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NUSOD 2007, University of Delaware

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Electronic structure of QD arrays: Application to intermediate-band solar cells

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Motivation

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Design of the solar cell with

POWER conversion efficiency >50%

with cost of <$100 per 1m^2 of panel !!!

This will make PV solar cell economically competitive

Today: $0.25-$0.65/kWh form PV vs. $0.04/kWh from coal

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Current status of solar technology

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Performance of photovoltaic and photochemical solar cells			
Type of cell	Efficiency (%)* Cell	Module	Research and technology needs
Crystalline silicon	24	10-15	Higher production yields, lowering of cost and energy content
Multicrystalline silicon	18	9-12	Lower manufacturing cost and complexity
Amorphous silicon	13	7	Lower production costs, increase production volume and stability
$CuInSe_2$	19	12	Replace indium (too expensive and limited supply), replace CdS window layer, scale up production
Dye-sensitized nanostructure materials	10-11	7	Improve efficiency and high-temperature stability, scale up production
Bipolar AlGaAs/Si photochemical cells	19-20	-	Reduce material cost, scale up
Organic solar cells	2-3	-	Improve stability and efficiency

M. Grätzel, Nature 415, 338 (2001)

Ultimate Efficiency Limits

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- Thermodynamic limit of Carnot engine: $\eta = 1 - T0/Ts$ ~ 95% (100% absorption)
- Thermodynamic limit of solar heath engine: $\eta = (1 - Ta^4/Ts^4)(1 - T0/Ts)$ ~ 85%
- Shockley-Queisser efficiency limit for single band semiconductor based on detail balance eq.:

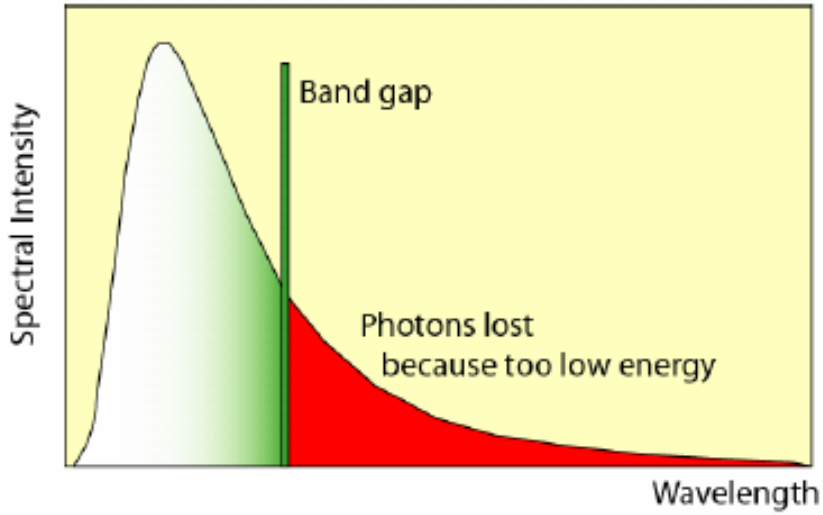
  ~31% (1 sun: Planck low) and ~41 (max conc.)

Origin of the solar cell losses:

a) Light with energy below Eg will not be absorbed

b) The photons with excess energy above Eg is lost in the form of heath

c) Single crystal GaAs solar cell ~ 25%(AM1.5)



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Tandem solar cells

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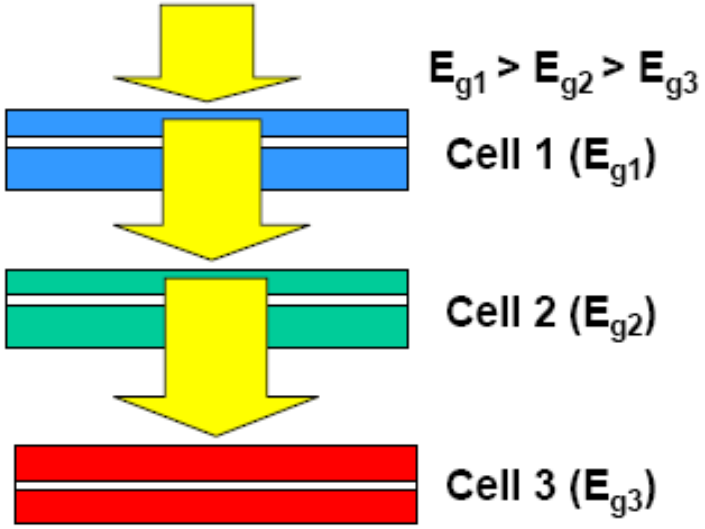
Multijunction or tandem cells:

- First approach to exceed single junction efficiency
- To achieve >50% efficiency need 3 or more tandems with different Eg's
- Significant technological problem to relax strain
- 75% efficiency achieved with 36 tandems



No of junctions	1 sun	Max conc.
1	30.8%	40.8%
2	42.9%	55.7%
3	49.3%	63.8%
∝	68.2%	86.8%

Intermediate band solar cells

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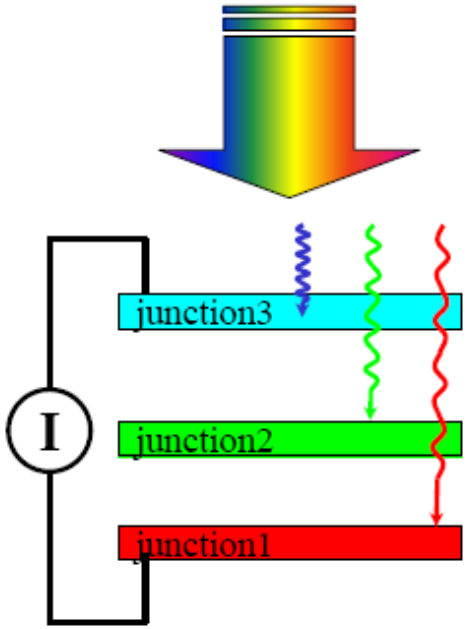
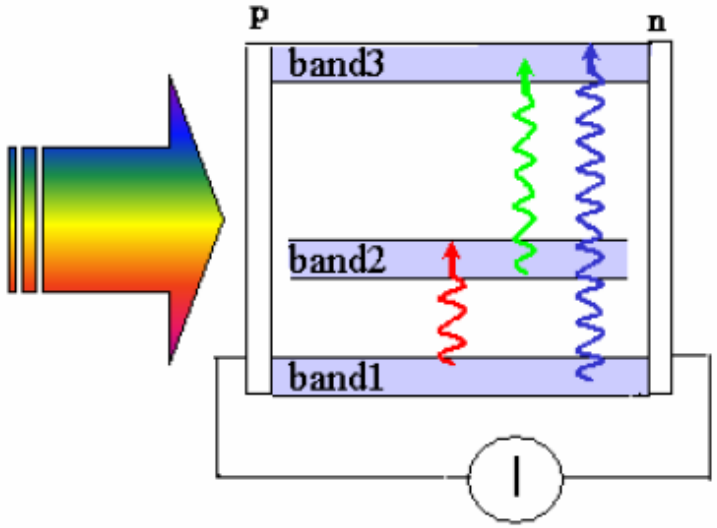


Luque & Marti



Multi-junction solar cell

- Each junction ⇒ single gap
- N- junctions ⇒ N- absorptions

Multi-band solar cell

- Single junction (no lattice mismatch)
- N- bands ⇒ N(N-1)/2 (gaps) absorptions
- Add 1 band ⇒ Add N- absorptions

Intermediate band solar cells

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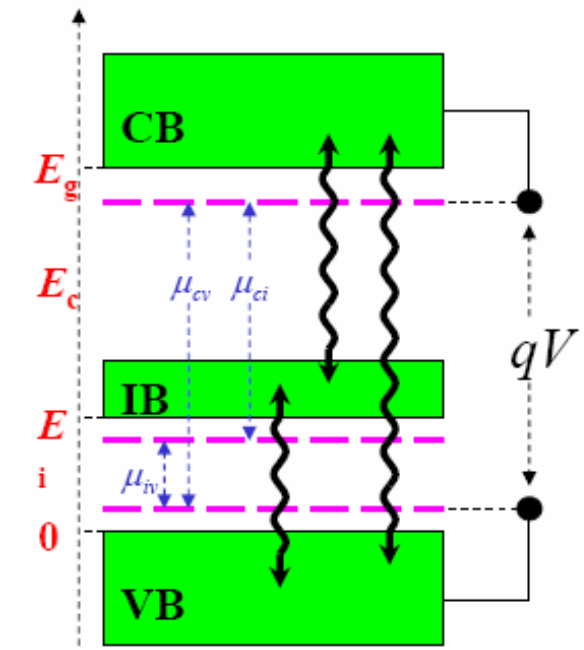

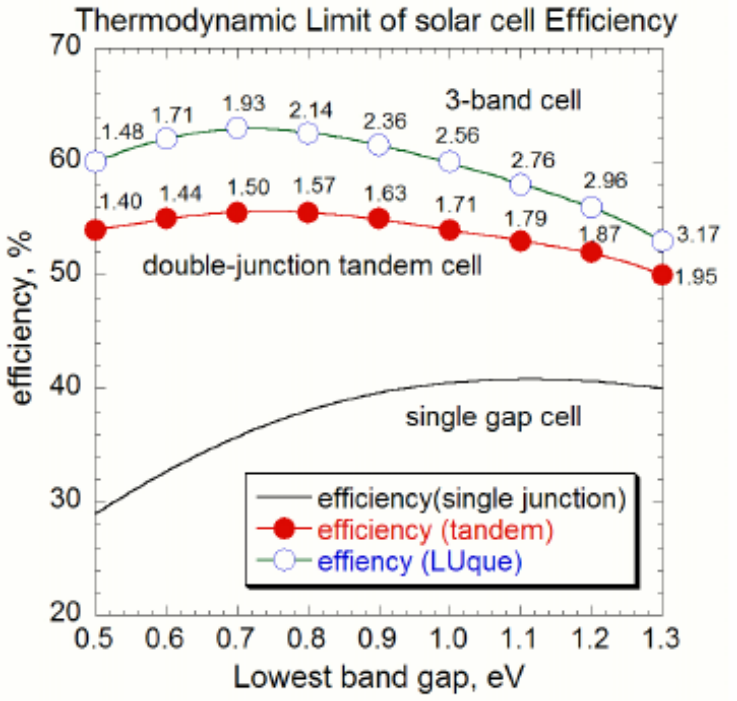

Intermediate band vs multi-junction solar cell

- Max. efficiency for 3 band cell ~66% (vs 55%)
- Max. efficiency for 4 band cell ~72% (vs 60%)
- Better performance than any other structure of similar complexity

A. Luque & A. Marti, Phys. Rev. Lett **78**, 5014 (1997)

Requirements & possible realization

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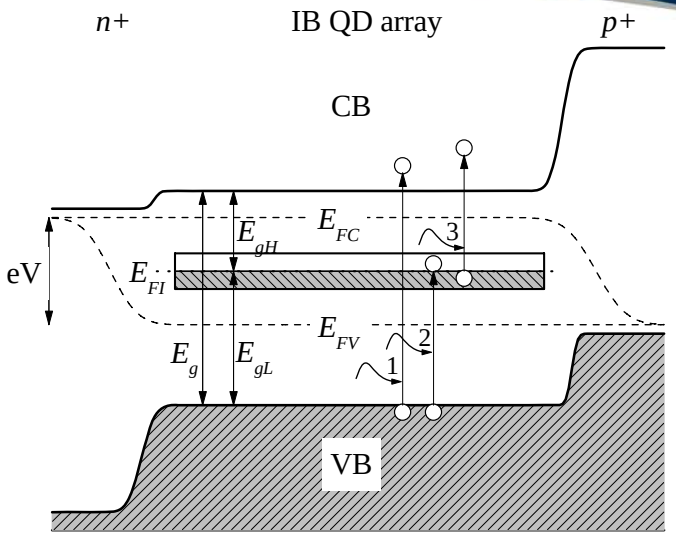
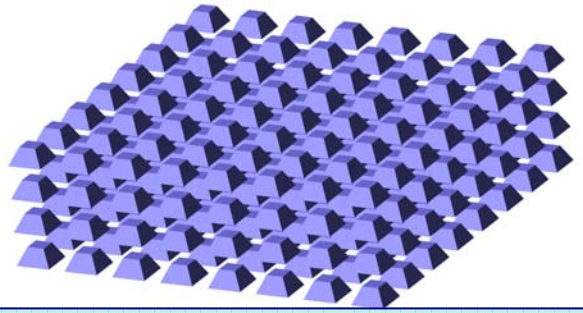
There are two ***contradictory requirements*** for IB:

(a) the IB should exhibit finite energy width so that it can be partially occupied and facilitate simultaneous excitaion from IB to CB and VB to IB

(b) the IB should be as narrow as possible to reduce carrier transport through the mini-band.





Arrays of QD ideal candidate for realisation of IB because of zero density of states between VB & IB & CB that increase radiative lifetime relative to relaxation time within bands

Current technology

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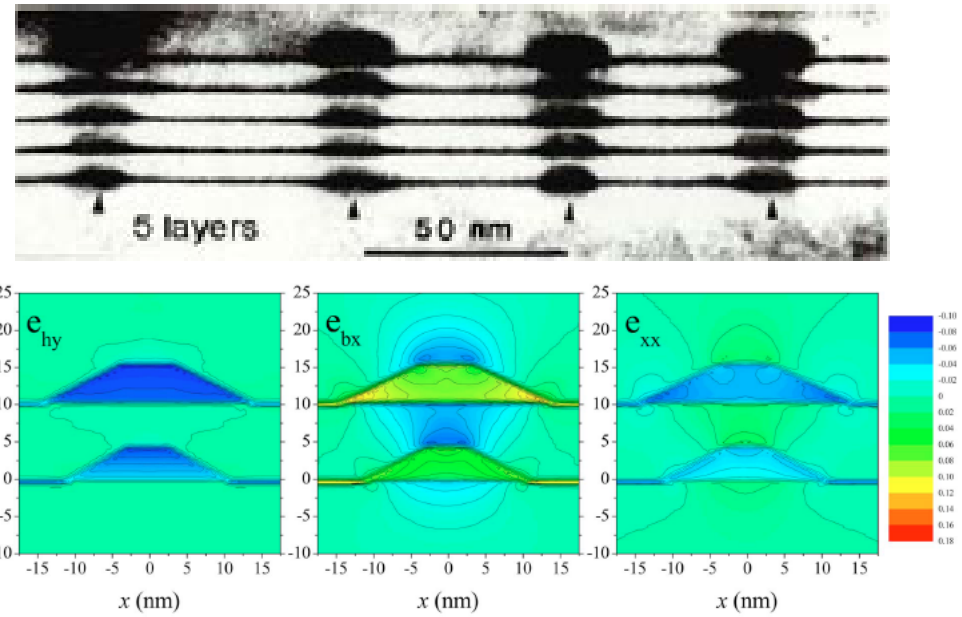
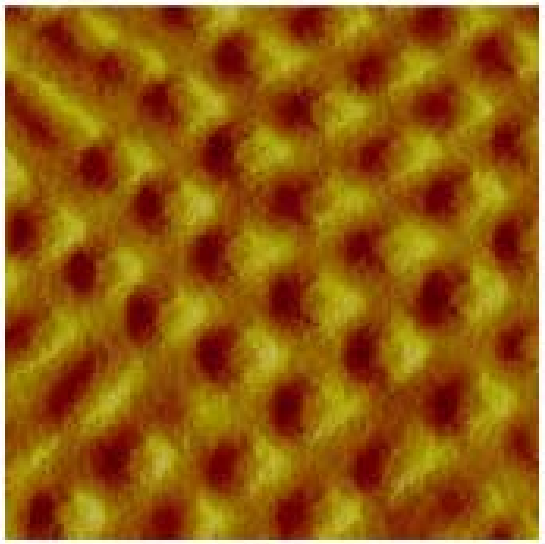
InAs QDs on GaAs(100)



InAs quantum dots



(500nm×500nm)

- Vertical ordering is provided by strain driven alignment
- Horizontal regularity of QD's is observed on high Miller indices growth (311)

Q. Xie, et al., Phys. Rev. Lett. **75**, 2542 (1995)
S. Tomic et al., J. Appl. Phys. **99**, 093522 (2006)
Y. Okada, private communication

Methodology

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Fourier transform of the strain tensor in analytical form for the cubic crystal symmetry and actual truncated-pyramid QD shape

Analytical expressions for the Fourier transform of the piezoelectric field

Plane Wave expansion (Fourier) method for the QD carrier spectra and wave functions from 8 band kp theory

Use of Hamiltonian C2 symmetry property: ~2×rank(4) diagonalizations instead ~rank(8) diagonalization (8 times sped up)

$H = H_k + H_e + V_{pz}$ **is dense matrix => NOT sparse**

Direct diagonalization by ScaLAPACK and MPI packages on HPCx

Methodology

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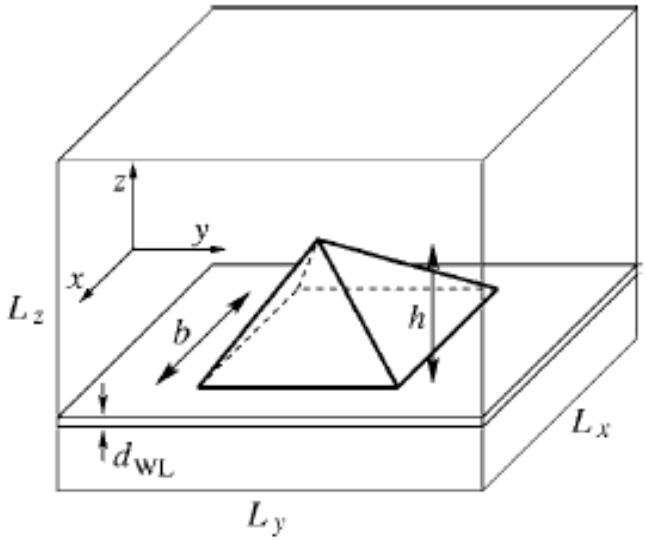
To calculate the electronic structure of an QD array the only modification to the basis set is

$$k_v \rightarrow k_v + K_v^{SL} \qquad v \in (x,y,z)$$

$$K_v^{SL} = a(\pi / L_v^{SL}) \qquad a \in [0,1]$$

This allows the sampling along the K points of a QD-SL to be done at several points at the cost of the single QD calculation at each K point.



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Scalability

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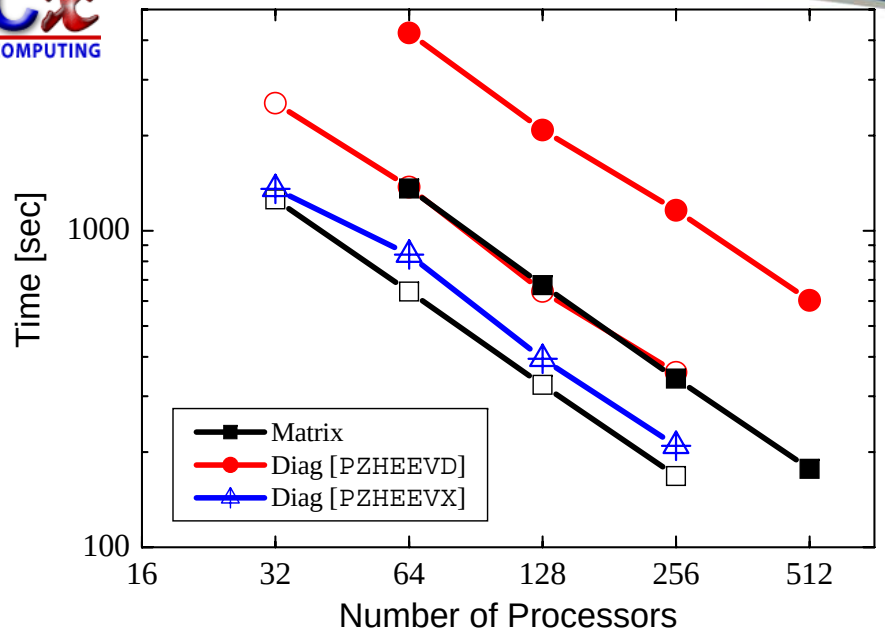
HPCx CAPABILITY COMPUTING



www.top500.org
Rank 65

- 160 IBM Power5 p5-575SMP nodes
- 2560 processors
- 15.3 TeraFlop/s peak
- 12.9 TeraFlops/s sustained
- 5.12 TByte of memory

N = (6,6,9) = 25688 (from 32 to 256)

Scalability = 1.93^3 (ideal 2^3)

N = (7,7,10) =37800 (from 64 to 512)
Scalability = 1.93^3 (ideal 2^3)

Located at STFC Daresbury Laboratory

S. Tomic, A. Sunderlans, I. Bush, *J. Mat. Chem* **16,** 1963 (2006)

Electronic structure of QD array

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Truncated pyramid QD shape

b = 120 A

h = 60 A

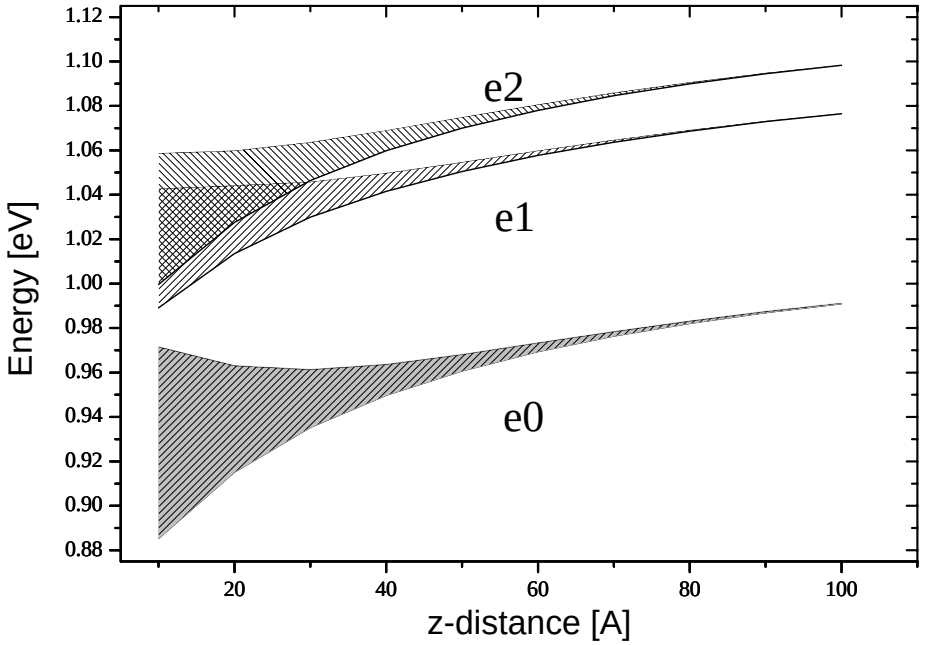
t = 0.5

$L_x = L_y = 200$ A

vertical distance:

$d_z = L_z - h \in [10,100]$ A

material: InAs/GaAs



band width = $E(K_z = \pi/L_z) - E(K_z = 0)$

e0 width: 86 meV @ $d_z = 10$ A

< 1 meV @ $d_z = 100$ A

Electronic structure of IB

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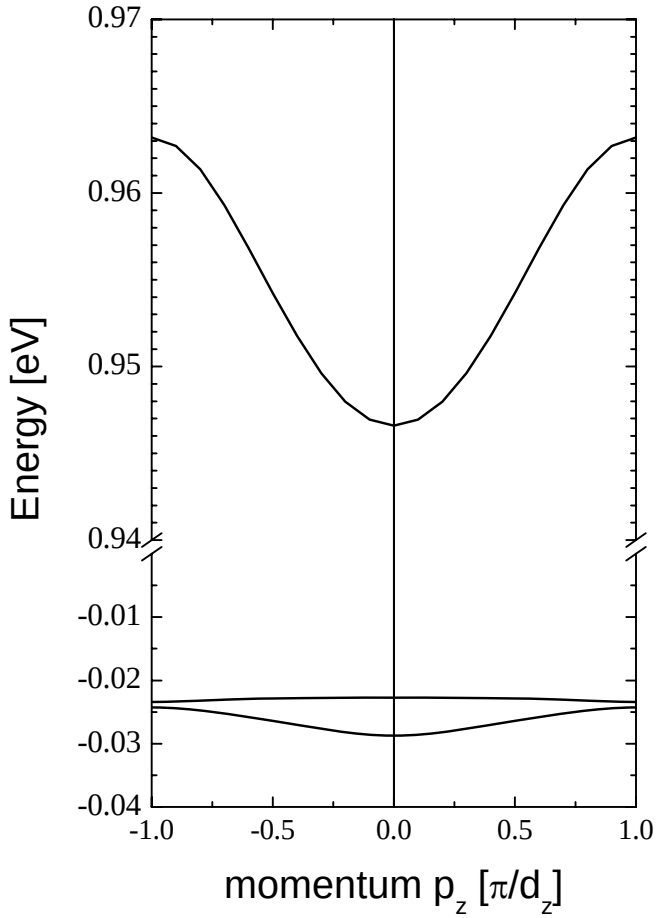
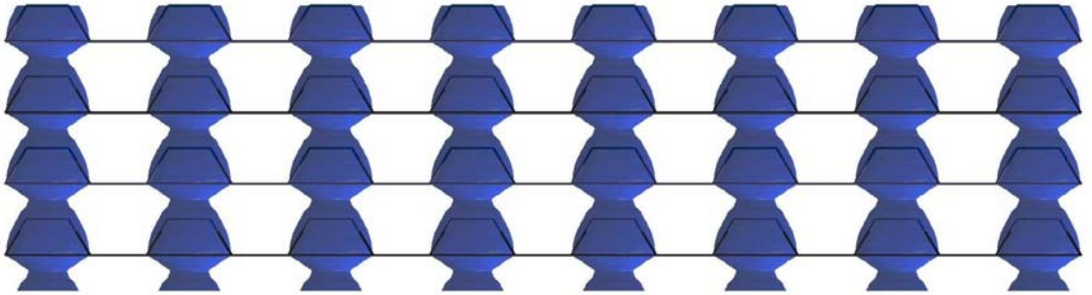


Electronic structure of 40 A vertically spaced QD array:

- e0 width 14 meV @ d_z = 40 A
- h0 & h1 almost dispersionless



Electronic structure of IB

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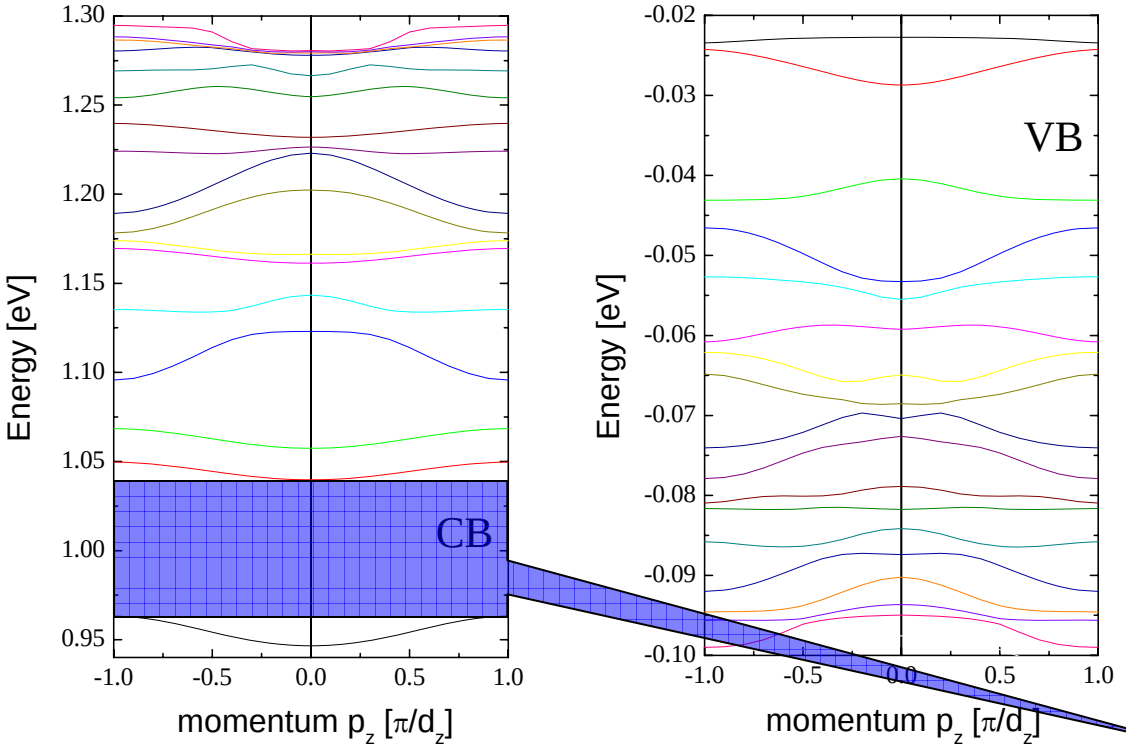
d_z = 40 A



In VB probably all minibands will merge because of small energy separation < ~kT (25 meV@ 300K)

In CB probably just one miniband : e0

In CB probably just 1 gap

First Brillouin zone

Radiative lifetime

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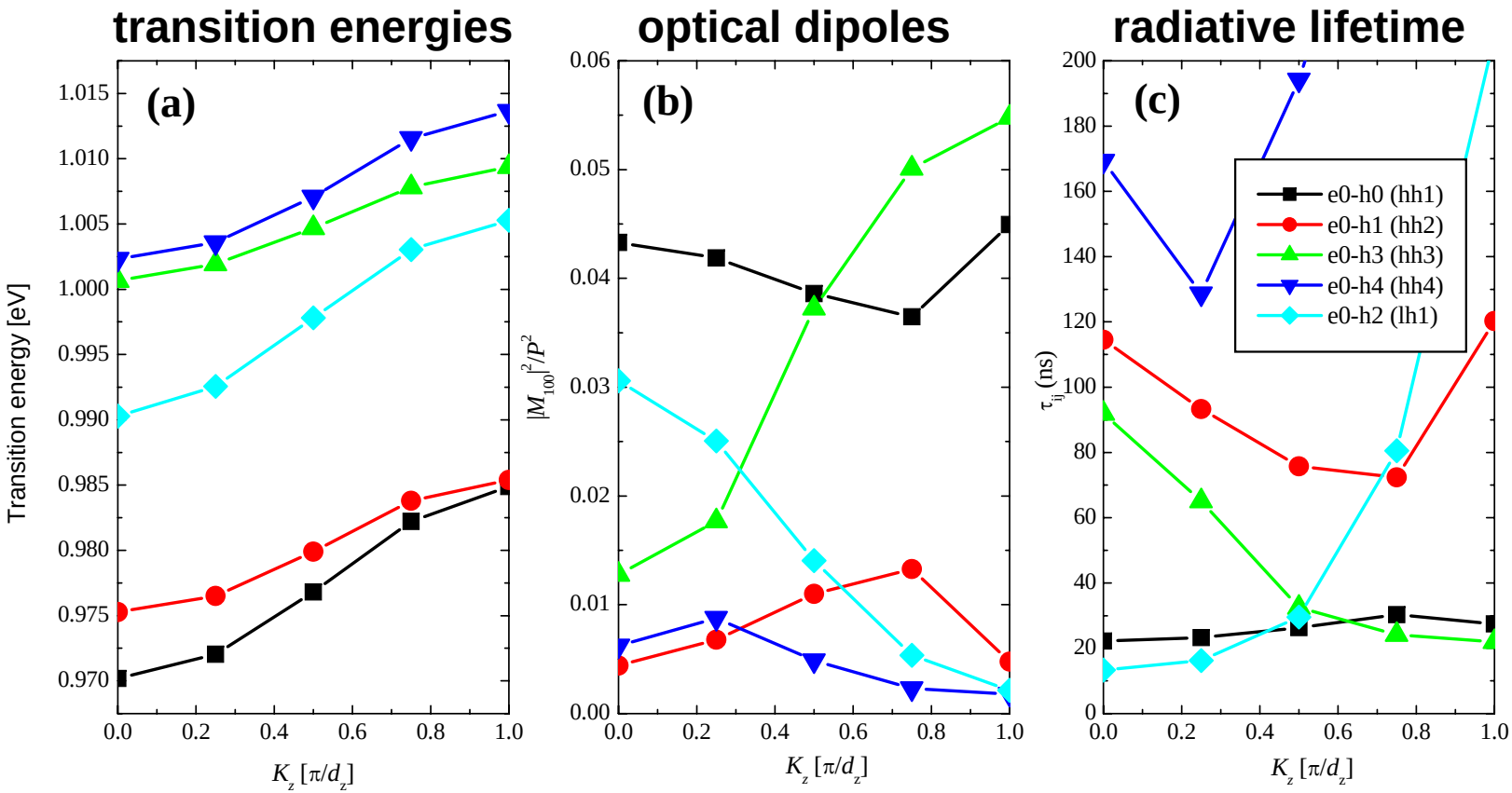
Fermi Golden rule:

$$\frac{1}{\tau_{if}^{\mathrm{rad}}} = \frac{(E_i - E_f)\bar{n}}{3\pi\varepsilon_0\hbar^2 c^3}(M_x^2 + M_y^2 + M_z^2)$$

~25 ns in QD-SL vs. ~2 ns in QD



Concussions

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- 8 band kp theory can successfully predict electronic and optical structure of QD arrays
- PW methodology with periodic boundary condition is particularly suited for QD array analysis
- Results on radiative lifetime in QD arrays show significant increase due to existence of the IB
- Model presented provides much realistic parameters for drift-diffusion equations and efficiency prediction

Future work should:

1) Identify relative role of the intermediate band:
   width, energy position, doping, etc.
2) Identify best material combination
3) Identify relative role of the structure/size imperfection