Thermal Transport in Nanostructures: A Multiscale Multiphysics Simulation Approach

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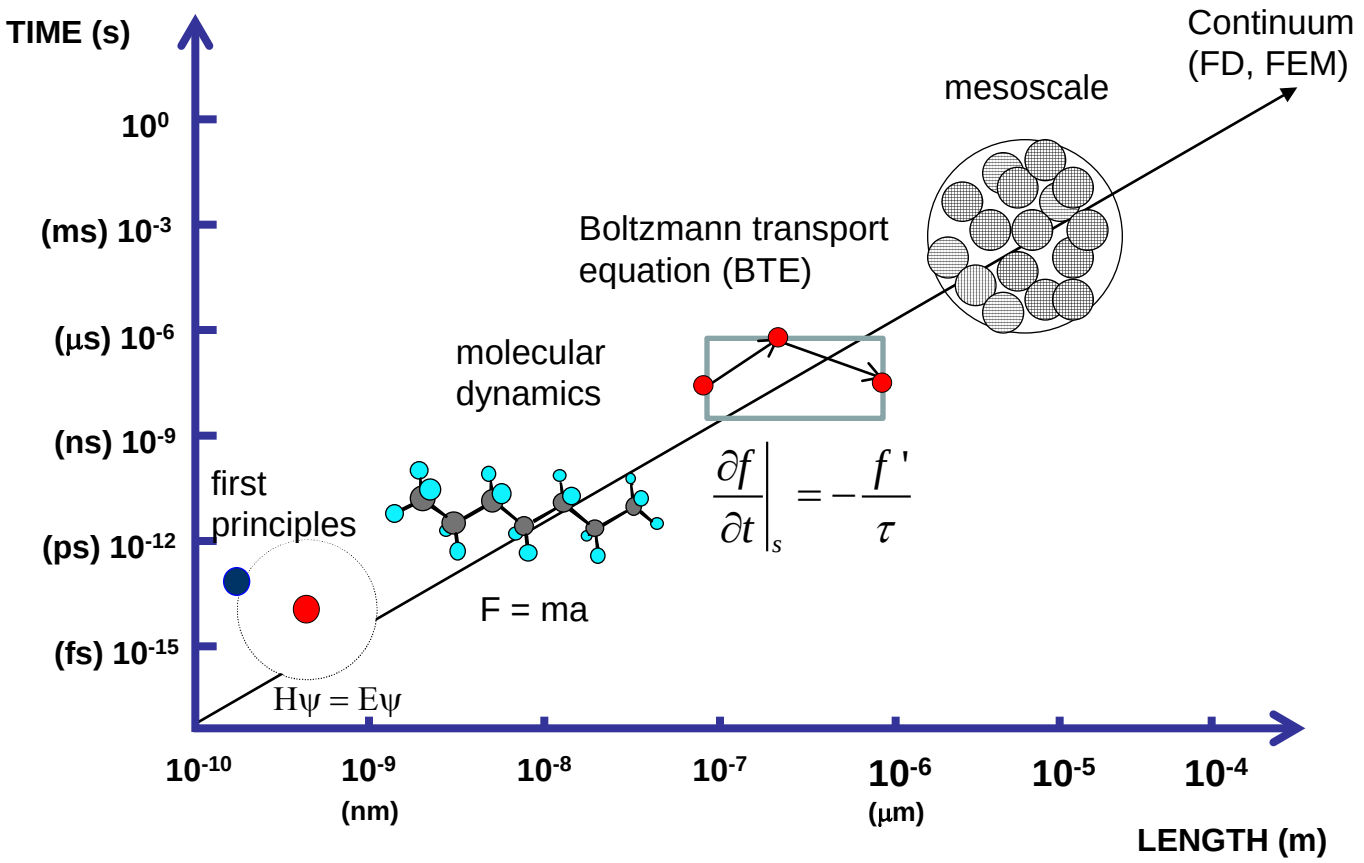
Outline

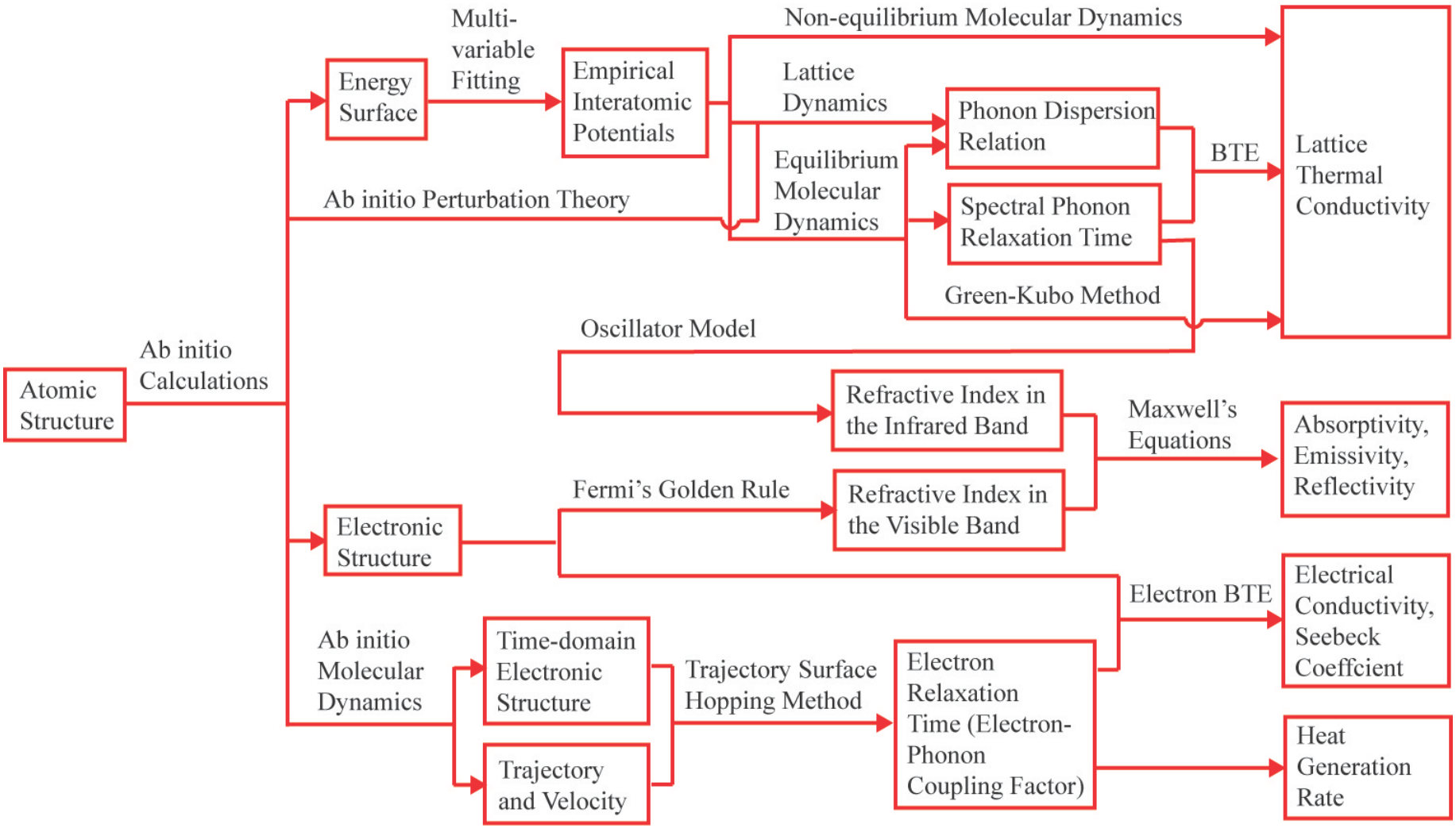
- *Overview of Multiscale Multiphysics Simulations*
- Thermal Transport and Thermal Rectification involving Interfaces
- Thermal Conductivity of Thermoelectric Bulk and Nanomaterials
- Enhancement of Optical Absorption in Nanotube/Nanowire Array Solar Thermal/Photovoltaic Materials
- Reduction of Heat Generation in Quantum Dot Photovoltaic Materials
- Summary and Acknowledgements

Length and Time Scales of Simulation Methods



- Parameters obtained in a smaller scale simulation can be fed into larger scale simulations.

Multiscale Multiphysics Schemes



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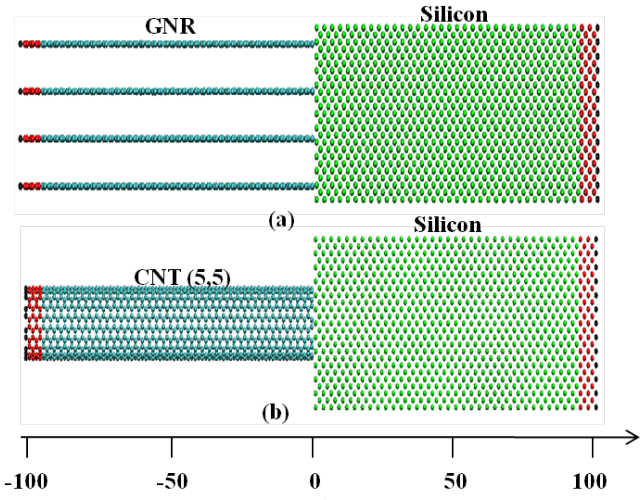
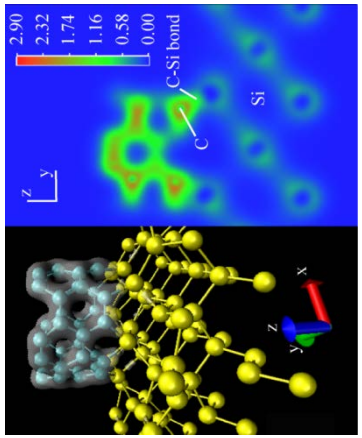
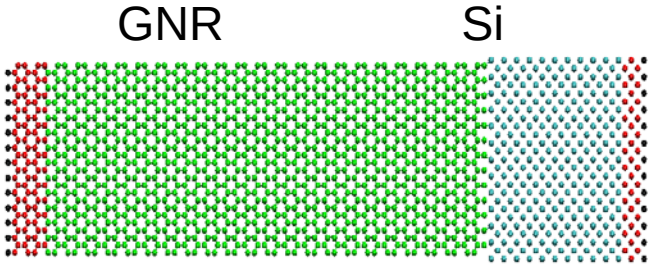
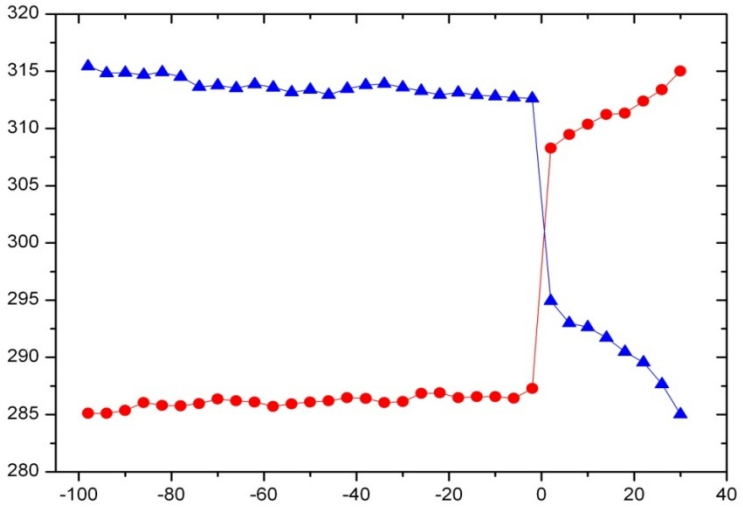
Vertical CNT-Si and GNR-Si Interfaces





- Interfacial bonding is covalent.





- The thermal conductance is

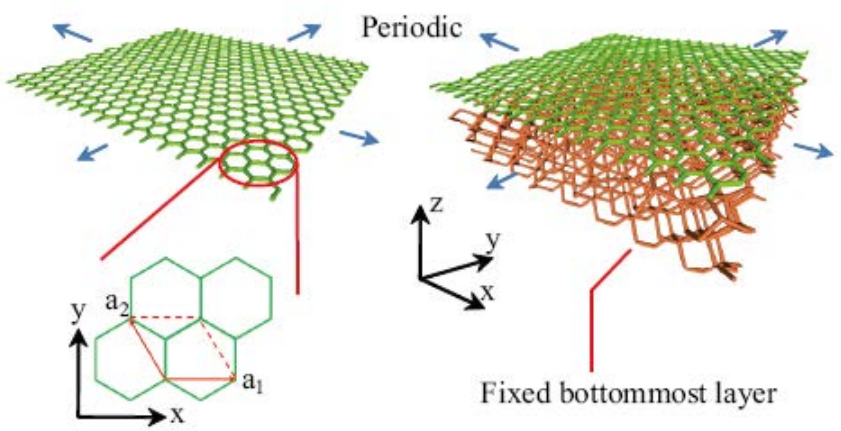
$$G_s = \frac{J}{A_s \Delta T}$$

A. Vallabhaneni, B. Qiu, J.N. Hu, Y.P. Chen, A.K. Roy, and X.L. Ruan, in review.

Graphene Supported on Substrate

- MD simulation domains

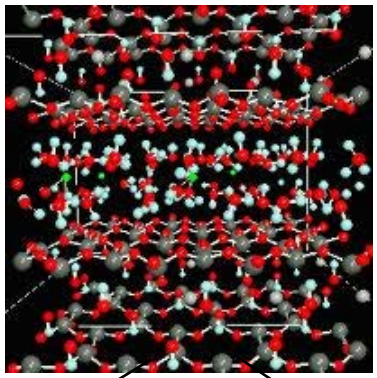
Single-layer Graphene



Single-layer Graphene

Silicon as substrate

- Spectral Energy Density



position velocity

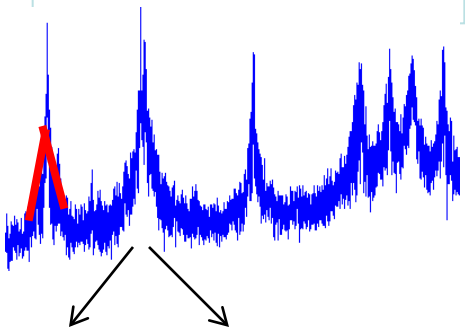
Atomic velocity

$$\Phi(\kappa,\omega)=\frac{1}{4\pi\tau_0 N_T}\sum_{\alpha}\sum_{b}^{B} m_b \left| \int_0^{\tau_0}\sum_{n_{x,y,z}}^{N_T} \dot{u}_\alpha\left(\begin{matrix}n_{x,y,z}\\ b\end{matrix};t\right) \times\exp\left[i\boldsymbol{\kappa}\cdot\mathbf{r}\left(\begin{matrix}n_{x,y,z}\\ 0\end{matrix}\right)-i\omega t\right] dt \right|^2 .$$

$$=\sum_{\nu}^{3n} C_0\left(\boldsymbol{\kappa}_\nu\right)\frac{\Gamma\left(\boldsymbol{\kappa}_\nu\right)/\pi}{\left[\omega_A\left(\boldsymbol{\kappa}_\nu\right)-\omega\right]^2+\Gamma^2\left(\boldsymbol{\kappa}_\nu\right)}$$

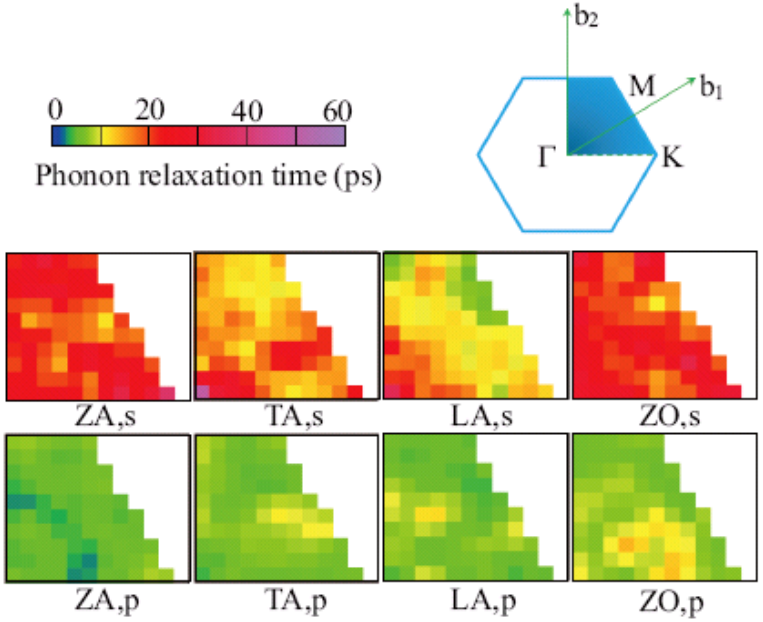
Lorentzian form

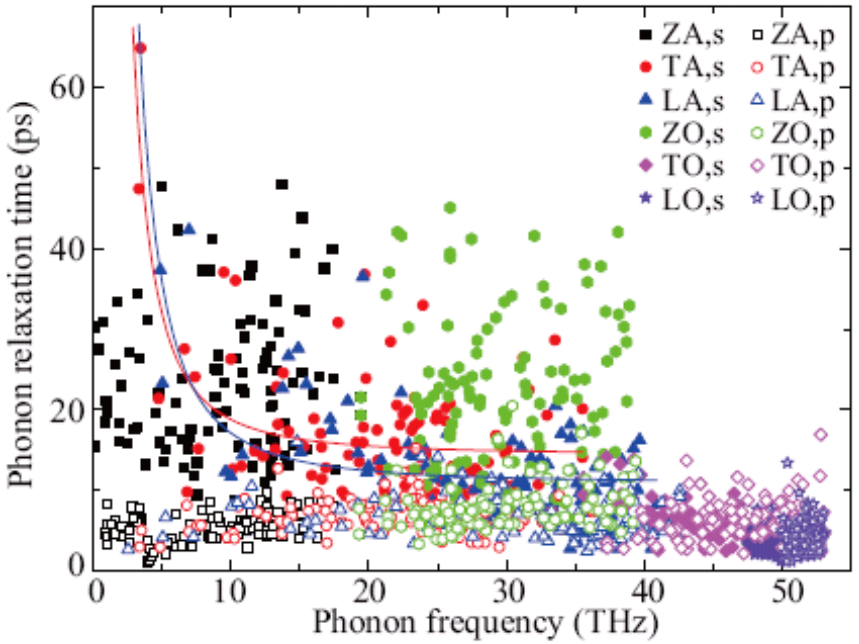
Spectral peaks fitting



κ, ω τ

Reduction of Phonon Relaxation Time τ

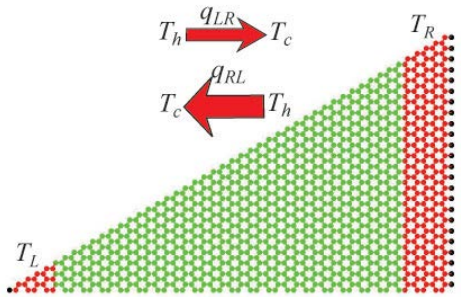




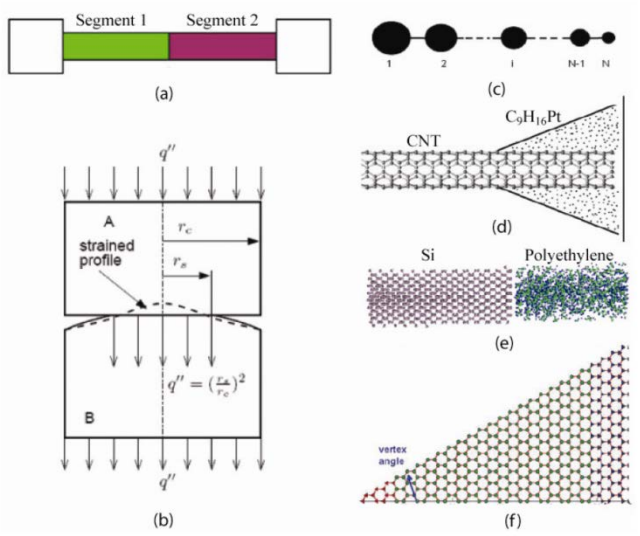
- Suspended graphene: τ below 70 ps
  - ZA, ZO : Large τ – weak coupling to in-plane phonons
    Not huge – higher order anharmonic interactions limiting τ
  - TA, LA: Larger τ at smaller **k**, smaller ω
- Supported graphene: τ of **all** phonon branches largely reduced
  - Symmetry broken: mirror symmetry, translational symmetry
  - Most drop in ZA

Qiu and Ruan, Appl. Phys. Lett., in press, 2012.

Thermal Rectification in Asymmetric Graphene Nanoribbons

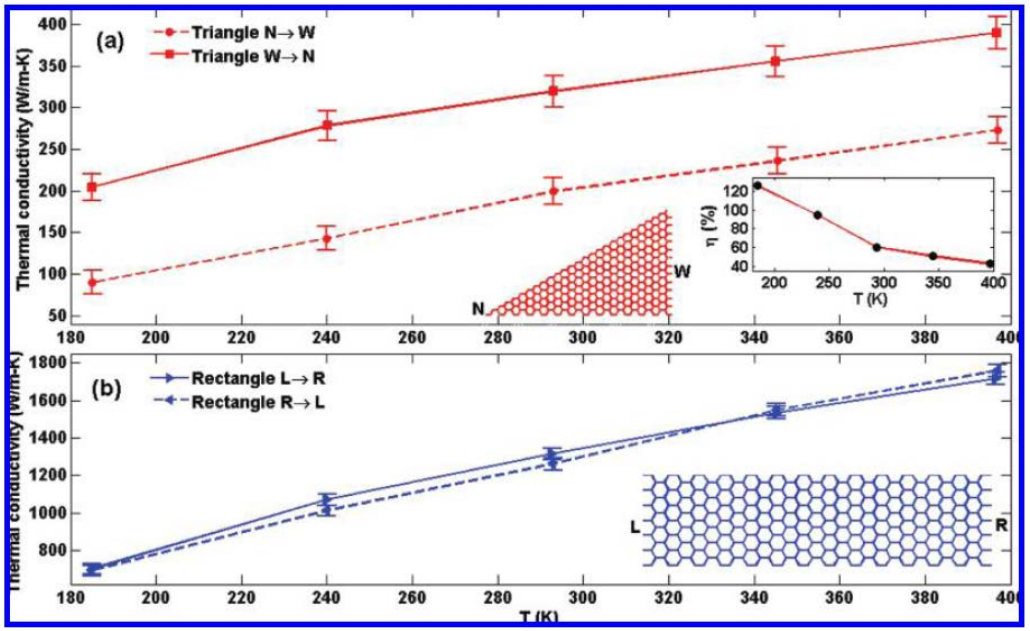


- Various thermal rectifiers



- Thermal rectification factor:

$$\eta = |q_{LR} - q_{RL}| / q_{min}$$



Hu, Ruan, and Chen, Nano Letters, 2009.

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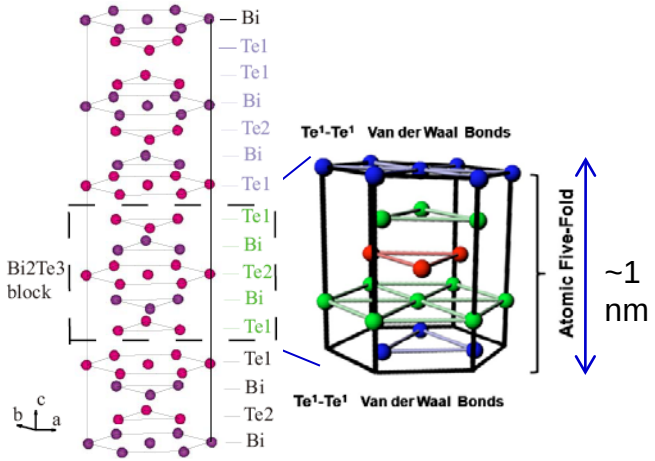
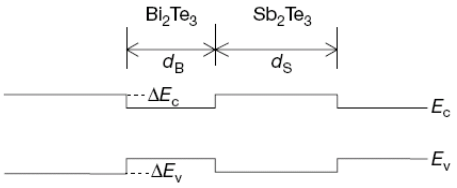
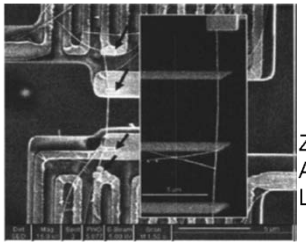
Bi_2Te_3 and PbTe Bulk and Nanostructures

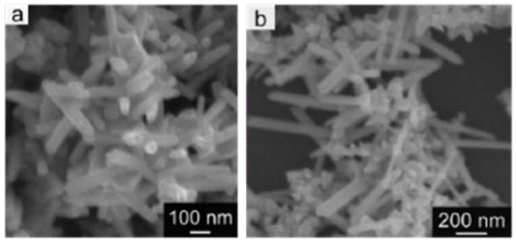




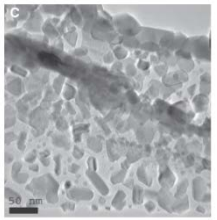
Venkatasubramanian *et al*, Nature, 2001



Zhou *et al*, Appl. Phys. Lett 2005

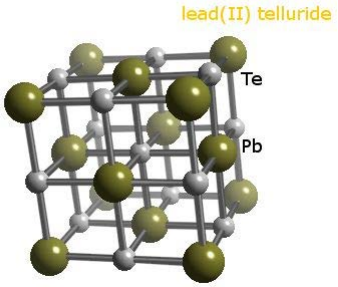


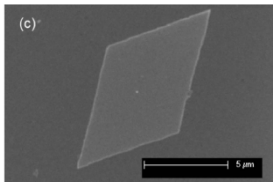
Purkayastha *et al*, Adv. Mater. 2006



Poudel *et al*, Science, 2008

- Bi_2Te_3: Layered structure similar to graphite. Van Der Waals and electrostatic interactions between quintuples.



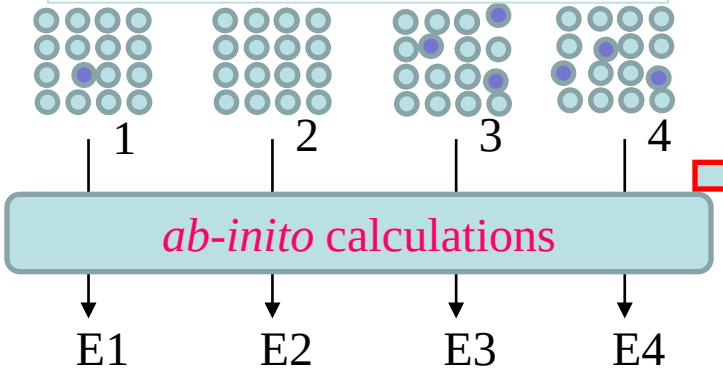


Teweldebrhan, Goyal, and Balandin, Nano Lett. 10, 1209, 2010.

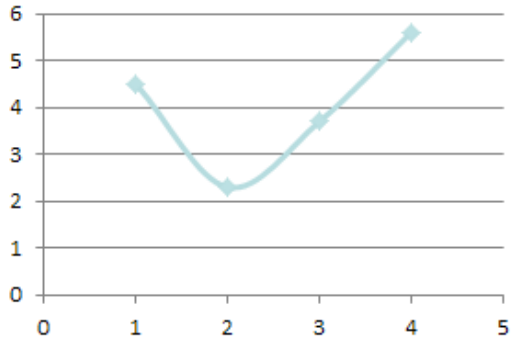
- PbTe: cubic structure similar to NaCl

Empirical Interatomic Potential Development

Configuration creation



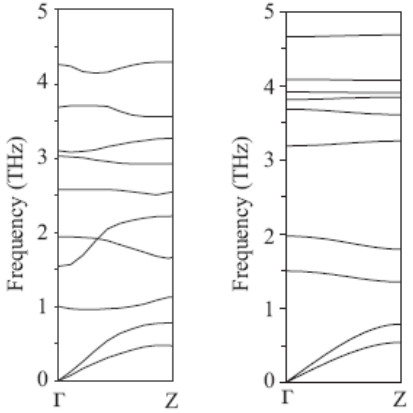
Energy surface generation



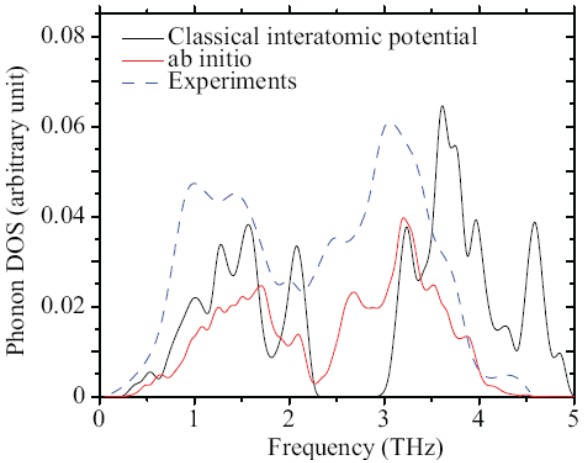
Potential parameterization

$$V(r) = D_e(1 - e^{-a(r-r_e)})^2$$

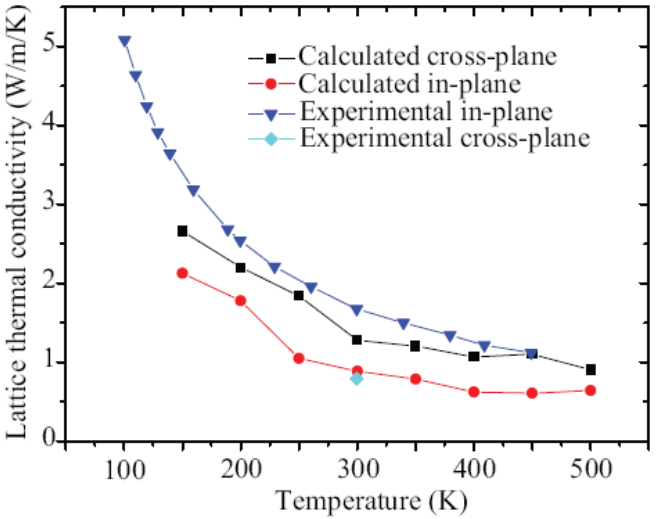
$D_e = ?, \alpha = ?, r_e = ?$



Phonon dispersion

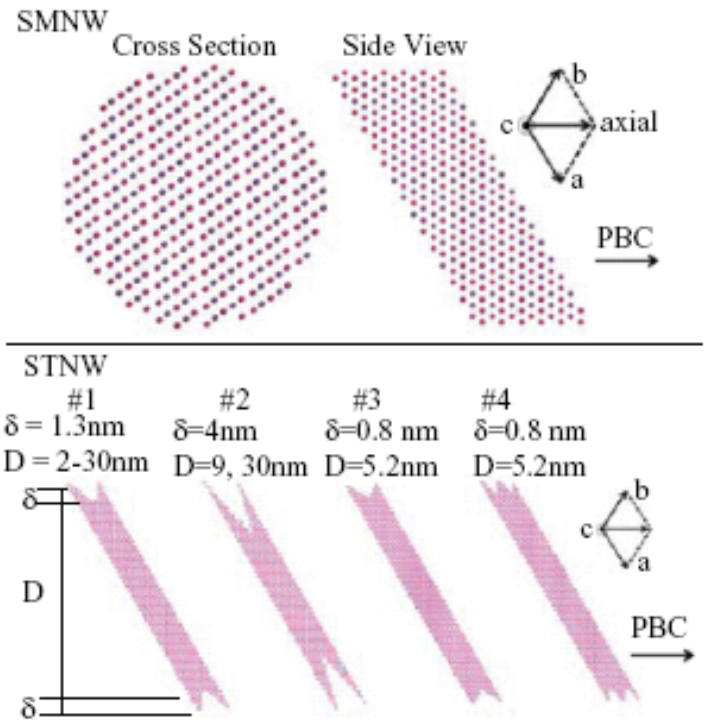


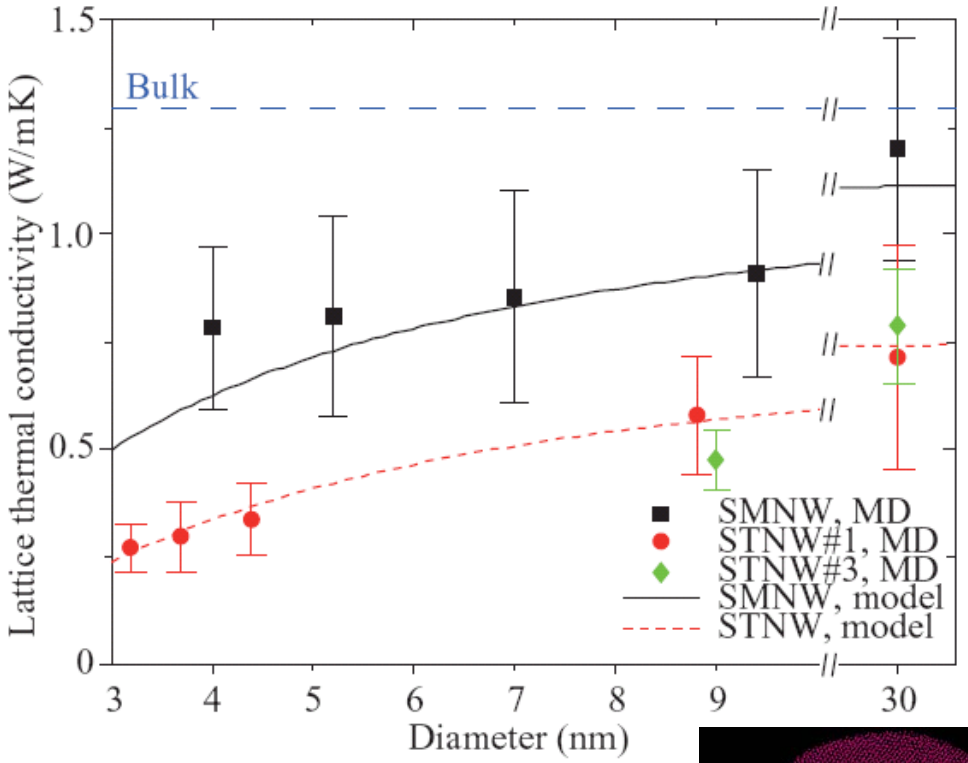
Phonon DOS



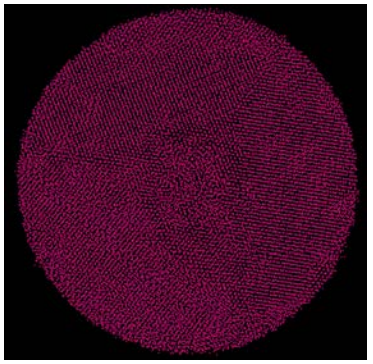
Qiu and Ruan, Phys. Rev. B, 2009.

Thermal Conductivity Reduction in Bi_2Te_3 Nanowires



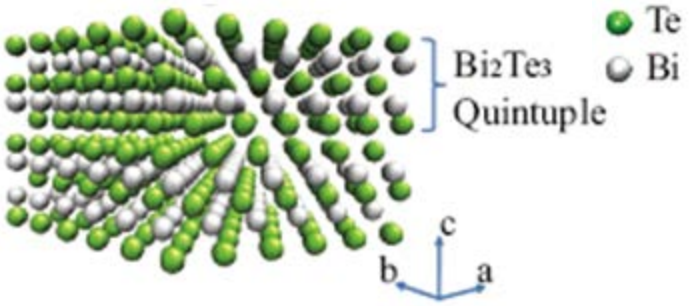


Qiu, Sun, and Ruan, *Phys. Rev. B*, **83**, 035312 (2011)



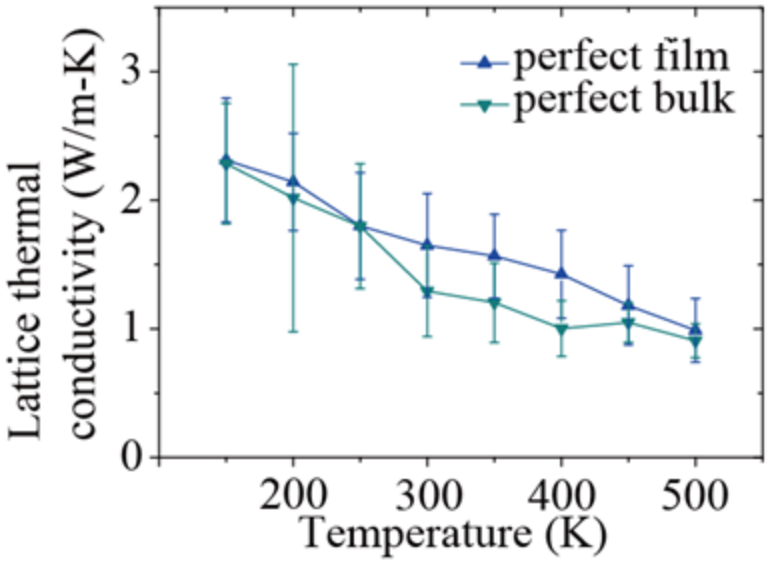
- Diameter decreases → thermal conductivity decreases
- Bulk → SMNW : 50% lower thermal conductivity
- SMNW → STNW: additional 35% lower thermal conductivity
- At 30 nm diameter, no significant reduction seen

Thermal Conductivity of Bi_2Te_3 Few-Quintuple Films



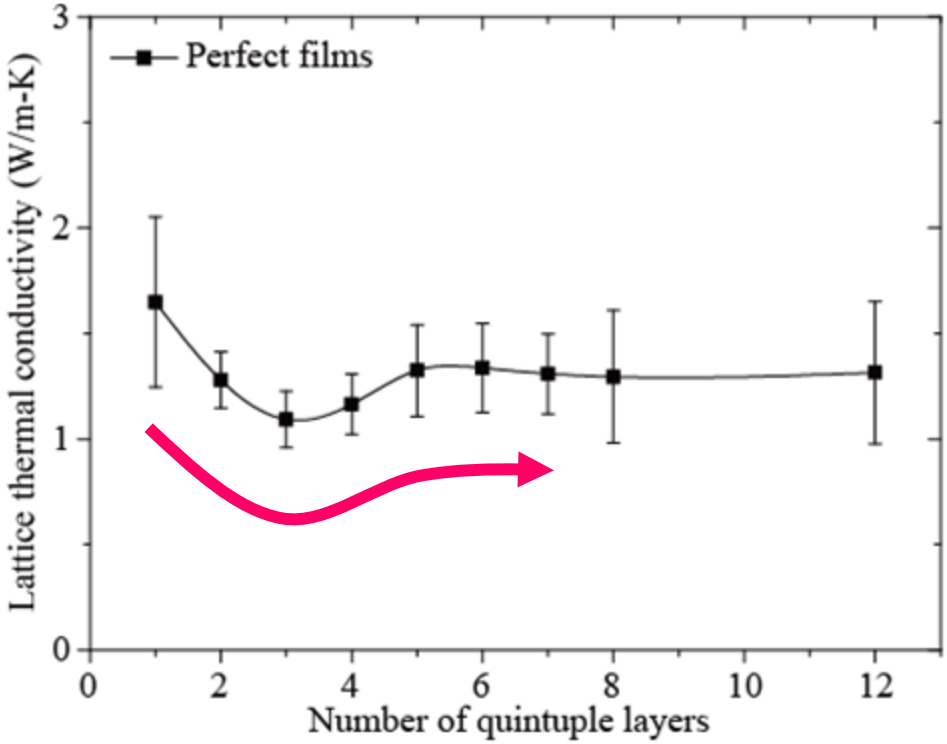
Temperature dependence

Single quintuple film



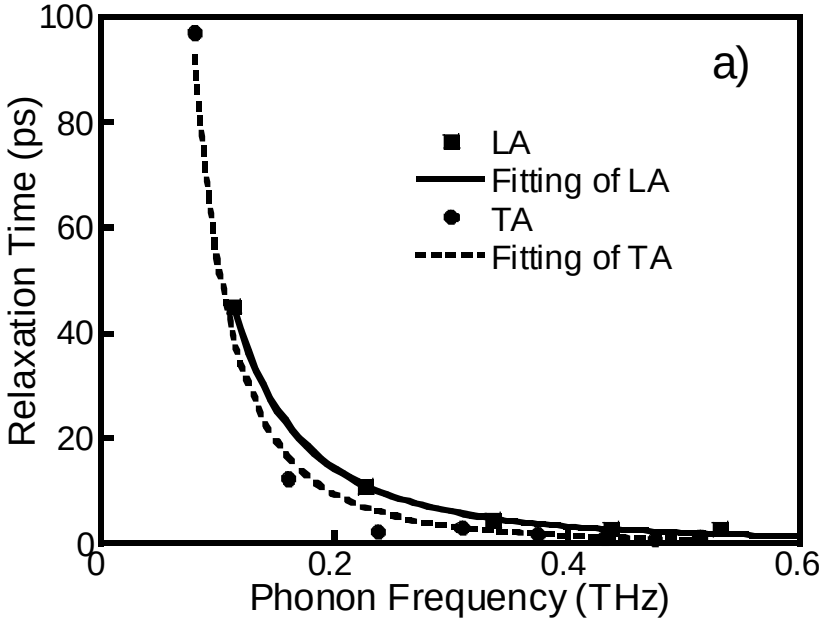
Thickness dependence

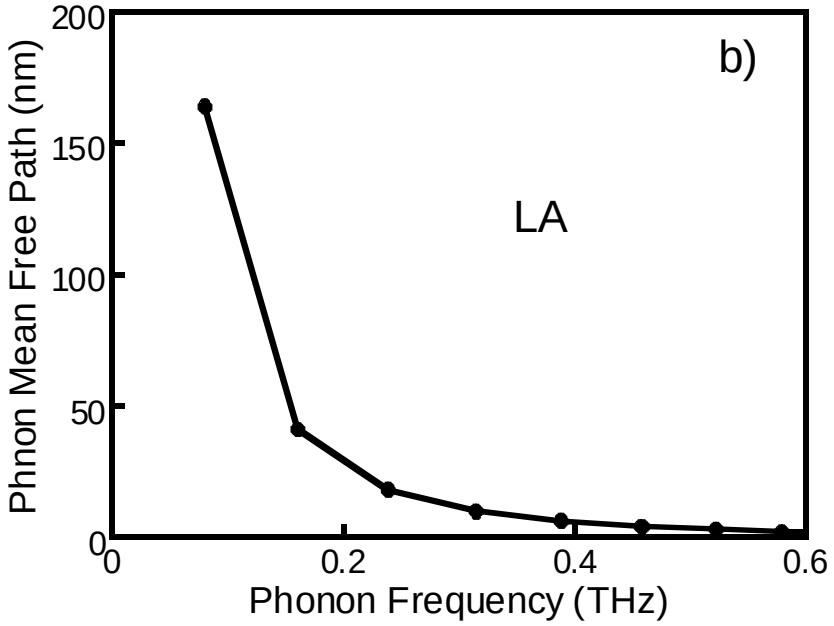
Room temperature



Qiu and Ruan, *Appl. Phys. Lett*. **97**, 183107 (2010)

Spectral Phonon Relaxation Time and MFP





- The phonon relaxation time agrees very well with ultrafast pump-probe measurements.

Wang, Qiu, McGaughey, Ruan, and Xu, J. Heat Transfer, in preparation, 2012.

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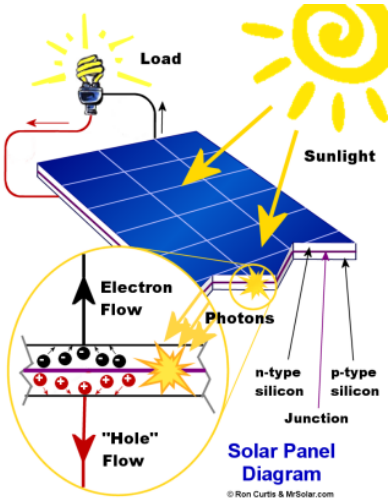
Solar Energy Harvesting and its Loss Mechanisms

http://deviceace.com/science/266/more-efficient-solar-cells-thanks-to-cnt-films-that-transmit-infrared-light.html



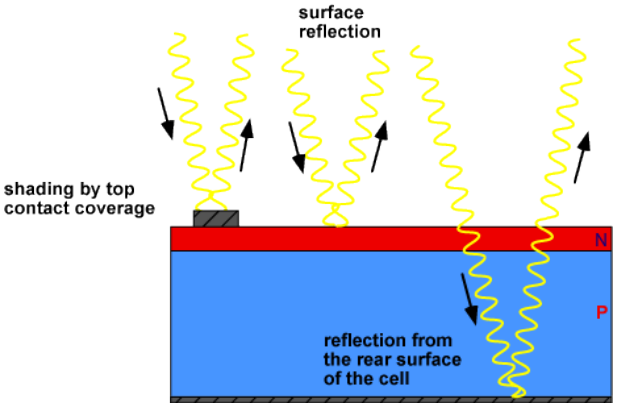
http://mrsolar.com

- Three steps:

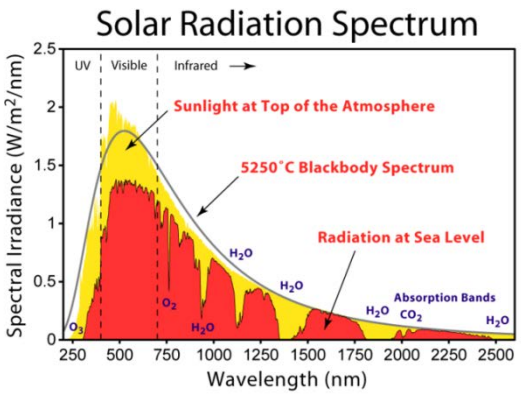
(1) Absorption of solar photons and creation of electron-hole pairs;

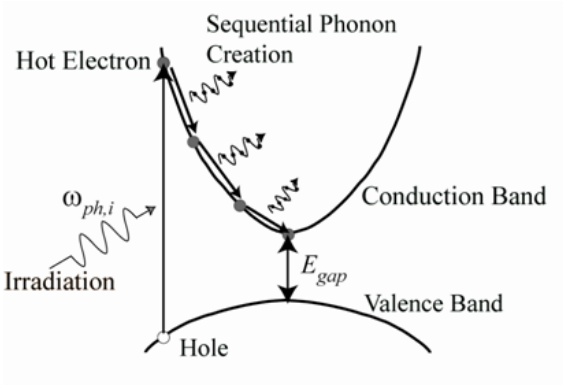
(2) Decay of hot carriers;

(3) Diffusion of carriers to electrodes.



- Loss mechanism 1: reflection





- Loss mechanism 2: hot carrier relaxation

Ground State Ab Initio Methodology

- Density functional theory to solve the Kohn-Sham equation

$$\left[-\frac{\hbar^2\nabla^2}{2m}+V_{KS}\right]\varphi_{nk}(\mathrm{r})=\varepsilon_{nk}\varphi_{nk}(\mathrm{r})$$

- Fermi's golden rule – imaginary part of the dielectric function

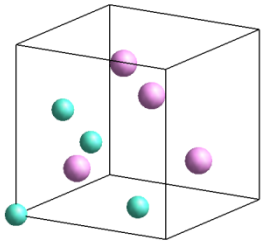
$$\epsilon''_{\alpha,\alpha}(\omega)=\frac{4\pi^2}{\Omega\omega^2}\sum_{i\in VB,j\in CB}\sum_{\mathbf{k}}w_{\mathbf{k}}\left|p^{\alpha}_{ij}\right|^2\delta(\epsilon_{\mathbf{k}j}-\epsilon_{\mathbf{k}i}-\omega)$$

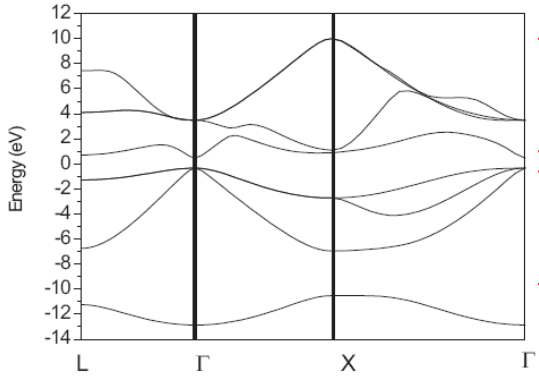
$p^a_{ij}=\langle\mathbf{k}j|p_a|\mathbf{k}i\rangle$ is the transition matrix element.

- Kramer-Kronig transformation – real part of the dielectric function

$$\epsilon'(\omega)=1+\frac{4}{\pi}\mathbf{P}\int_0^{\infty}d\omega'\frac{\omega'\epsilon''(\omega')}{\omega'^2-\omega^2}$$

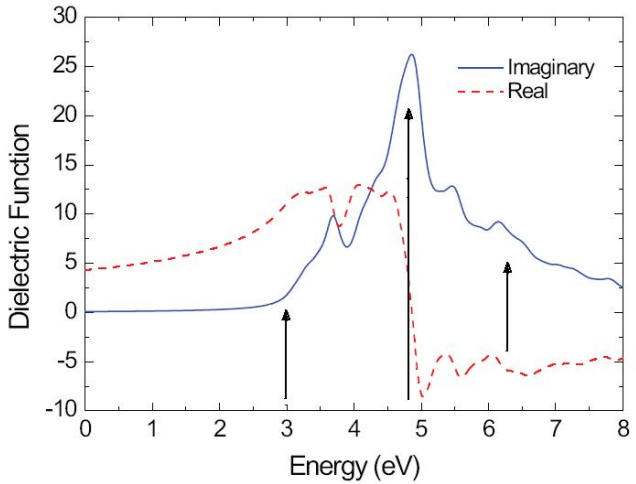
Benchmark Work on Bulk GaAs

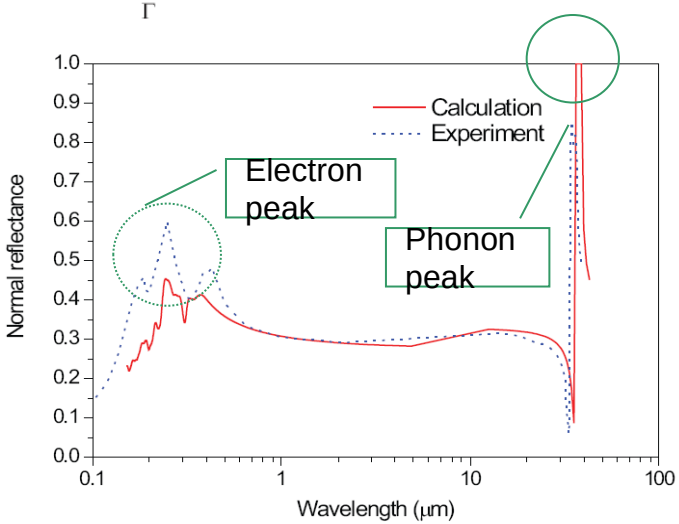




Conduction band (CB)

Valence band (VB)

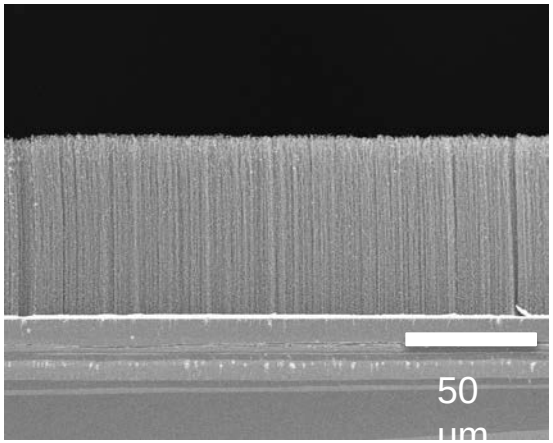


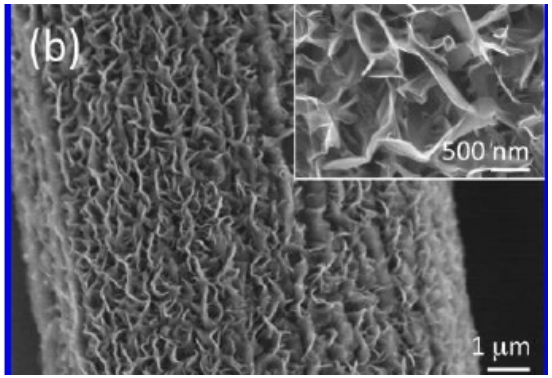


H. Bao and X.L. Ruan, *Int. J. Heat Mass Transfer* **53**, 1308-1312, 2010.

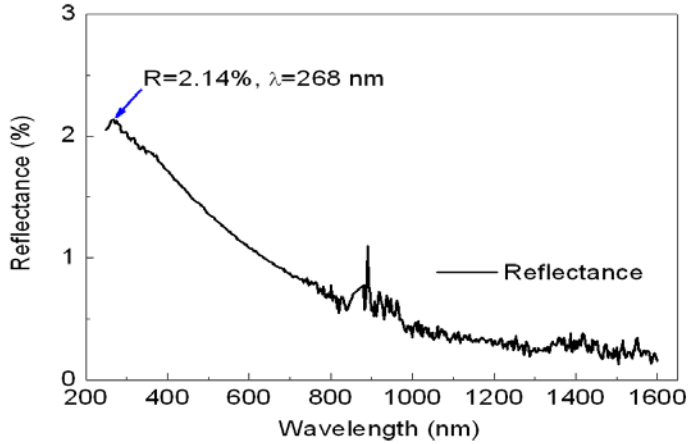
- Electron peak: photon-electron interaction
- Phonon peak: photon-phonon interaction

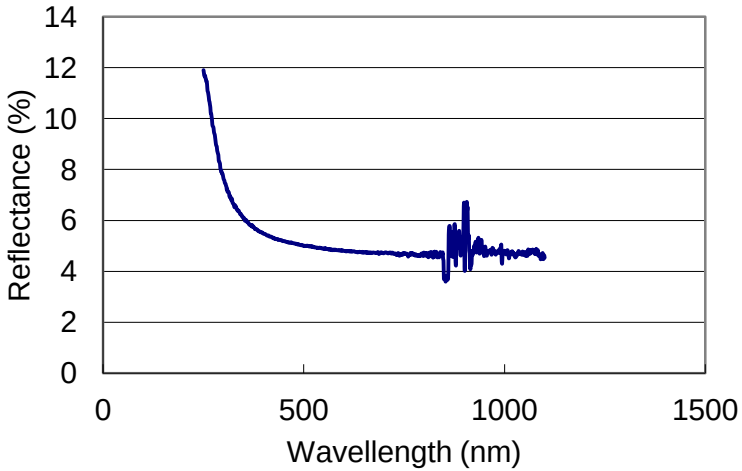
Ultra-Low Reflectance in CNT and Graphene Arrays





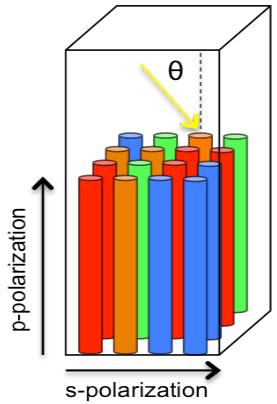
Samples provided by Timothy Fisher at Purdue

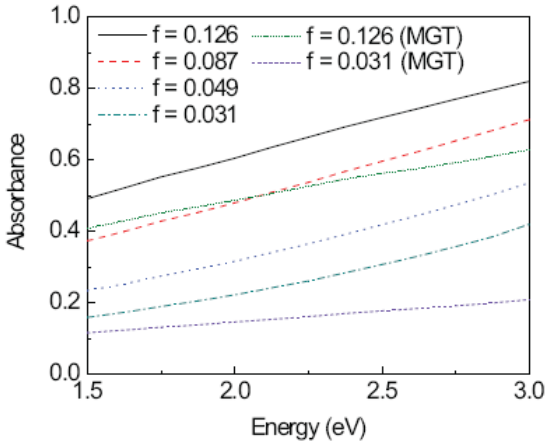


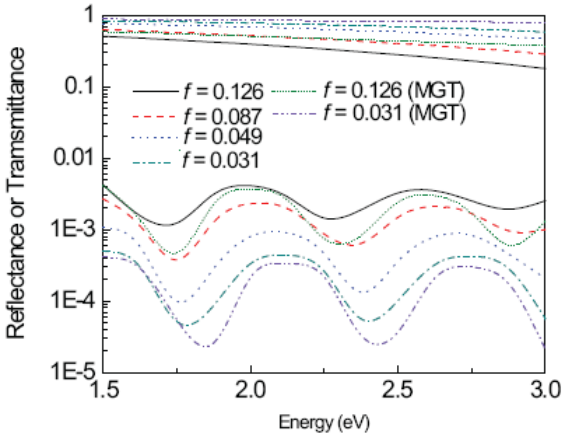


- Vertical CNT and graphene arrays are excellent solar thermal absorbers.

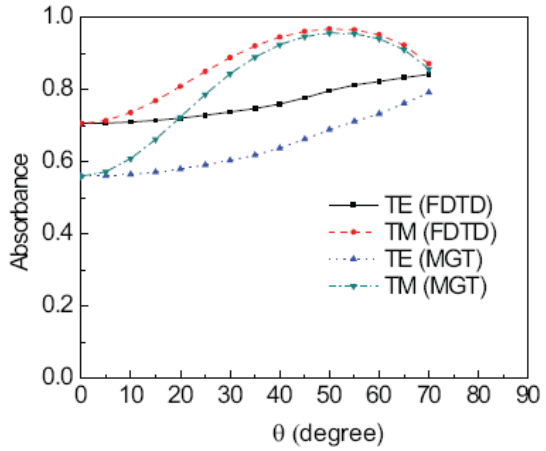
Finite Difference Time Domain (FDTD) Simulations

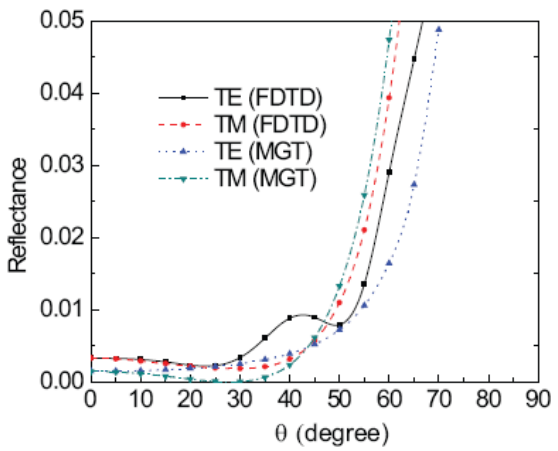






- The absoptance increases with filling fraction
- The absorptance first increases and then decreases with increasing incident angle.
- The effective medium theory is not accurate.

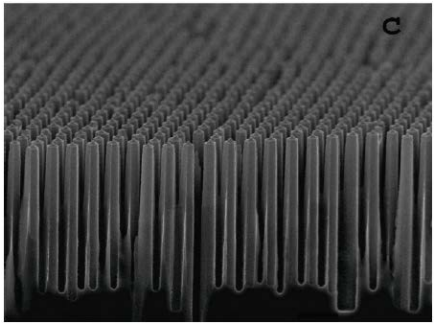




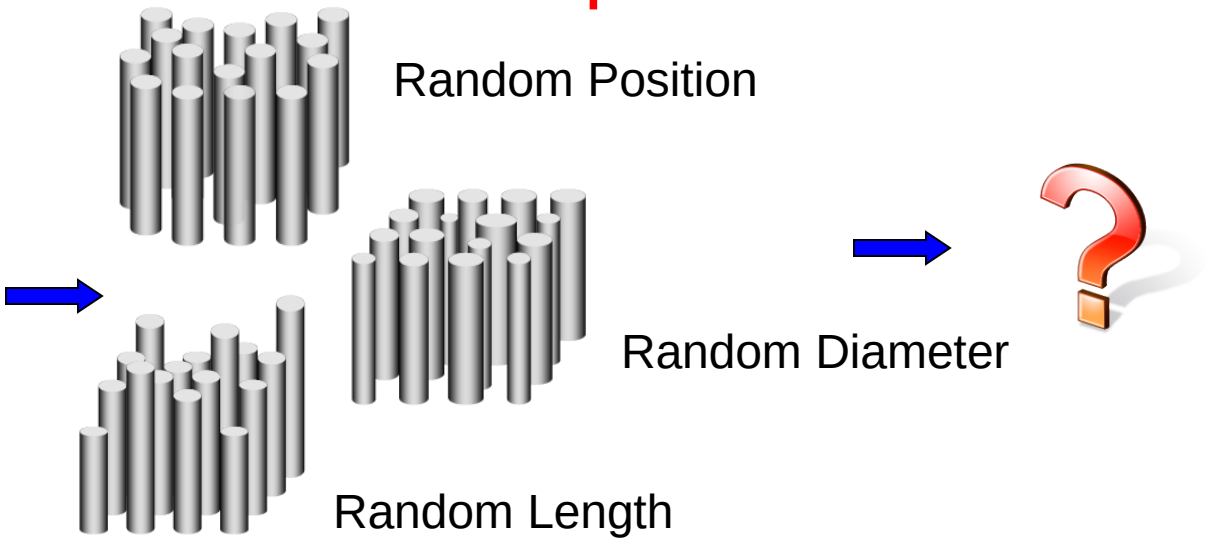
Bao, Ruan, and Fisher, Optics Express, **18**, 6347-6359, 2010.

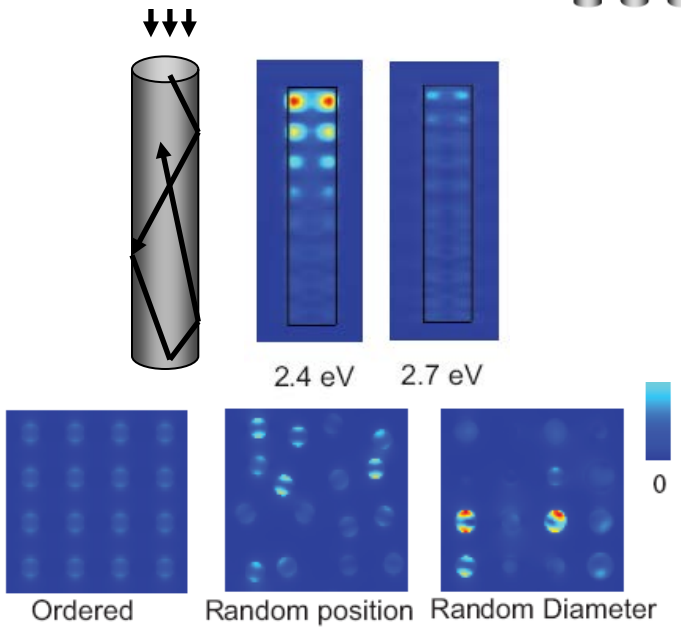
Randomness Enhances Absorption!

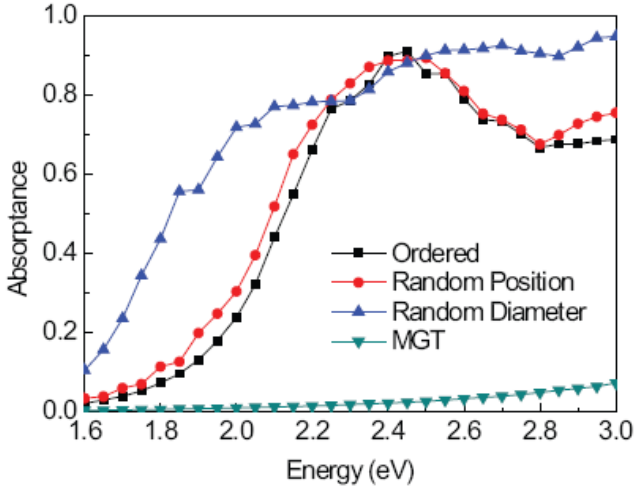
Experiment



Garnett et al., Nano Lett, 2010





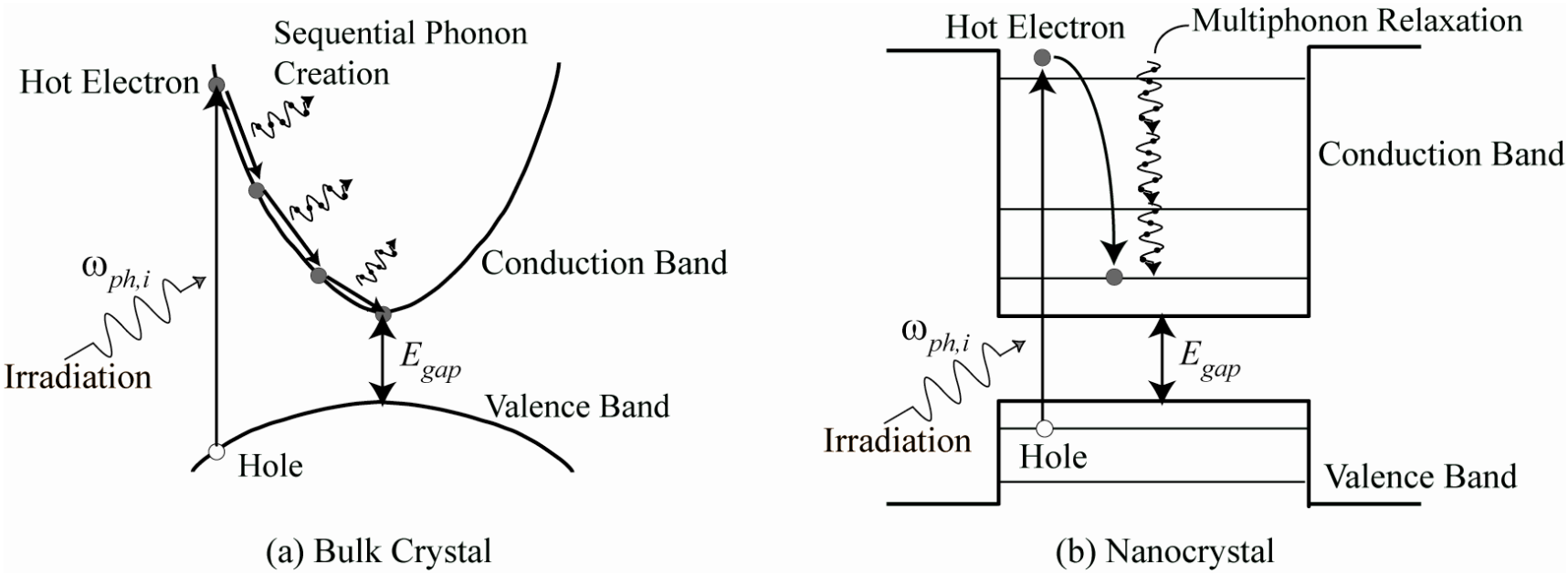


Bao and Ruan, *Opt. Lett*. **35**, 3378-3380, 2010.

Outline

- Overview of Multiscale Multiphysics Simulations
- Thermal Transport and Thermal Rectification involving Interfaces
- Thermoelectric Properties of Bulk and Nanomaterials
- Enhancement of Optical Absorption in Nanotube/Nanowire Array Solar Thermal/Photovoltaic Materials
- *Reduction of Heat Generation in Quantum Dot Photovoltaic Materials*
- Summary and Acknowledgements

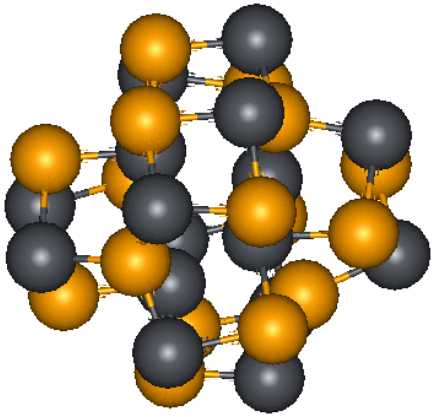
Reduced Electron-Phonon Coupling in QDs

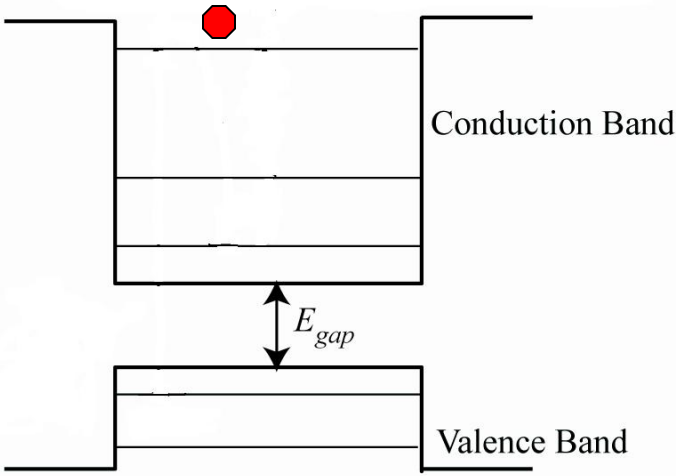


- Electron-phonon scattering is expected to be more difficult to occur in quantum dots because: (1) multiphonon process is a higher order process, (2) energy conservation is more difficult.

Non-Adiabatic Molecular Dynamics

- MD with **electron-phonon coupling calculations**
- Trajectory surface hopping within time-dependent density functional theory (TDDFT)





NA coupling strength:

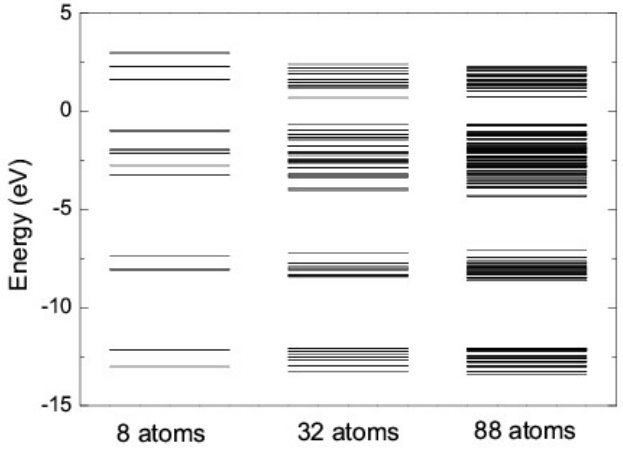
$$\mathbf{d}_{km} \cdot \dot{\mathbf{R}} = -i\hbar \left\langle \hat{\varphi}_k \left| \frac{\partial}{\partial t} \right| \hat{\varphi}_m \right\rangle$$

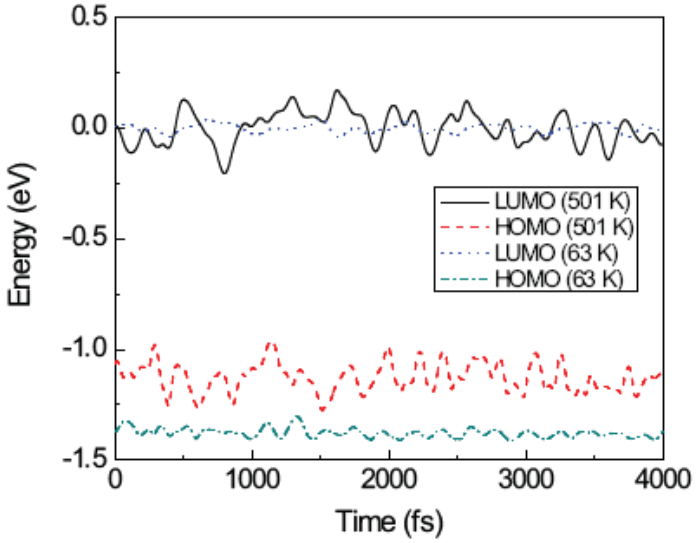
Transition probability:

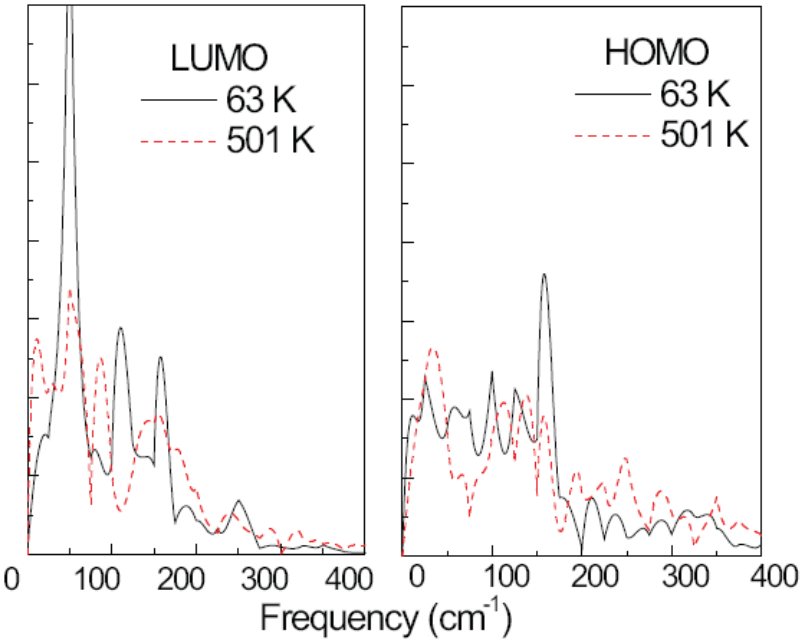
$$\mathrm{d}P_{km} = \frac{b_{km}}{a_{kk}} \mathrm{d}t$$

$$b_{km} = -\mathbf{Re}(a_{km}^{*} \mathbf{d}_{km} \cdot \dot{\mathbf{R}}); \; a_{km} = c_k c_m^{*}$$

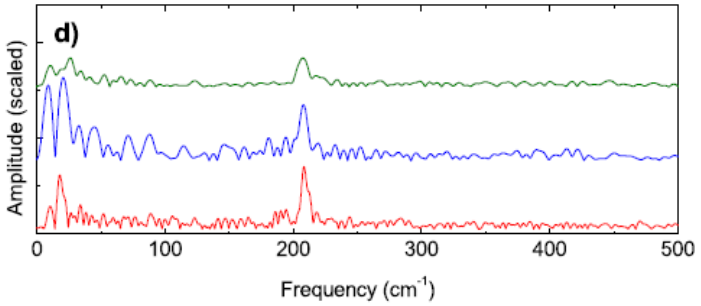
Band Structures and E-P Coupling Spectra for PbSe Quantum Dots





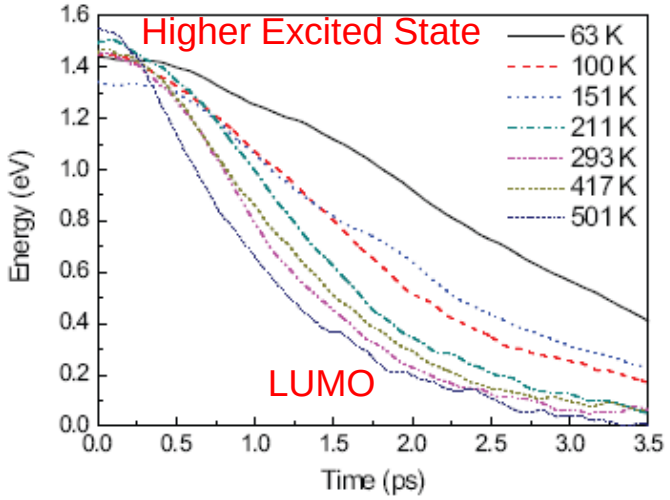


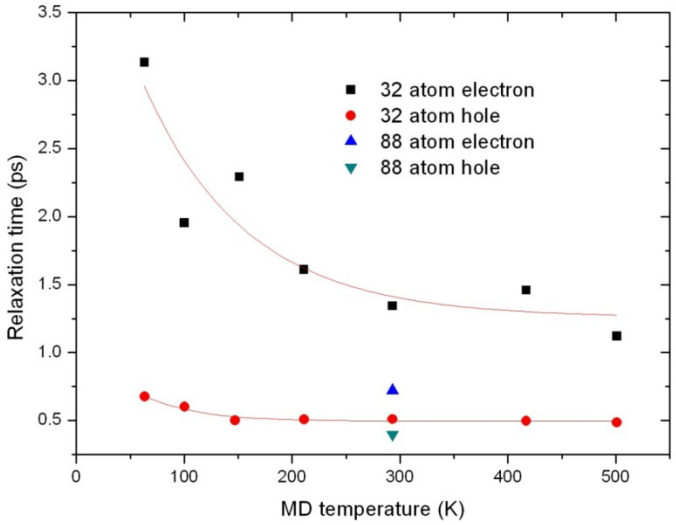
Bao, Habenicht, Prezhdo, and Ruan, Phys. Rev. B 79, 235306-1-7, 2009.



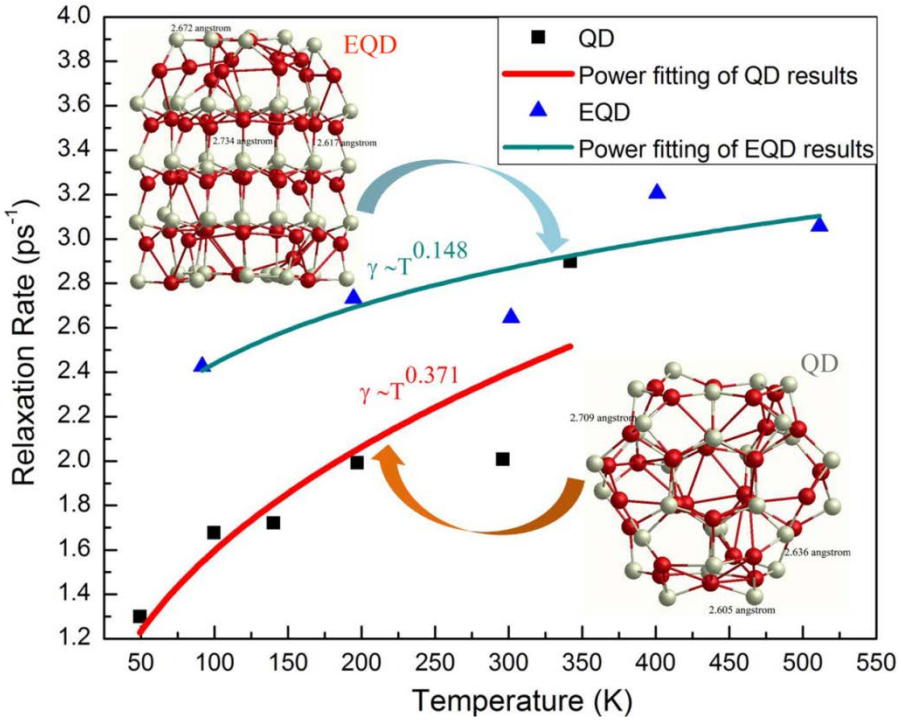
Sagar et al, PRB, 77, 235321 (2008)

Effects of Temperature, Size, and Shape





Bao, Habenicht, Prezhdo, and Ruan, Phys. Rev. B 79, 235306-1-7, 2009.



Chen, Bao, Tan, Prezhdo, and Ruan, *J. Phys. Chem. C*, 2011.

- The relaxation time is in the picosecond order. It decreases with increasing size and temperature.

Summary

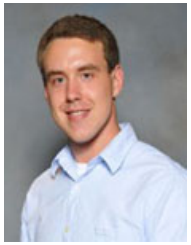
- Multiscale multiphysics approaches bridging different length and time scales can link macroscopic level thermal properties with atomic level structure. It is essential for a fundamental understanding of structure-property relationship.

- Thermal transport control that is not possible at the bulk phase can be realized at the nanoscale, by taking advantage of boundary, interface, and quantum confinement effects.

- Nanoscale control of thermal transport may significantly enhance the efficiencies of energy transfer and conversion processes.

Acknowledgements

- Graduate students:


Aaron


Ajit


Bo


Hua


Kelly


Bhagirath


Liang


Yan

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