Predicting the electronic and optical properties of nano-materials from first-principles

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Outline

- **Motivation**
- GW-BSE method applied to nanosystems.
- Challenges and bottlenecks
- Getting Physics from computation.

Motivation

- Nanomaterials have unique and tunable electronic properties. Promising for future electronic/optical devices.
- Want to predict the electronic, quasiparticle and optical properties of nanomaterials without adjustable parameters

Nano Tunable Properties

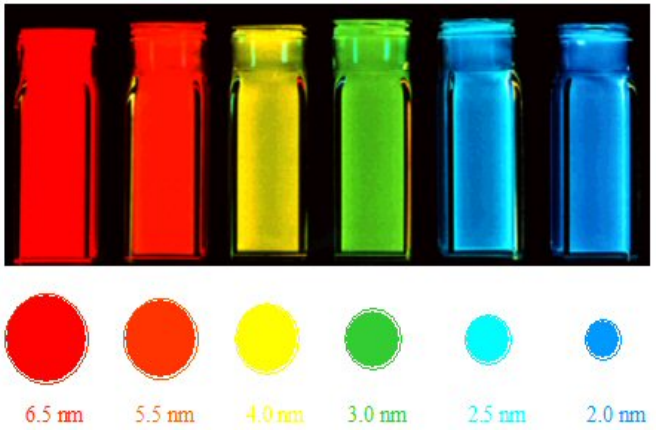
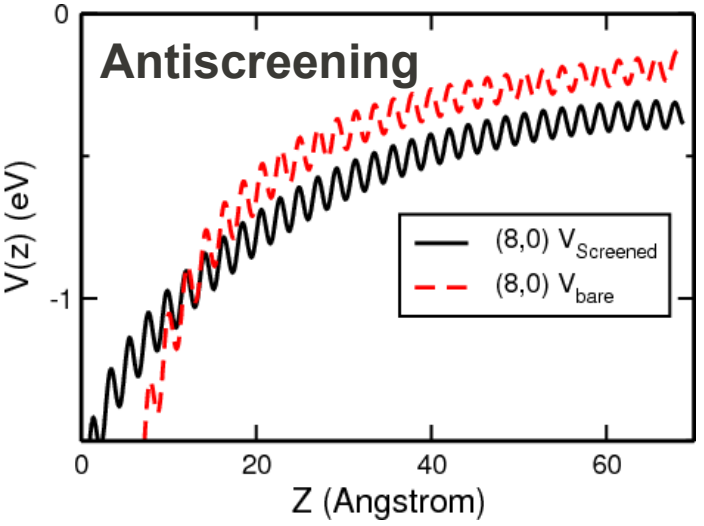


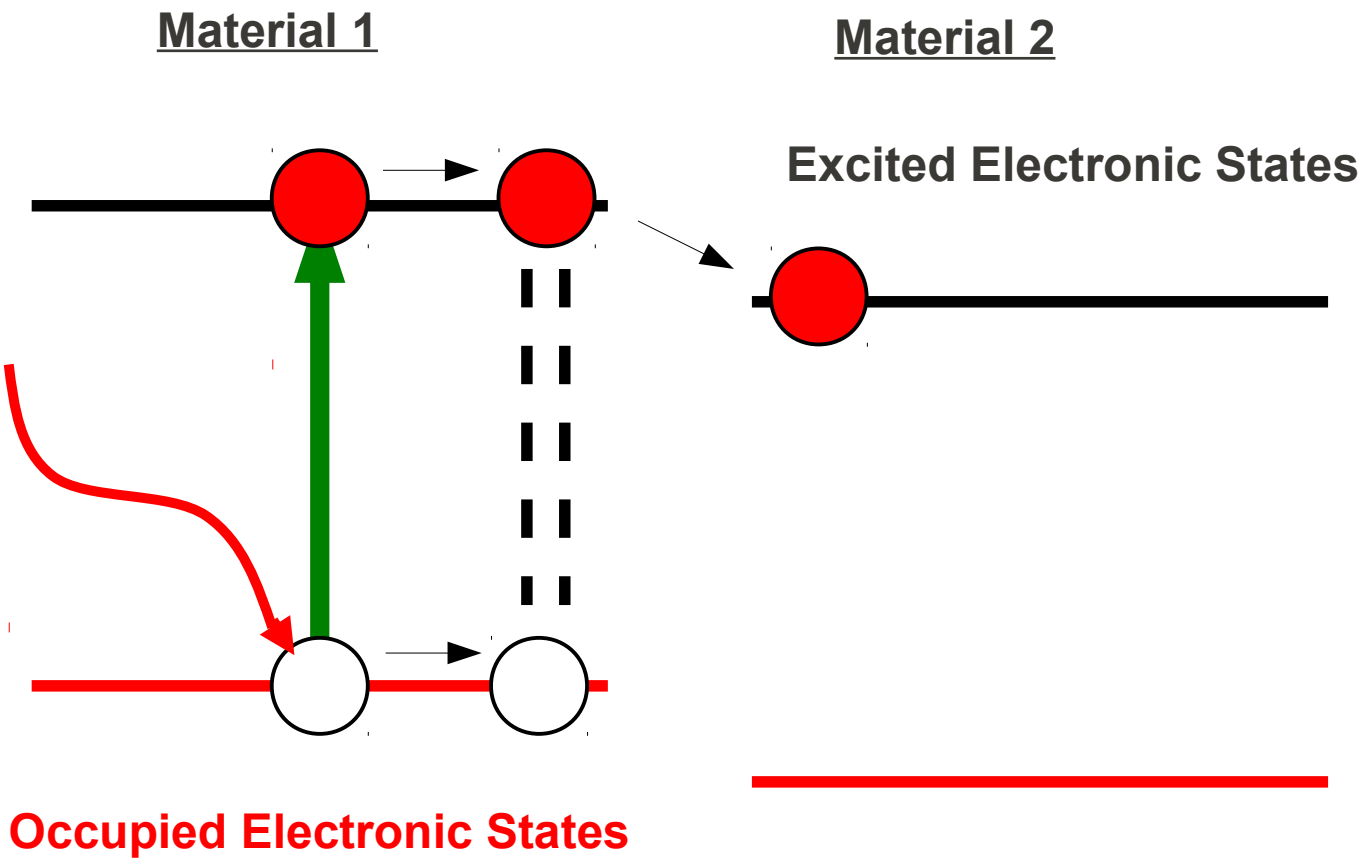
Photo from Vanderbilt Mat. Phys. Group

New Physics at Nanoscale:

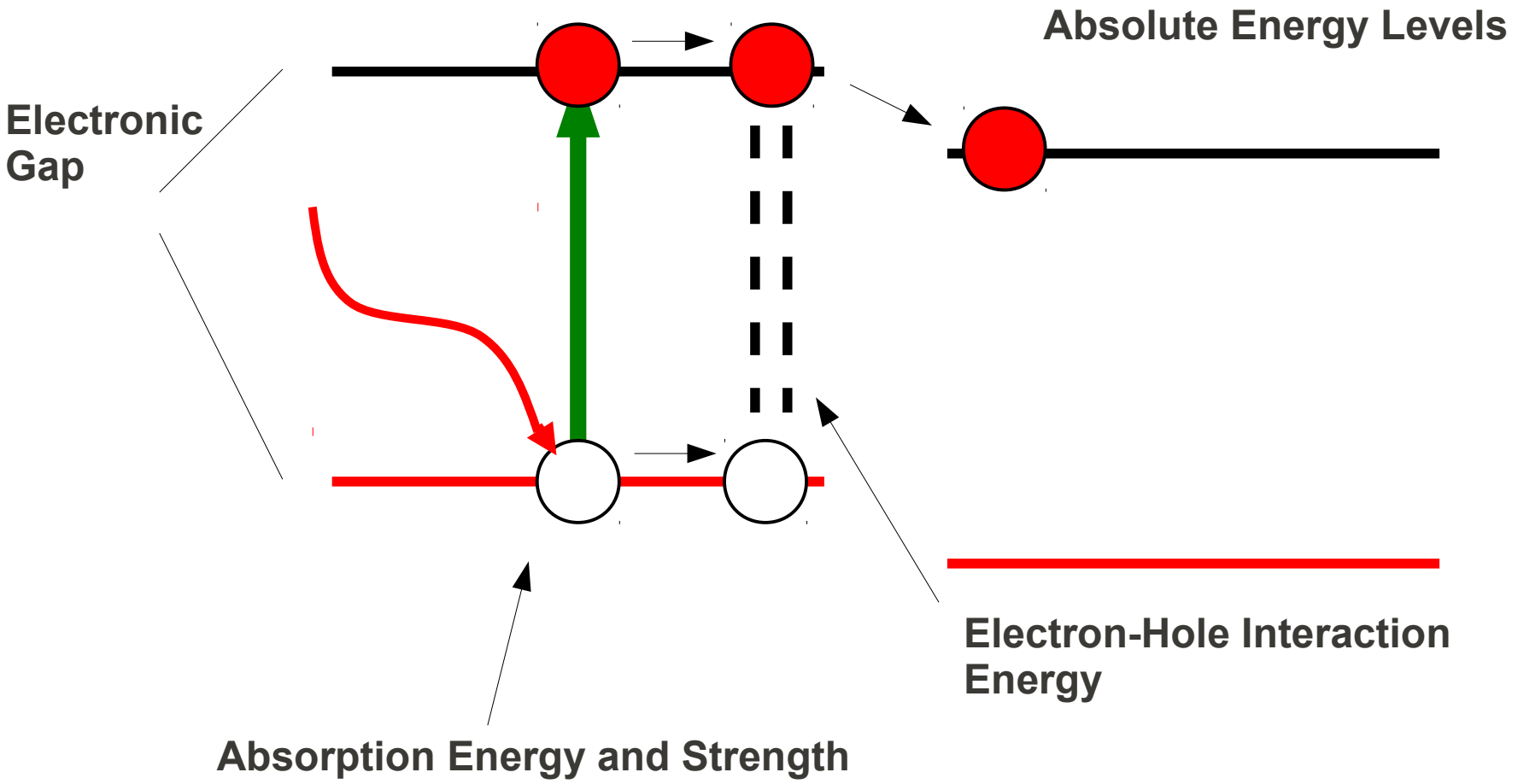


Nano Optics

Photovoltaics – Quantities to Predict



Photovoltaics – Quantities to Predict



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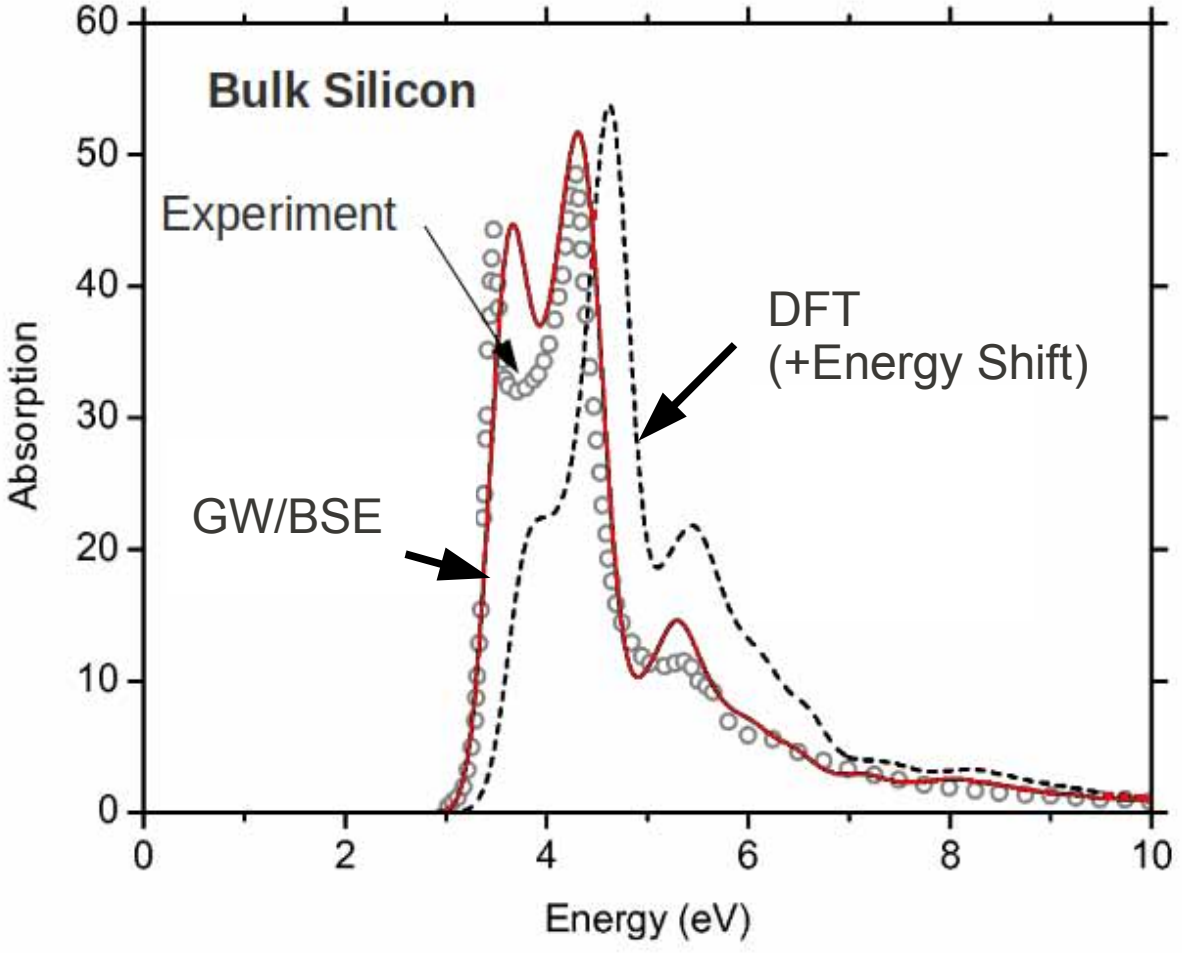
GW-BSE Method Overview

Density Functional Theory:
(Ground State Properties)

GW:
(Quasiparticle Properties)

Bethe-Salpeter Equation
Optical Properties



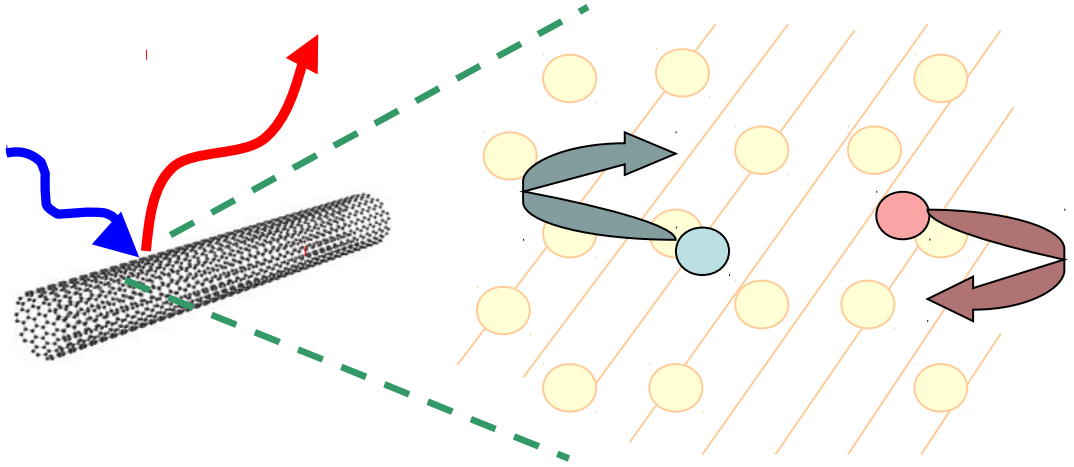
Expt. G.E. Jellison, M.F. Chisholm, S.M. Gorbatkin, Appl. Phys. Lett. 62, 3348 (1993).

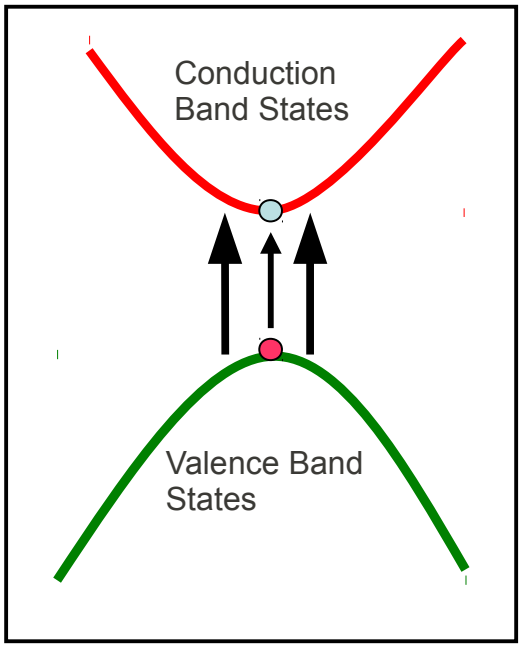
Excitons and the Bethe-Salpeter Equation

$$|N,S\rangle = \sum_{v}^{hole} \sum_{c}^{elec} A^{S}_{vc} a^{+}_{v} b^{+}_{c} |N,0\rangle + \ldots$$

$$\left(E_{ck\,QP} - E_{vk\,QP}\right) A^{S}{}_{vck} + \sum_{k'v'c'} \langle vck | K^{eh} | v'c'k' \rangle A^{S}{}_{v'c'k'} = \Omega^{S} A^{S}{}_{vck}$$

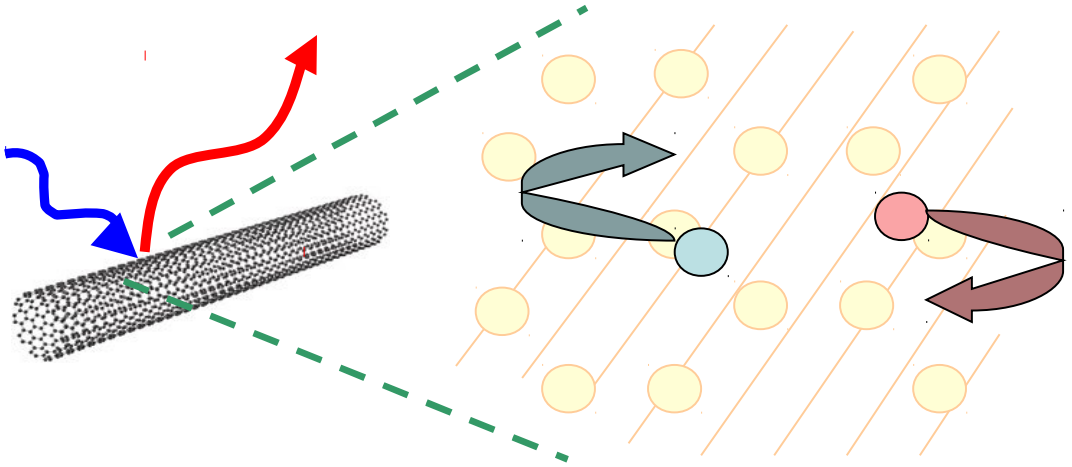
Hydrogenic like – electron-hole interaction Kernel

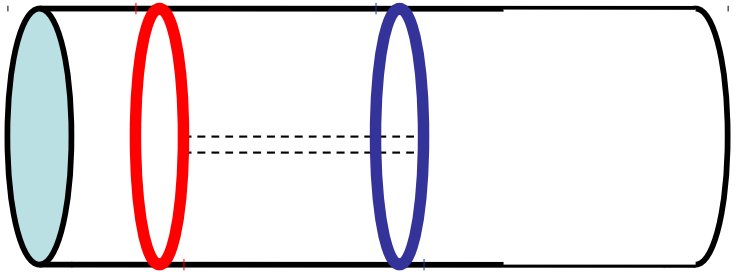




$$\varepsilon_2(\omega) = \frac{16\pi^2 e^2}{\omega^2} \sum_{S} |\langle N,0|e\cdot v|N,S\rangle|^2 \delta(\Omega_S - \hbar\omega)$$

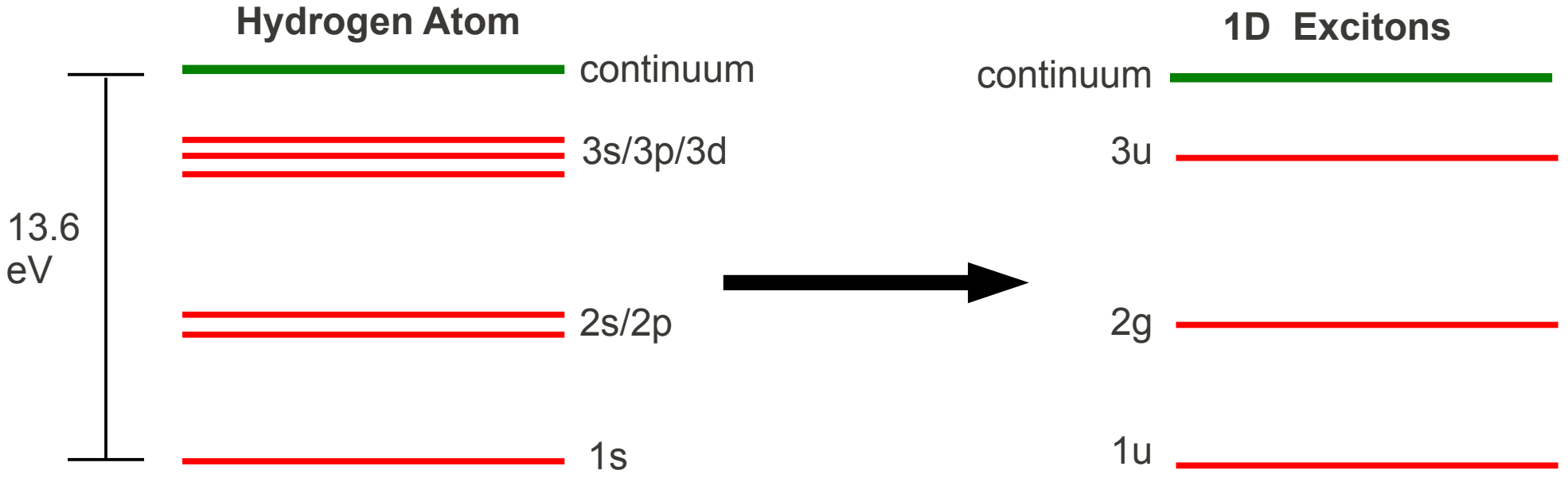
Excitons





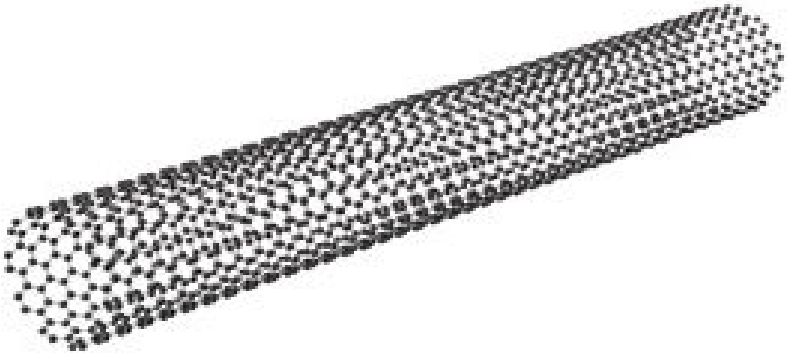
Interact via a Hydrogenic like Coulomb Interaction – but screened:

$$v(\mathbf{r},\mathbf{r}') = \frac{\varepsilon^{-1}(\mathbf{r},\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$



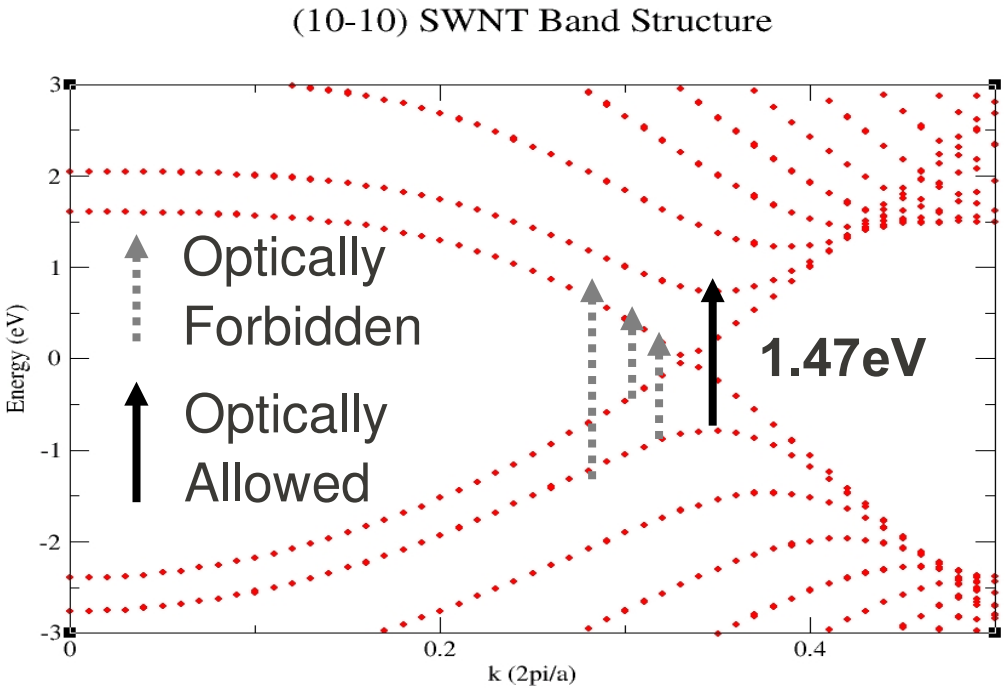
Metallic Nanotubes

- Single Walled Carbon Nanotubes are 1D materials made by rolling up a Graphene sheet.



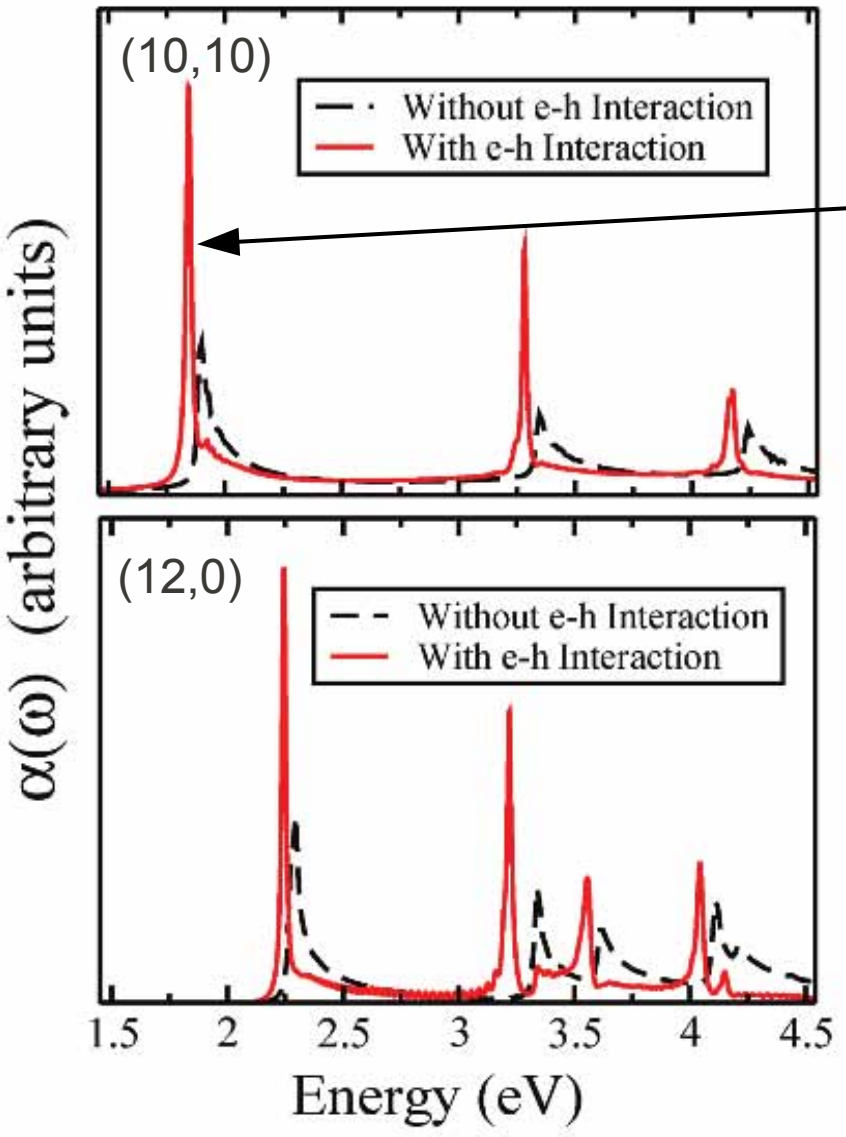
- Electronic properties can be controlled by physical parameters. (Rolling angle & diameter).

- Axial rotation symmetry commutes with the k-point. Leads to selection rules.



Absorption Spectra of Metallic Tubes

Optical absorption vs. Photon Energy



- Peak from a single eigenvalue.
- Exciton binding energy - 0.06 eV.
- The onset is calculated to be 1.84 eV.

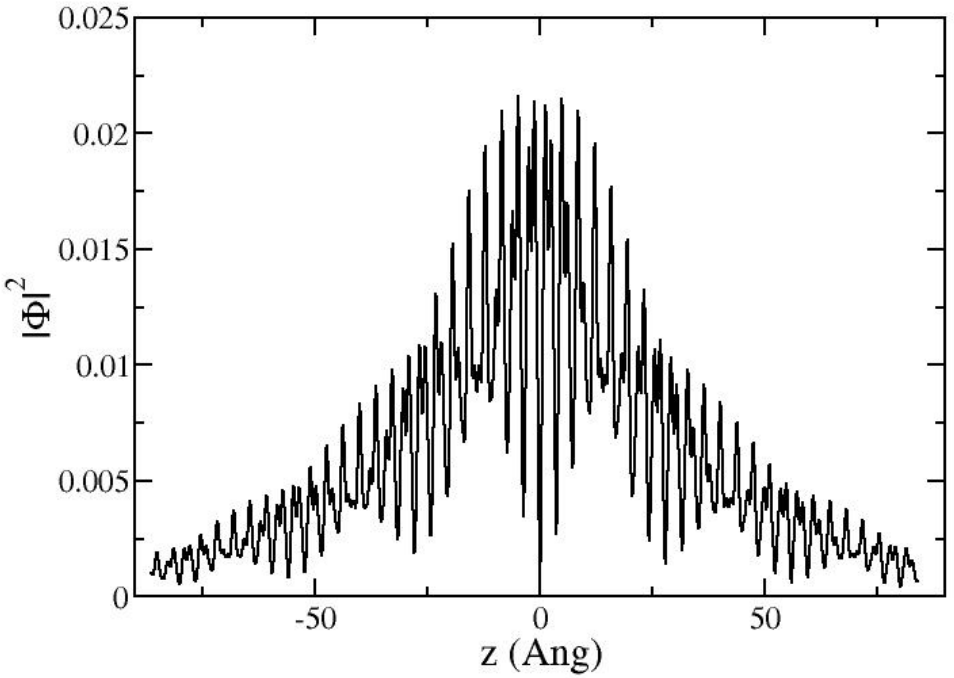
Experimental value*: 1.89 eV

(Experiment) Fantini, C.; Jorio, A.; Souza, M.; Strano, M. S.; Dresselhaus, M. S.; Pimenta, M. A. Phys. Rev. Lett. 93, 147406. (2004)

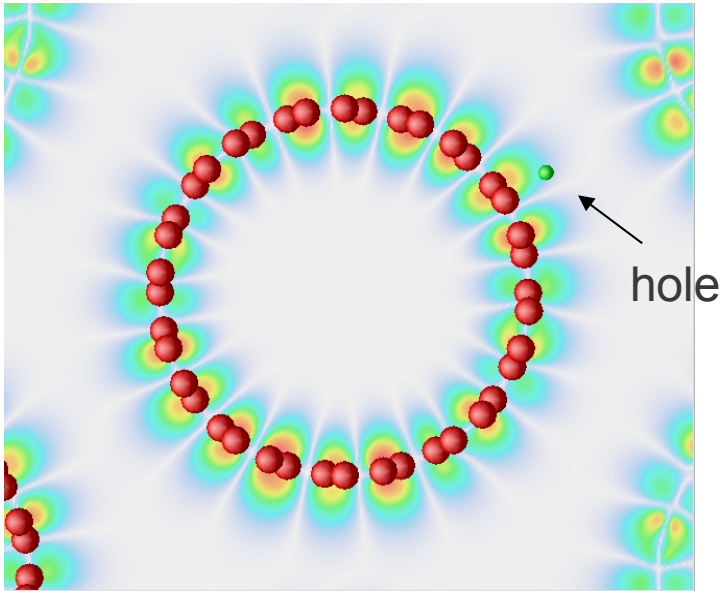
(Theory) J. Deslippe, D. Prendergast, CD Spataru, S.G. Louie, Nano Lett. 7 (6) 1626-1630, (2007)

Excitons in Metallic Tubes

Electron Wavefunction vs. Distance From Hole



Radial Distribution of Electron



Exciton is localized over 160 Å along tube axis but spans the whole tube in the radial direction.

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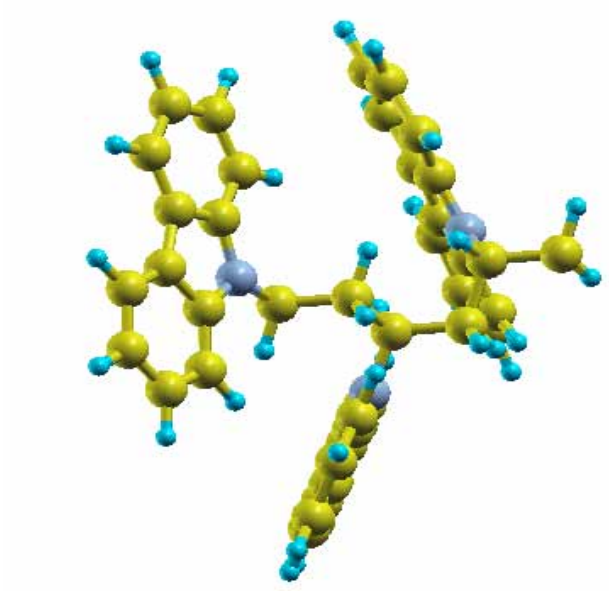
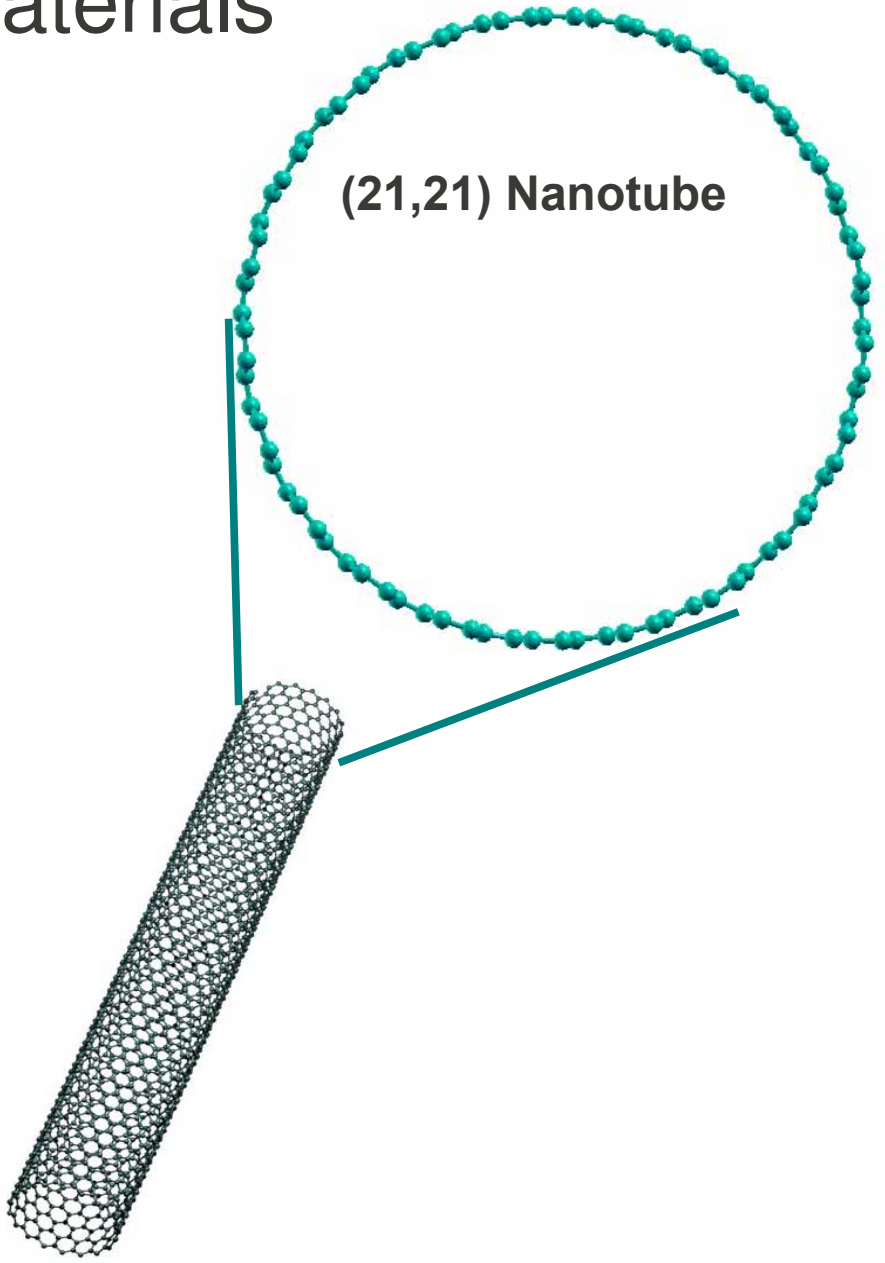
Challenges/Opportunities in Applying the Method to Nano-Materials

- Size (Atoms, Basis, Bands)
- Interfaces/Vacuum/Absolute Energies
- Very scalable on HPCs

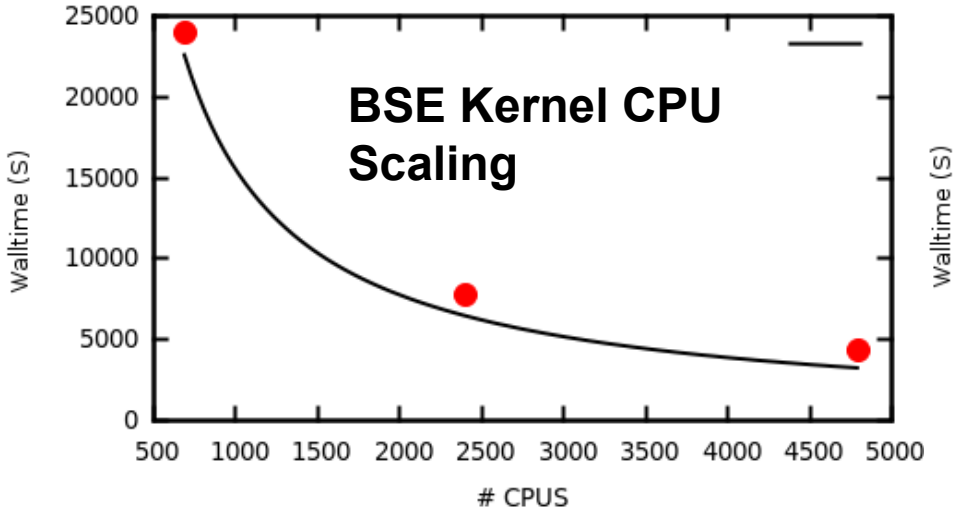


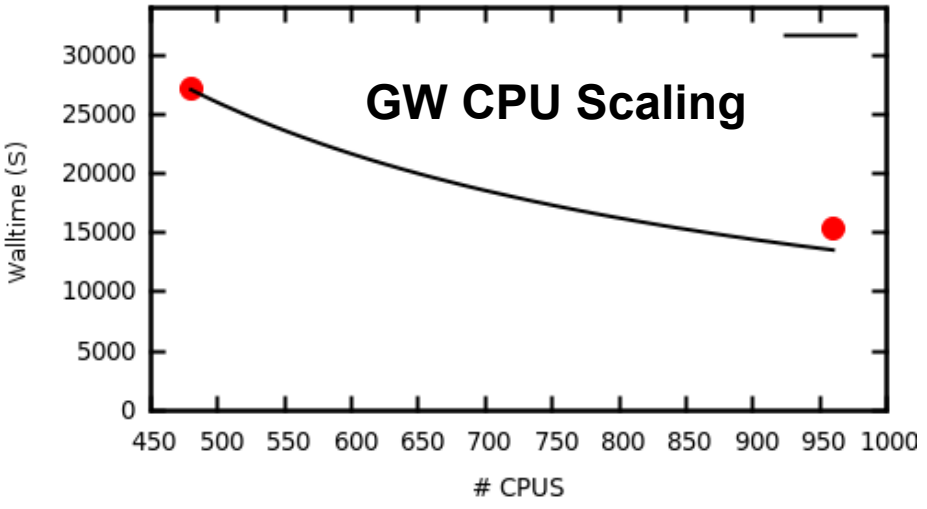


Overcoming Obstacles

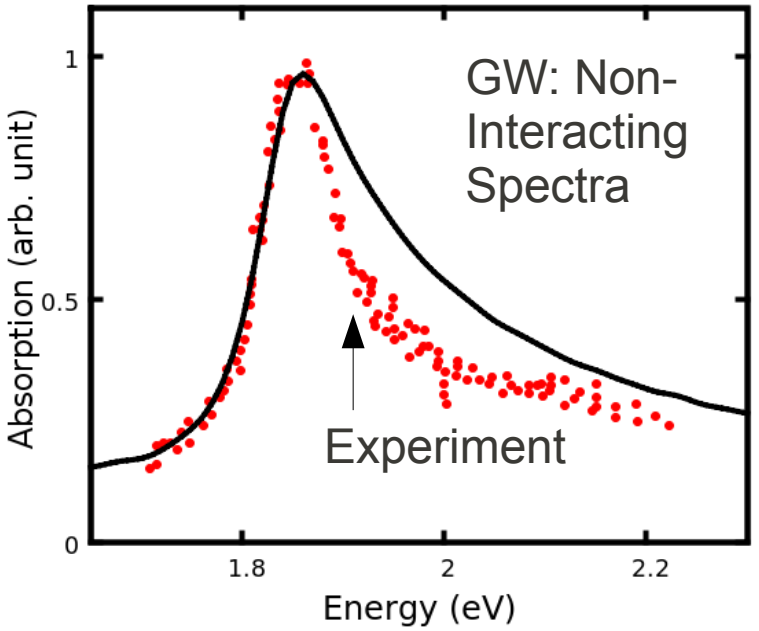
- Distribution of ~500GB of memory.
- Distribute work for nearly linear scaling up to 10,000 CPUs
- Optimize bottlenecks.

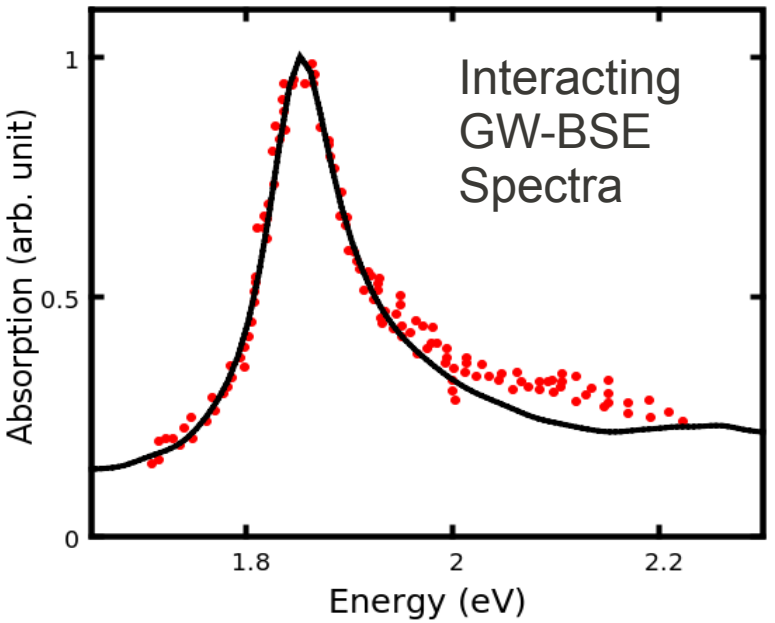
BerkeleyGW (COMING SOON)





The Pay-Off





Strong evidence of bound exciton origin of optical absorption spectra.

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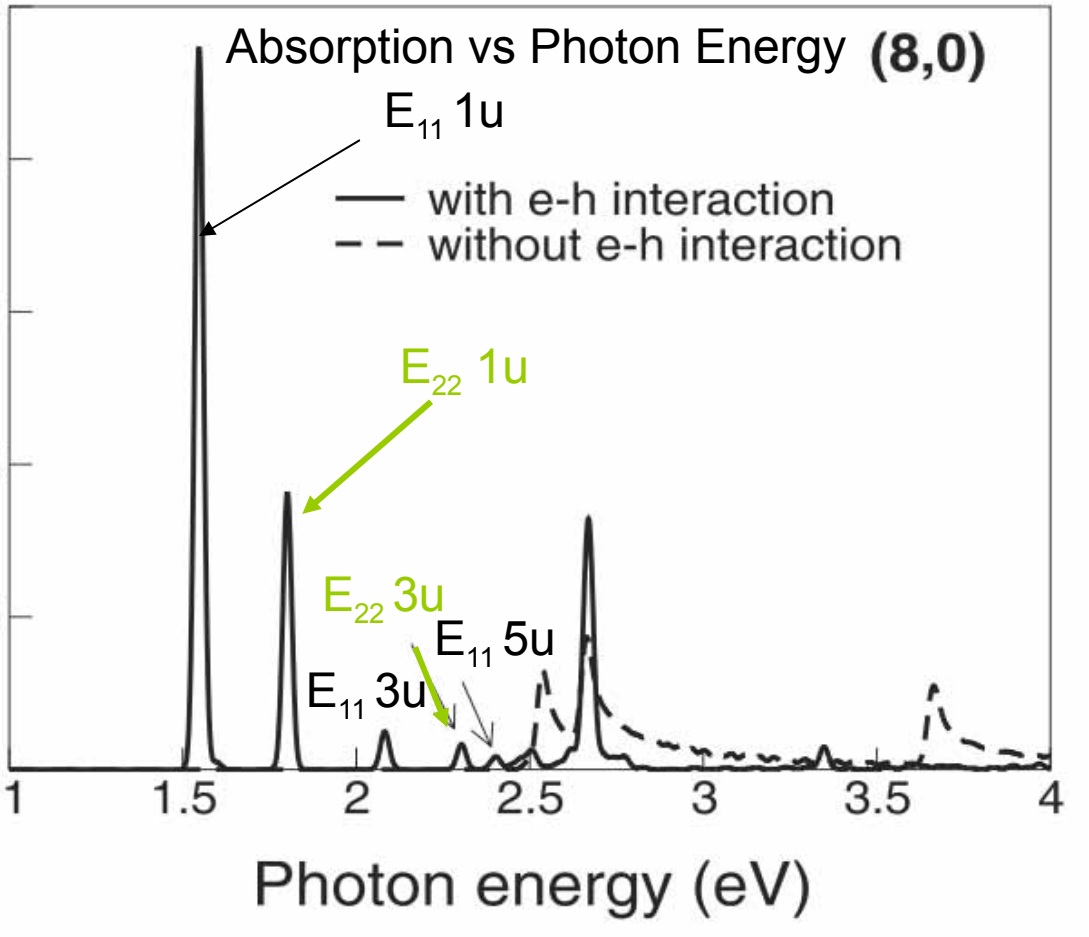
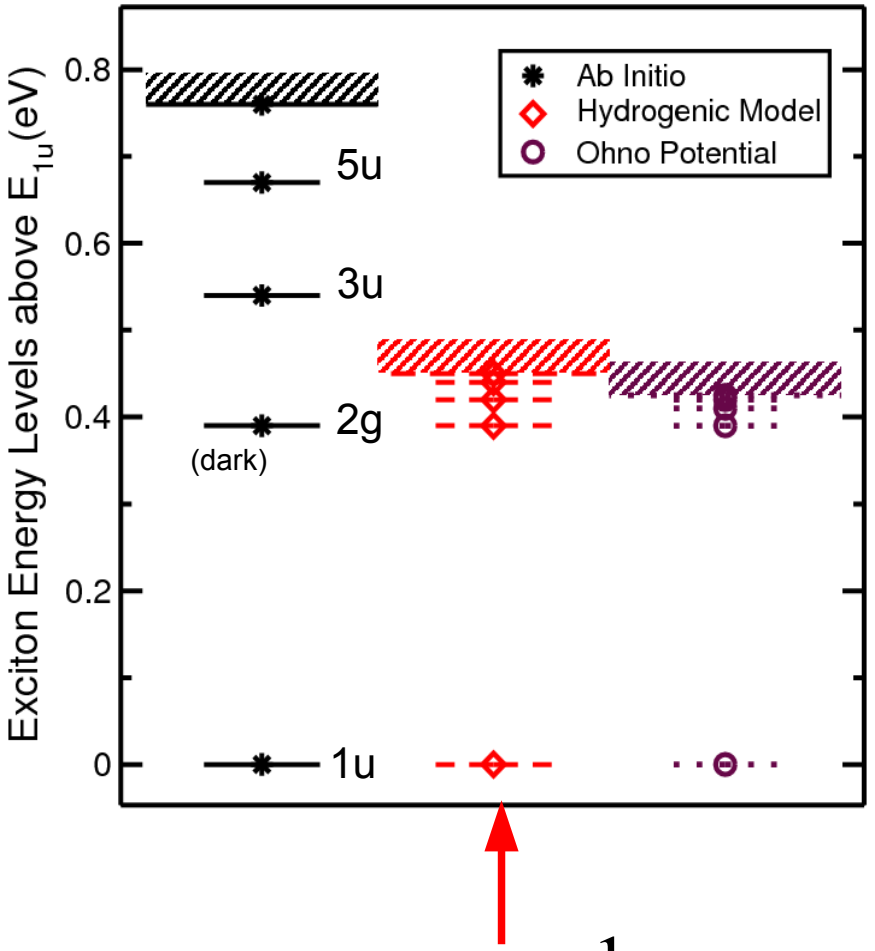
Semiconducting Tubes





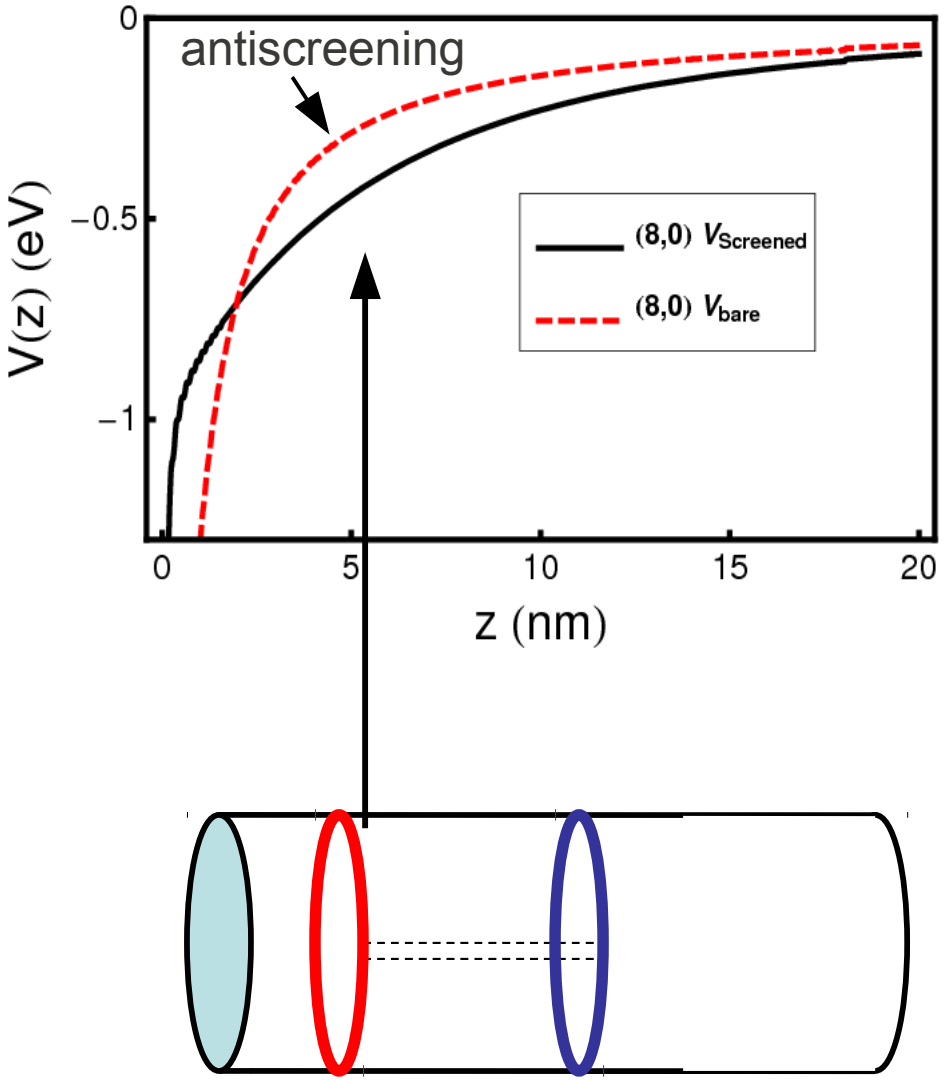
$$V_H(z) = \frac{1}{\varepsilon(|z| + z_0)}$$

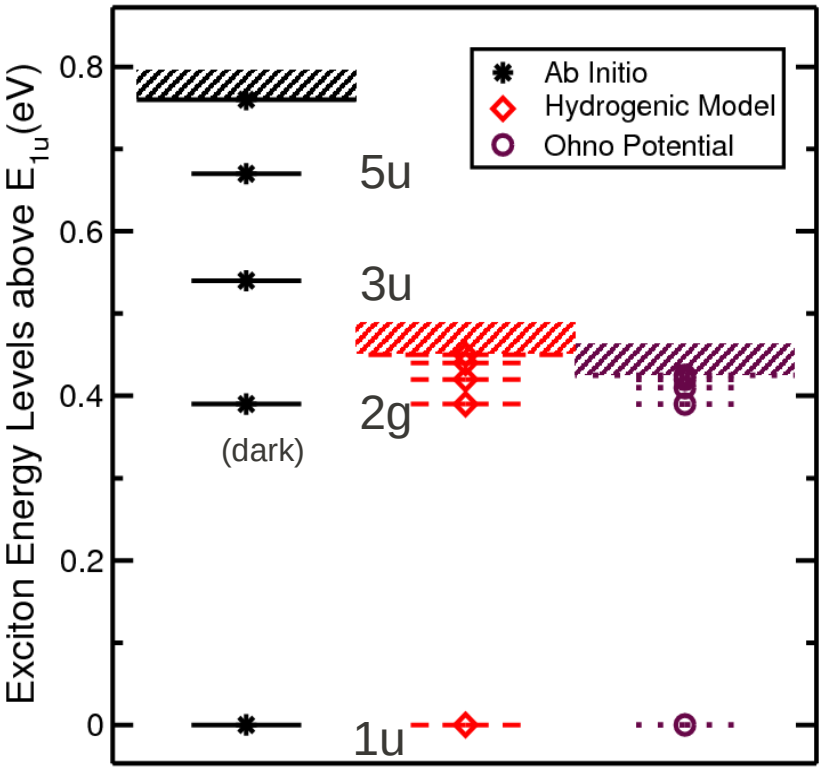
***C.D. Spataru, S. Ismail-Beigi, L.X. Benedict, S.G. Louie. PRL 077402 (2004)**

***J. Deslippe, C.D. Spataru, D. Prendergast, S.G Louie. Nano Letters. 7 1626 (2007)**

Simple models have very different excitation specra than ab intio.

Antiscreening





- Screened electron-hole interaction enhanced for separations greater than tube diameter.
- Increases binding energies for 2g, 3u, … relative to 1u
- Confirmed by experiment – **J. Lefebvre P. Finnie. Nano Letters 8 1890 (2008).**

***J. Deslippe, M. Dipoppa, D. Prendergast, M. Moutinho, R. Capaz, S.G. Louie Nano Letters. (2009) - nl802957t**

Acknowledgments

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Li Yang
Catalin Spataru
Rodrigo Capaz
Feng Wang
Cheol-Hwan Park

Thank you Krell!

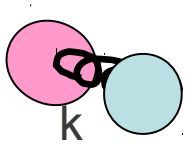
DOE CSGF

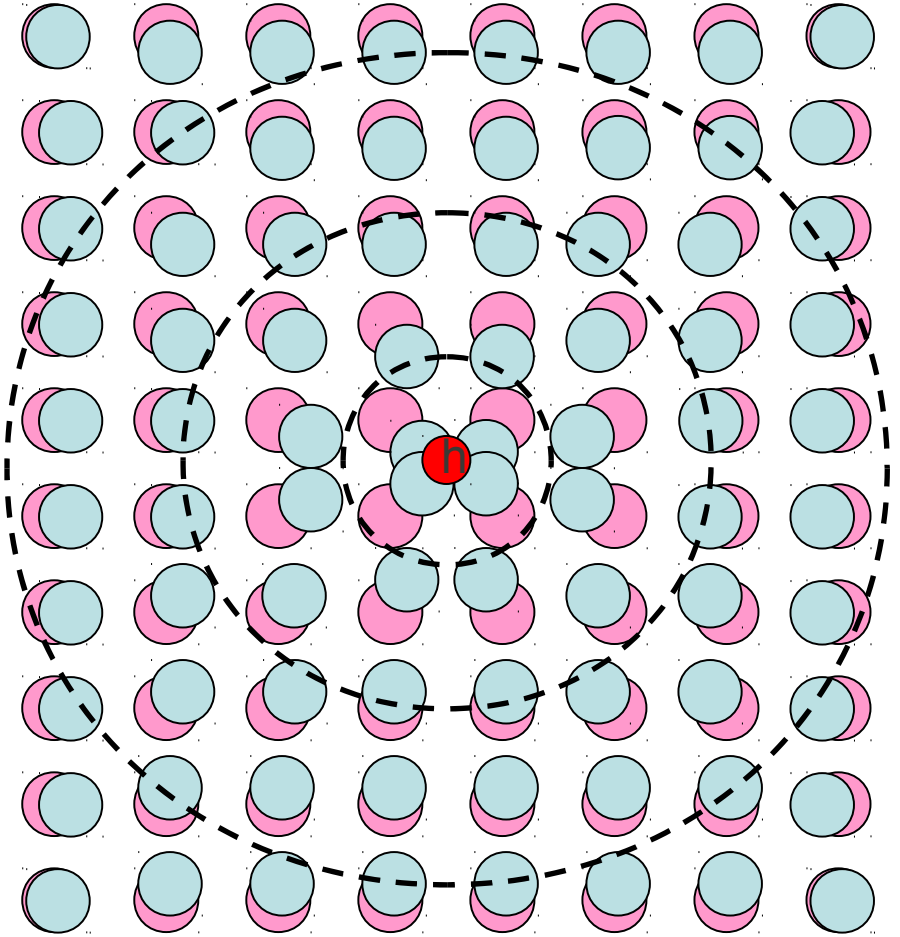
Supplemental Slides

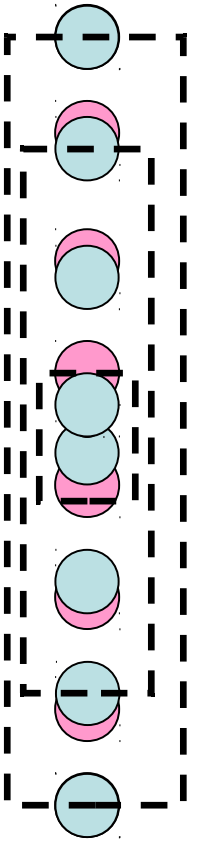
Why Anti-Screening? Part 1

3D – Total induced charge in sphere constant with sphere size.

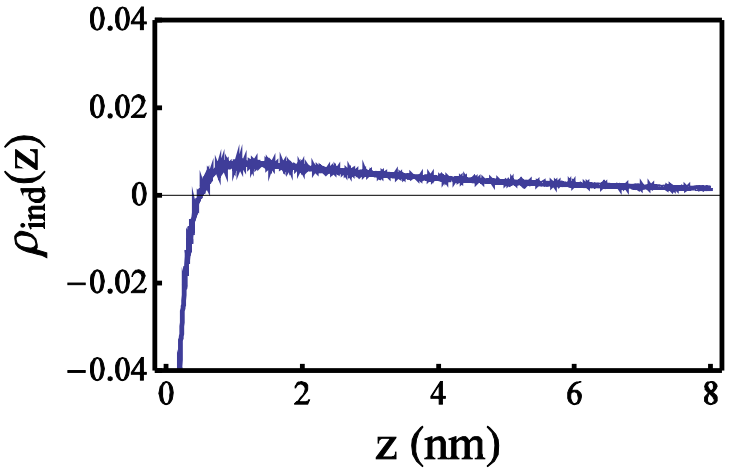
1D – Total induced charge in pillbox goes to zero.



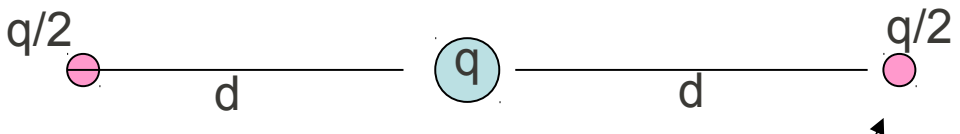


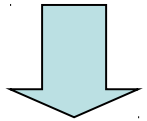


Why Antiscreening? Part 2



Model of Induced Charge





Screening Charges

Antiscreening charges

V(z) from induced Charge