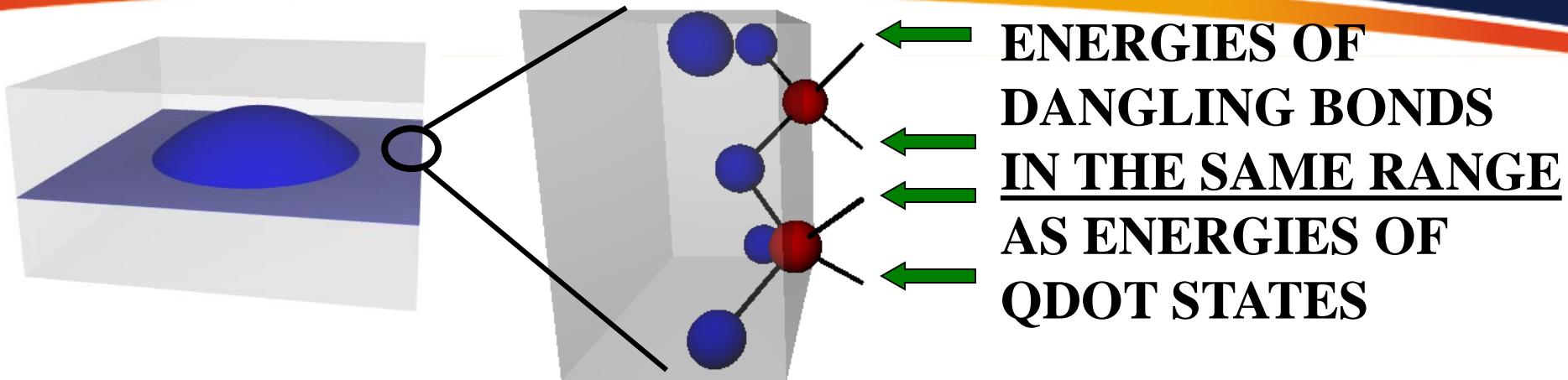


DFT RESULTS FROM
A. ZUNGER et al., APL 81, 4377



BOUNDARIES OF THE COMPUTATIONAL BOX

BOUNDARIES



1. ROTATE THE sp^3 BASIS INTO HYBRIDIZED ORBITALS

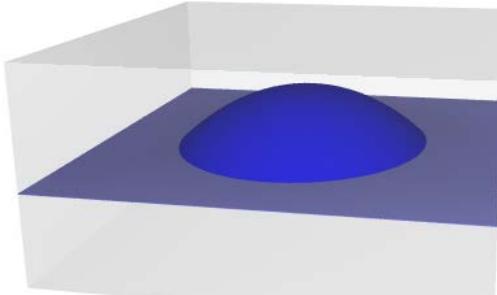
$$|sp_1^3\rangle = \frac{1}{2}(|s\rangle + |p_x\rangle + |p_y\rangle + |p_z\rangle) \quad |sp_2^3\rangle = \frac{1}{2}(|s\rangle + |p_x\rangle - |p_y\rangle - |p_z\rangle)$$

$$|sp_3^3\rangle = \frac{1}{2}(|s\rangle - |p_x\rangle + |p_y\rangle - |p_z\rangle) \quad |sp_4^3\rangle = \frac{1}{2}(|s\rangle - |p_x\rangle - |p_y\rangle + |p_z\rangle)$$

2. IDENTIFY DANGLING BONDS, SHIFT THEIR ENERGIES

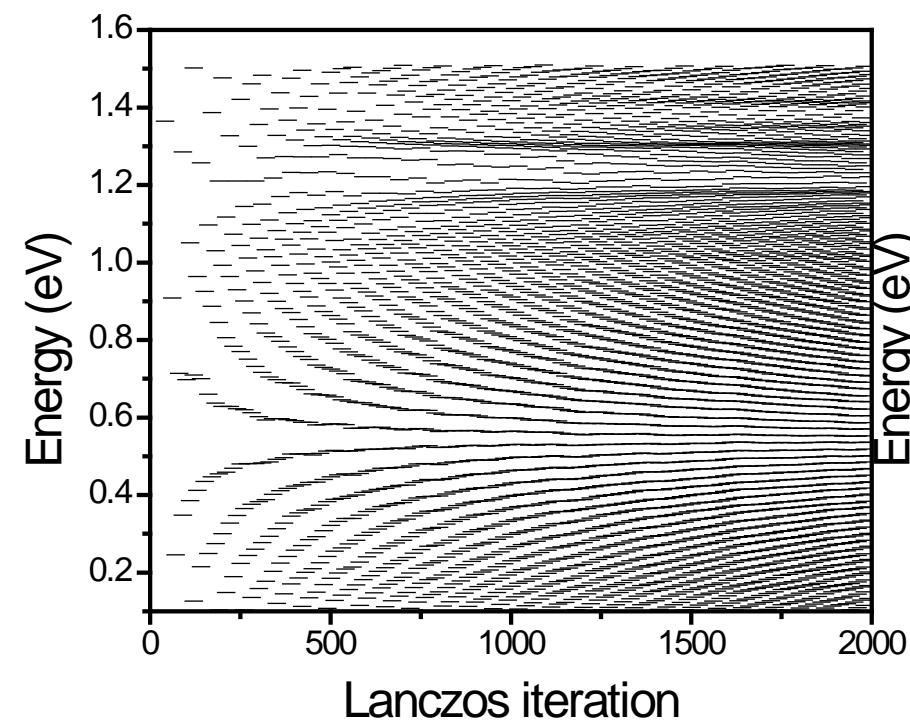
3. ROTATE HYBRIDIZED ORBITALS BACK TO sp^3 BASIS

BOUNDARIES

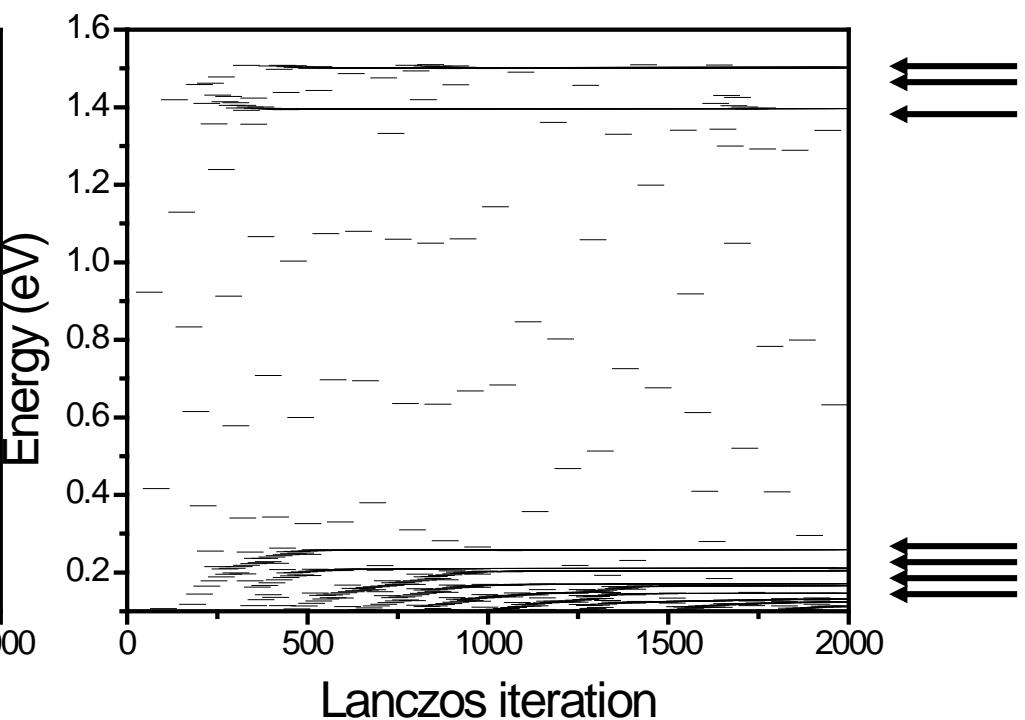


IS IT IMPORTANT?

LANCZOS ITERATIVE DIAGONALIZATION

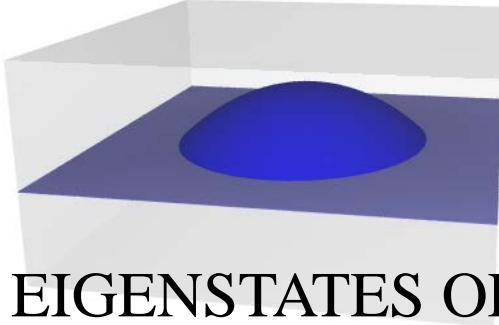


NO SURFACE PASSIVATION



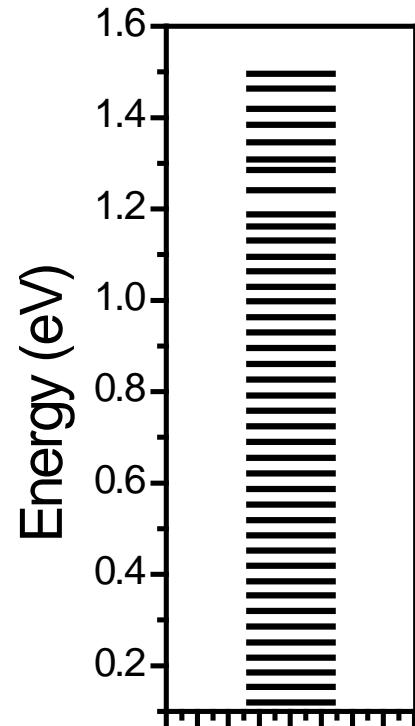
SURFACE PASSIVATION

BOUNDARIES

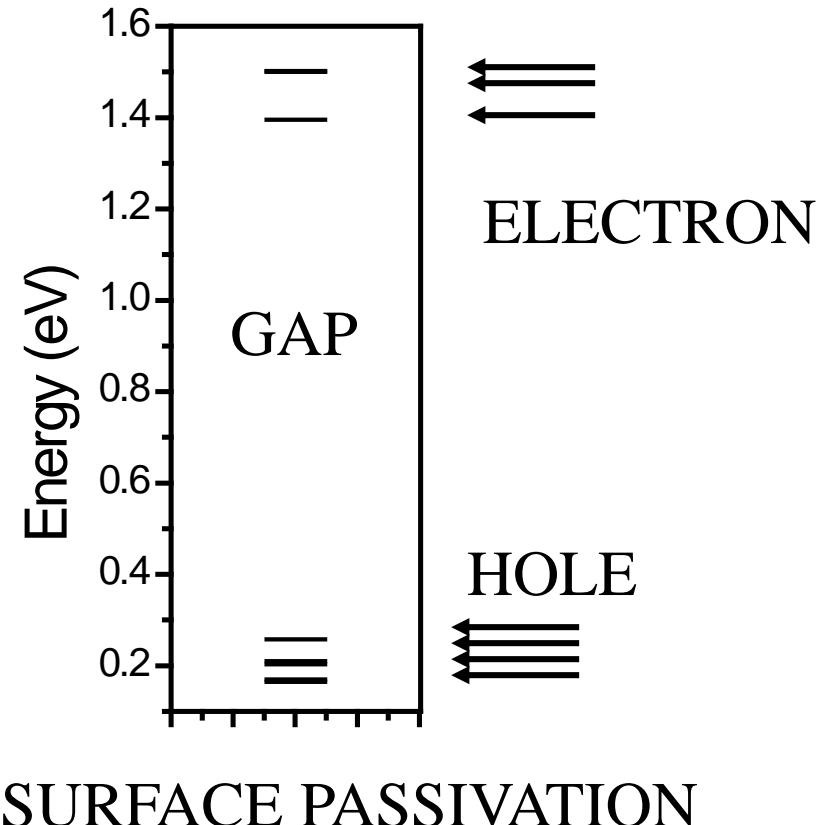


IS IT IMPORTANT?

EIGENSTATES OF HAMILTONIAN



NO SURFACE PASSIVATION



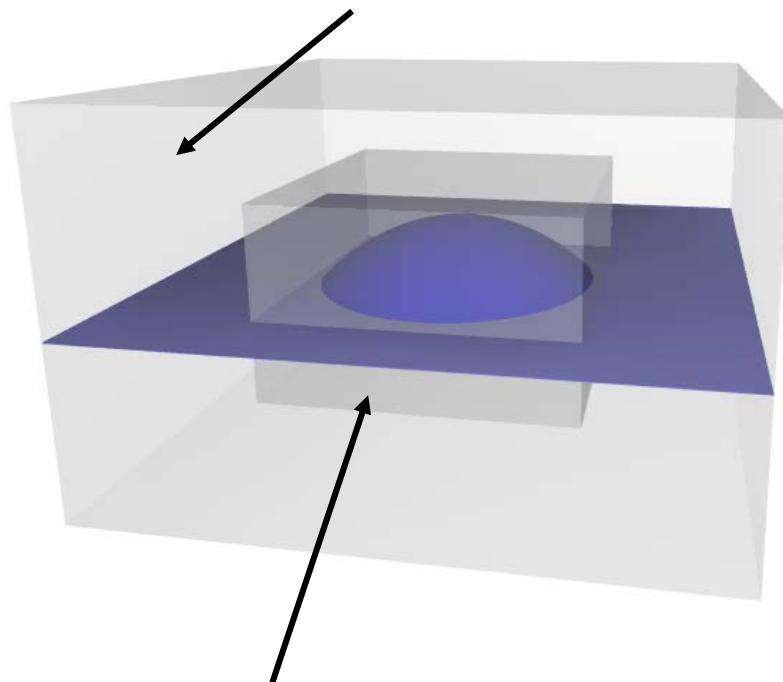
SURFACE PASSIVATION



ORGANIZATION OF COMPUTATIONAL DOMAINS

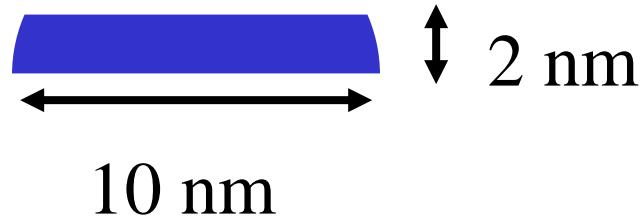
COMPUTATIONAL DOMAINS

EXTERNAL DOMAIN – STRAIN ONLY



INTERNAL DOMAIN – STRAIN + ELECTRONIC

LONG-RANGE NATURE OF STRAIN



TEST SYSTEM:
TRUNCATED LENS

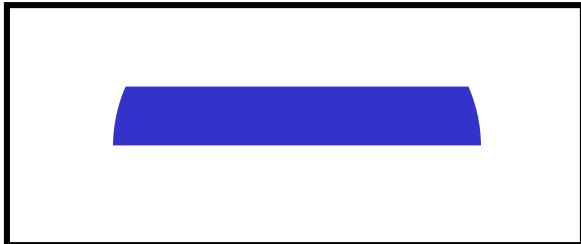
LONG-RANGE NATURE OF STRAIN



TEST SYSTEM:
TRUNCATED LENS

ELECTRONIC DOMAIN: 18 nm x 18 nm x 12 nm
CONTAINING 170 000 ATOMS

LONG-RANGE NATURE OF STRAIN



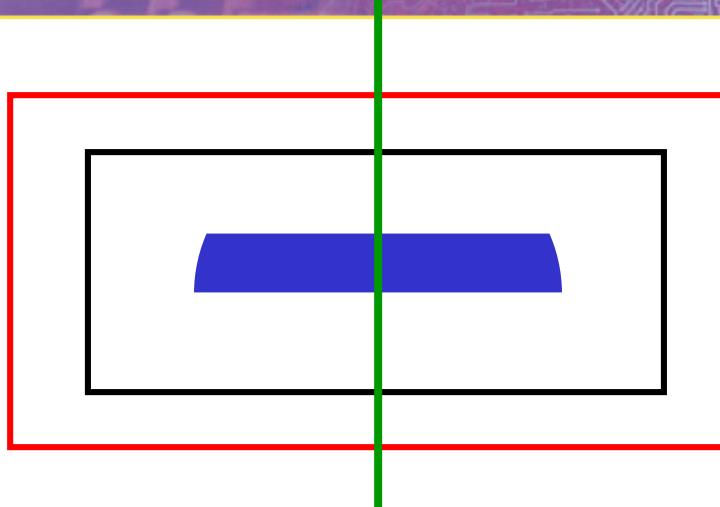
TEST SYSTEM:
TRUNCATED LENS

ELECTRONIC DOMAIN: 18 nm x 18 nm x 12 nm
CONTAINING 170 000 ATOMS

STRAIN DOMAIN:

- 1) IDENTICAL TO ELECTRONIC DOMAIN
- 2) 34 nm x 34 nm x 35 nm, CONTAINING 1.8 MILLION ATOMS

LONG-RANGE NATURE OF STRAIN

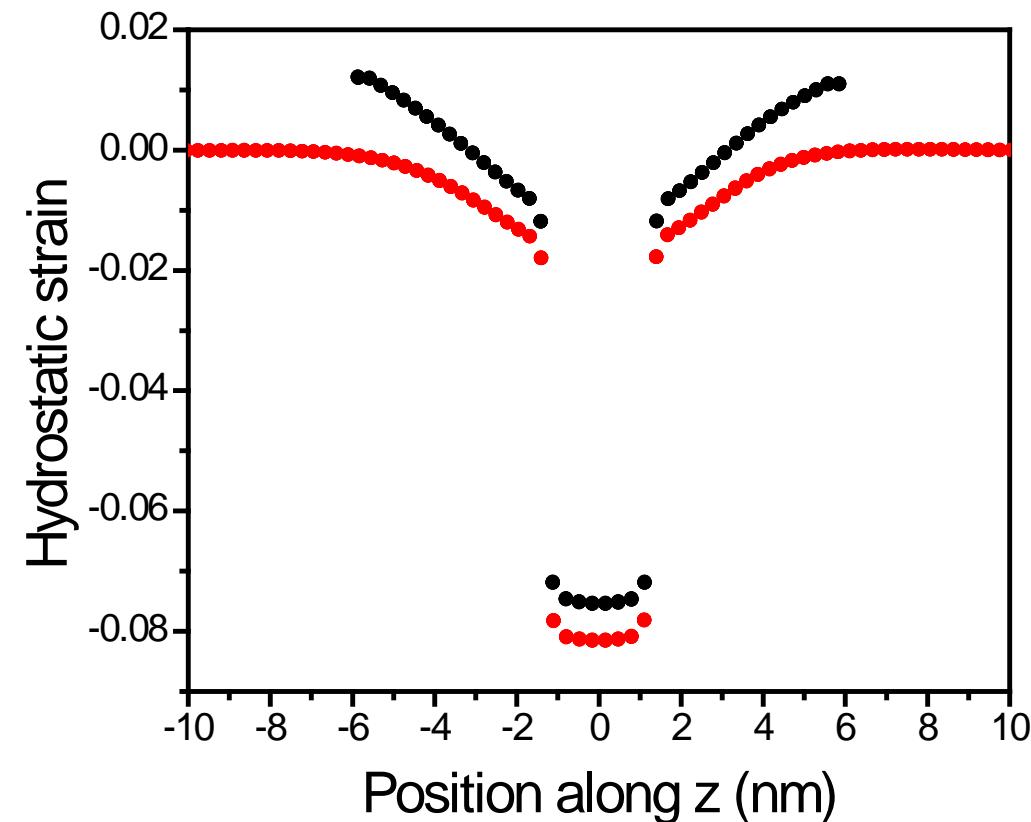


SMALL DOMAIN:

STRAIN NONZERO AT BOUNDARIES

HYDROSTATIC STRAIN ABOUT
1% LOWER IN THE DOT

HYDROSTATIC STRAIN

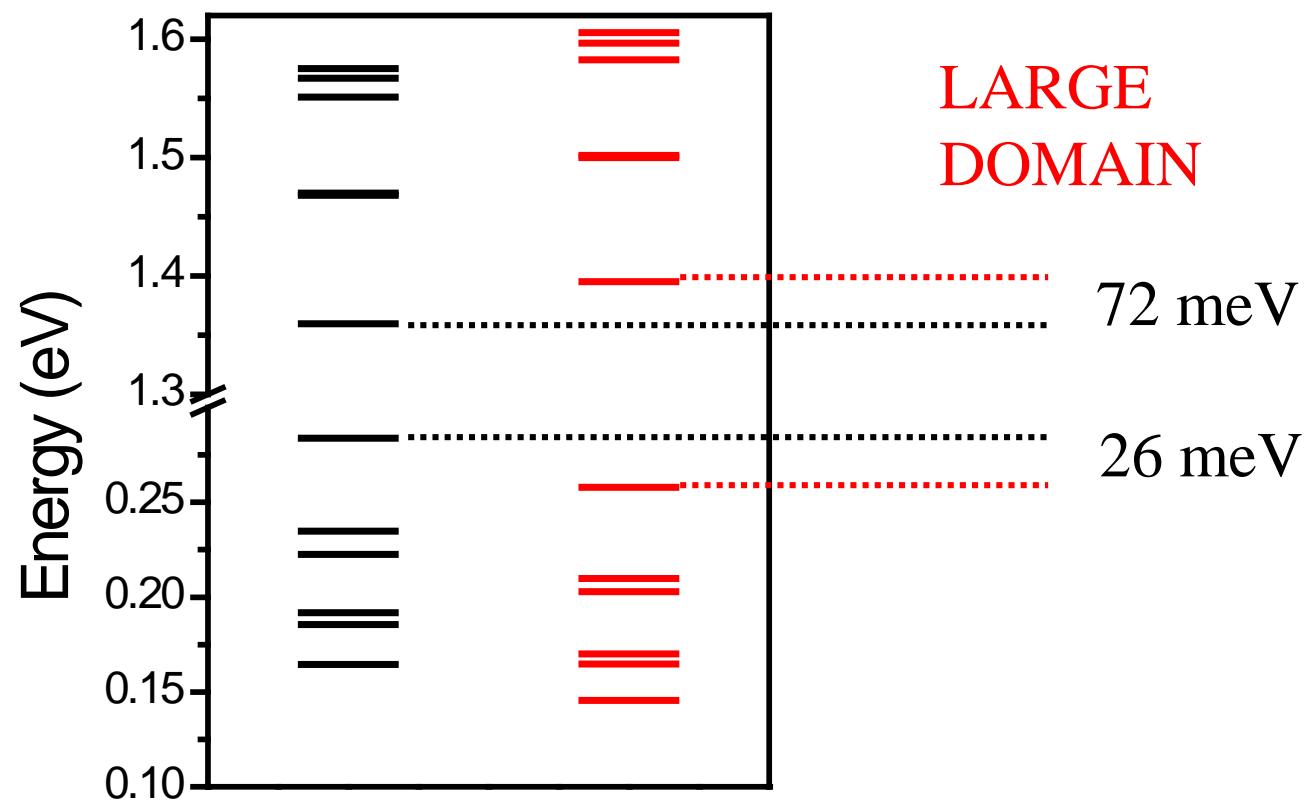


LONG-RANGE NATURE OF STRAIN



SINGLE-PARTICLE ENERGIES

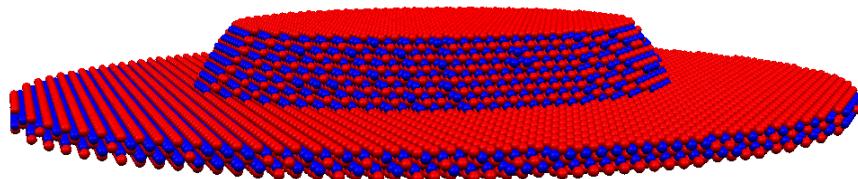
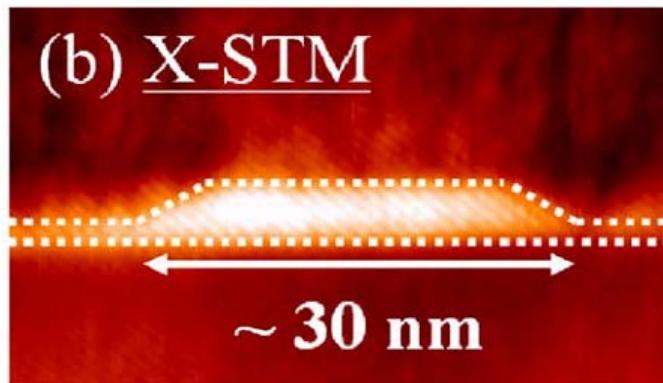
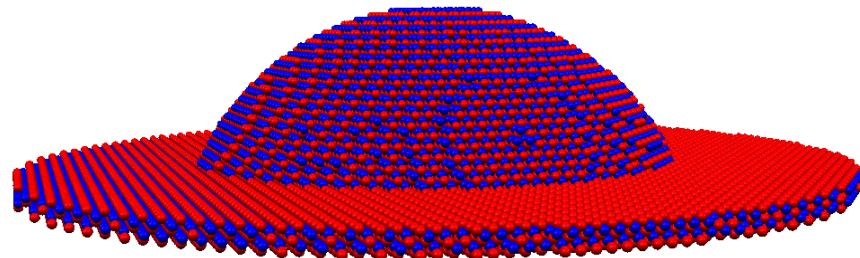
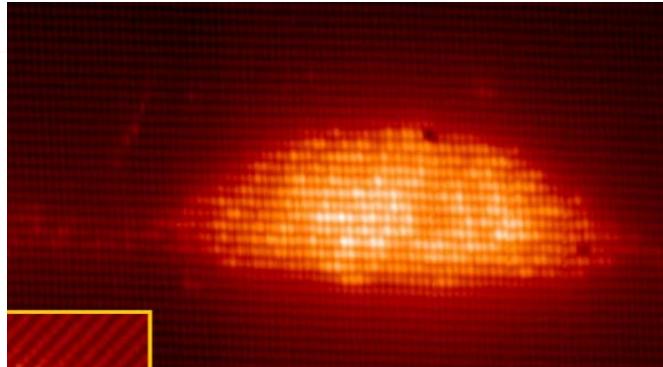
SMALL
DOMAIN





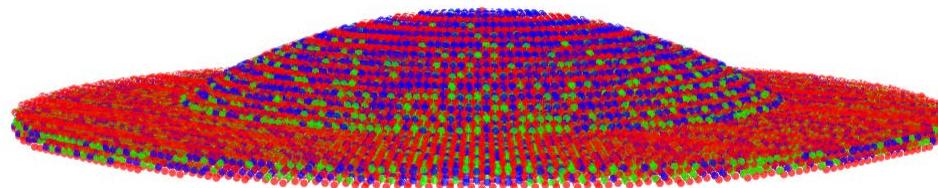
SYSTEMS OF INTEREST

QNANO: BUILDING QUANTUM DOT WITH ATOMS



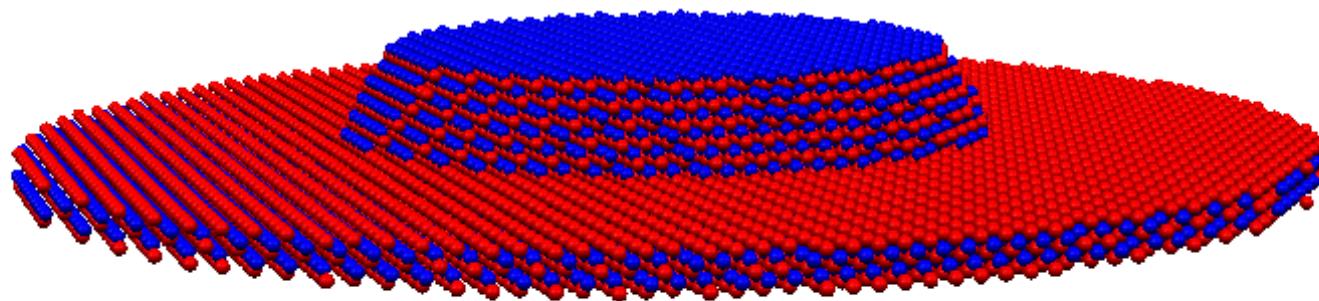
Alloy (vertical gradient)

P. Koenraad's group X-STM



InAs/GaAs In-FLUSH GROWN DOT

IMS In-flush growth technique – Wasilewski et al J.Cryst.Growth,1999

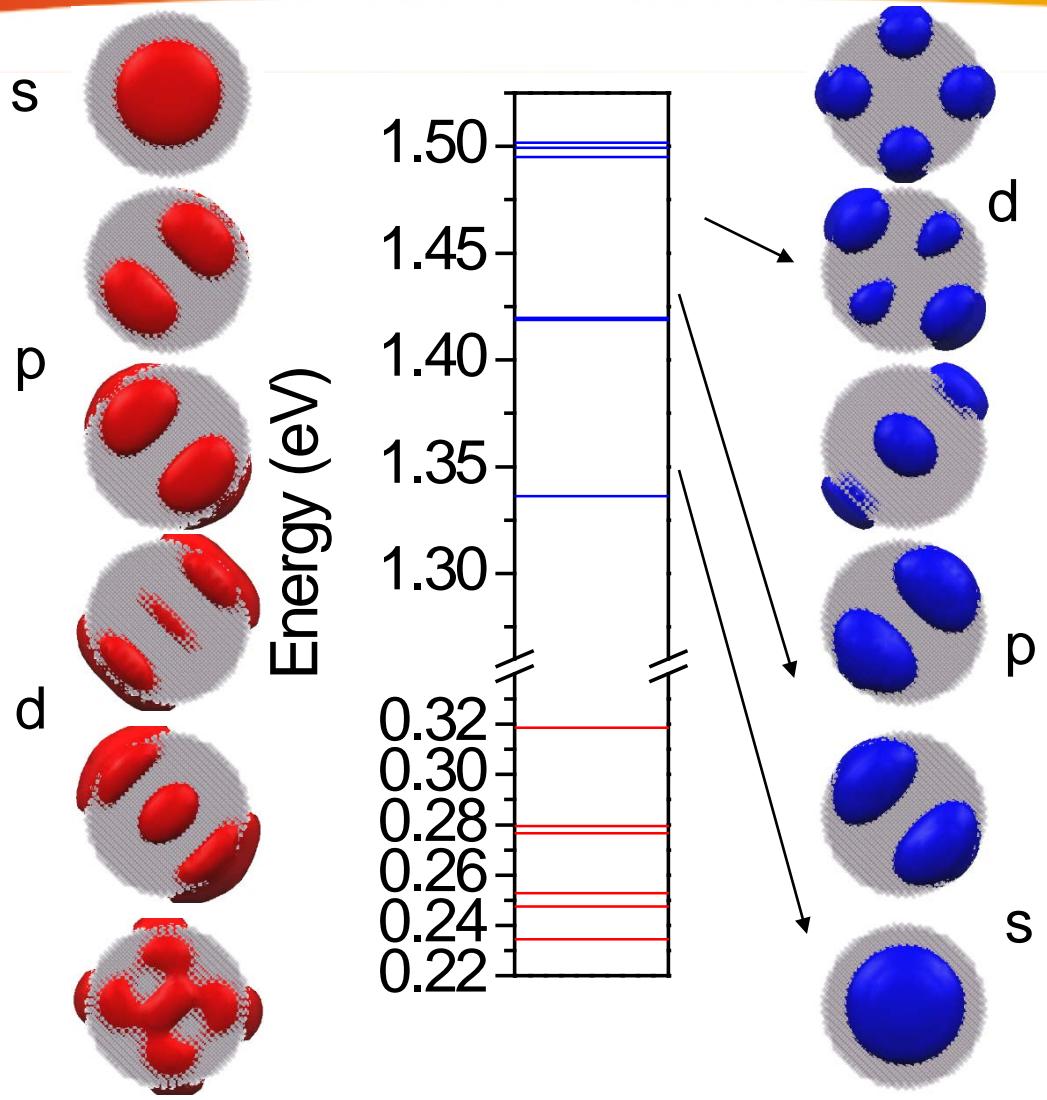


QD diameter 12.5 nm, height 2.0 nm

VFF domain: 32.2 mln atoms

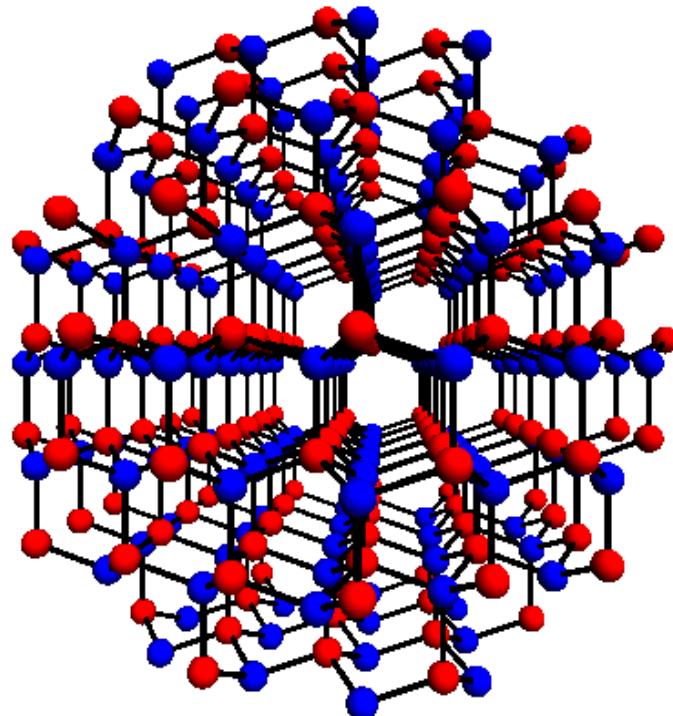
TB domain : 0.298 mln atoms

InAs/GaAs In-FLUSH GROWN DOT

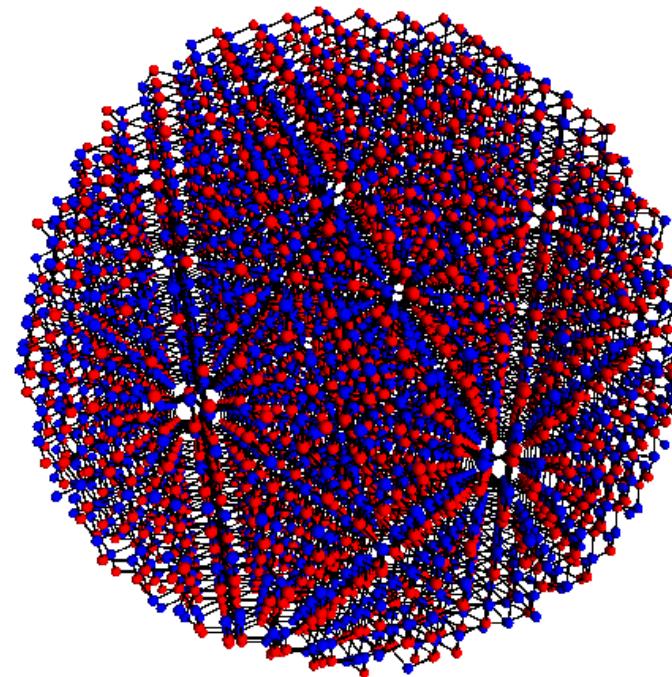


**SHELL
STRUCTURE
OF HOLES!**

CdSe NANOCRYSTALS



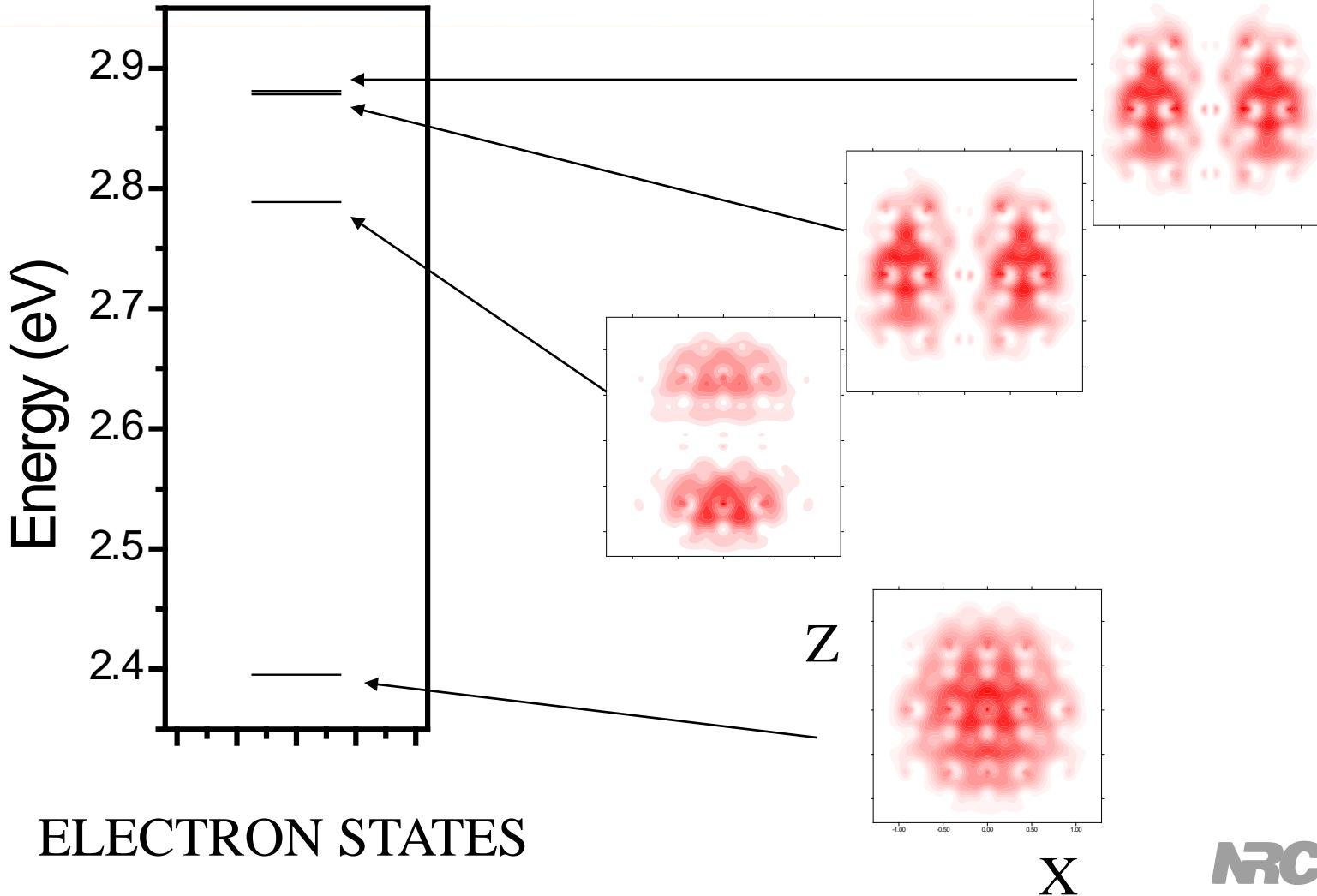
DIAMETER 2.5nm
~300 ATOMS



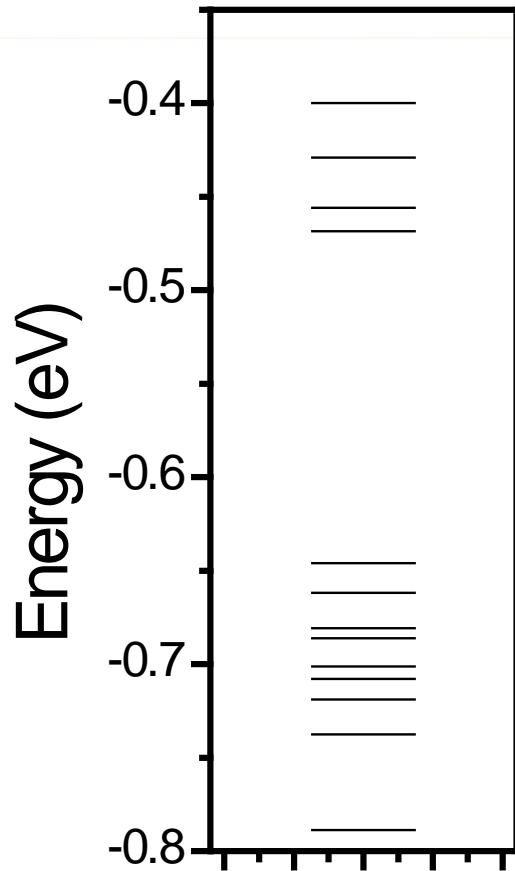
DIAMETER 7nm
~6500 ATOMS

WURTZITE CRYSTAL STRUCTURE

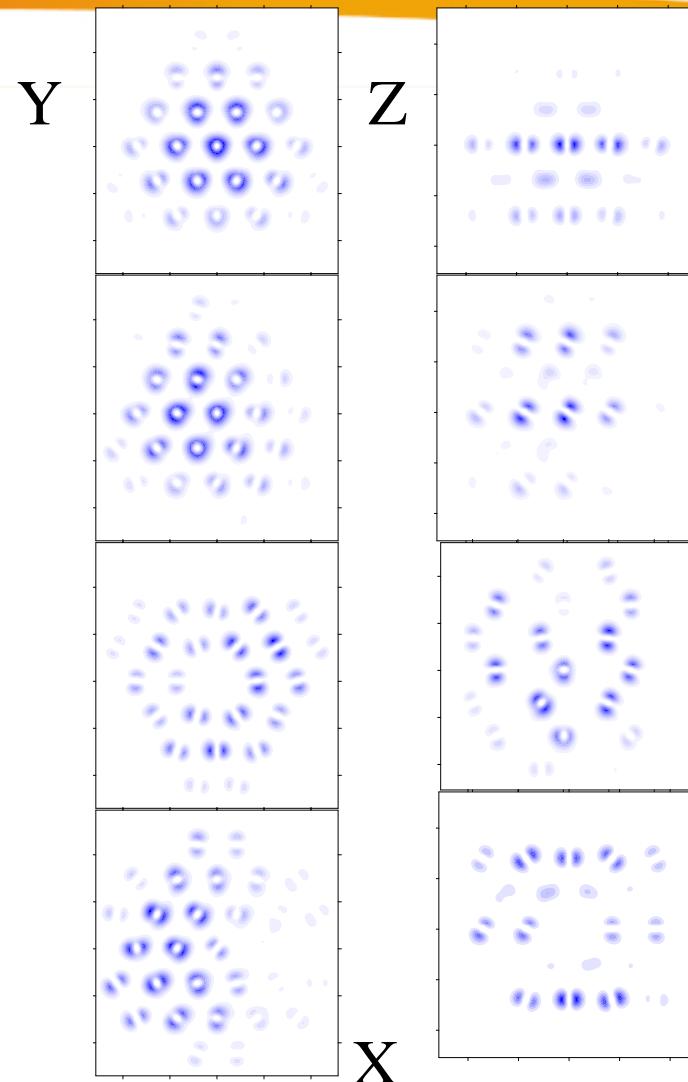
CdSe NANOCRYSTAL, DIAMETER 3nm



CdSe NANOCRYSTAL, DIAMETER 3nm



HOLE STATES



SYMMETRY IS NOT CLEAR

X

X

NRC-CNRC



SYSTEMS OF CONFINED INTERACTING ELECTRONS AND HOLES

INTERACTING ELECTRONS AND HOLES

HAMILTONIAN:

$$\hat{H} = \sum_i E_i^{(e)} c_i^+ c_i + \frac{1}{2} \sum_{ijkl} \langle i, j | V_{ee} | k, l \rangle c_i^+ c_j^+ c_k c_l$$

ELECTRON SP STATES,
E-E INTERACTION

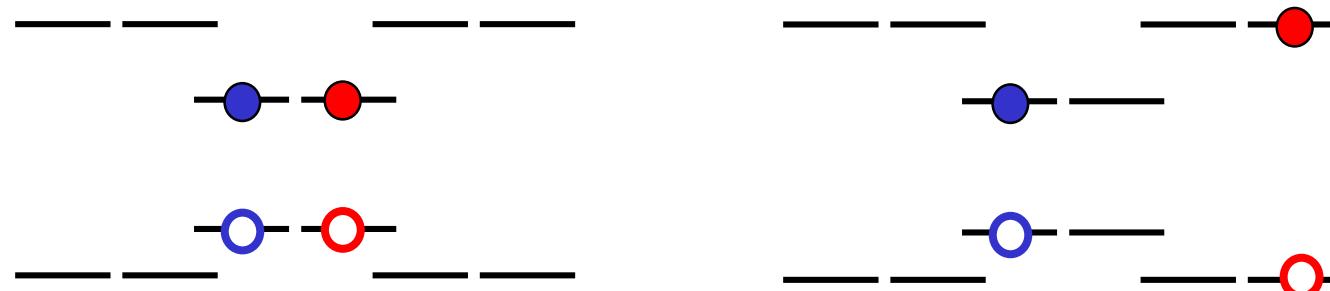
$$+ \sum_j E_j^{(h)} h_j^+ h_j + \frac{1}{2} \sum_{ijkl} \langle i, j | V_{hh} | k, l \rangle h_i^+ h_j^+ h_k h_l$$

HOLE SP STATES,
H-H INTERACTION

$$- \sum_{ijkl\sigma\sigma'} (\langle i_e, j_h | V_{eh,dir} | k_h, l_e \rangle - \langle i_e, j_h | V_{eh,xchg} | l_h, k_e \rangle) c_i^+ h_j^+ h_k c_l$$

ELECTRON-HOLE
INTERACTION

BASIS OF ELECTRON-HOLE CONFIGURATIONS



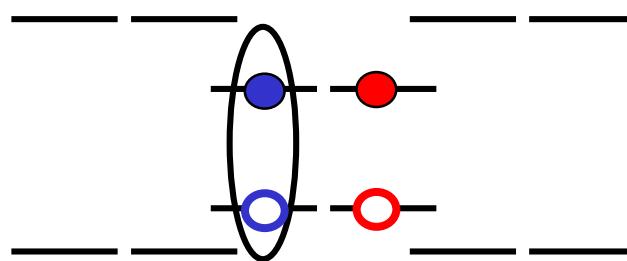
OPTICAL SPECTRA

EMISSION SPECTRA – FERMI'S GOLDEN RULE

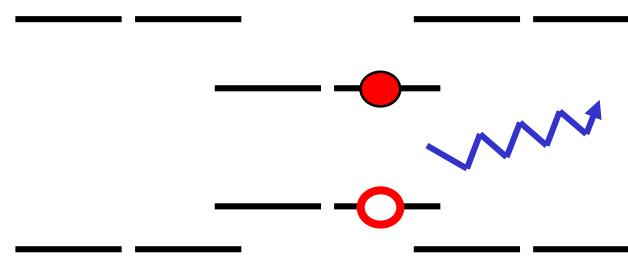
$$I(\omega) = \sum_f \left| \langle f, N-1 | P^- | i, N \rangle \right|^2 \delta(E_i - E_f - \hbar\omega)$$

$$P^- = \sum_{lm} \langle l_e | \vec{\epsilon} \cdot \vec{r} | m_h \rangle c_l h_m$$

EXAMPLE: BIEXCITON



INITIAL STATE $|XX\rangle$



FINAL STATE $|X\rangle$



COULOMB MATRIX ELEMENTS

COULOMB MATRIX ELEMENTS

ONSITE

$$\begin{aligned} \langle \Psi_i \Psi_j | V | \Psi_k \Psi_l \rangle = & \sum_{R_1} \sum_{\alpha_1 \alpha_2 \alpha_3 \alpha_4} a_{R_1 \alpha_1}^{(i)*} a_{R_1 \alpha_2}^{(j)*} a_{R_1 \alpha_3}^{(k)} a_{R_1 \alpha_4}^{(l)} \langle R_1 \alpha_1, R_1 \alpha_2 | \frac{e^2}{1 \cdot |\vec{r}_1 - \vec{r}_2|} | R_1 \alpha_3, R_1 \alpha_4 \rangle + \\ & + \sum_{R_1} \sum_{R_2 \neq R_1} \sum_{\alpha_1 \alpha_2 \alpha_3 \alpha_4} a_{R_1 \alpha_1}^{(i)*} a_{R_2 \alpha_2}^{(j)*} a_{R_2 \alpha_3}^{(k)} a_{R_1 \alpha_4}^{(l)} \langle R_1 \alpha_1, R_2 \alpha_2 | \frac{e^2}{\varepsilon |\vec{r}_1 - \vec{r}_2|} | R_2 \alpha_3, R_1 \alpha_4 \rangle \end{aligned}$$

LONG-RANGE

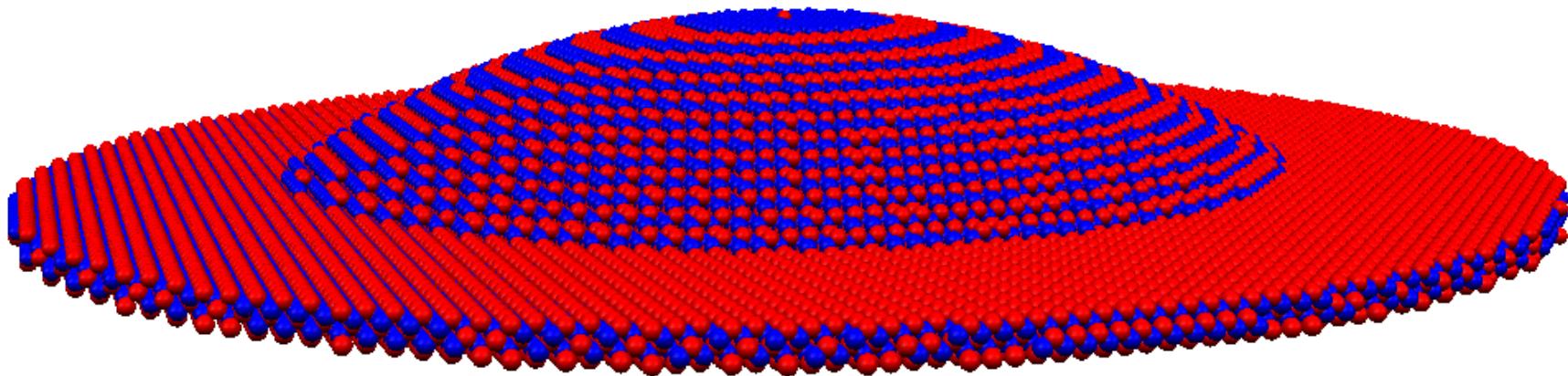
OFFSITE ELEMENT:

$$\begin{aligned} \langle R_1 \alpha_1, R_2 \alpha_2 | \frac{1}{|\vec{r}_1 - \vec{r}_2|} | R_2 \alpha_3, R_1 \alpha_4 \rangle \approx & \int d\vec{r}_1 \int d\vec{r}_2 \frac{\varphi_{\alpha_1}^*(\vec{r}_1 - \vec{R}_1) \varphi_{\alpha_2}^*(\vec{r}_2 - \vec{R}_2) \varphi_{\alpha_3}(\vec{r}_2 - \vec{R}_2) \varphi_{\alpha_4}(\vec{r}_1 - \vec{R}_1)}{\varepsilon |\vec{R}_1 - \vec{R}_2|} \\ & = \frac{\delta_{\alpha_1, \alpha_4} \delta_{\alpha_2, \alpha_3}}{\varepsilon |\vec{R}_1 - \vec{R}_2|} \end{aligned}$$

MULTI-EXCITON COMPLEXES

MULTI-EXCITON COMPLEXES IN A BENCHMARK QD

For pseudopotential calculations see
L. He and A. Zunger Phys. Rev. B 73, 115324 (2006)

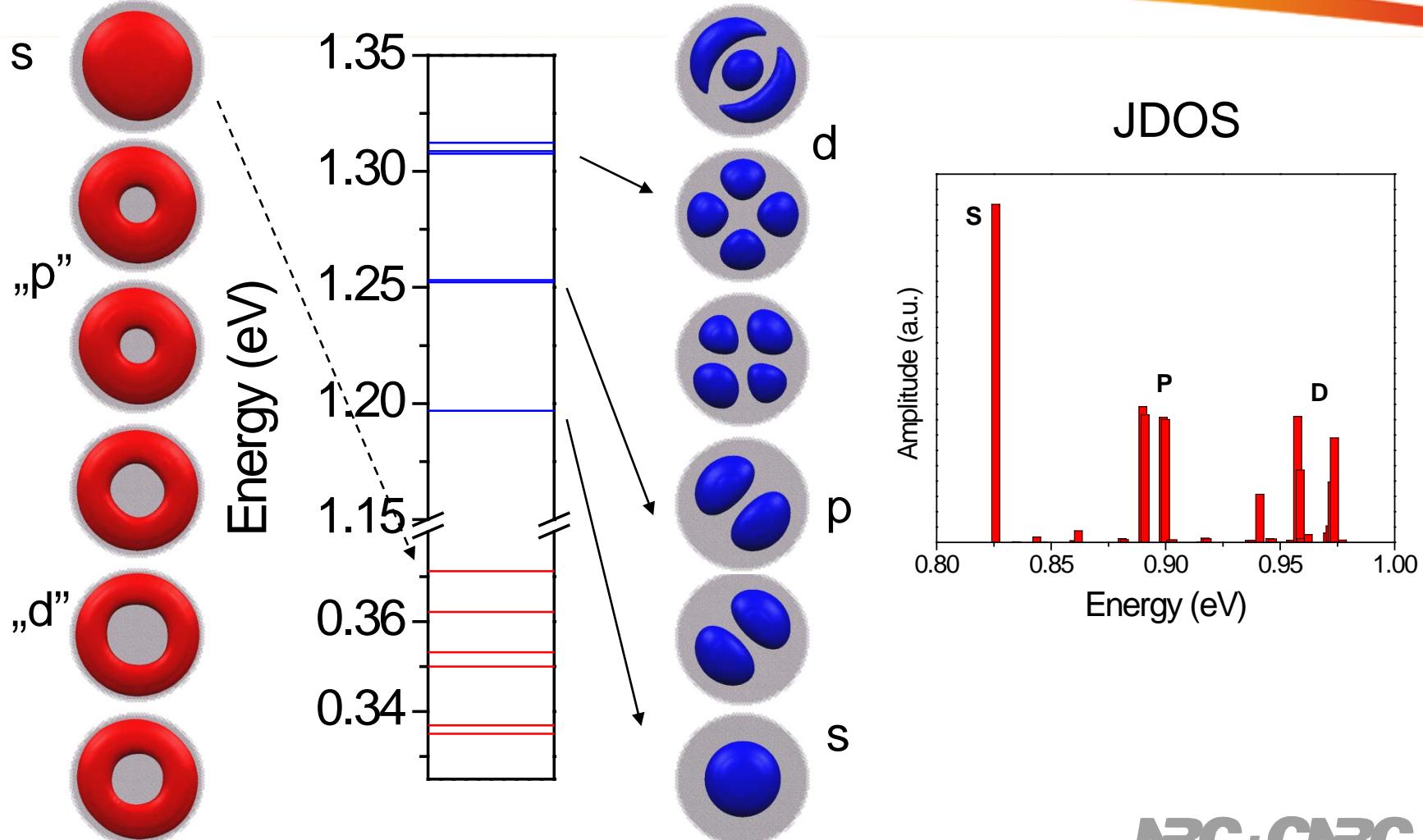


QD diameter 25nm, height 3.5 nm

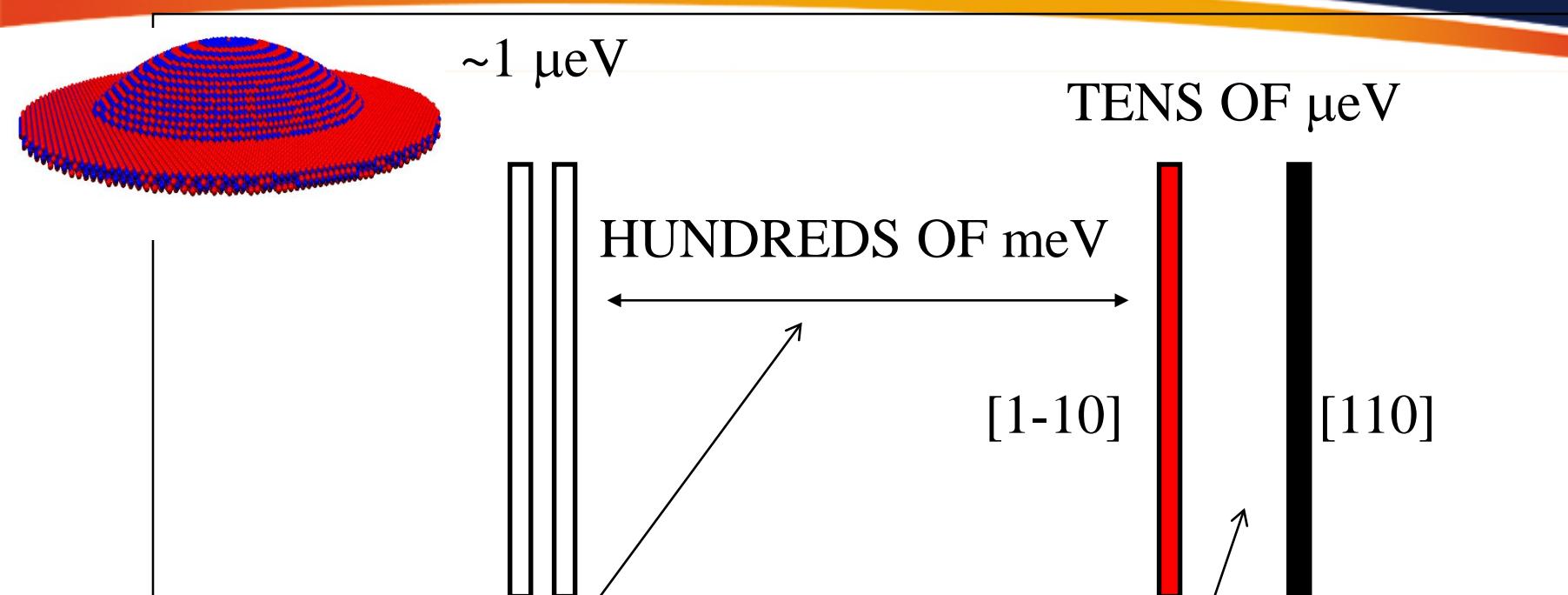
VFF domain: 50.3 mln atoms

TB domain : 0.561 mln atoms

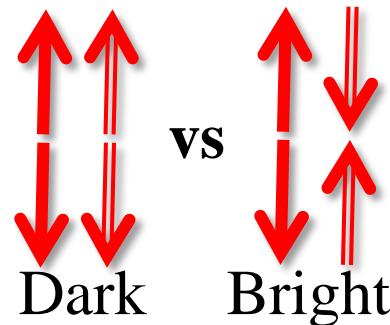
SINGLE PARTICLE STATES AND OPTICAL DENSITY OF STATES



EXCITON FINE STRUCTURE



Electron-hole exchange splitting



Anisotropic EH exchange splitting.
But QD is cylindrical...

Cylindrical symmetry reduced by crystal lattice

MULTIEXCITON EMISSION SPECTRA

