Reengineering the Optics of Quantum Dots with Mechanical Strain

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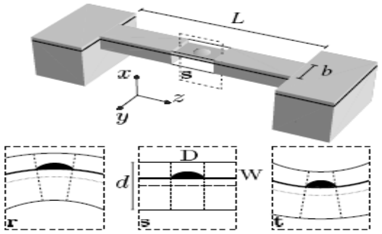

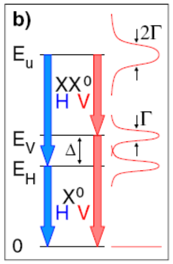

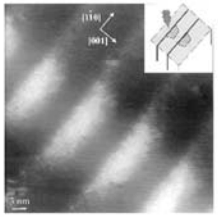

NIST

JQI Joint Quantum Institute

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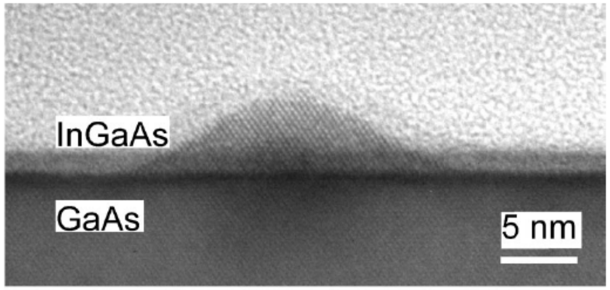
Strained quantum dots



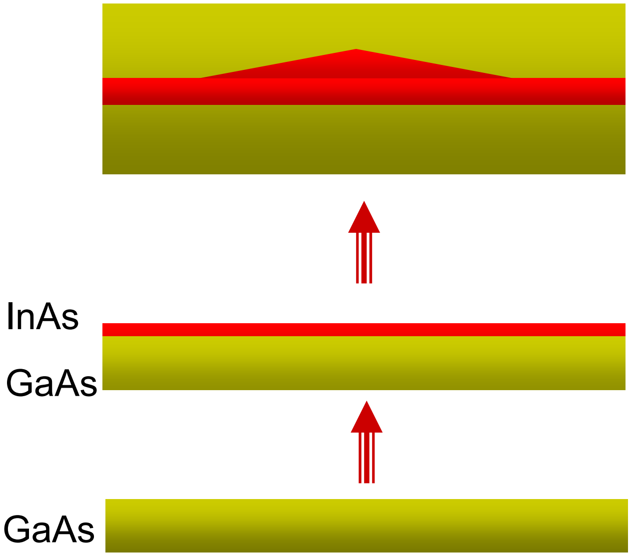
Self-assembled quantum dots

… molecular beam *epitaxial* growth

… self-organizing: a_{InAs} = 6.04 Å, a_{GaAs} = 5.65 Å

… growth driven by interfacial strain

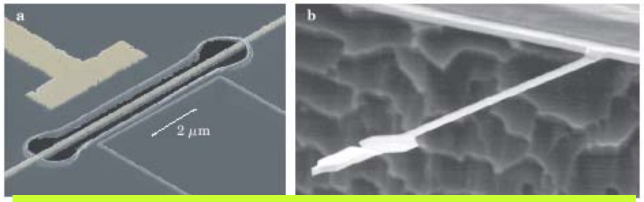
… integrability



Externally imposed strain to modulate dots and coupled dots

Internal Strain

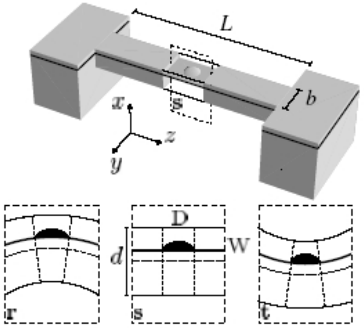
- Interfaces in core/shell nanocrystals and self-assembled QDs



(Physics Today, July 2005)

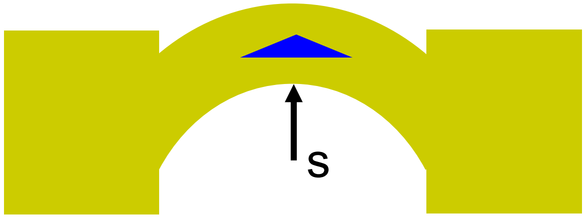
External strain

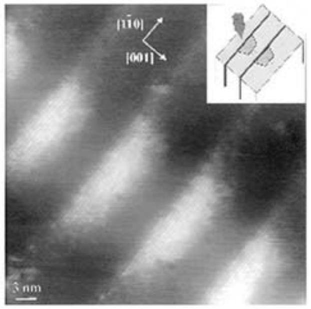
- Stressors above self-assembled QDs
- Vibrations in nanomechanical cantilevers or bridges with embedded self-assembled dots
- Surface acoustic waves interacting with QDs



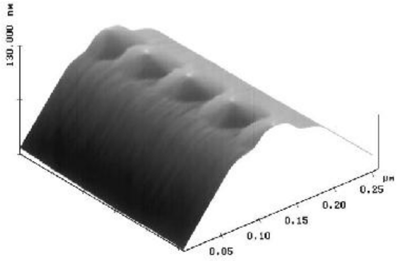
(Zoller group, PRL 2004)

How much control over SADs or couple QDs?





B. Grandidier, et al., Phys. Rev. Lett. **85**, 1068 (2000)



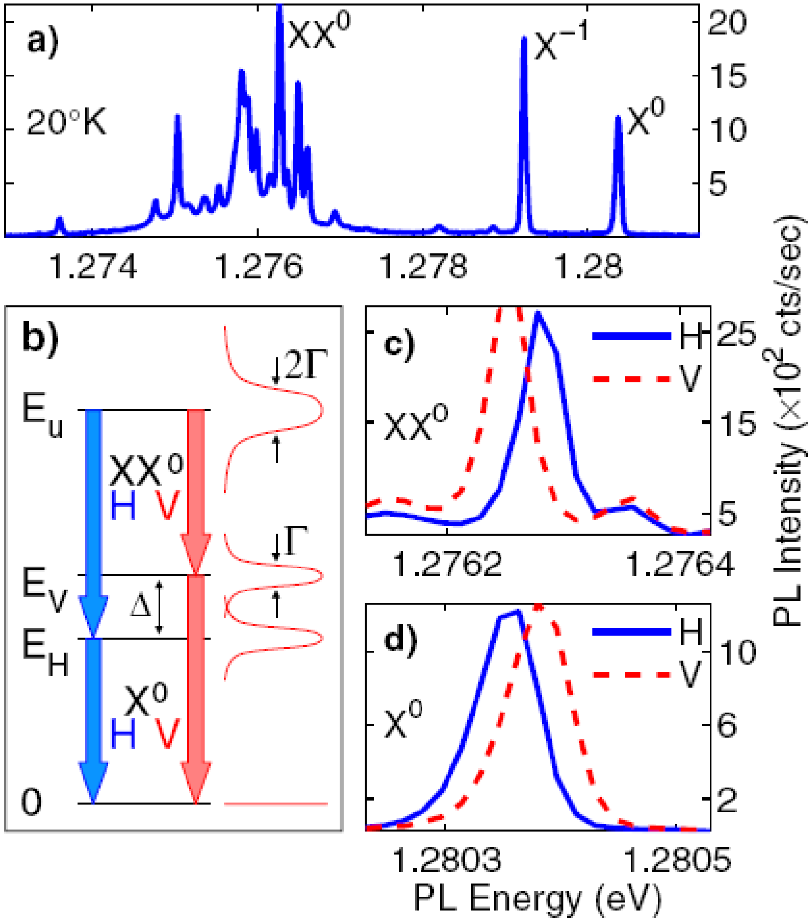
R.L. Williams, et al. J. Crystal Growth **223**, 321 (2001)

Entanglement

Key resource of quantum science

- Anisotropic splitting
- Exchange (shape anisotropy)
- State control
  - –Annealing
  - –Electric fields
  - –Magnetic fields
- Alloy effects as big as shape effects

  [Mlinar and Zunger, PRB 79, 115416 (2009)]

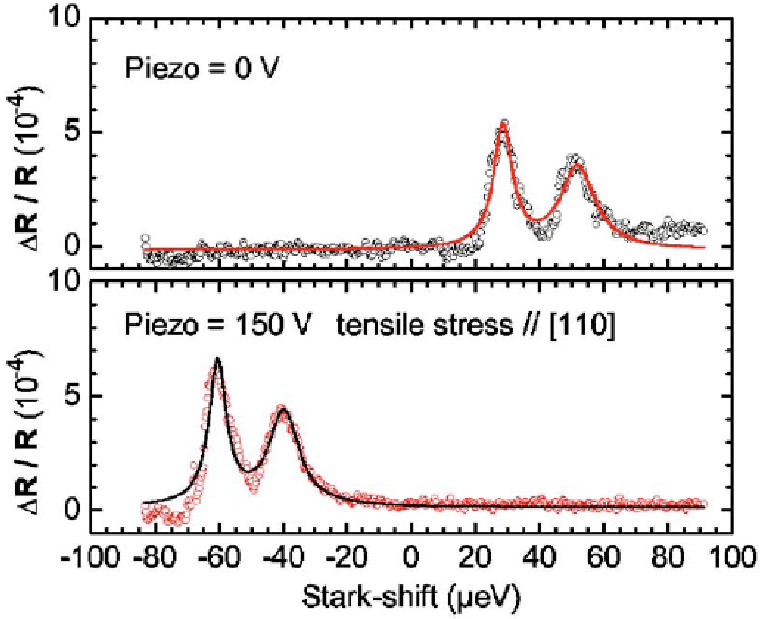


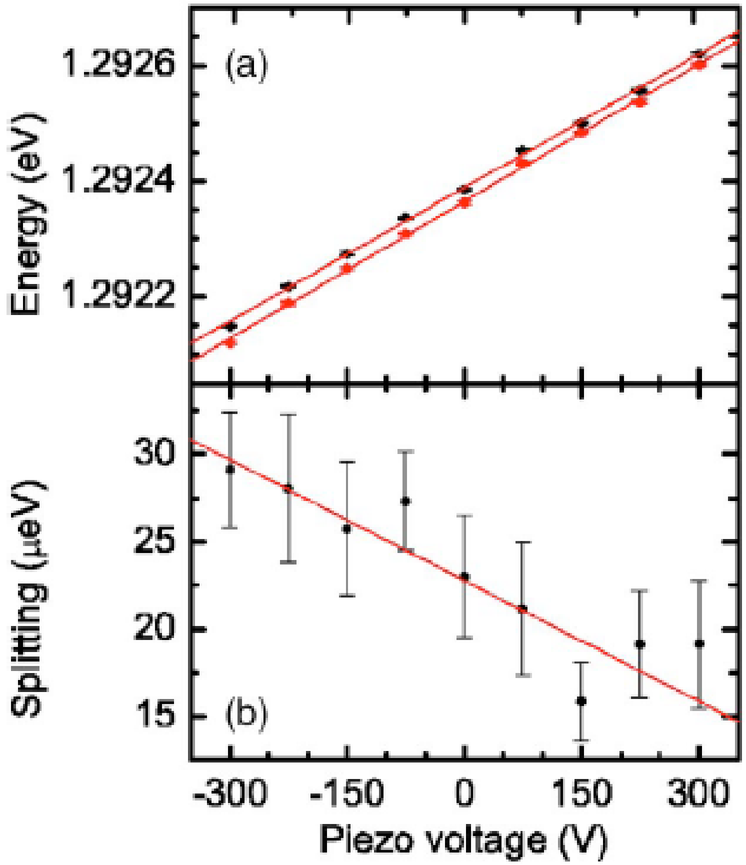
Goal: control states with external strain…what is possible

Uniaxial stress on excitons in quantum dots

[Seidl, et al., APL 88, 203113 (2006)]

Control splitting with external strain



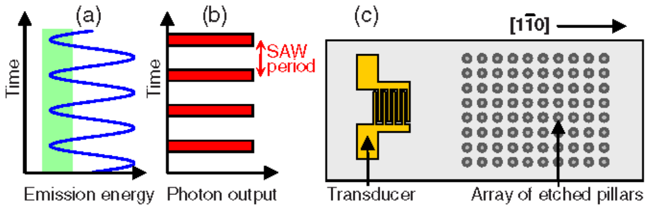


Modulation of single dot levels with surface acoustic waves

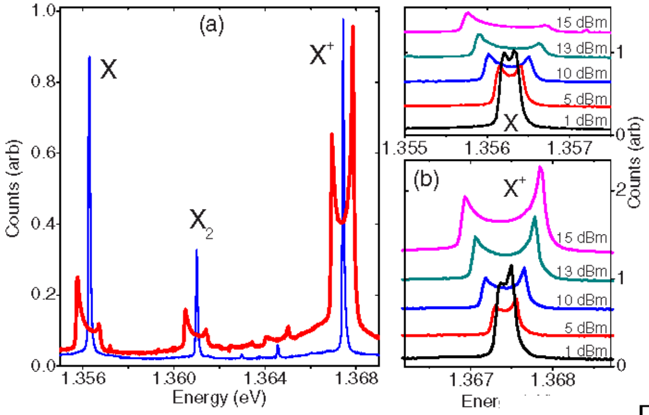
[Gell, et al., APL 93, 081115 (2008)]

Control energy levels with SAWs: single photon source

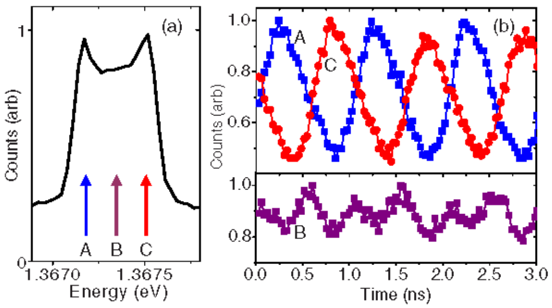
Scheme



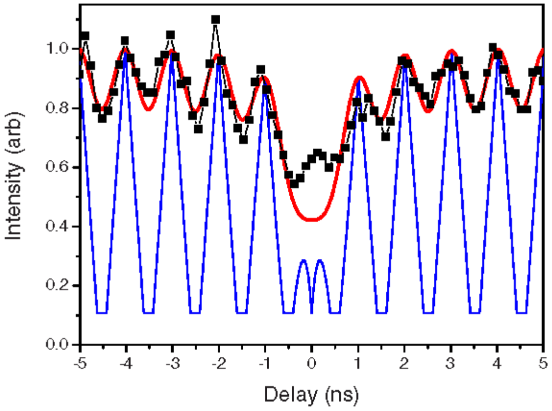
Modulation



Dynamics



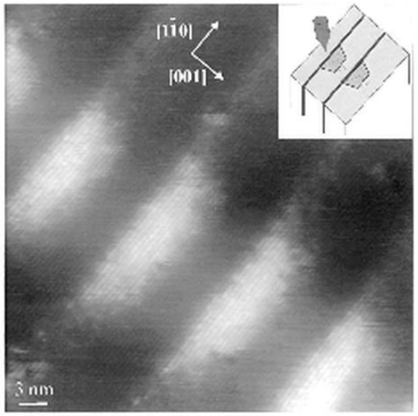
Antibunching



Coupled quantum dots

1D coupled dots

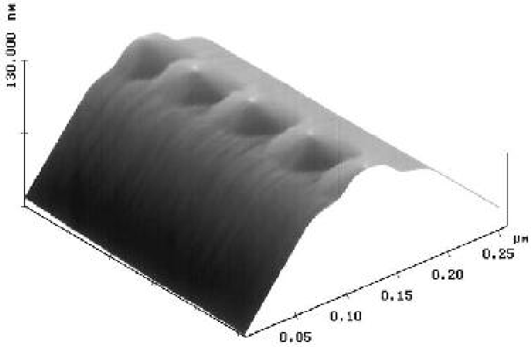
… vertically stacked self-assembled dots



B. Grandidier, et al., Phys. Rev. Lett. **85**, 1068 (2000)

… laterally coupled dot

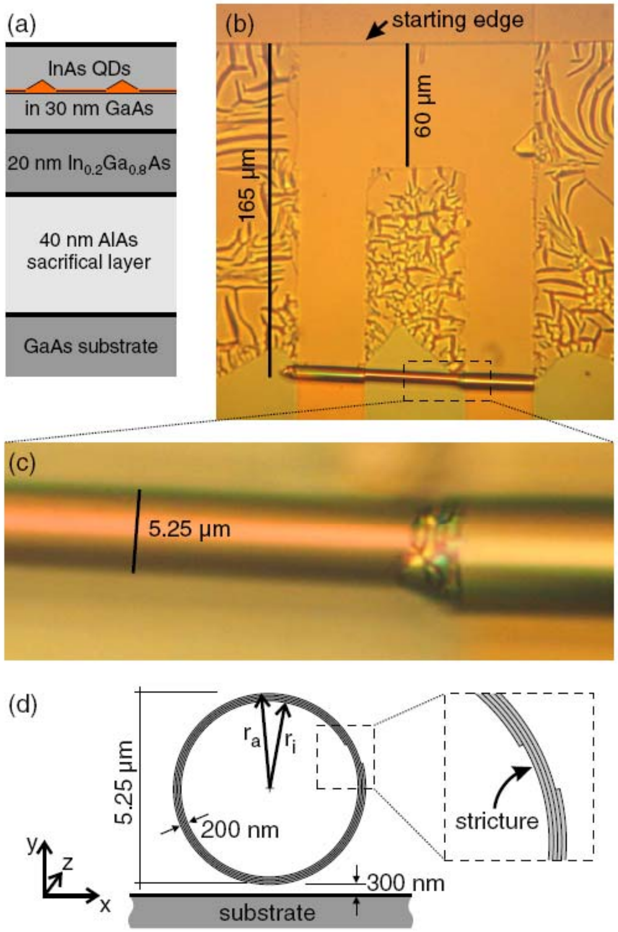
(by accident or design)

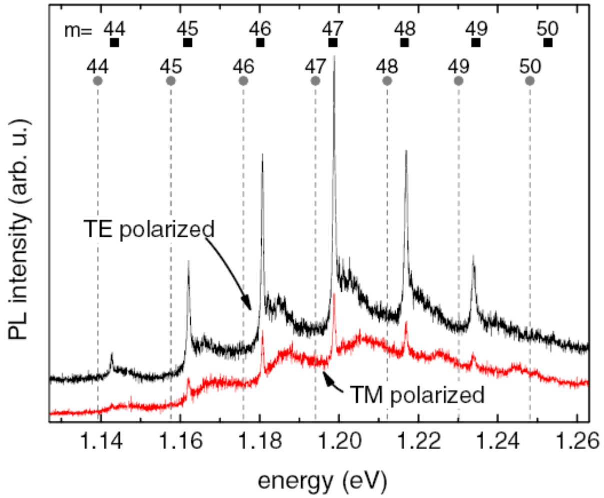


R.L. Williams, et al. J. Crystal Growth **223**, 321 (2001)

Novel optical cavities: Rolled up structures

[Kipp et al. PRL **96**, 77403 (2006)]



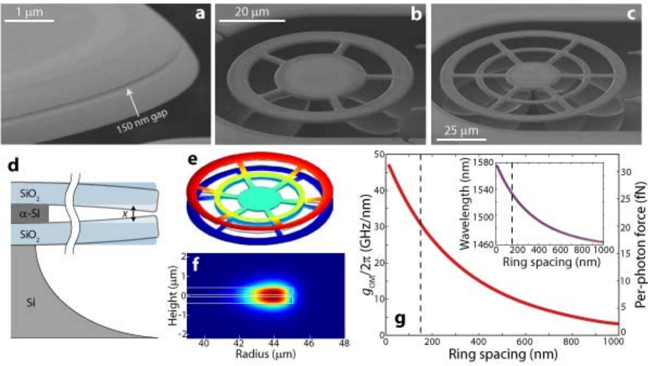


Modulating QD properties with rolled up waveguides: radius of curvature ~ μm

S = a: radius of curvature ~1μm

Seidl et al.: 1 GPa uniaxial strain at breaking…~10 meV shifts for GPa strains

… more uses for nanomechanical/QD hybrids



Local strain gauge for

- … nanomechanical energy harvesting
- … mass sensing
- … motion sensing

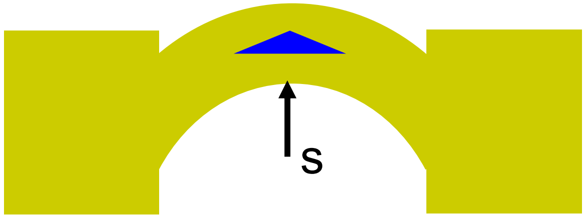
Mechanical computing

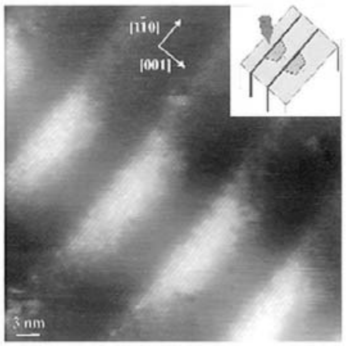
- … nanomechanical memory
- … coupling to nanoelectronics and nanooptics
- … coupling to solid state QI systems

Optomechanics [Painter group, Caltech: Rosenberg et al., arXiv:0905.3336v1]

Outline

- Modeling nanomechanical/QD hybrids
  - Atomistic tight-binding models for QDs
  - Valence force field
  - Model for external strain
- Self-assembled dots under external strain
  - Electron and hole levels, band gaps
  - Strain relaxation
  - Charge redistribution
- Vertical and lateral double dots
  - Electron and hole levels
  - Strain-induced tunneling
- Excitons
  - Fine structure
  - Strain-induced mixing
  - Polarization rotation





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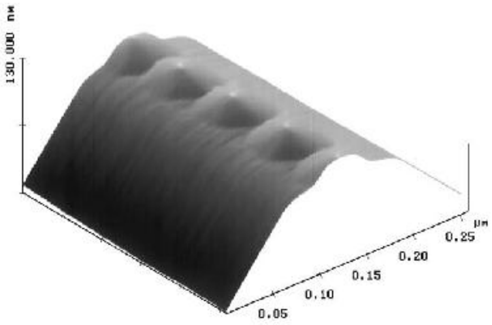
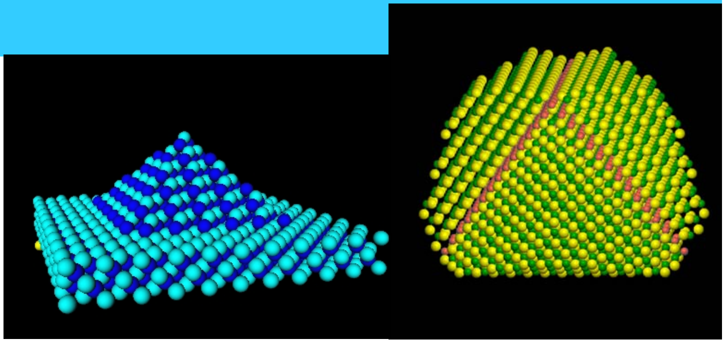


R.L. Williams, et al. J. Crystal Growth **223**, 321 (2001)

Empirical Tight-Binding Calculations: Atomistic Approach



Atomic geometry
… precise definition of size, shape, surfaces

Model: sp^{3}s* or sp^{3}s*d^5 atomic orbitals, nearest-neighbor coupling, spin-orbit

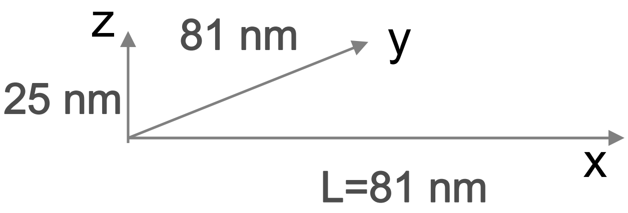
Strain: empirical valence force field

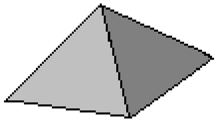
Empirical tight-binding parameters
… sp^{3}s*: Vogl, Hjalmarson and Dow, J. Phys. Chem **44**, 365 (1983) + so
… sp^{3}s*: CdS, ZnS: Lippens and Lannoo, PRB **39**, 10935 (1989)
… sp^{3}s*d^5: Jancu et al., PRB **57**, 6493 (1998)
… sp^{3}s*d^5 : Sapra, Shanti, and Sarma, PRB **66**, 205202 (2002)

Optical response: dipole matrix elements from Slater orbitals, on-site only

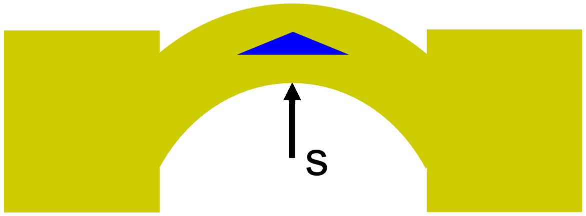
Self-assembled quantum dots in nanomechanical oscillators

Results for small bridge with pyramidal QD

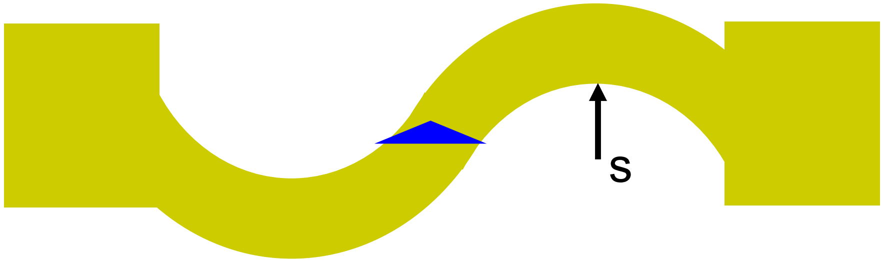




height 3 nm
base 7 nm

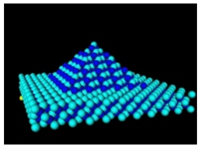


Symmetric bend (biaxial deformation)



Antisymmetric bend (shear)

- atomistic tight-binding theory: spin-orbit interaction, no piezo
- relaxation: valence force field
- external strain: <u>fixed</u>, bent bridge
- internal lattice relaxation due to InAs/GaAs lattice mismatch and applied strain
- excitons: configuration interaction approach



Caveats

One QD size (small ~1/2 size)

Symmetric QD

Strain applied only along x (100)

… minimal piezoelectric effect (ignore)

… nanomechanical structures but not SAWs

Static distortion

… scattering of nanomechanical modes by QDs (??)

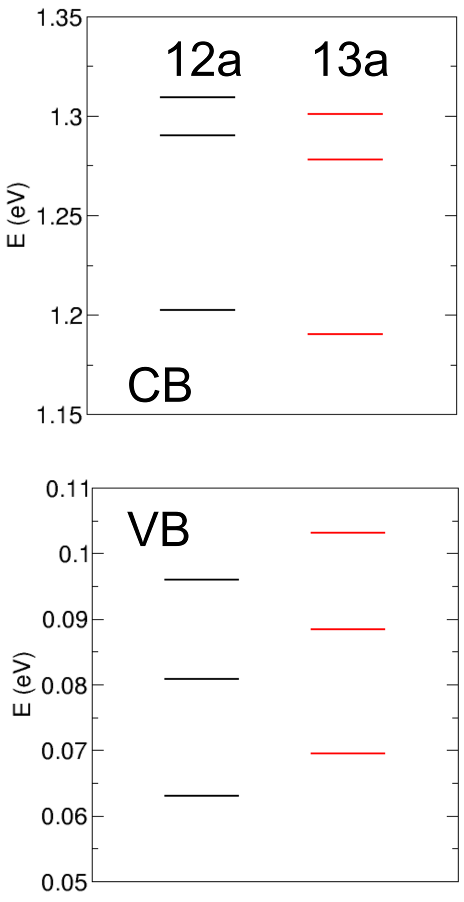
One type of boundary condition

… sensitivity to boundary conditions (?)

Internal biaxial strain

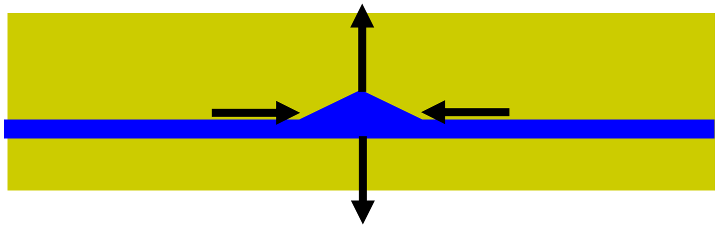
Compression in x,y ($\Delta V > 0$)
Expansion in z ($\Delta V < 0$)

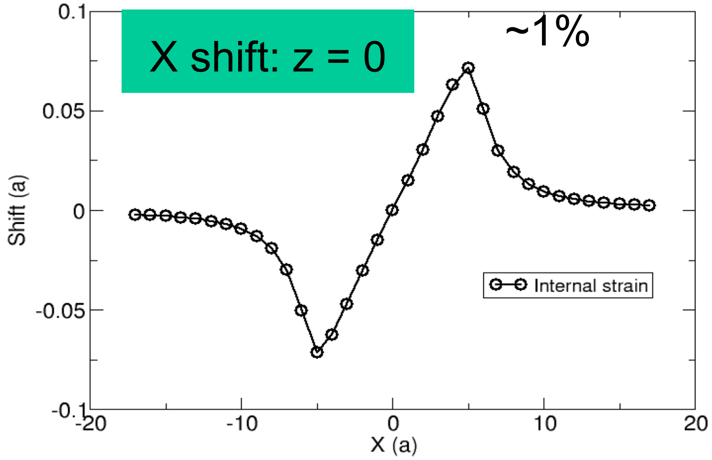
Bound states



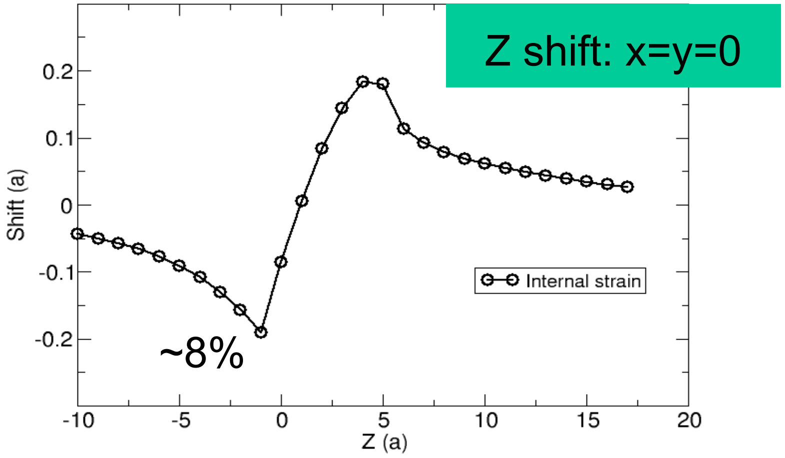
Lattice relaxation [7% mismatch]

$a_{GaAs} = 5.65$ Å; $a_{InAs} = 6.06$ Å





X shift: z = 0



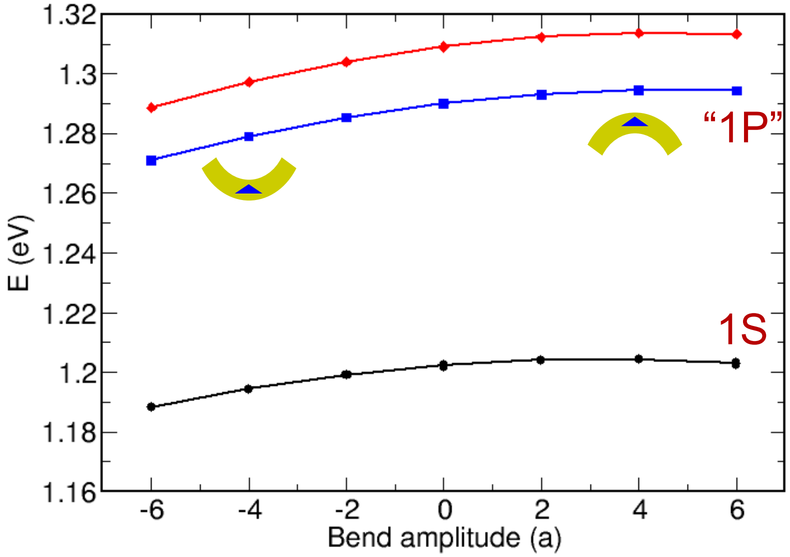
Z shift: x=y=0

Manipulating electron and hole energies

Electrons

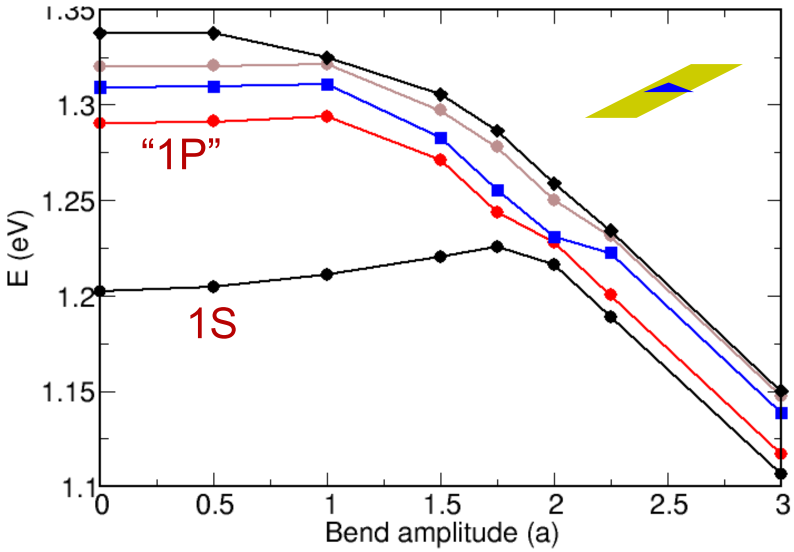
Biaxial deformation

- “Rigid” shift with fixed level ordering and state symmetries
- Shift: 1-10 meV
- Electron and “hole” shift in same way



Shear

- Quadratic increase
- Mixing, collapse to wetting layer
- Electron and “hole” shift in opposite way



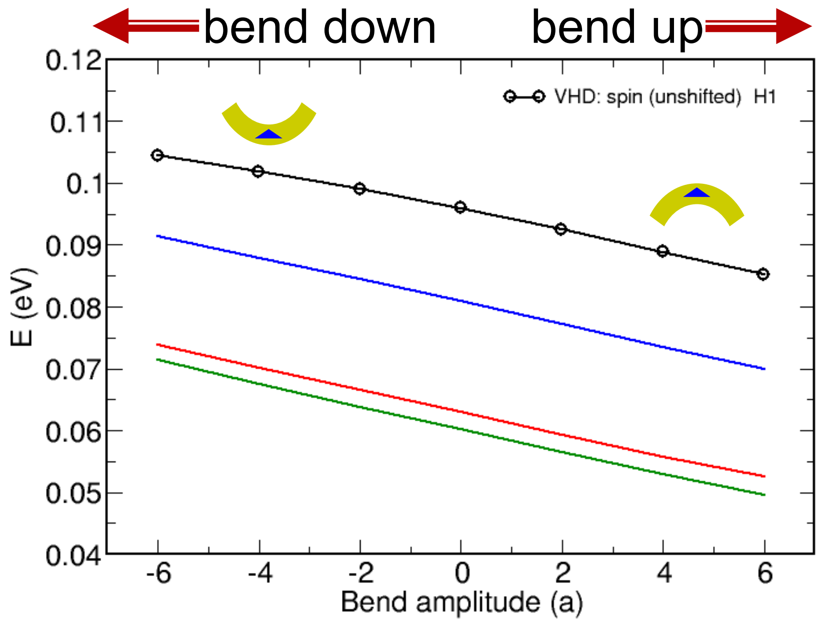
“Analog” to DC Stark effect: same or opposite charge

InAs QDs in GaAs bridges: coupling to bending modes

Holes

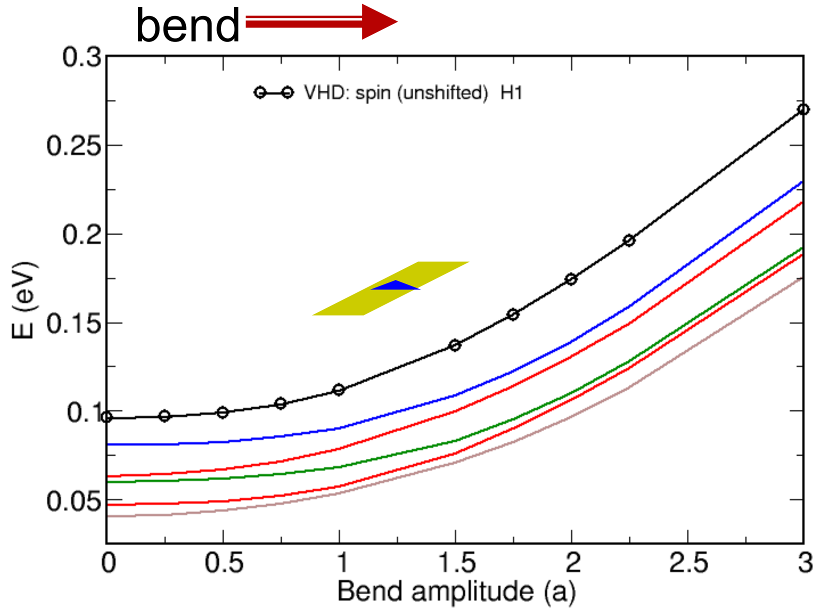
Biaxial deformation

- “Rigid” shift with fixed level ordering and state symmetries
- Electron and “hole” shift in same way



Shear

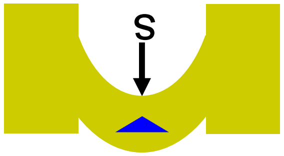
- Quadratic decrease
- Mixing



InAs QDs in GaAs bridges: coupling to bending modes

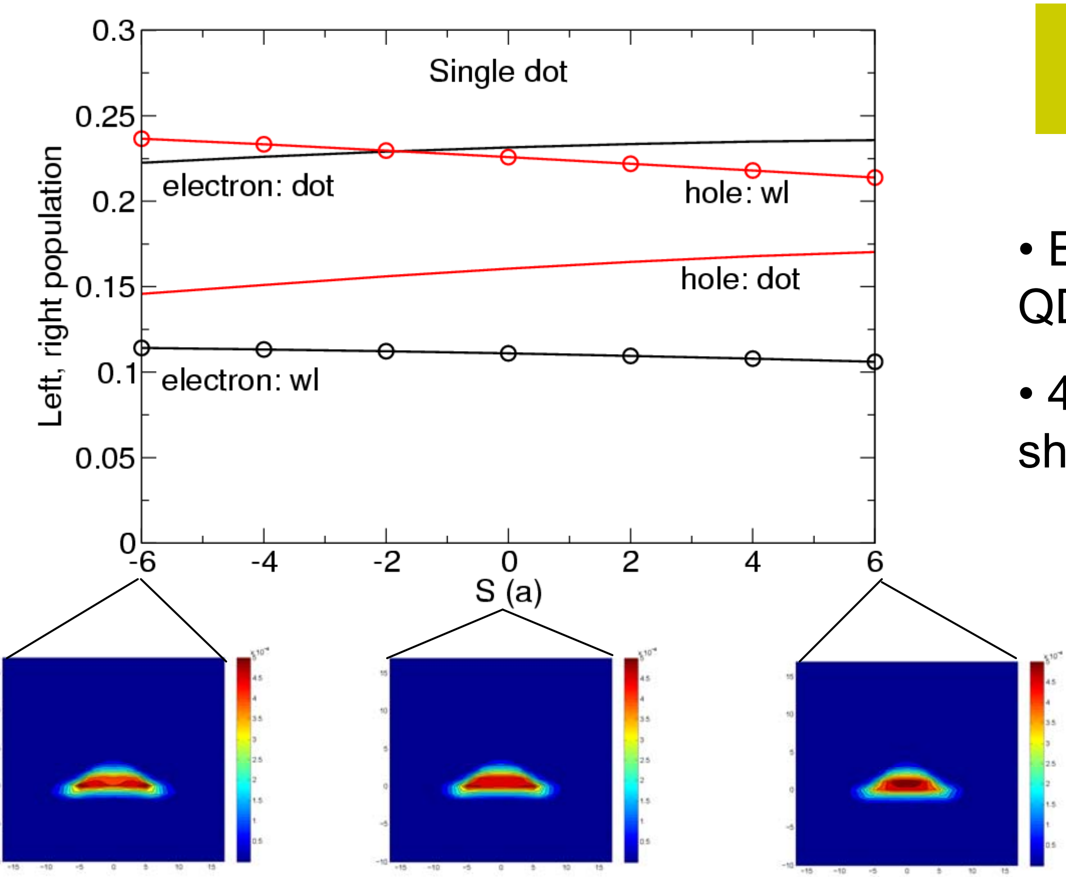
•Electron localized to QD

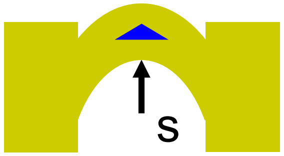
•Hole more localized in WL under the dot



• E and h: **down**, out of QD, into WL

• 4-10% charge shifts





• E and h: **up**, into QD, out of WL

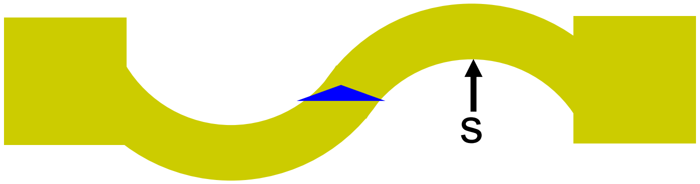
• 4-10% charge shifts

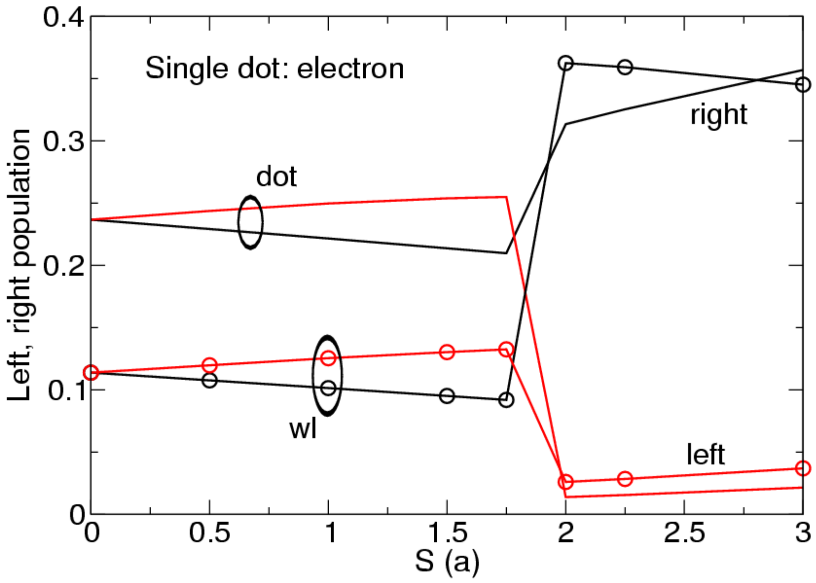
Hole ground state

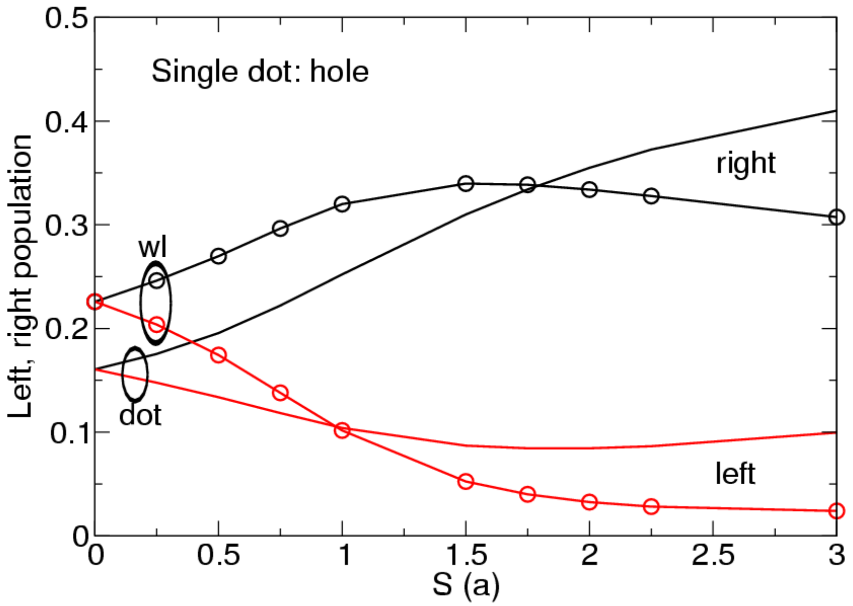
“Analog” to DC Stark effect

InAs QDs in GaAs bridges: coupling to shear modes

Electron and hole ground state







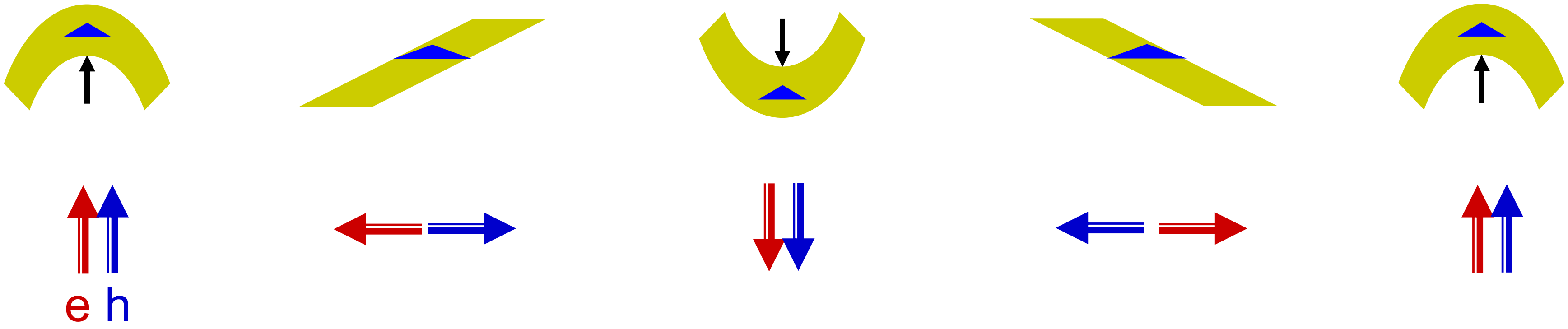
E: pushed **left**, then **right** and into **WL**

H: pushed **right** and into **dot,**

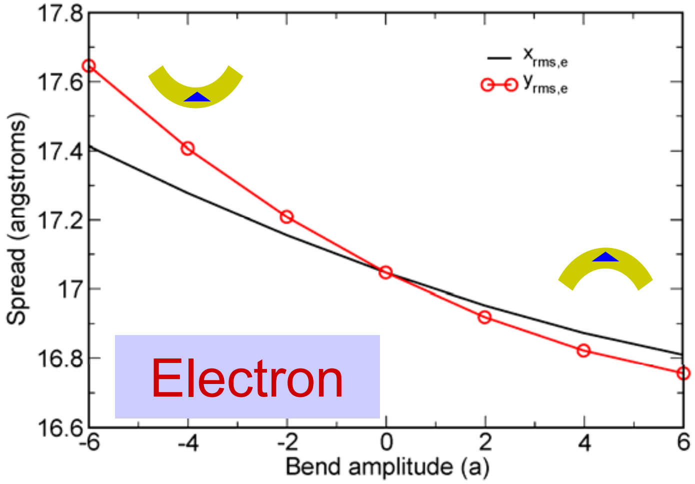
50% polarization

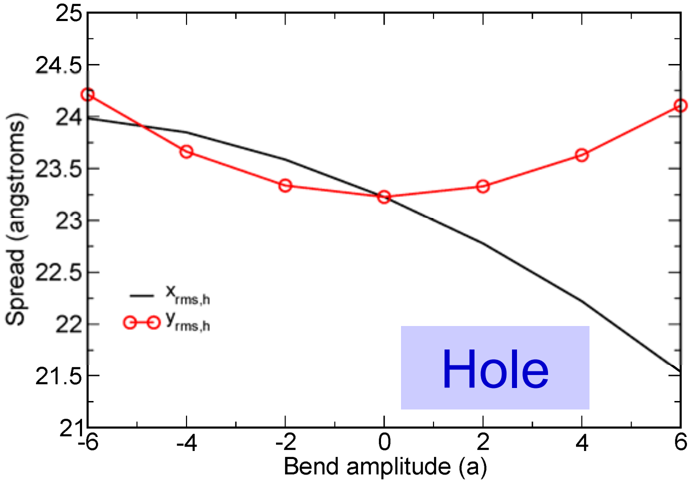
Manipulating electron and hole states

“Analog” to DC Stark effect: counter-rotating e and h



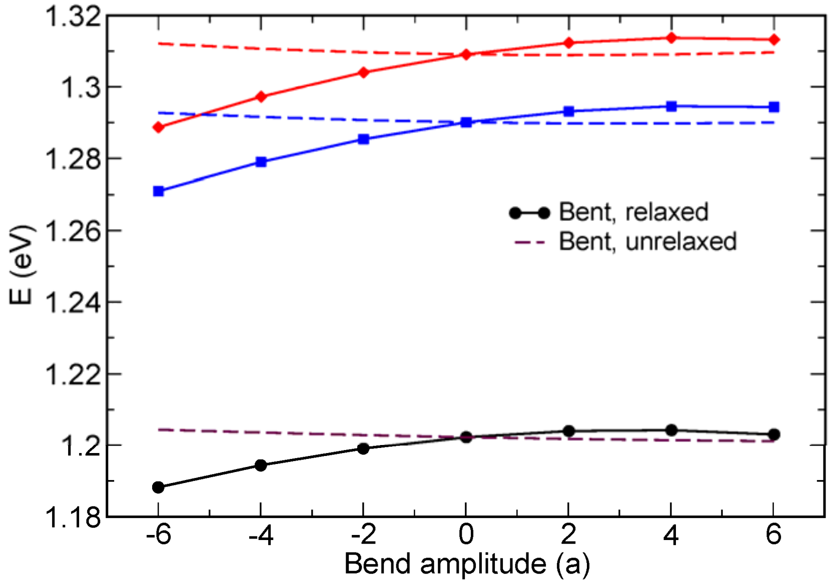
Asymmetric stretching or squeezing



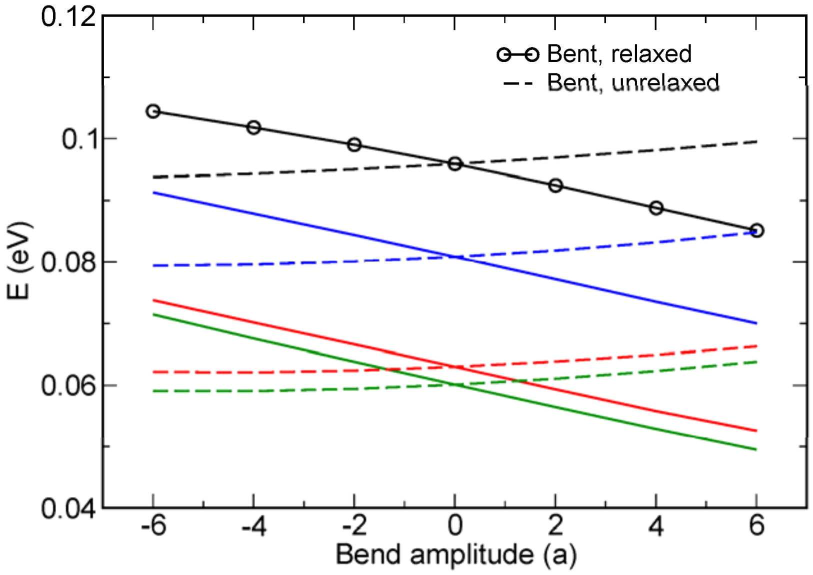


Importance of internal relaxation: symmetric bend

Electrons



Holes

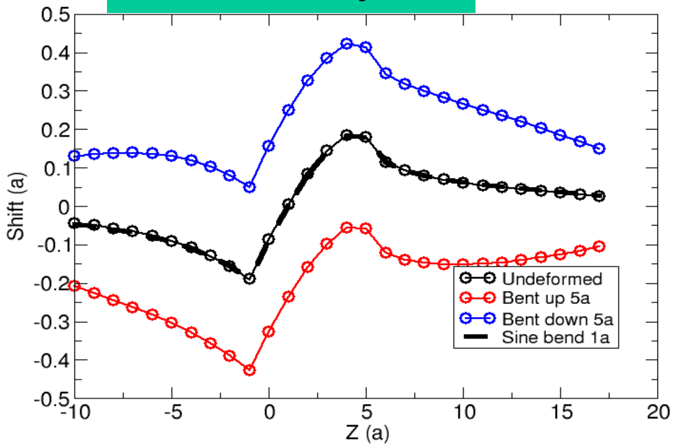


Biaxial deformation: internal relaxation is critical

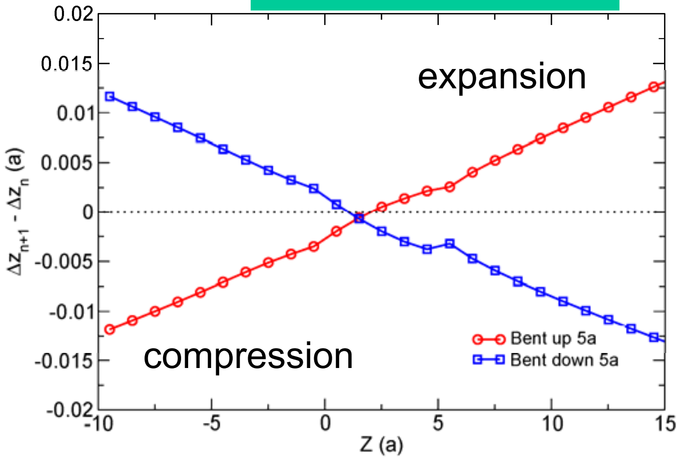
Importance of relaxation

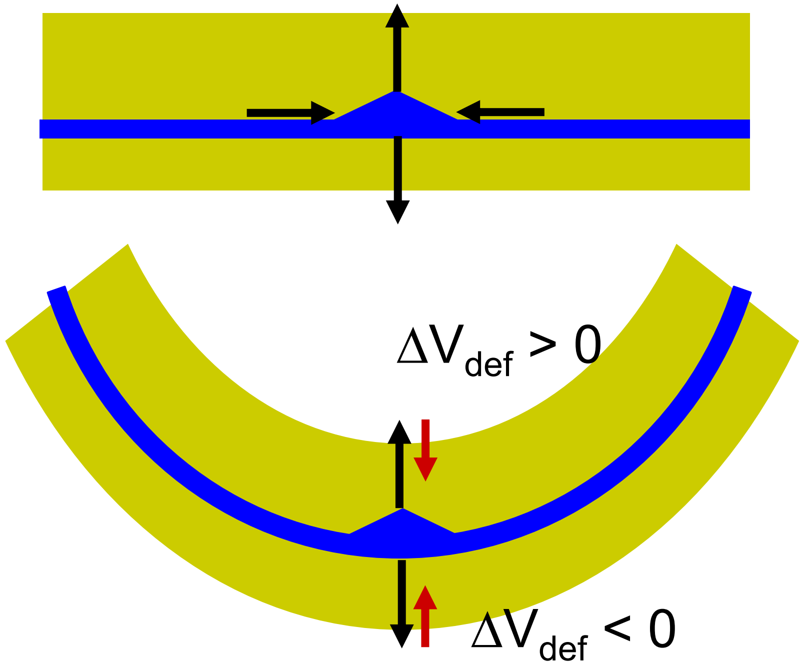
Lattice deformation

Z shift: x=y=0



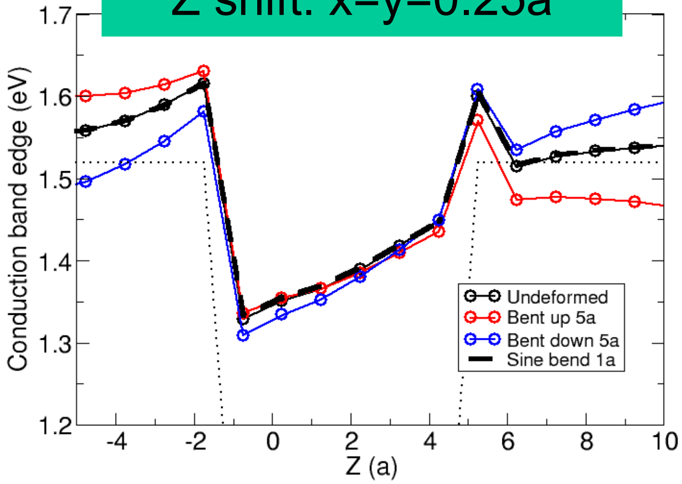
δZ shift: x=y=0





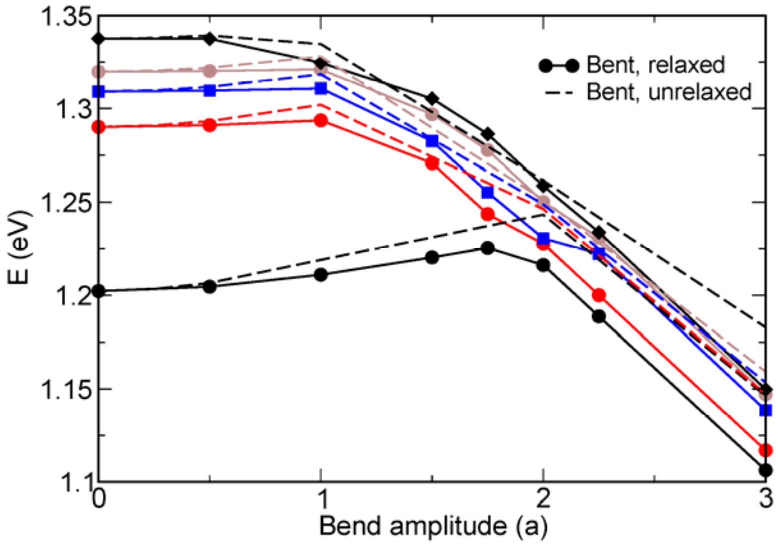
Conduction band edge

Z shift: x=y=0.25a

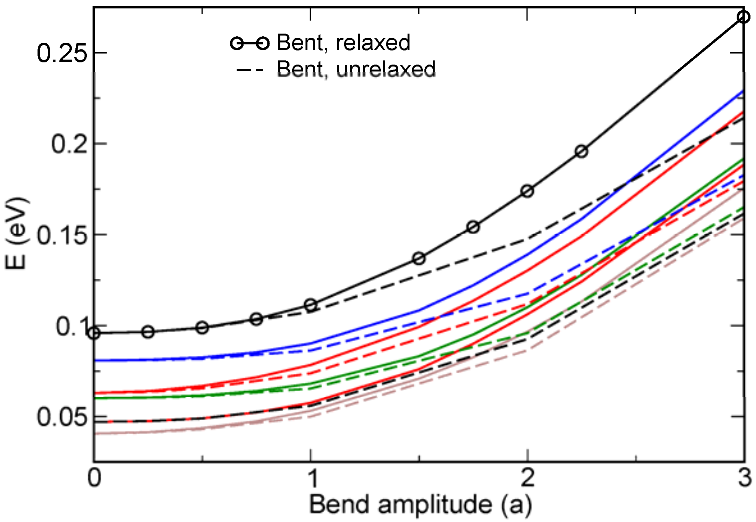


Importance of internal relaxation: shear bend

Electrons



Holes

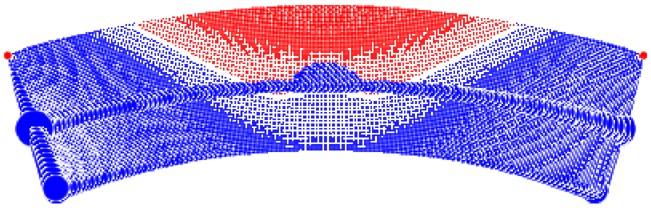


Shear deformation: bending shear is dominant

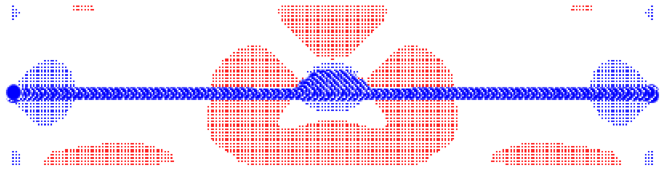
Sensitivity to boundary conditions

Strain distribution (Trε) for up and down bendings

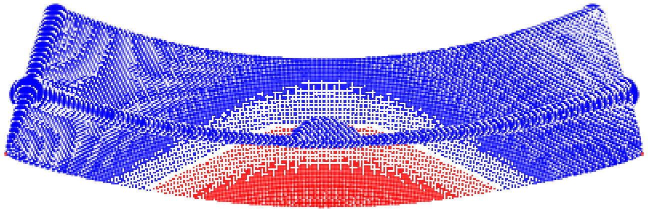
bent up



unbent



bent down



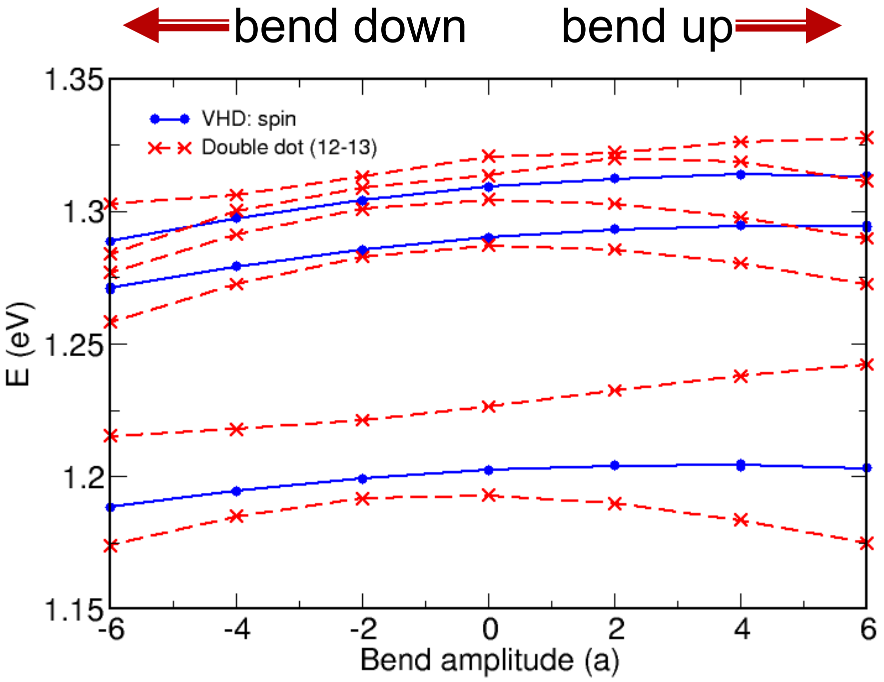
blue – compression

red - expansion

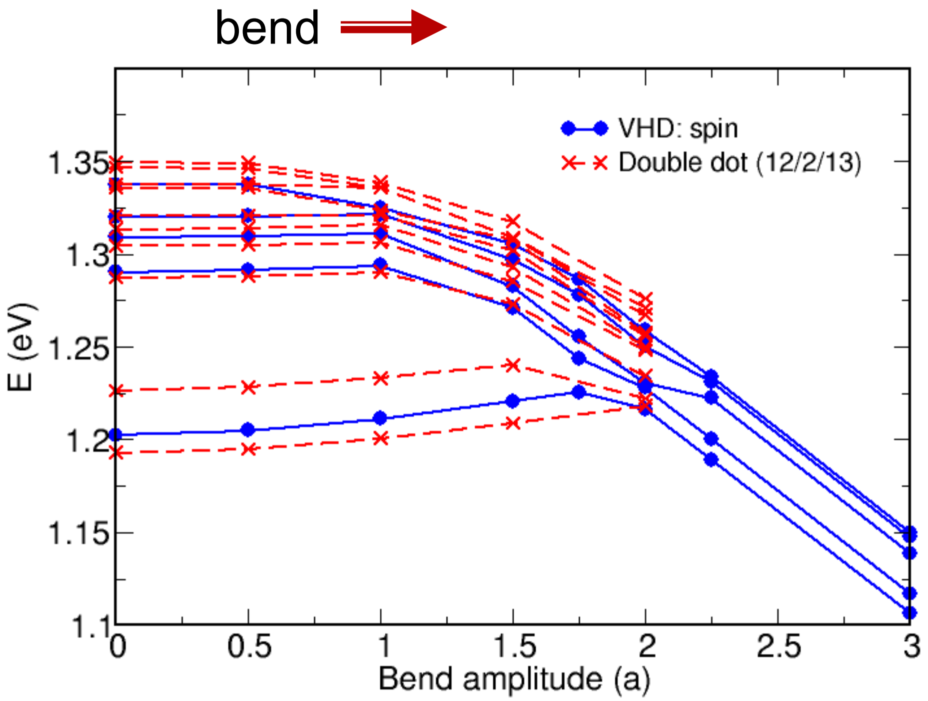
Transferring electrons and holes in coupled dots

Vertical double dot: electrons

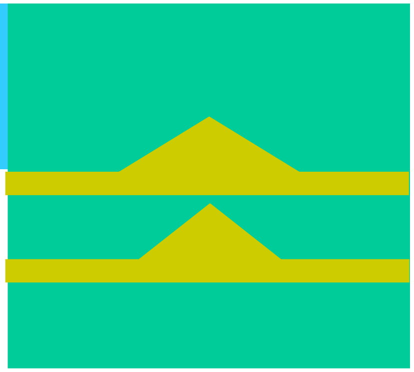
Biaxial deformation



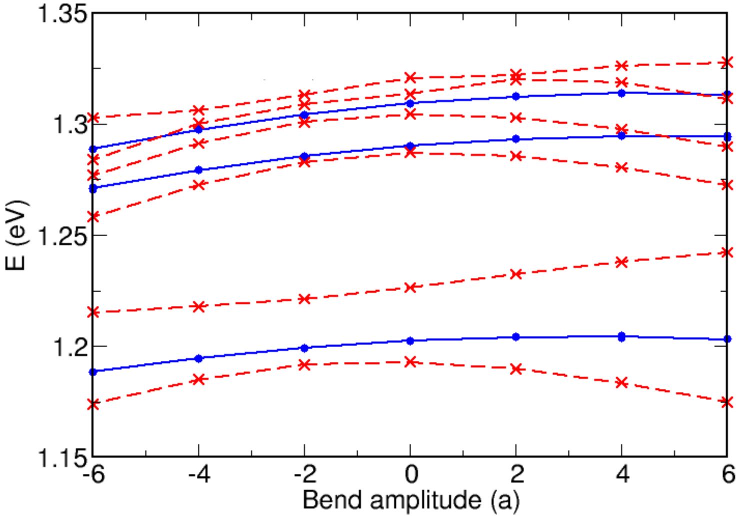
Shear



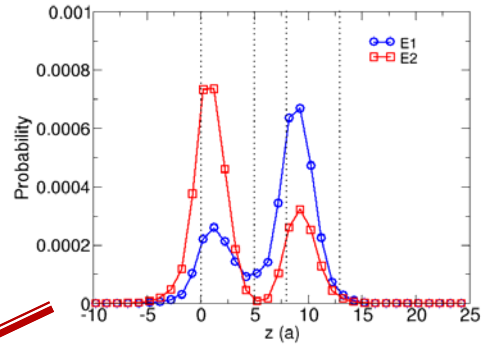
Manipulating inter-dot transfer



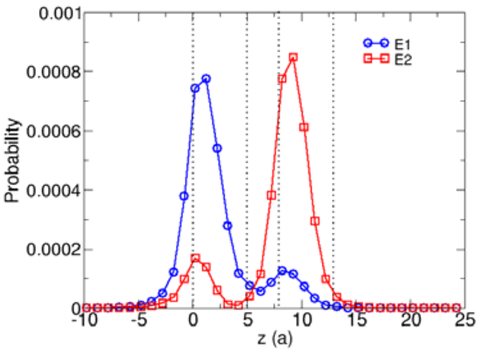
Biaxial deformation: electrons



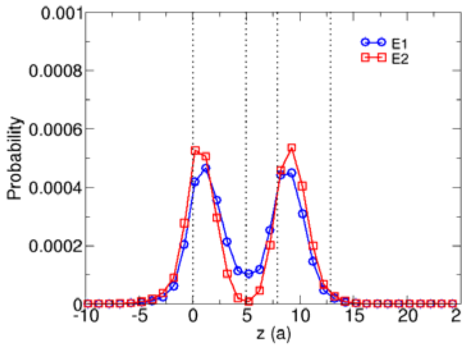
Flat



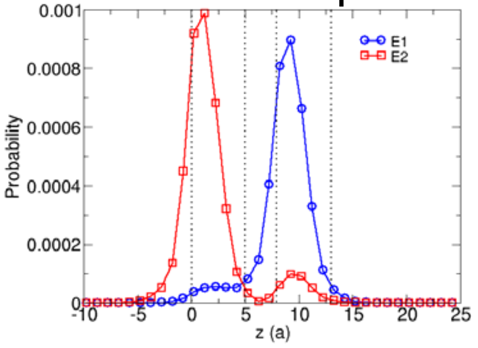
Down 6a



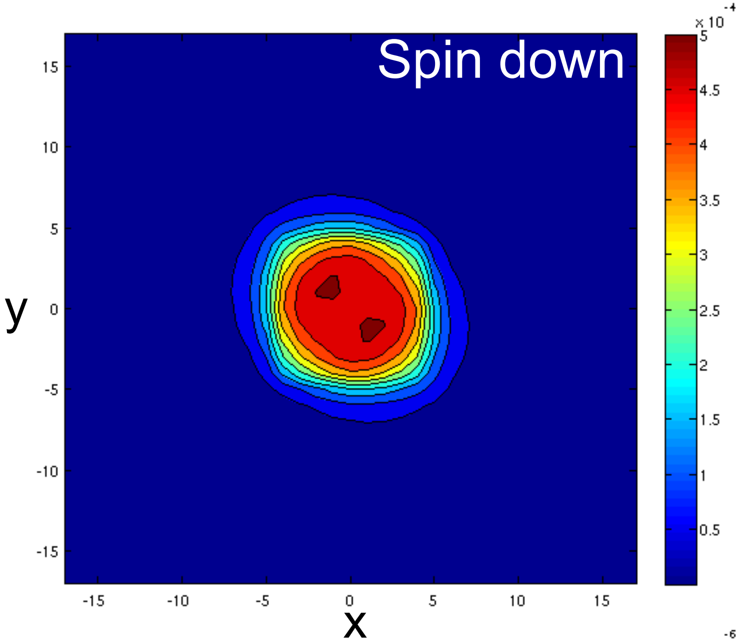
Down 2a

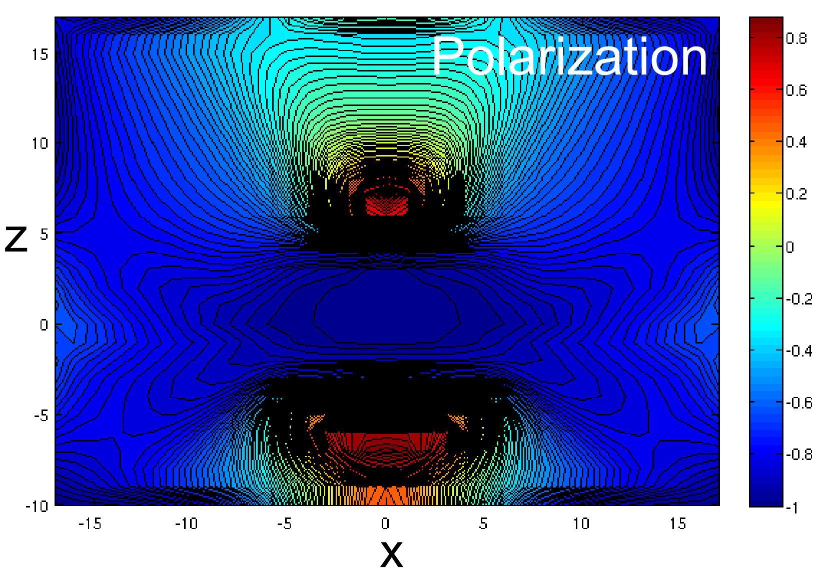


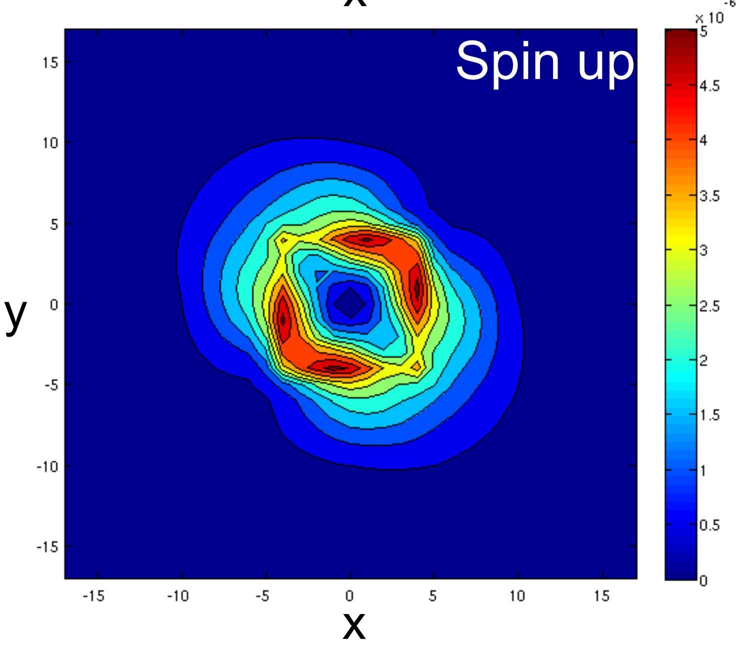
Up 6a

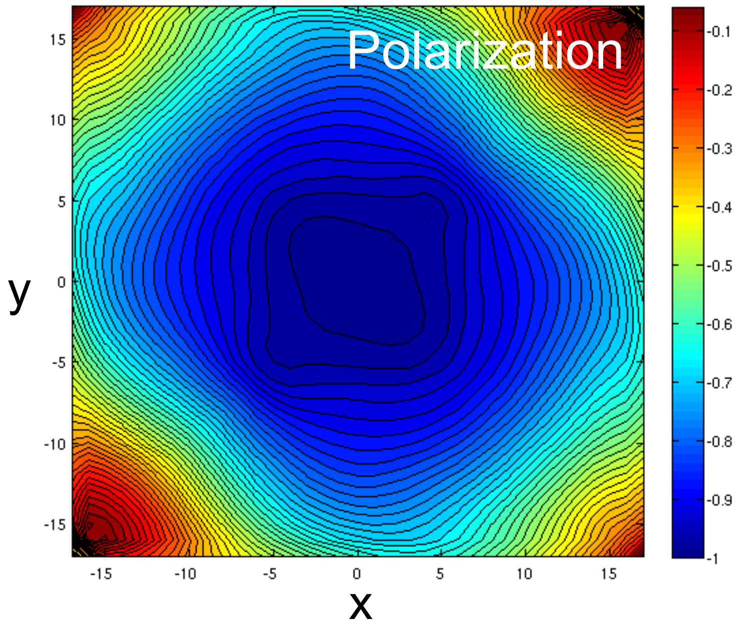


Hole ground state: spin distribution in an unbent dot

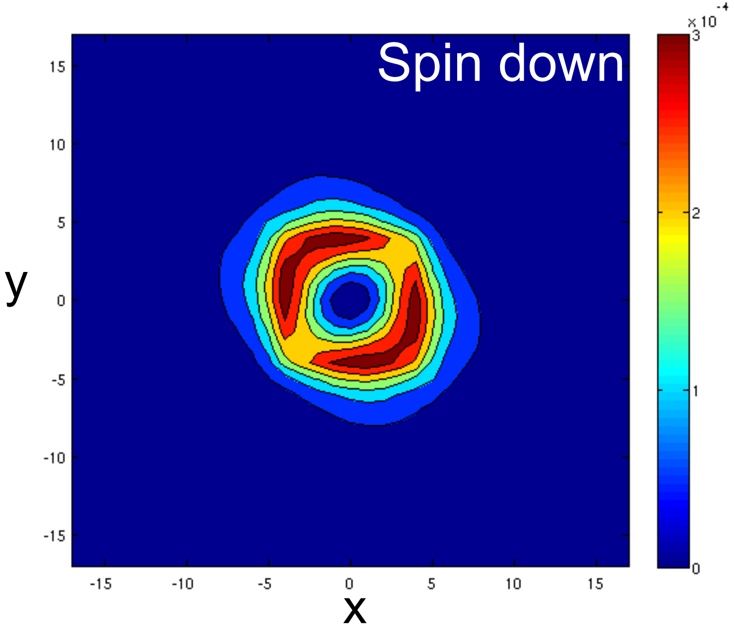


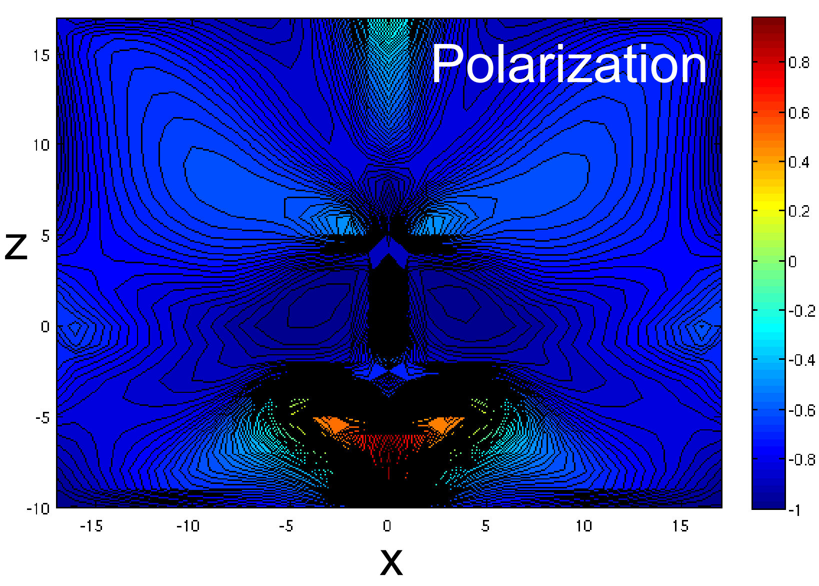


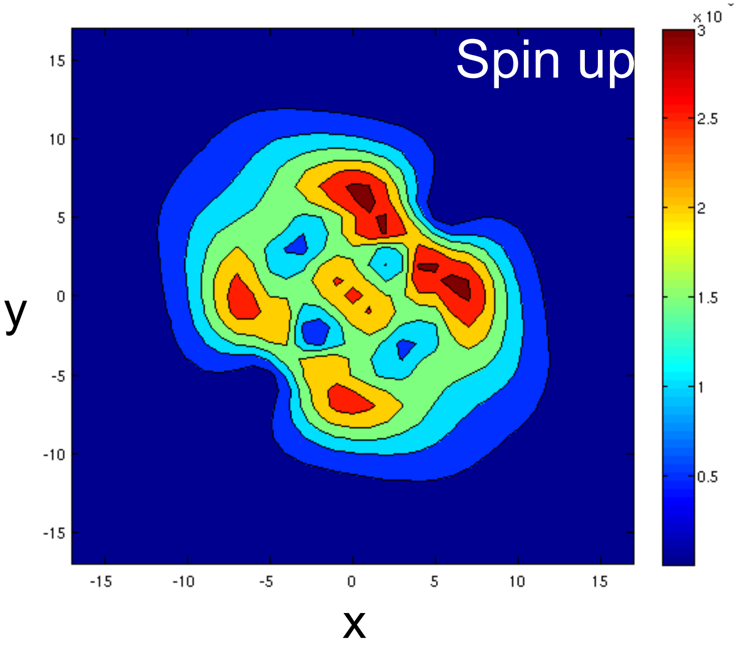


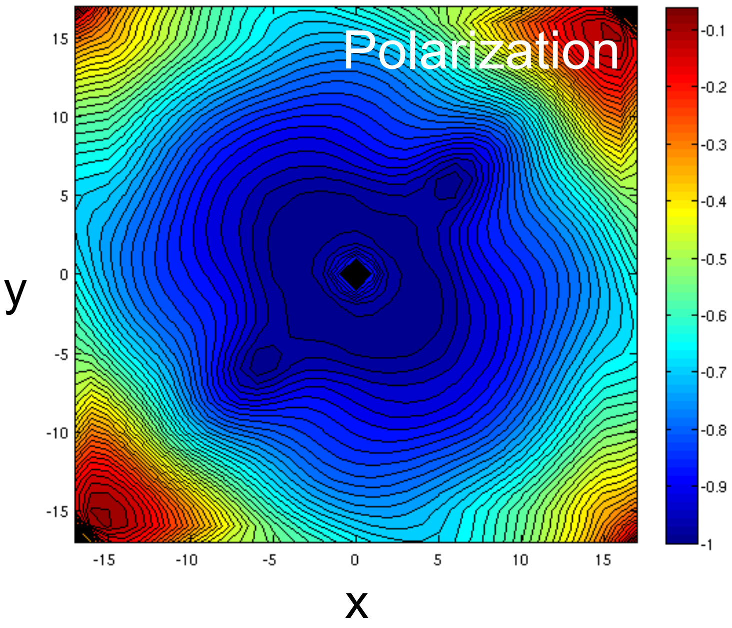


Hole excited state: spin distribution in an unbent dot



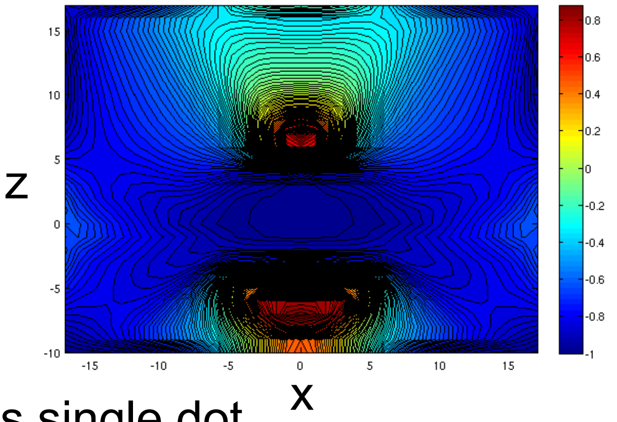


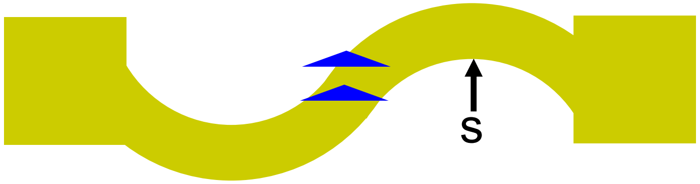




Spin-dependent coupling ??

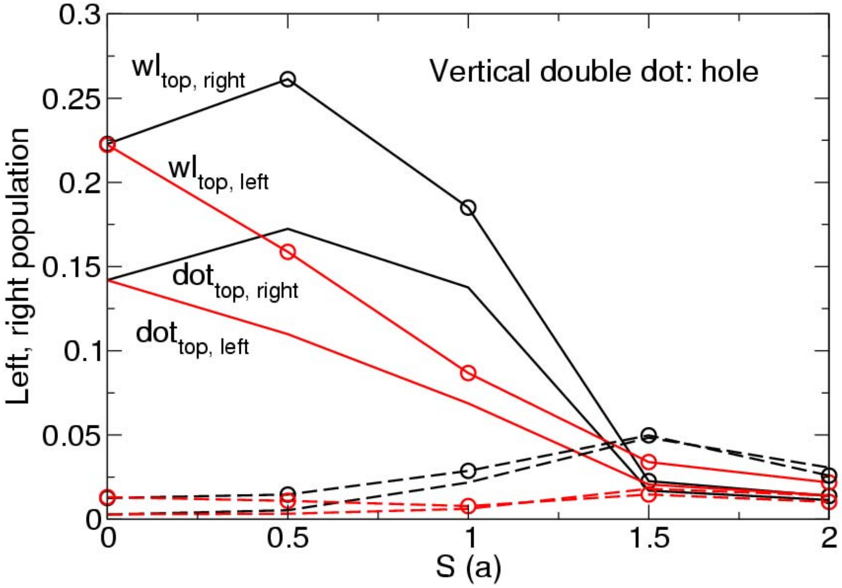
Vertical double dot: holes



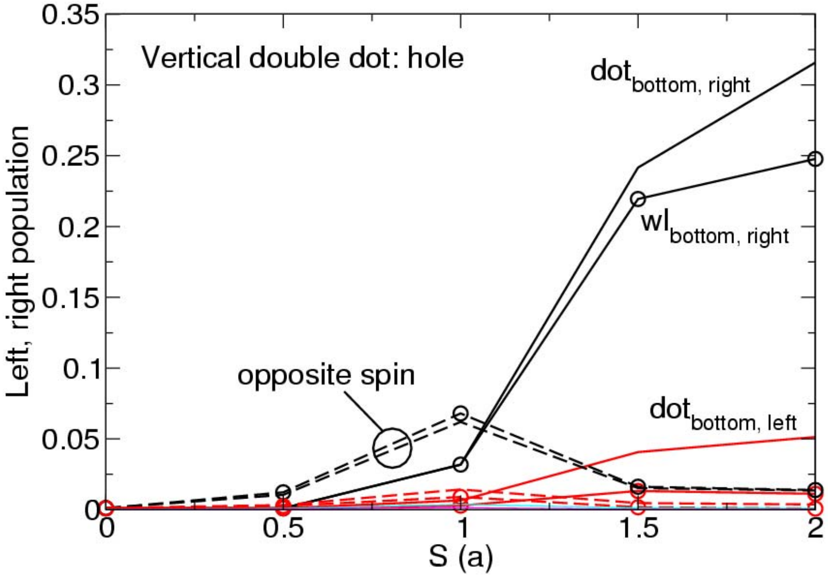


- Initially, follows single dot
- **Strong** transfer top to bottom dot
- Spin mixed from other dot

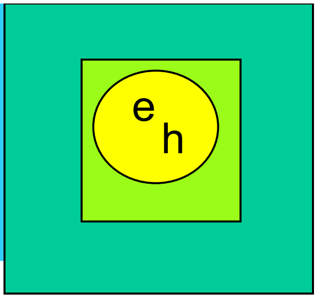
Top dot



Bottom dot

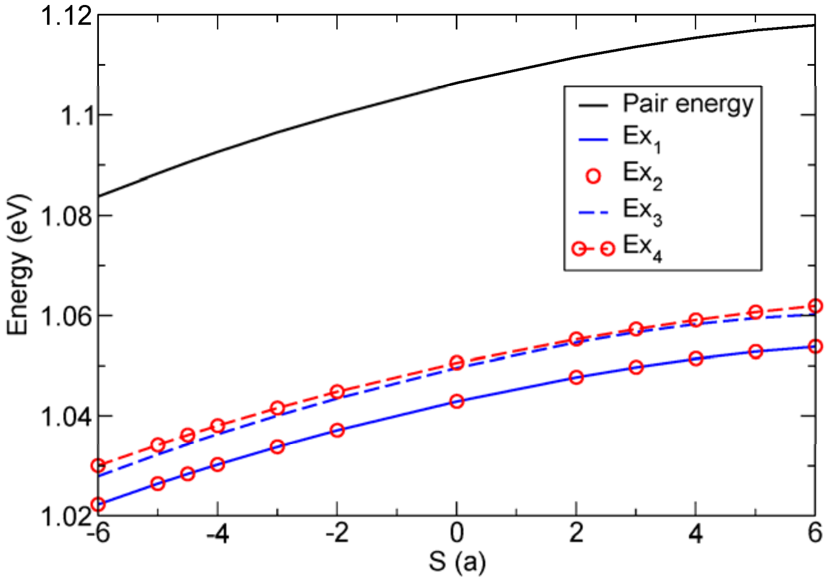


Excitons: biaxial deformation



Exciton follows pair ground state

Binding can increase or decrease by bending (charge shifting)



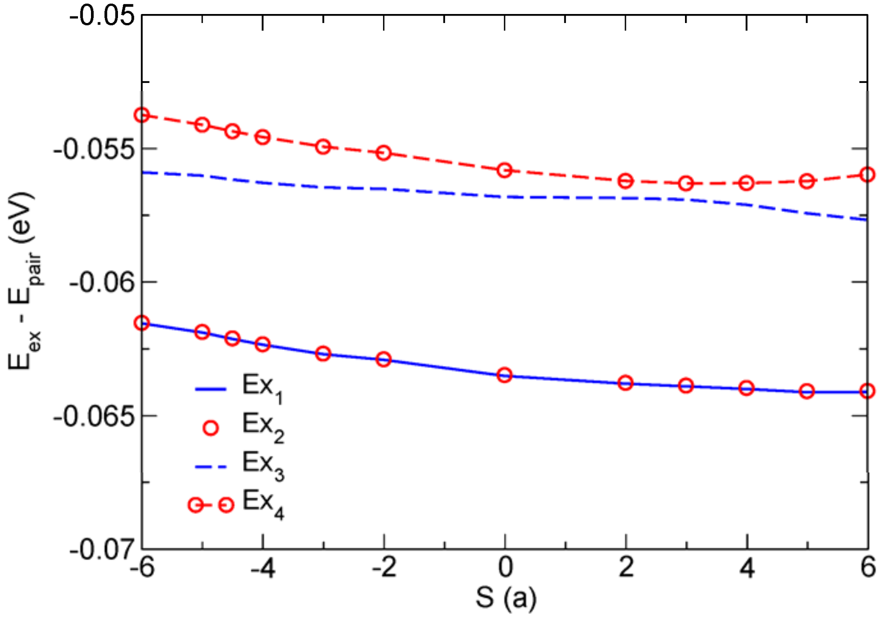
<u>Fine structure</u>

Ex_1 and Ex_2 are dark (“triplet”)

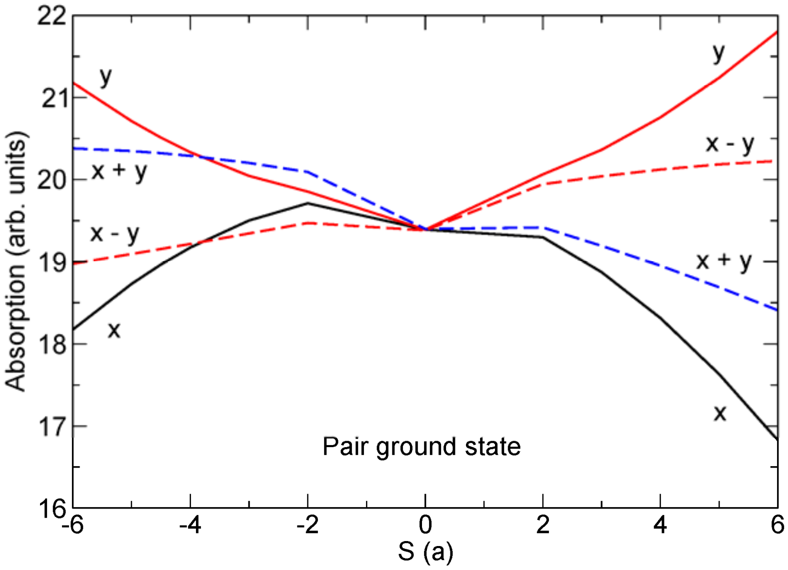
Ex_3 and Ex_4…exchange split bright states (“singlet”)

Ex_3 and Ex_4…asymmetric exchange

Bend-induce level crossing and polarization rotation



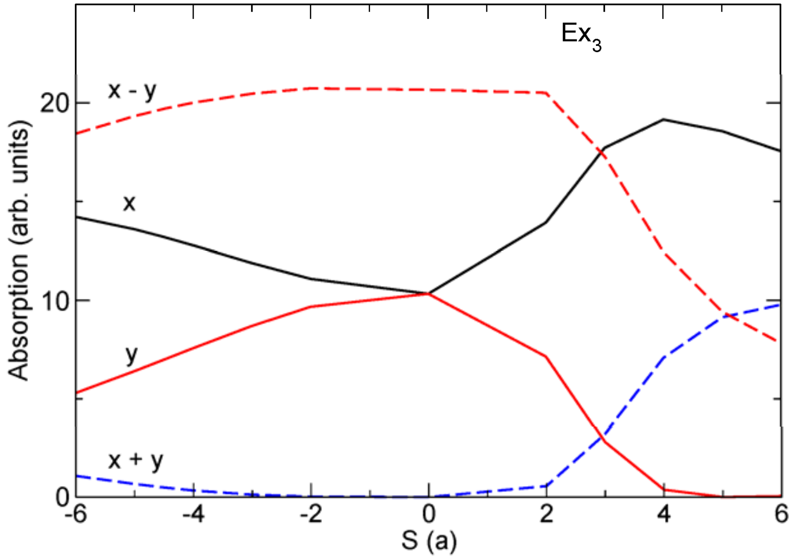
Excitons: biaxial deformation

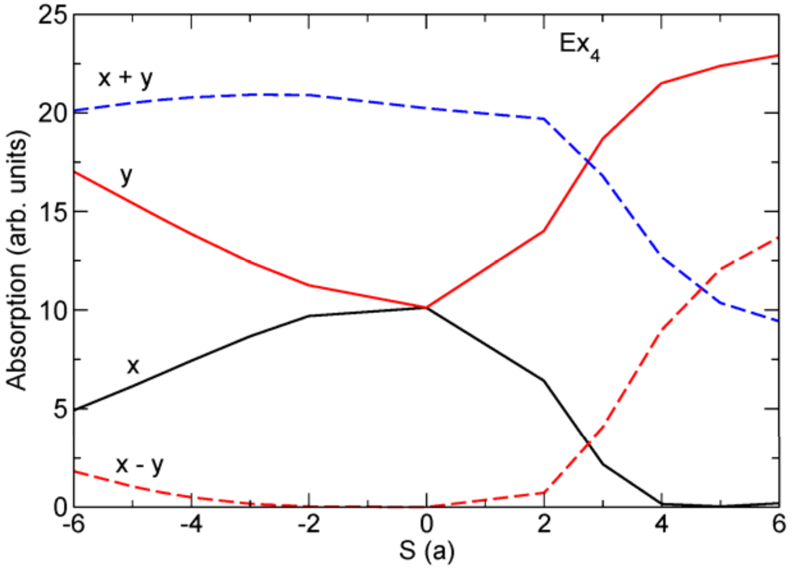


Pair ground state weakly polarized, strain polarizes along y

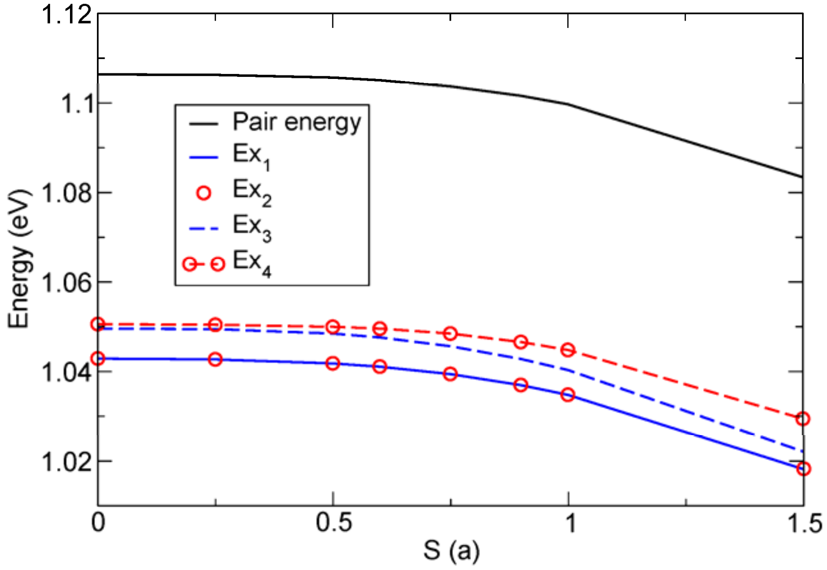
$Ex_{3(4)}$ polarized along x-y (x+y) in unbent structure

$Ex_{3(4)}$ polarized along x (y) by bending





Excitons: shear



Exciton follows pair ground state

Binding increases by bending (hole squeezing)

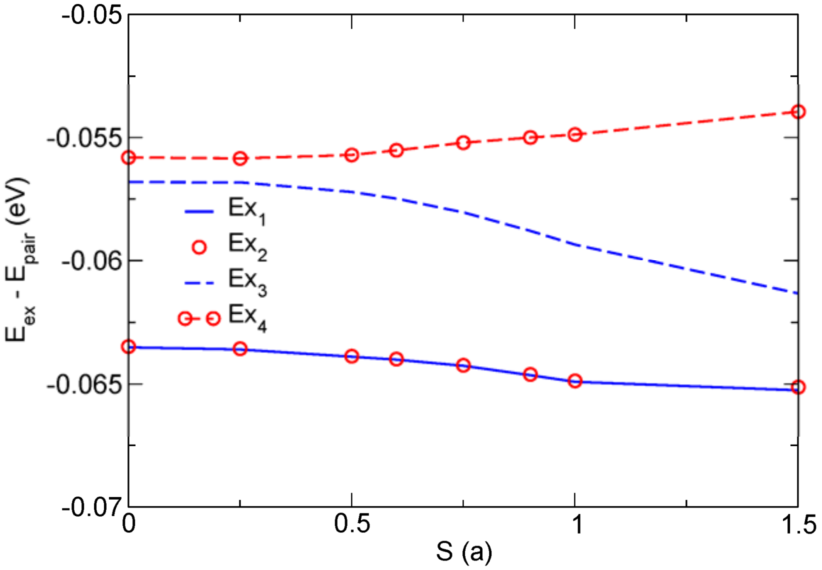
<u>Fine structure</u>

Ex$_1$ and Ex$_2$ are dark ("triplet")

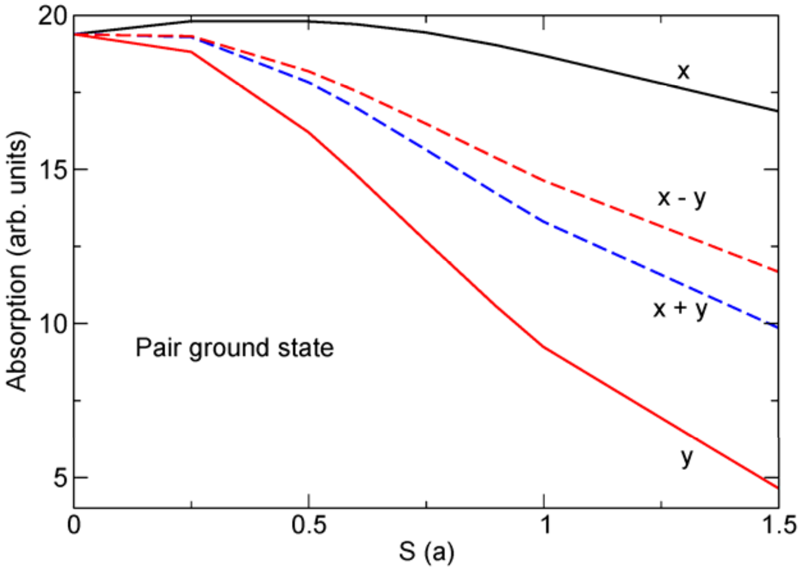
Ex$_3$ and Ex$_4$…exchange split bright states ("singlet")

Ex$_3$ and Ex$_4$…asymmetric exchange and strong coupling

Bend-induce level crossing and polarization rotation



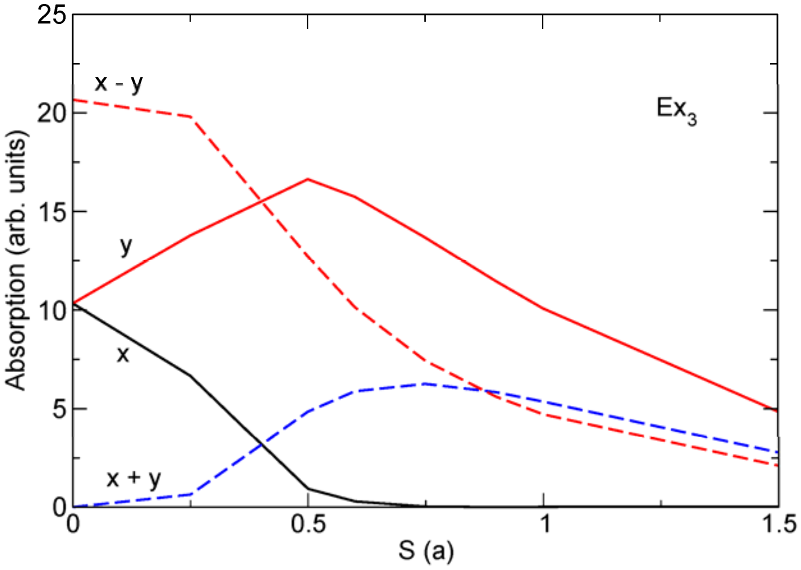
Excitons: shear

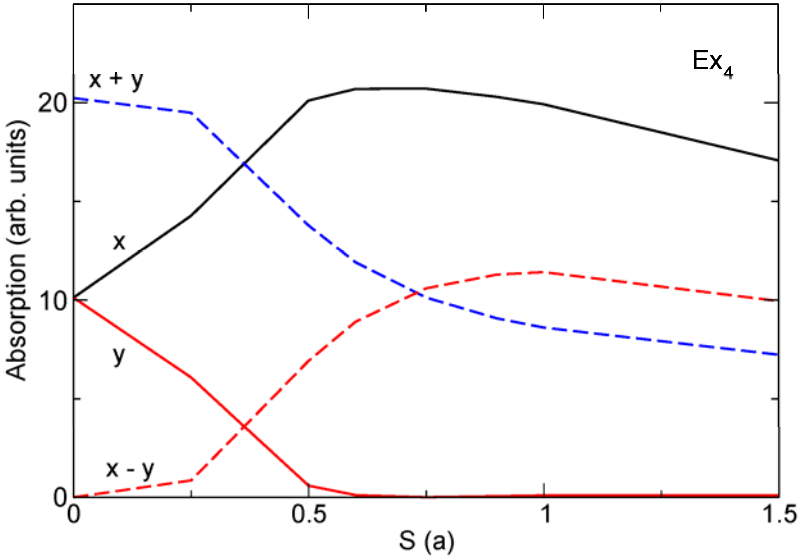


Pair ground state weakly polarized, strain polarizes along x

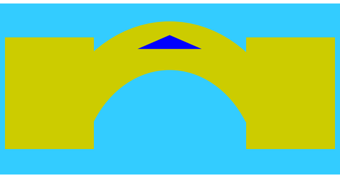
$Ex_{3(4)}$ polarized along x-y (x+y) in unbent structure

$Ex_{3(4)}$ polarized along y (x) by bending (reverse of symmetric bend)

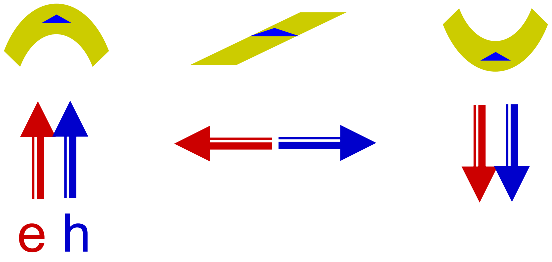




Final comments

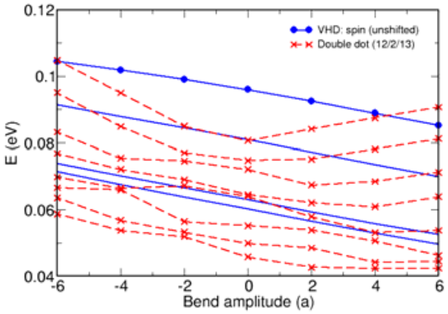


Strained QDs



- DC Stark field (imperfect) analog
  - Biaxial deformation:like E_z
    - E and h shift the same way
  - Shear bends: like E_x
    - E and h shift the opposite way
  - Internal relaxation and state distortion

- Excitons
  - Dominated by e/h shift
  - Fine structure: mixing, level crossing and polarization rotation



- Coupled dots
  - Strain-induced state crossing and transfer
  - Spin-dependent interdot coupling??