Phonon Transport Across Interfaces

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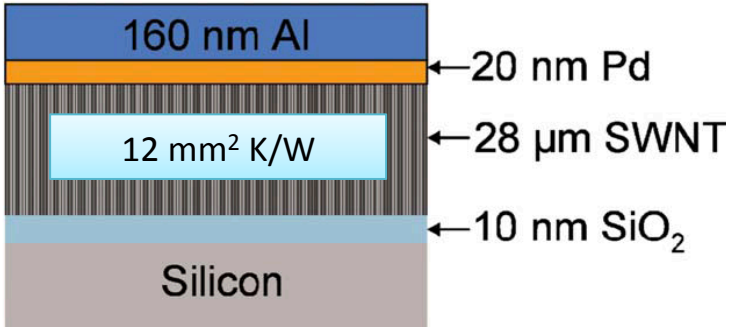
CNT TIM Structures and Performance



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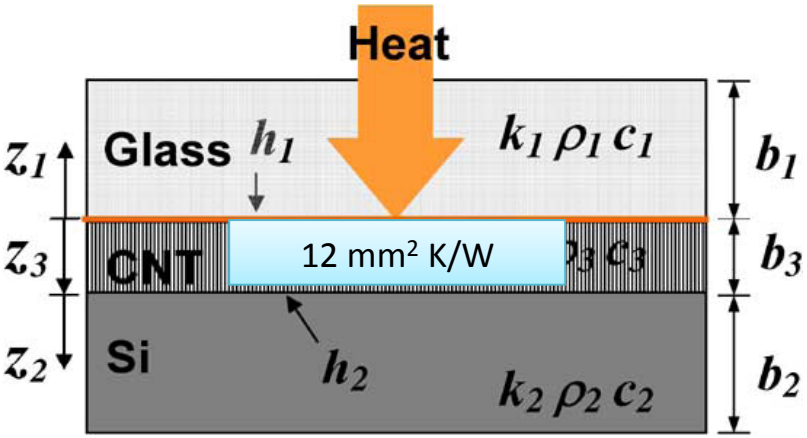
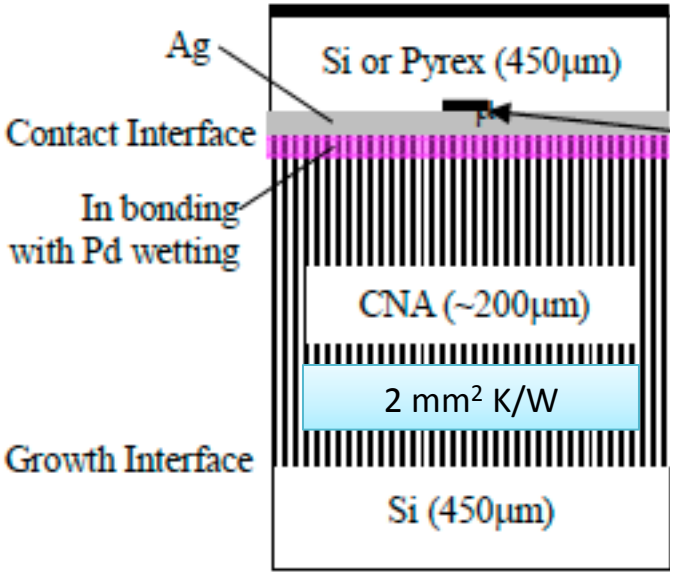


Tong et al. *IEEE Trans. on Comp. Pack. Tech.*, **30(1)**, 92 (2007)



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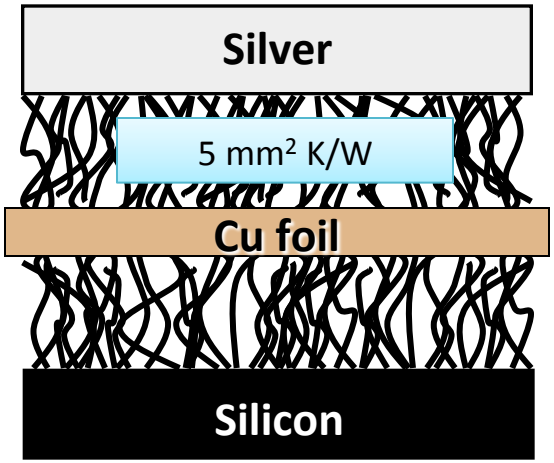
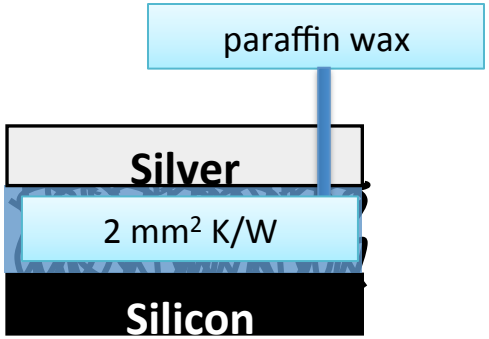
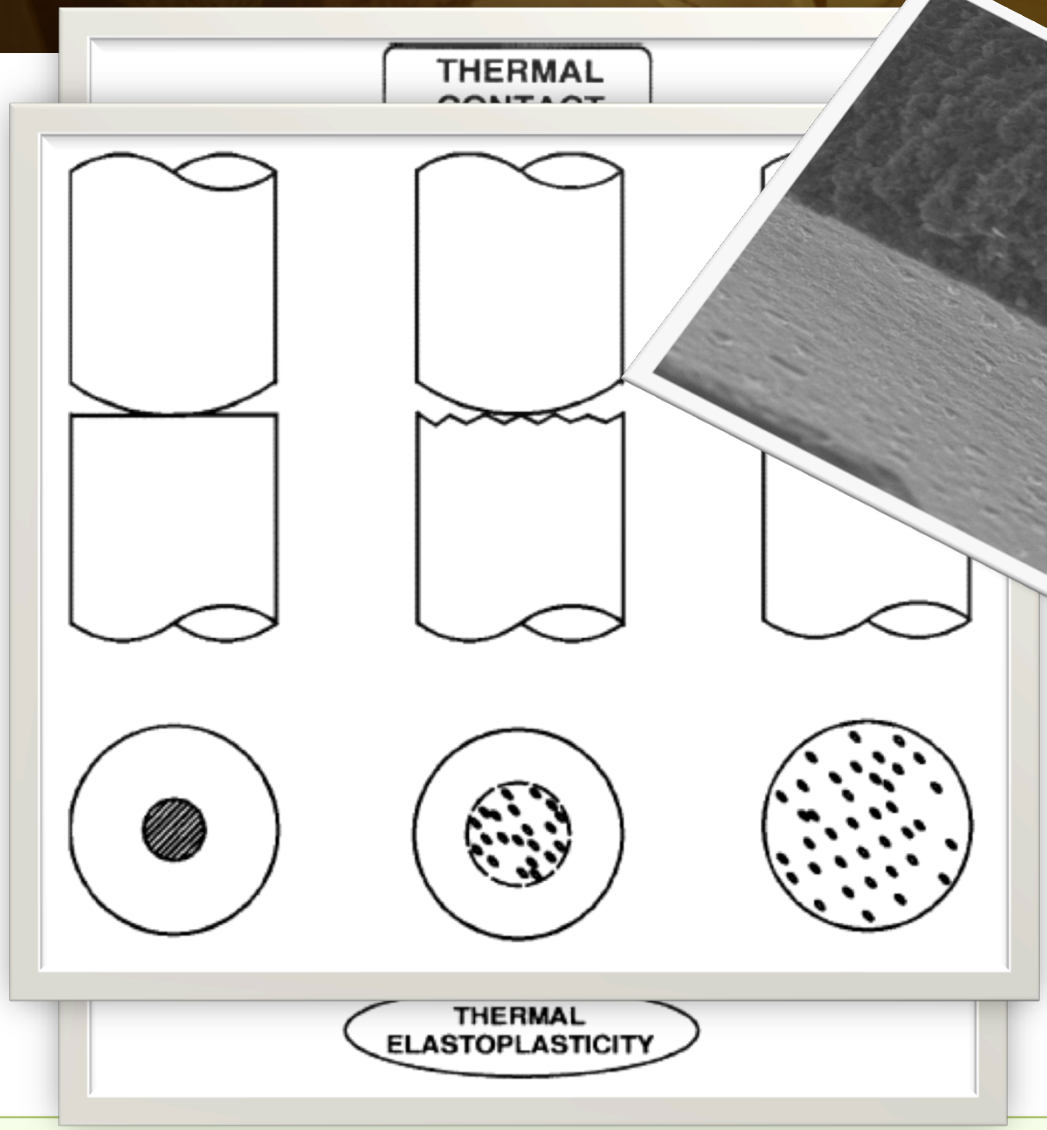


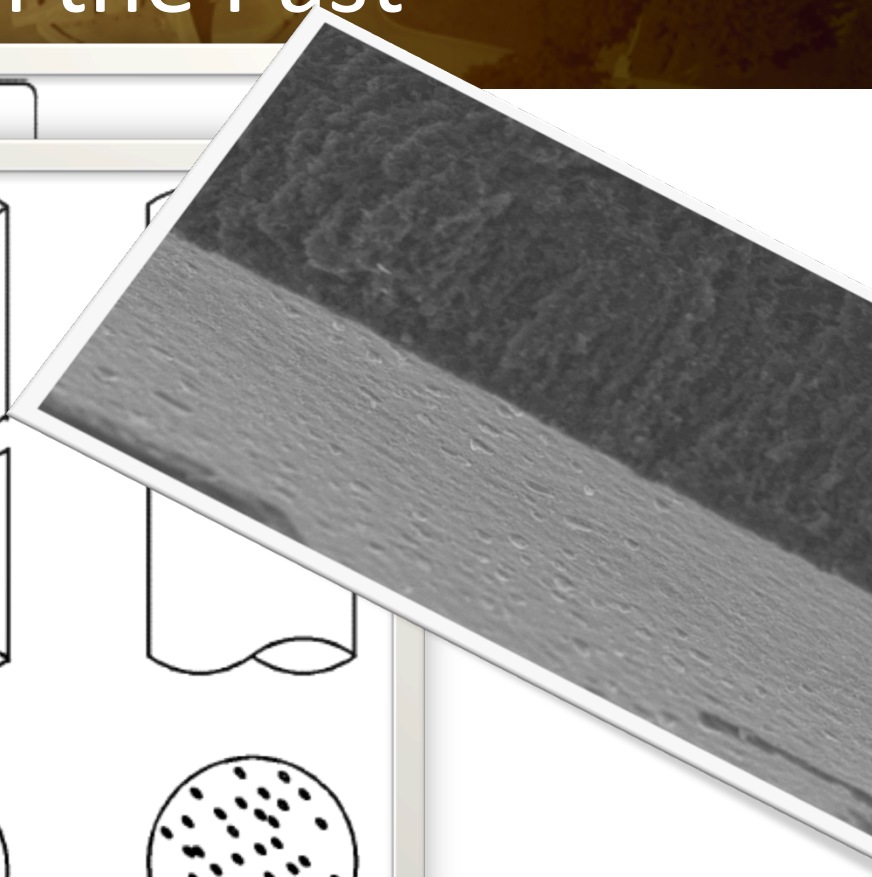
Cola et al. *Appl. Phys. Lett.*, **90**, 093513 (2007)



Cola et al. *Proc. of ASME Summer Heat Trans. Conf.*, HT**56483** (2008)

Learning from the Past





M. Yovanovich, "Four Decades of Research on Thermal Contact, Gap, and Joint Resistance in Microelectronics," *IEEE CPT*, 28, 182 (2005)

The ‘School’ Part

Displacement of a Loaded String

See Greenberg, *Foundations of Applied Mathematics*, Prentice-Hall, 1978.

- Consider a generic ODE of the form

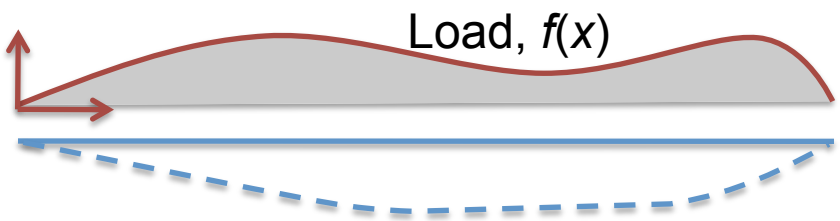
$$u'' + k^2 u = f(x)$$

  - Where the prime (‘) denotes a derivative
- With boundary conditions (u is displacement)

$$u(x=0) = a$$

$$u(x=L) = b$$

- Example: a loaded string ($k = a = b = 0$, u is displacement)

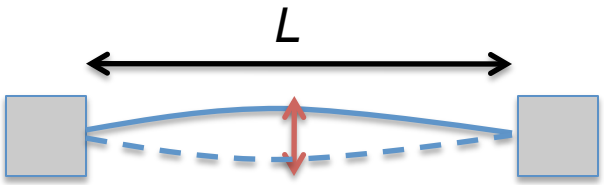


Note: in this case, f is the downward force per unit length per force of string tension

Example 2: Dynamic String

- Now consider a vibrating string with fixed-end boundary conditions

$$\frac{\partial^2 y}{\partial x^2} = \frac{\mu}{T}\frac{\partial^2 y}{\partial t^2}$$



- Where *y* is displacement, μ is mass density of the string, and *T* is tension
- We are interested in the natural frequencies of vibration, and therefore assume:

$$y(x,t) = u(x)\cos(\omega t)$$

Conversion of PDE to ODE

- The governing equation then becomes

$$\frac{d^2u}{dx^2} + \omega^2 \frac{\mu}{T} u = 0$$

- With boundary conditions

$$u(0) = u(L) = 0$$

- Compare to our general equation for static displacement:

$$u'' + k^2 u = f(x)$$

Vibrating String Solution

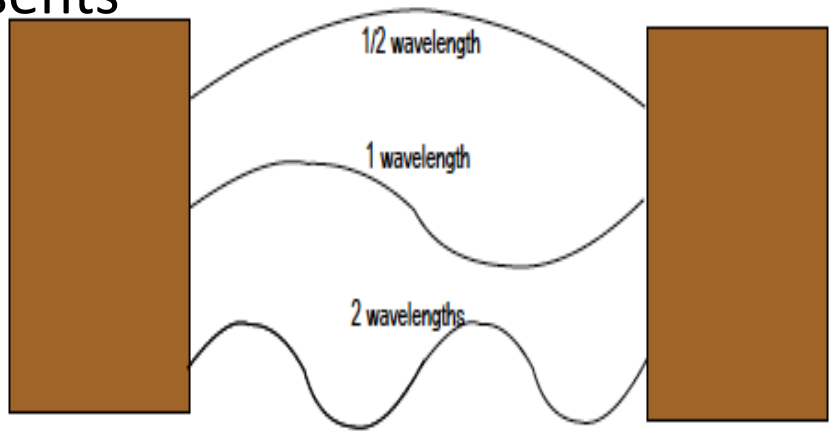
- We find that many different wavelengths λ_n will satisfy a solution of the form

$$u_n(x) = A_n \sin\left(\frac{n\pi x}{L}\right) = A_n \sin\left(\frac{2\pi x}{\lambda_n}\right), n = 1,2,3,\ldots$$

  – Where $\lambda_n = 2L/n$, and each n represents a different mode of vibration

- Let v be a speed defined by the tension and mass density

$$v = \sqrt{\frac{T}{\mu}}$$



See AP French, *Vibrations and Waves*, 1971

Vibrating String, cont'd

- The corresponding frequencies for each *n* are

$$\omega_n = \frac{n\pi}{L} v = \frac{2\pi}{\lambda_n} v$$

- And the overall solution becomes

$$y_n(x,t) = u_n(x)\cos(\omega_n t) = A_n \sin\left(\frac{2\pi x}{\lambda_n}\right)\cos(\omega_n t) = A_n \sin\left(K_n x\right)\cos(\omega_n t)$$

  – Where the last equality defines the wavevector

$$K_n = \frac{2\pi}{\lambda_n}$$

Progressive Waves

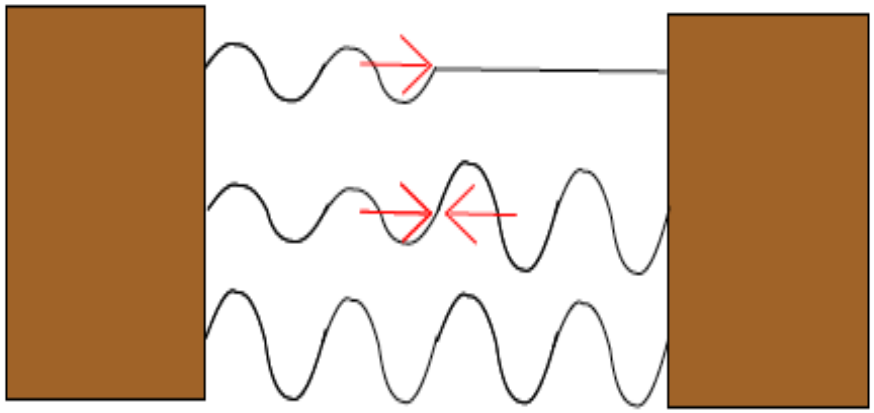
- Consider the trigonometric identity

$$\sin\theta\cos\varphi = \frac{1}{2}\left[\sin(\theta-\varphi)+\sin(\theta+\varphi)\right]$$

- Applied to our previous displacement result…

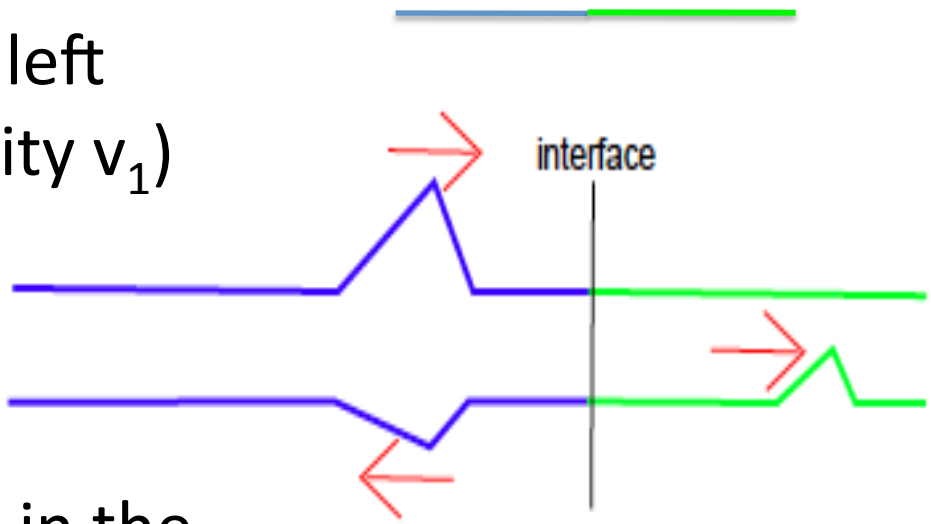
$$y_n(x,t) = A_n \sin\left(K_n x\right)\cos(\omega_n t)$$

$$= \frac{A_n}{2}\left[\underbrace{\sin\left(K_n x-\omega_n t\right)}_{rightward}+\underbrace{\sin\left(K_n x+\omega_n t\right)}_{leftward}\right]$$



An Interface between Different Strings under the Same Tension

- Consider an arbitrary, rightward displacement pulse from the left string (mass density μ_1, velocity v_1) defined as $f_1(t - x/v_1)$
  - A portion of this pulse will <u>reflect</u> at the interface, becoming a leftward wave in the left string, $g(t + x/v_1)$
  - The remainder will transmit into the right string as a rightward wave $f_2(t - x/v_2)$



Interface, cont'd

- The transverse displacements in each string are

$$y_1(x,t) = f_1\left(t - \frac{x}{v_1}\right) + g\left(t + \frac{x}{v_1}\right)$$

$$y_2(x,t) = f_2\left(t - \frac{x}{v_2}\right)$$

- With boundary conditions

$$y_1(0,t) = y_2(0,t) \quad \text{and} \quad \frac{\partial y_1}{\partial x}(0,t) = \frac{\partial y_2}{\partial x}(0,t)$$

Interface Solution

- Solving for f_2 and g_1 in terms of f_1

$$\left.\begin{aligned} f_2(t) &= \frac{2v_2}{v_2+v_1} f_1(t) \\ g(t) &= \frac{v_2-v_1}{v_2+v_1} f_1(t) \end{aligned}\right\} g(t) = \frac{v_2-v_1}{2v_2} f_2(t)$$

- Observations
  - If $v_1=v_2$, then nothing is reflected (all transmitted)
  - If $v_2=0$ (infinite mass), all is reflected

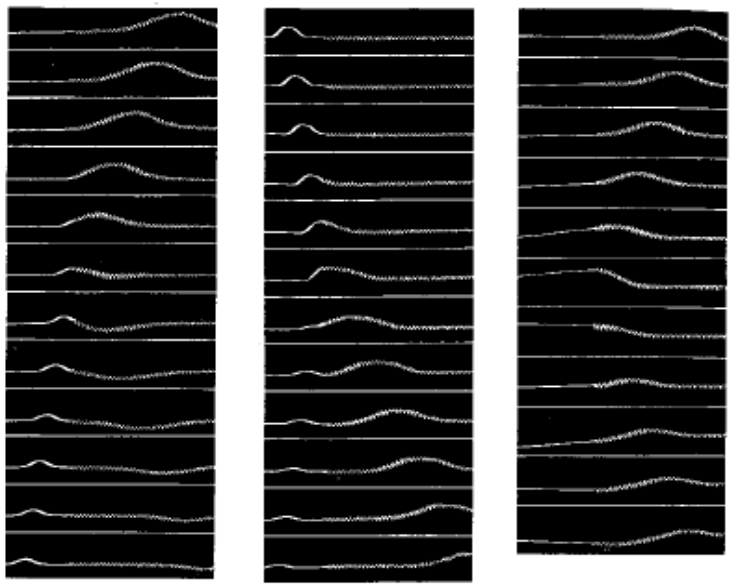


Fig. 8-4 Photographs of pulses encountering the boundary between two media. (a) Pulse passing from a light spring (right) to a heavy spring. At the junction the pulse is partially transmitted and partially reflected. You will note that the reflected pulse is upside down. (b) Pulse passing from a heavy spring (left) to a light spring. At the junction the pulse is partially transmitted and partially reflected. The reflected pulse is right side up. (c) Pulse on a spring reflected from a junction with a very light thread. The whole pulse returns right side up. The blurring of pictures indicates that the particles of the thread are moving at high speed as the pulse passes. Can you determine the direction of this motion in each of the frames? (Photographs from Physical Science Study Committee, Physics, *Heath, Boston, 1965.)*

AP French, *Vibrations and Waves*, 19

Interface Energy Flow

- So far, we've focused on displacements (*y, f, g*) and velocities (*v*), but our prime focus is energy, specifically the <u>rate</u> of energy flow

$$P = \frac{1}{2}\mu y_{\max}^2 v$$

- The ratio of energy reflected at the interface to that incident becomes

$$\frac{P_g}{P_{f1}} = \left(\frac{g}{f_1}\right)^2 = \left(\frac{v_2 - v_1}{v_2 + v_1}\right)^2$$

Interfaces and Transmission

- Most commonly, an acoustic impedance is defined, as

$$Z = \frac{T}{v} = \sqrt{T\mu}$$

- Then, the normal-direction boundary transmittance $\tau_{b,1\text{-}2}$ becomes

$$\tau_{b,1-2} = 1 - \left(\frac{g}{f_1}\right)^2 = 1 - \left(\frac{Z_1 - Z_2}{Z_1 + Z_2}\right)^2 = \frac{4Z_1Z_2}{\left(Z_1 + Z_2\right)^2}$$

  – This is called the Acoustic Mismatch Model (AMM) (Little, W. A., 1959, Can. J. Phys. 37, 334)

The Green's Function

See Greenberg, *Foundations of Applied Mathematics*, Prentice-Hall, 1978.

- Again consider the generic ODE of the form

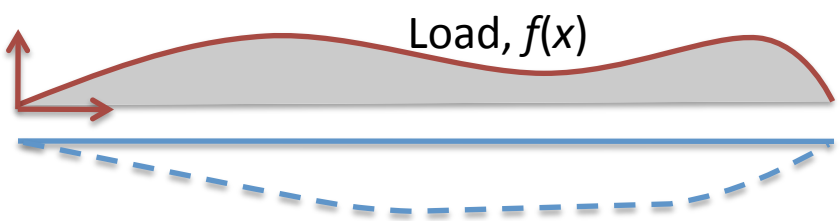
$$u'' + k^2 u = f(x)$$

  - Where the prime (') denotes a derivative
- With boundary conditions (u is displacement)

$$u(x = 0) = a$$

$$u(x = l) = b$$

- Example: a loaded string ($k = a = b = 0$, u is displacement)



Note: in this case, f is the downward force per unit length per force of string tension

The Inner Product Solution

- Definition of an inner product

$$(f,g) \equiv \int_0^l f(x)g(x)dx$$

- Linear operator, *L*

$$Lu = u'' + k^2 u$$

- Seek function *G* to be used in

$$(Lu,G) = \text{boundary terms} + (u,L^*G)$$

  - Where L^* is the <u>adjoint</u> of the operator *L*
  - And the equality arises from integration by parts

$$\int_a^b u\,dv = uv\Big|_a^b - \int_a^b v\,du$$

The Key Step

- If we can somehow 'contrive' a way for (u,$L*G$) to give us the function u, then we will have solved the problem because (Lu,G) is also ($f(x)$,G) from the original ODE [$Lu=f(x)$]
- The contrivance is to make $L*G$ a Dirac delta function, $\delta(\xi-x)$, where ξ is the variable of integration

$$(u, L*G) = \int_0^l u(\xi)\delta(\xi - x)\,d\xi = u(x)$$

- Resulting in…

Differentiation w.r.t. ξ

$$(Lu,G) = \int_0^l G(\xi,x)f(\xi)\,d\xi = G(l,x)u'(l) - bG_\xi(l,x) - G(0,x)u'(0) + aG_\xi(0,x) + \boxed{u(x)}$$

Note: the Green's function G depends on both ξ (the internal variable of integration) and x, which is the real-space variable upon which $f(x)$ and $u(x)$ depend

Cleaning Up…

- The (general) <u>adjoint</u> problem for *G* is

$$L^* G = G_{\xi\xi} + k^2 G = \delta(\xi - x)$$

  - i.e., same as the *Lu=f* problem, but with a delta function source term
  - <u>Big idea</u>: this change generalized the problem--we can calculate a *G* for a given operator *L*, and then use integrals of *G•f* for <u>any</u> problem
- Some extraneous boundary terms still remain
  - Choose *G* such that *G*(0,*x*) = *G*(*l*,*x*) = 0

$$(Lu,G) = \int_0^l G(\xi,x) f(\xi)\, d\xi = \underbrace{G(l,x)}_{0} u'(l) - bG_\xi(l,x) - \underbrace{G(0,x)}_{0} u'(0) + aG_\xi(0,x) + u(x)$$

Solution

- Need to solve the adjoint problem to find *G*, then substitute into equation for *u*(*x*)
- For $x \neq \xi$ the solution is simple ($G'' + k^2 G = 0$)

$$G(\xi, x) = \begin{cases} A \sin k\xi + B \cos k\xi & 0 \leq \xi < x \\ C \sin k\xi + D \cos k\xi & x \leq \xi < l \end{cases}$$

- And with the BCs on *G*,

$$G(\xi, x) = \begin{cases} A \sin k\xi & 0 \leq \xi < x \\ E \sin k(\xi - l) & x \leq \xi < l \end{cases}$$

Solution, cont'd

- Must address the 'jump' across the delta fn

$$\int_{x-}^{x+} (G_{\xi\xi} + k^2 G)\, d\xi = \int_{x-}^{x+} \delta(\xi - x)\, d\xi \equiv 1$$

- Leads to

$$G\Big|_{x-}^{x+} = 0 \quad \text{(continuity)}$$

$$G_{\xi}\Big|_{x-}^{x+} = 1 \quad \text{(jump)}$$

- Substitute to find *A* and *E*

$$G(\xi, x) = \begin{cases} \dfrac{\sin k(x-l)}{k \sin kl} \sin k\xi & 0 \le \xi < x \\ \dfrac{\sin kx}{k \sin kl} \sin k(\xi - l) & x \le \xi < l \end{cases}$$

Final Result

- Substitute *G* and *f* into original integral and solve for *u*(*x*), which is the solution of our original problem
  - *Lu(x) = f(x)*
  - with boundary conditions *u*(0)=*a and u*(l)=*b*

$$u(x) = \int_0^l G(\xi,x) f(\xi) d\xi + bG_\xi(l,x) - aG_\xi(0,x)$$

Connecting Continuous with Discrete Waves

Continuous

$$\tau_{b,1-2} = 1 - \left(\frac{g}{f_1}\right)^2 = \left(\frac{f_2}{f_1}\right)^2$$

- Energy transmissivity proportional to (displacement)^2

Discrete

- Green's functions are proportional to displacement
- Then the transmission function in trace form is proportional to (displacement)^2

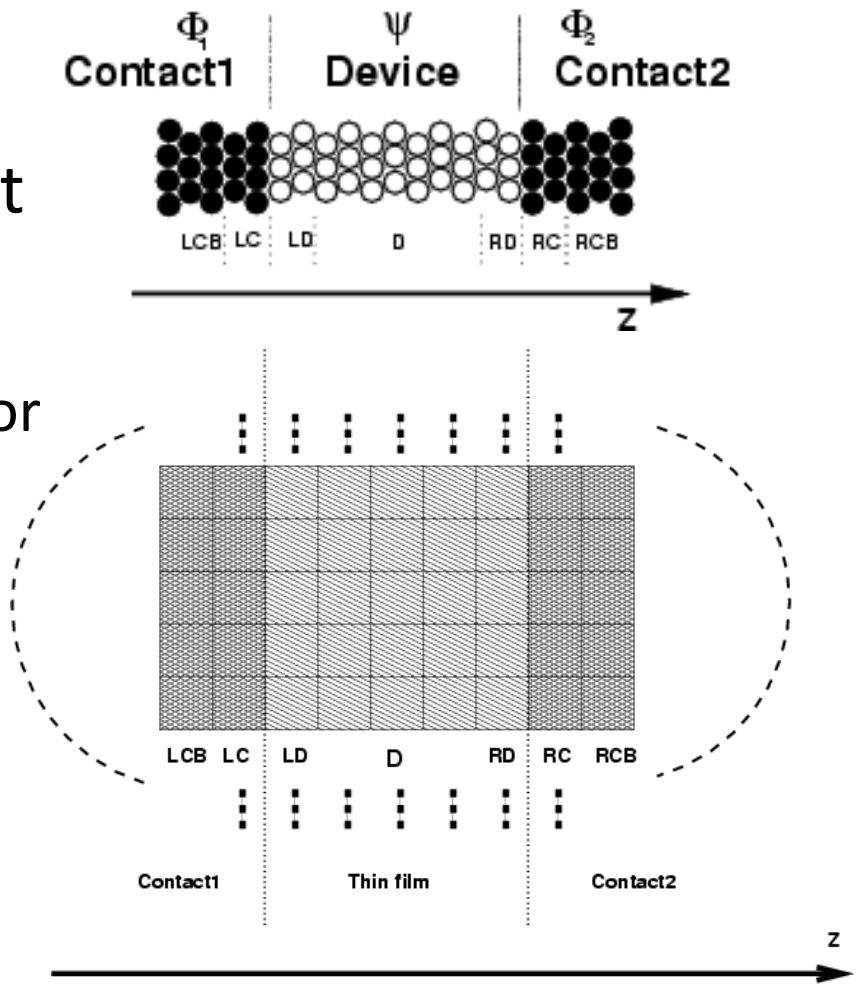
$$\Xi(\omega) = Trace\left[\Gamma_1 \mathbf{G} \Gamma_2 \mathbf{G}^T\right]$$

$$= Trace\left[\Gamma_2 \mathbf{G} \Gamma_1 \mathbf{G}^T\right]$$

Atomistic Green's Functions

Transmission Function Ξ for Phonons

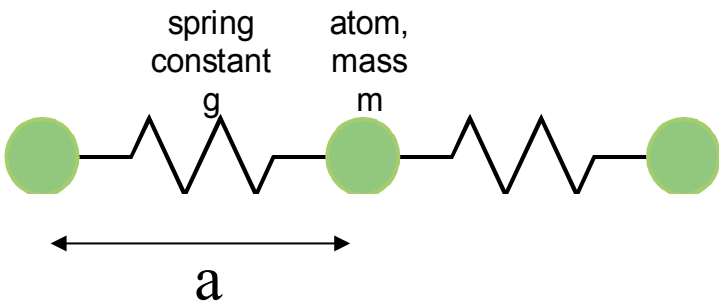
- Includes effects of bulk contacts through self-energy matrices
- Suitable for ballistic transport
  - Nanoscale devices at room temperature, or
  - Low-temperature conditions, or
  - Scattering dominated by boundaries and interfaces
- Required inputs
  - Equilibrium atomic positions
  - Inter-atomic potentials
  - Contact temperatures



Lattice Dynamics

- Equation of motion for a 1D atomic chain

$$m\frac{d^2x_n}{dt^2} = -g\left\{2x_n - x_{n-1} - x_{n+1}\right\}$$



- Plane wave assumption

$$x_n(t) \sim \exp\left\{i\left(Kna - \omega t\right)\right\}$$

- Combine

$$-\omega^2 x_n = -\frac{g}{m}\left\{2x_n - x_{n-1} - x_{n+1}\right\}$$

- Re-arrange and write in matrix form

$$\left[\omega^2\mathbf{I} - \mathbf{H}\right]\mathbf{x} = 0$$

$\mathbf{I}$ is the identity matrix

Harmonic (Force-Constants) Matrix

$$\mathbf{H} = \begin{bmatrix} -2f & f & \\ f & -2f & f \\ & f & -2f \end{bmatrix}$$

f is spring constant divided by atomic mass

1. **H** is not the same dynamical matrix used to determine dispersion curve (that matrix is the Fourier transform of **H**).
2. **H** is symmetric.
3. Sum of all elements in any row or sum of all elements in any column is zero, except in the first and last row/column.

Green's Functions

- In general, systems of equations can be written in operator form

$$\mathbf{L}[\mathbf{x}] = \left[\omega^2 \mathbf{I} - \mathbf{H}\right]\mathbf{x} = 0$$

- *Green's functions* are often used in such situations to determine general solutions of (usually) linear operators

Green's Functions, cont'd

- The Green's function **g** is the solution that results from the addition of a perturbation to the problem

$$\mathbf{L}[\mathbf{g}] = \delta$$

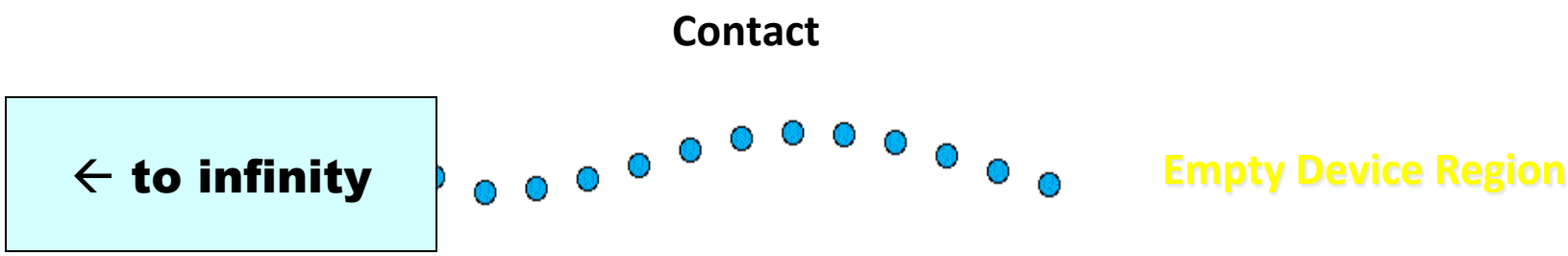
- In the present (matrix) problem, the *unperturbed Green's function* becomes

$$\mathbf{g} = \left[\left(\omega^2 + \delta i\right)\mathbf{I} - \mathbf{H}\right]^{-1}$$

  – Where δ is called the broadening constant, and i is the unitary imaginary number

Uncoupled Contacts

- In our context, Green's functions first appear in the two contact regions
- Imagine first that each contact is unconnected to the 'device' but extends to infinity in the other direction



Uncoupled Contact Green's Functions

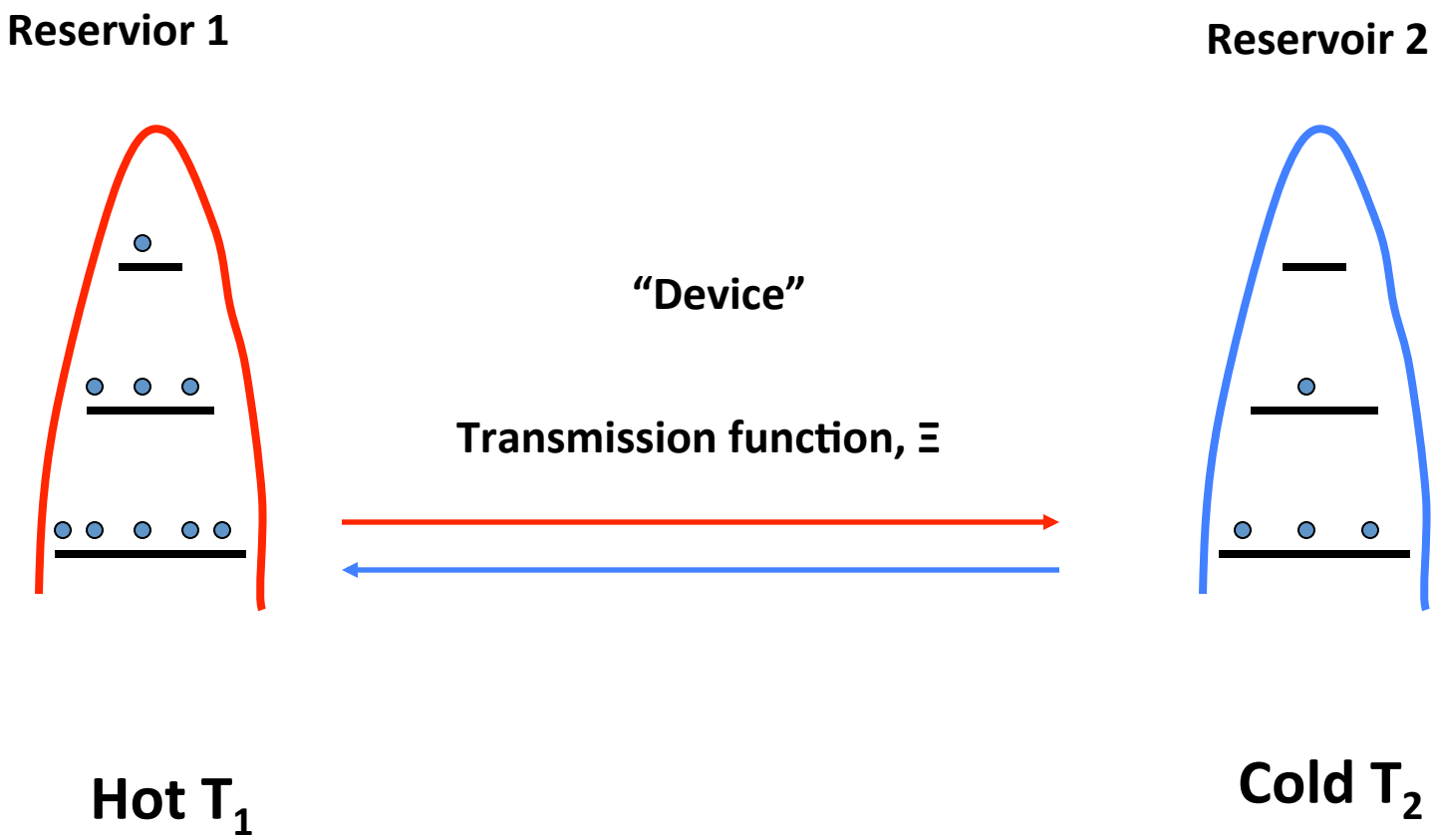
- Now, we form the uncoupled Green's functions for each contact ($\mathbf{g_1}$ and $\mathbf{g_2}$)

$$\mathbf{g}_1 = \lim_{\delta \to 0} [(\omega^2 + \delta i)\mathbf{I} - \mathbf{H}_1]^{-1}$$

$$\mathbf{g}_2 = \lim_{\delta \to 0} [(\omega^2 + \delta i)\mathbf{I} - \mathbf{H}_2]^{-1}$$

- Where the $\mathbf{H_i}$'s are the harmonic matrices for each contact

Phonon Transport through a "Device" between Two Contacts



Assembling the System

- We now need to *couple* the two contacts with the device
- The overall matrix equation becomes

Harmonic matrix for contact 1

Harmonic matrix for the device

Uncoupled displacement of contact 1

Effects of device on contact displacements

$$\begin{bmatrix} \omega^2\mathbf{I} - \mathbf{H}_1 & -\tau_1^\dagger & 0 \\ -\tau_1 & \omega^2\mathbf{I} - \mathbf{H}_d & -\tau_2 \\ 0 & -\tau_2^\dagger & \omega^2\mathbf{I} - \mathbf{H}_2 \end{bmatrix} \begin{Bmatrix} \Phi_1^R + \chi_1 \\ \psi \\ \Phi_2^R + \chi_2 \end{Bmatrix} = 0$$

Device displacement

Harmonic matrix for contact 2

Uncoupled displacement of contact 2

Device Green's Function

- We can model connections between different materials through the use of a different Green's function **G**

$$\mathbf{G} = \left[\omega^2 \mathbf{I} - \mathbf{H_d} - \underbrace{\boldsymbol{\tau}_1 \mathbf{g_1} \boldsymbol{\tau}_1^T}_{\Sigma_1} - \underbrace{\boldsymbol{\tau}_2 \mathbf{g}_2 \boldsymbol{\tau}_2^T}_{\Sigma_2} \right]^{-1}$$

superscript "*T*" = conjugate transpose

  - This matrix function includes self-energy matrices ($\boldsymbol{\Sigma}_1$, $\boldsymbol{\Sigma}_2$) that involve unperturbed Green's functions (**g**'s) associated with contacts (i.e., boundaries) in a transport problem
  - τ matrices handle connections between different system elements (materials, interfaces)
  - The full derivation is beyond the scope of this presentation, so we will simply use the results for computational purposes
- Very efficient in the ballistic regime but requires significant effort to implement 'internal' scattering

Relation between Transmission and Green's Functions

- Some definitions of convenience

$$\mathbf{A}_j = i\left[\mathbf{g}_j - \mathbf{g}_j^{\,T}\right]$$

$$\mathbf{\Gamma}_j = \boldsymbol{\tau}_j \mathbf{A}_j \boldsymbol{\tau}_j^{\,T}$$

Phonon escape rate

- The transmission function

$$\Xi(\omega) = Trace\left[\mathbf{\Gamma}_1 \mathbf{G} \mathbf{\Gamma}_2 \mathbf{G}^T\right] = Trace\left[\mathbf{\Gamma}_2 \mathbf{G} \mathbf{\Gamma}_1 \mathbf{G}^T\right]$$

From $\mathbf{g}_1$ and τ_1

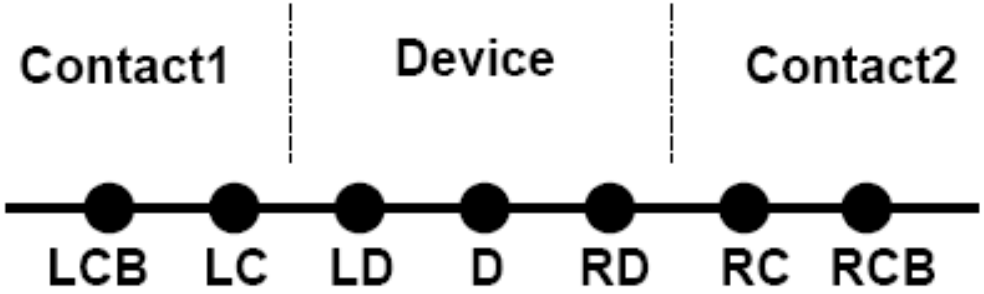
Device Green's function

From $\mathbf{g}_2$ and τ_2

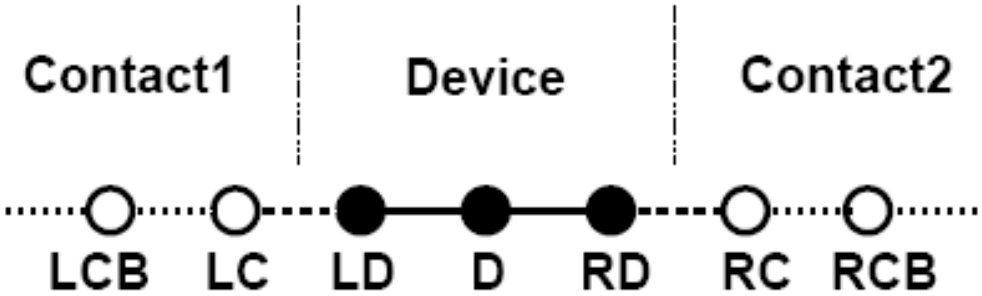
1D Atomic Chain

Student: Wei Zhang
Collaborator: N. Mingo

- Can be visualized as an atomic chain between two isothermal contacts (Note: contacts are still atomic chains in this example)



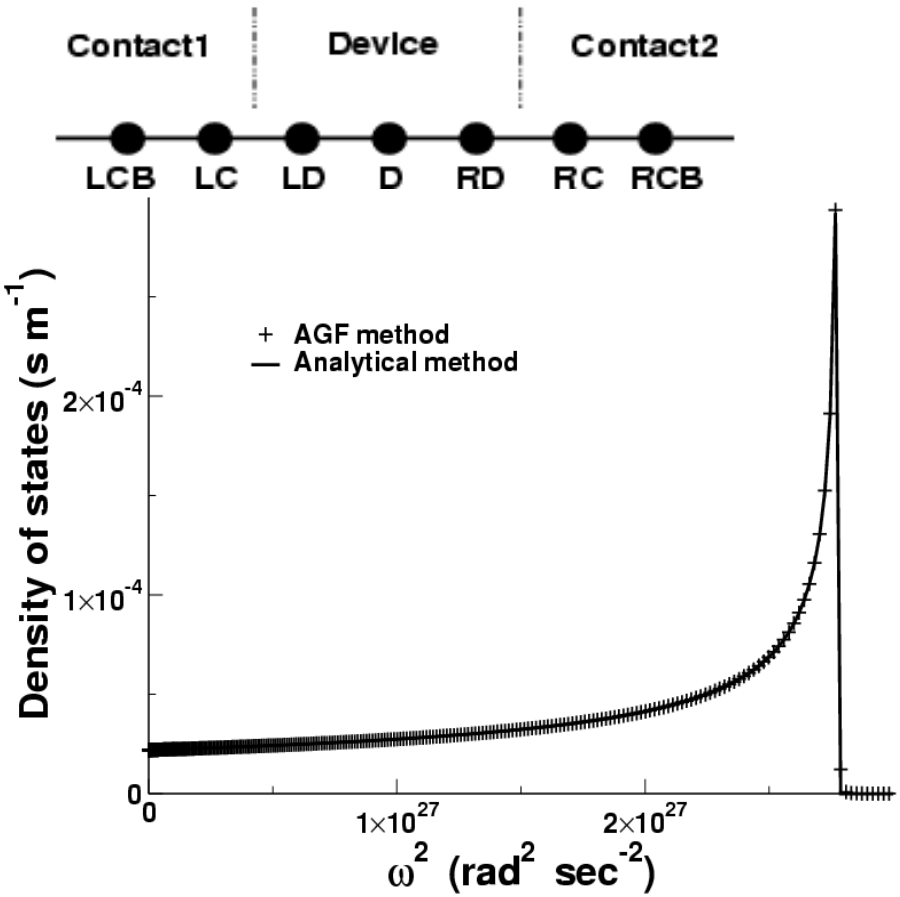
(a) Schematic diagram of a homogeneous chain



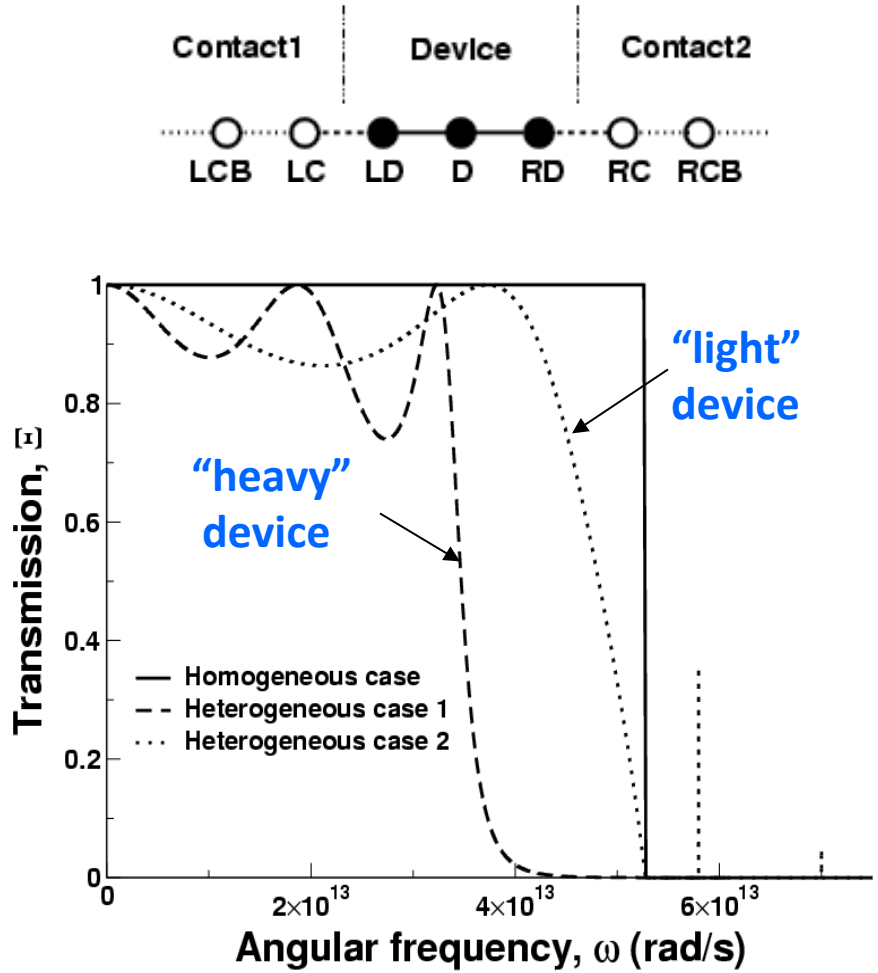
(b) Schematic diagram of a heterogeneous chain.

See Zhang et al. Num Heat Trans B 51 333-349(2007)

Results for Simple Atomic Chains



Homogeneous chain density of states



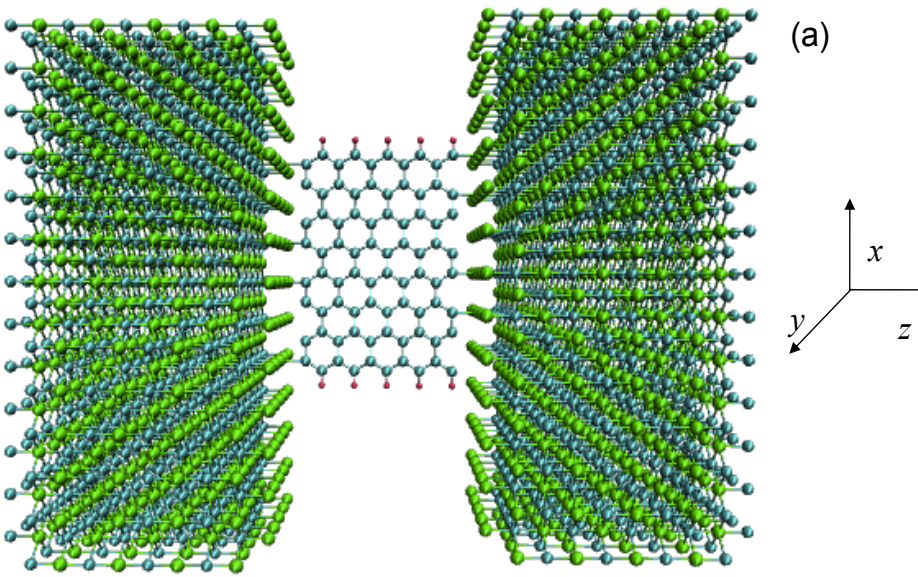
Homogeneous vs heterogeneous

Some Examples

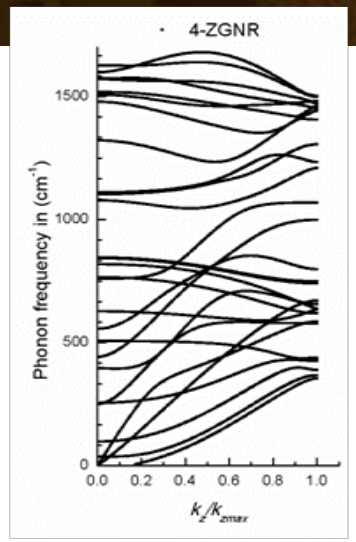
3D-2D Interface Modeling

Student: Zhen Huang
Faculty collaborator: J. Murthy

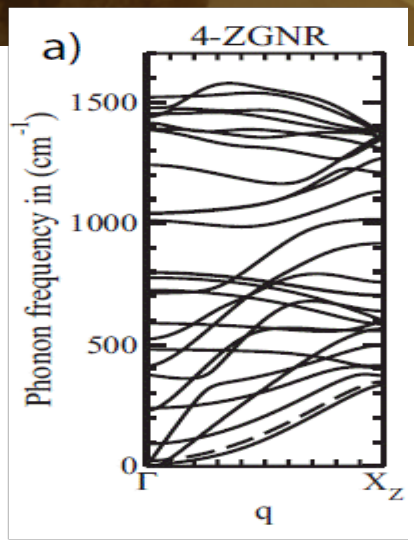
- TiC is a compound metal with two face centered cubic cells offset by a half-body diagonal
- Graphene nanoribbon (GNR) armchair edge is strained to match the lattice constants of TiC
- GNR edges are hydrogen-passivated
- Ti-face of TiC is assumed to be covalently bonded to GNR
- H atoms do not interact with TiC contacts
- Inter-layer interaction is ignored
- Force constants between Ti and C still valid at interface



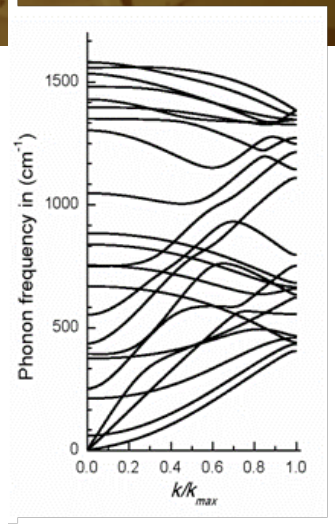
Thermal Conductance of GNRs



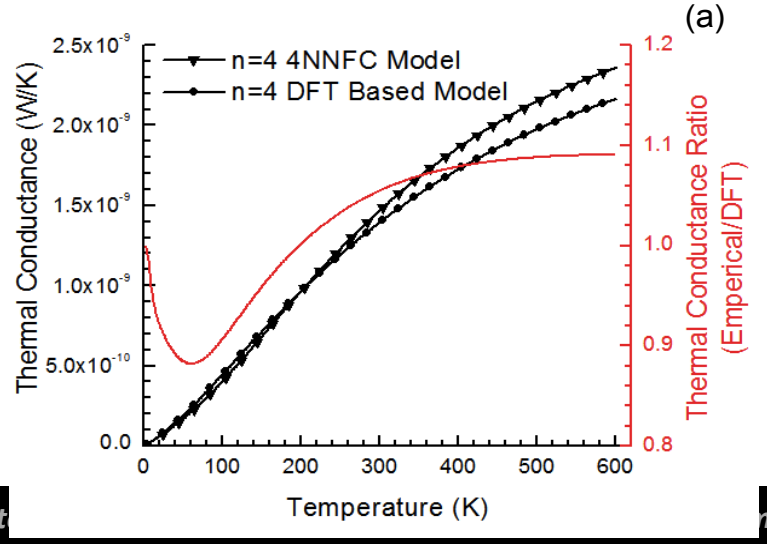
Phonon dispersion curve based on optimized IFCs

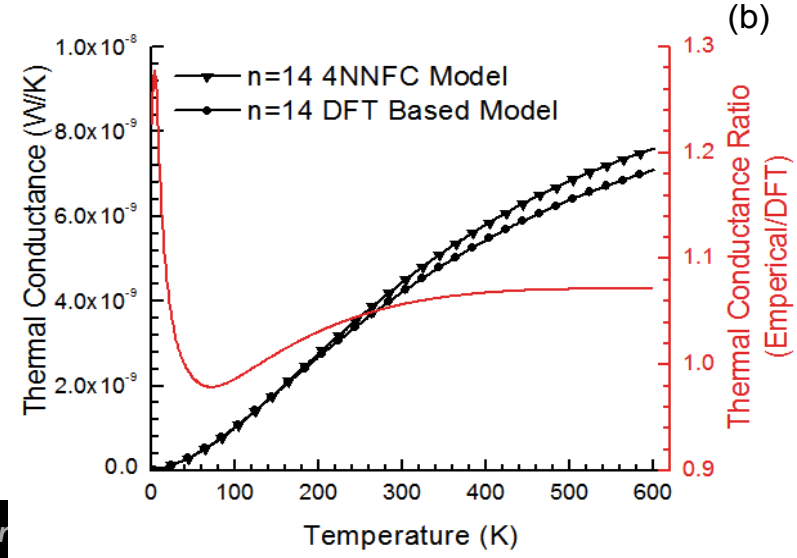


Gillen *et al.* Phys. Status Solidi B **246** 2009 DFT results

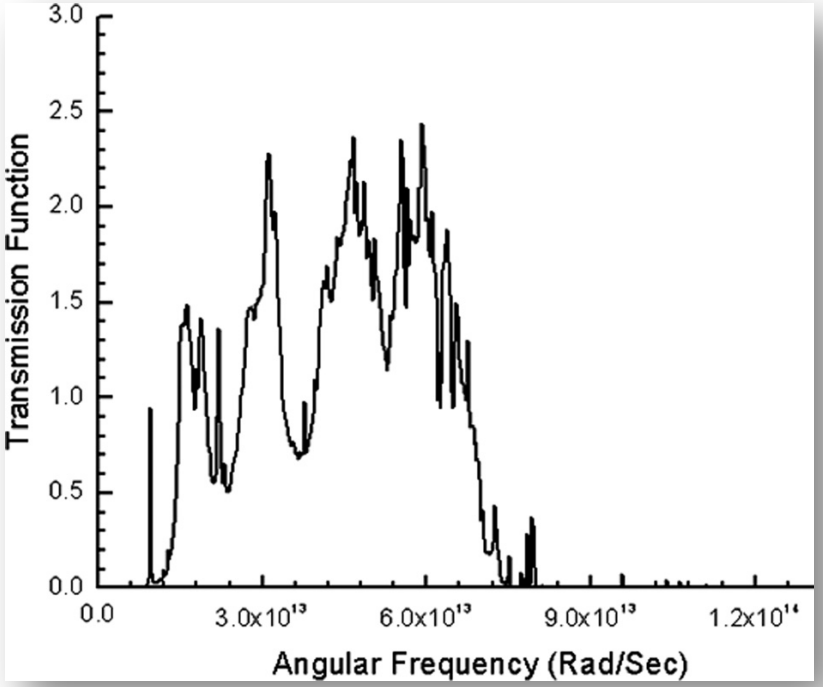


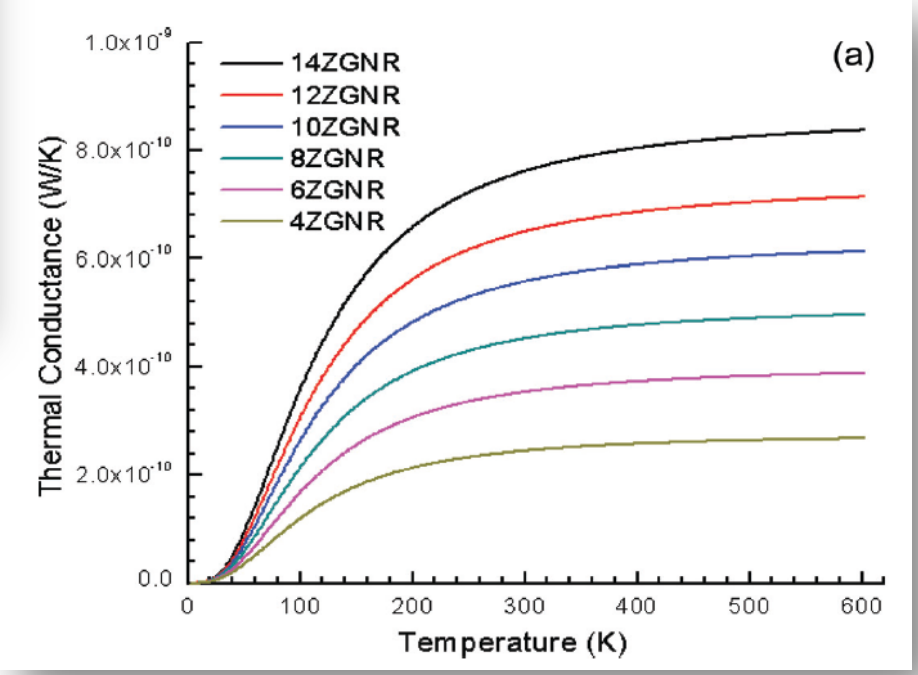
Empirical 4NNFC model result





Spectral and Total Transmission for TiC-GNR Interface Structure

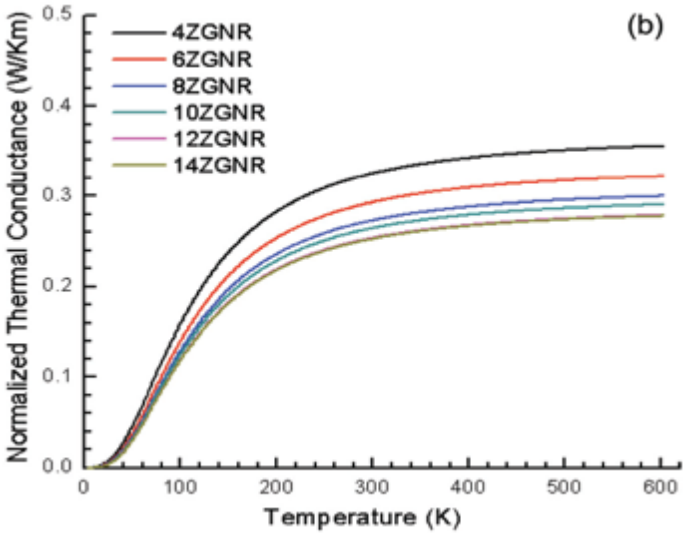


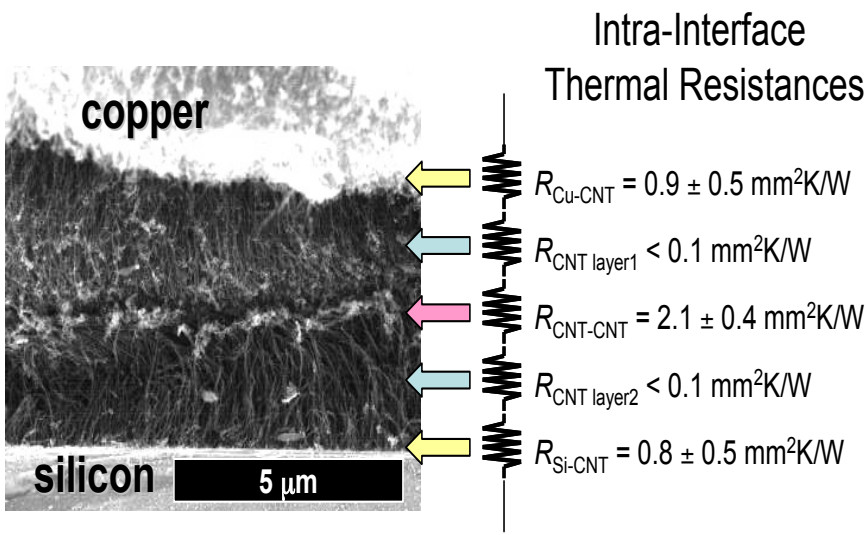


Z. Huang, T.S. Fisher, J.Y. Murthy, J Appl Phys, **109**, 074305, 2011.

Thermal Conductance of TiC-GNR

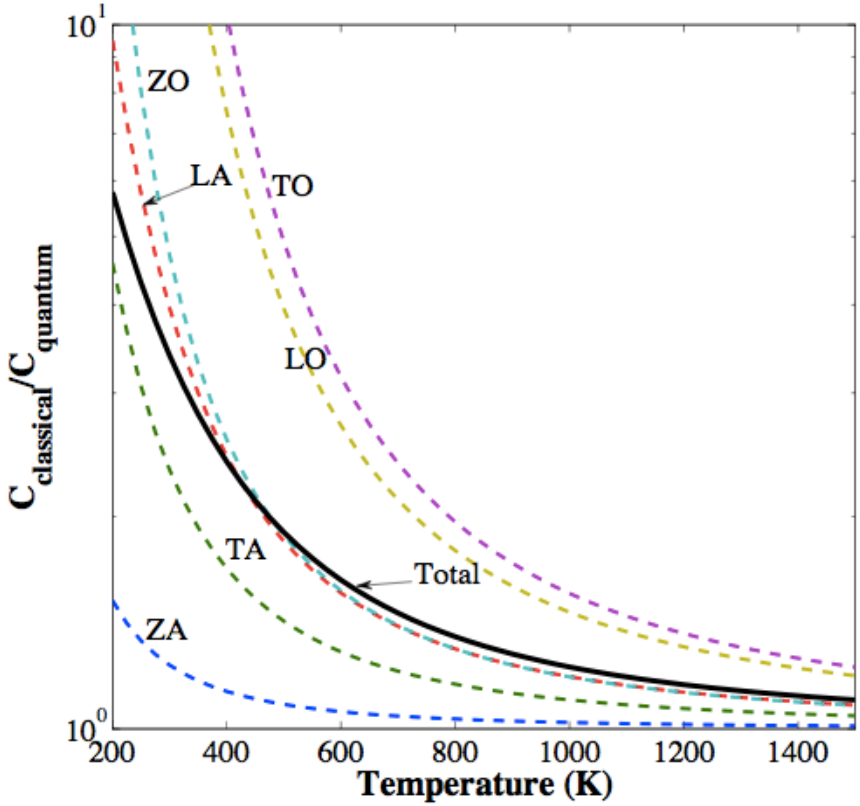
- GNR width-normalized conductance converges at 0.25 W/mK
- Average diameter of MWCNTS is 30 nm (Xu and Fisher, Int. J. H&M Trans 2006), with a circumference ~100 nm
- Density of arrays ~10^8 / mm^2
- Estimated thermal resistance for such array is 0.4 mm^2K/W
- **The interface alone produces a thermal resistance equivalent to a CNT length 1000 times its diameter**

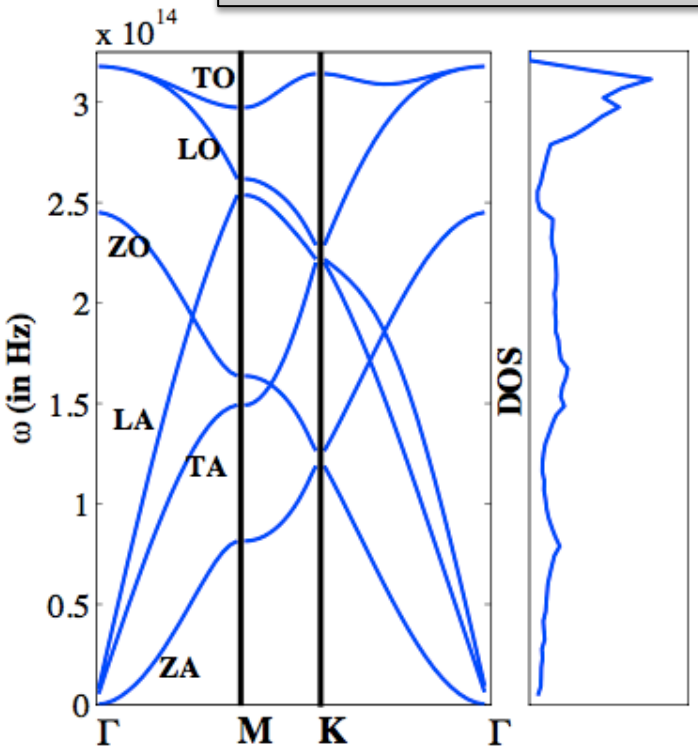




Graphene BTE: Classical vs. Quantum Specific Heat

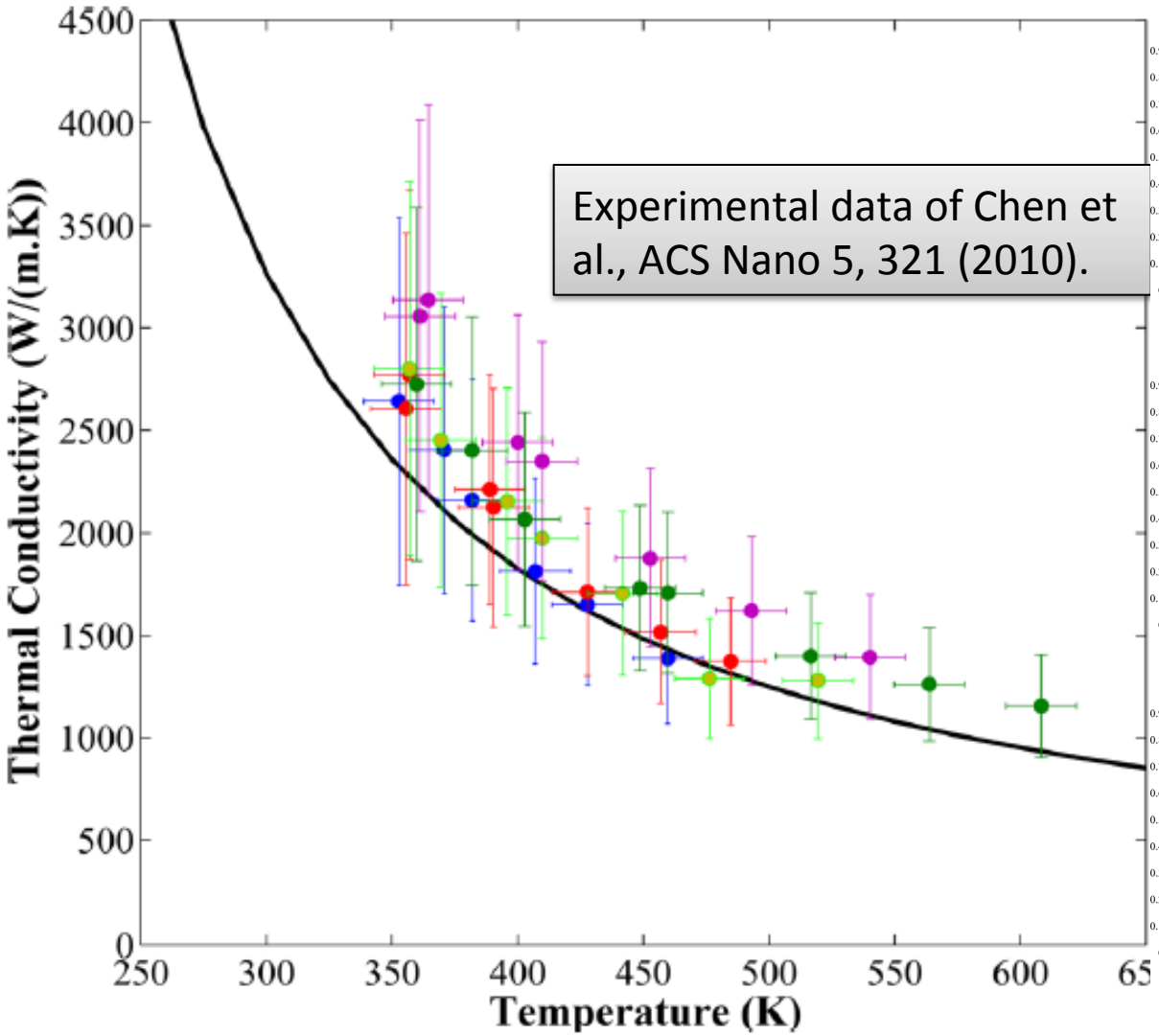
Student: Dhruv Singh
Faculty collaborator: J. Murthy



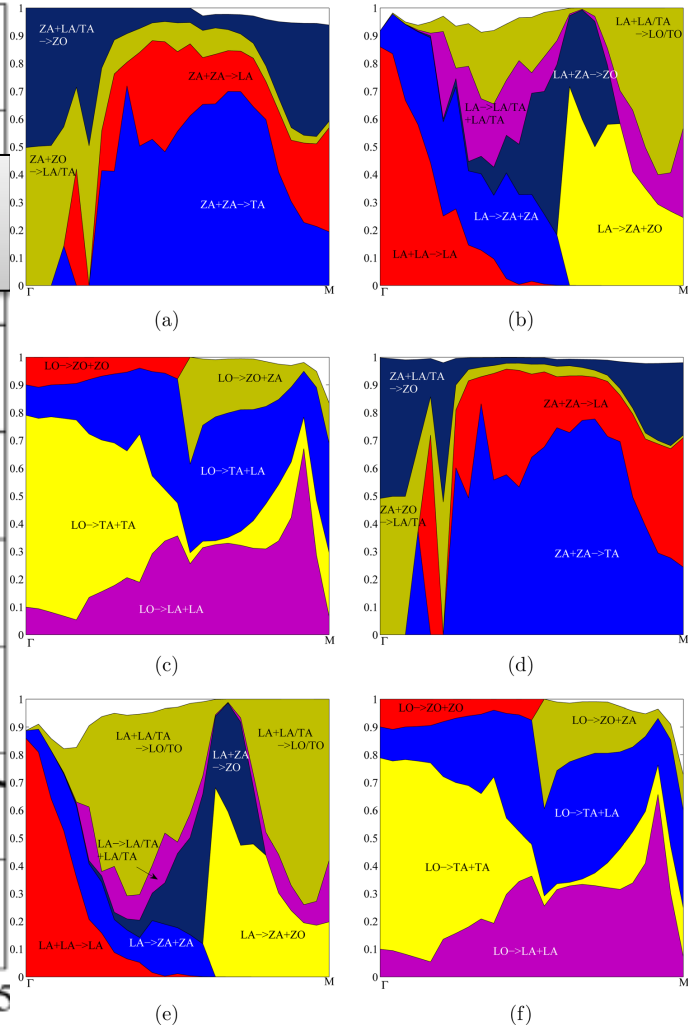


- Significant over-prediction of phonon specific heat at 300 K (~ 5 times)
- A satisfactory estimate is not obtained until 1000 K
- Not a bad approximation for ZA phonons (low frequencies)

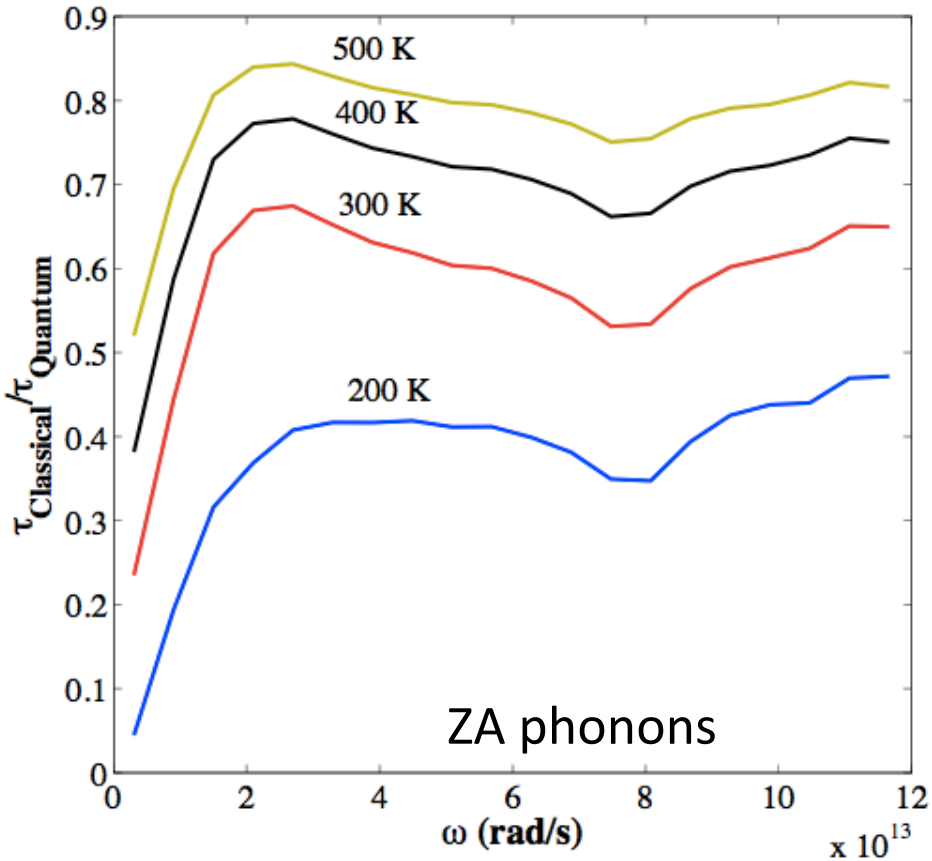
Full BTE Simulation of Graphene k

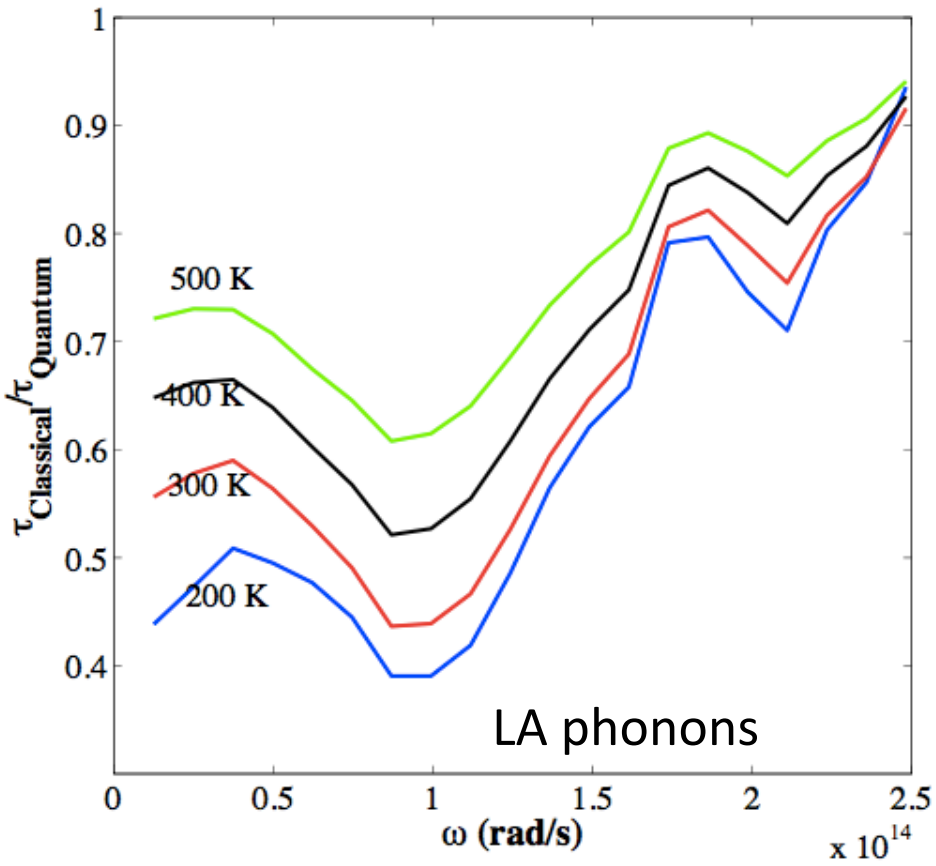


Experimental data of Chen et al., ACS Nano 5, 321 (2010).



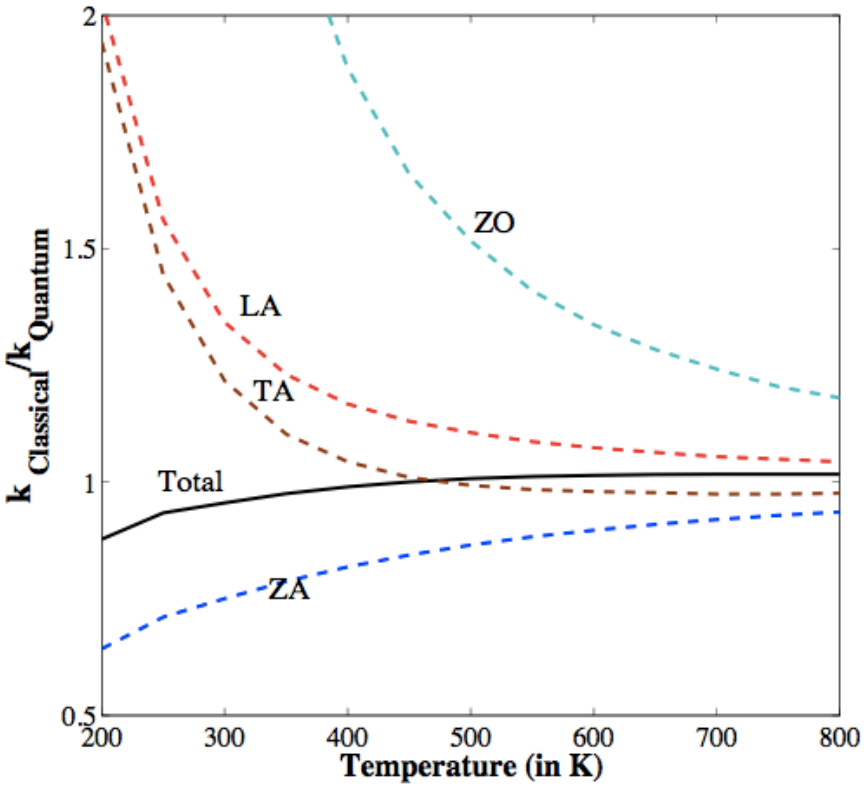
Classical vs. Quantum Relaxation Time





- Phonon relaxation times are underpredicted (classical approximation to n_0 > Bose-Einstein value)
- At 300 K, the relaxation times using the classical approximation are smaller by a factor of 2
- Works for high frequencies (as they interact with low frequency phonons)

Classical vs. Quantum Thermal Conductivity



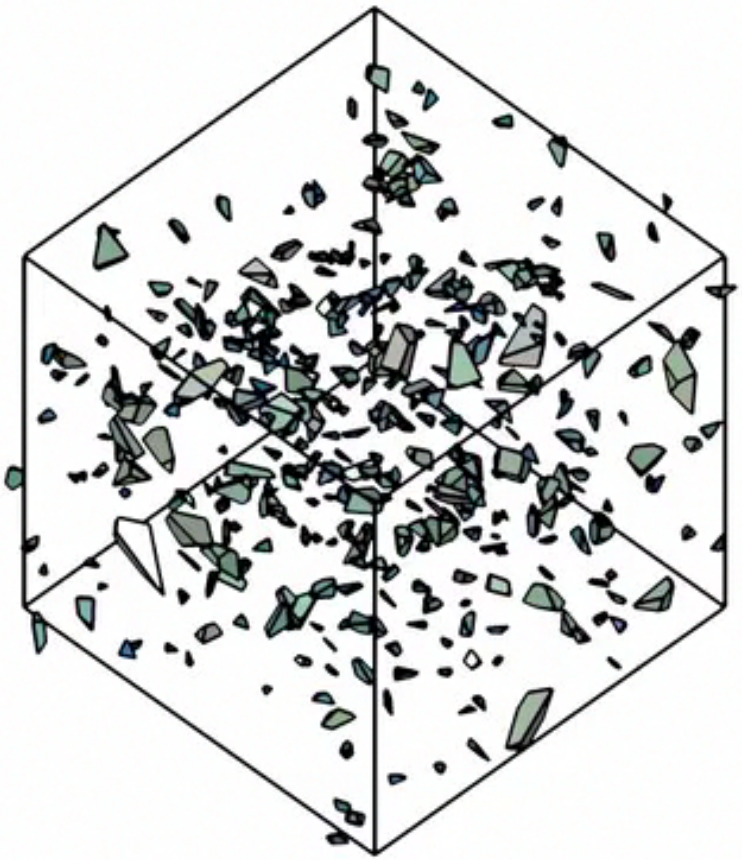
D. Singh, J.Y. Murthy, T.S. Fisher, *J Appl Phys*, **110** 113510, 2011.

- Classical theory ascribes lesser contribution to ZA phonons and much higher values to ZO phonons
- Quantum corrections necessary to MD simulations
- Specific heat overestimated, relaxation time underestimated: net thermal conductivity error is reduced by coincidence
- Modewise values are incorrect: order of magnitude larger for optical phonons at 300 K

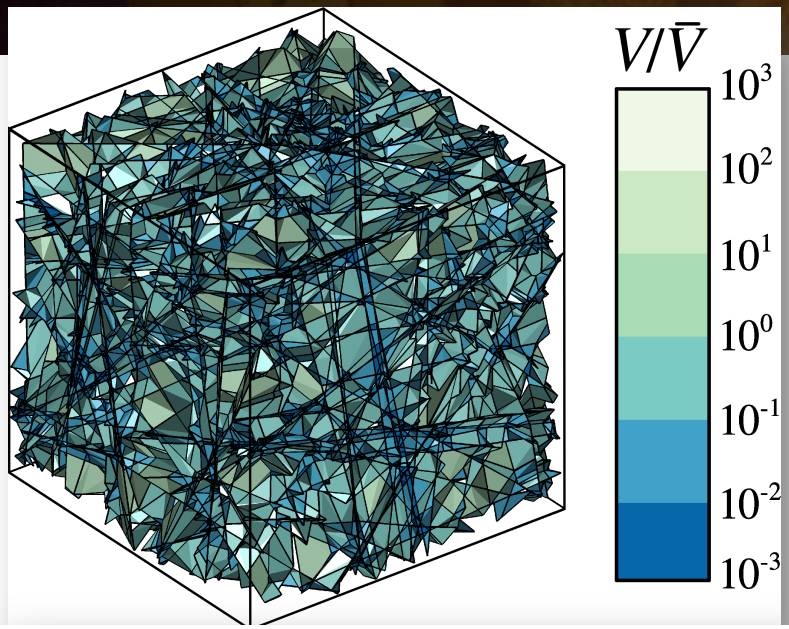
Heat Transfer in Heterogeneous Media

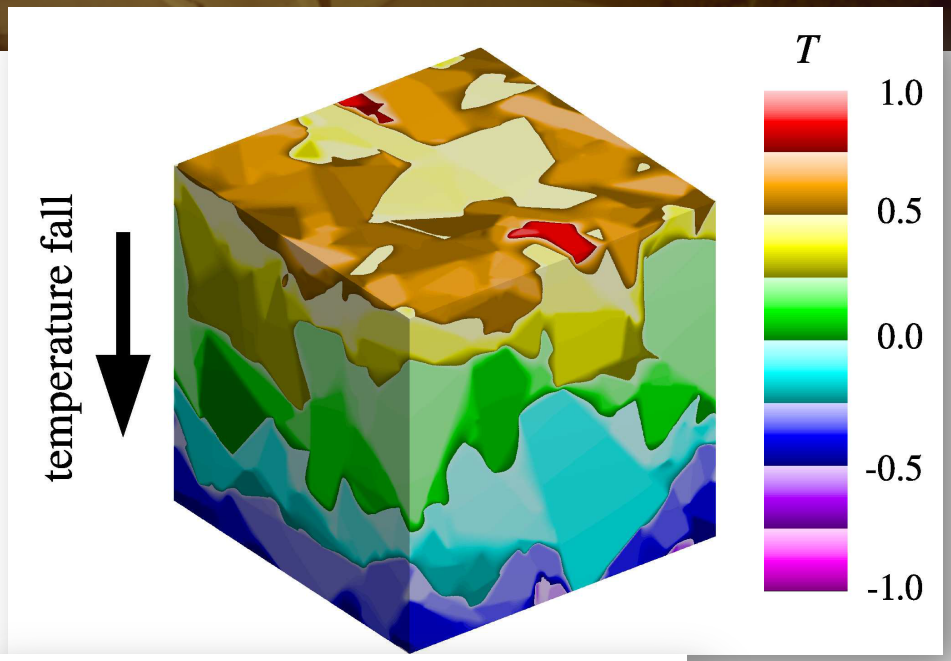
Student: Kyle Smith

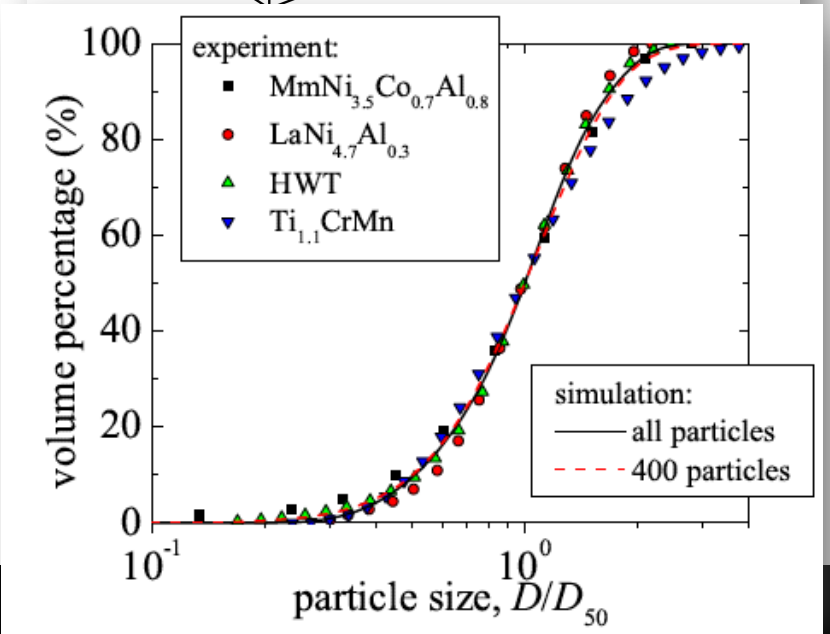
- Many emerging thermal/ power applications of granular materials
  - Batteries
  - Thermoelectrics
  - Metal hydrides (thermal and fuel storage)
  - Thermal/electrical interfaces

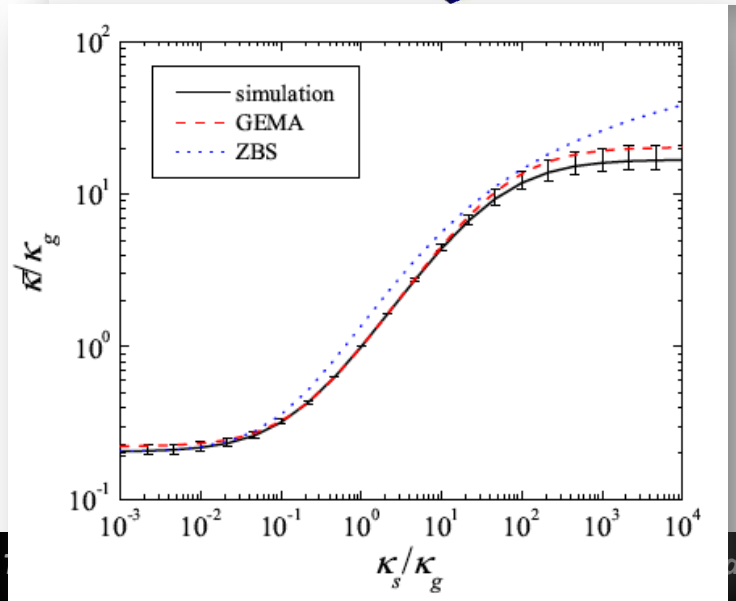


Particle Assembly and Conduction



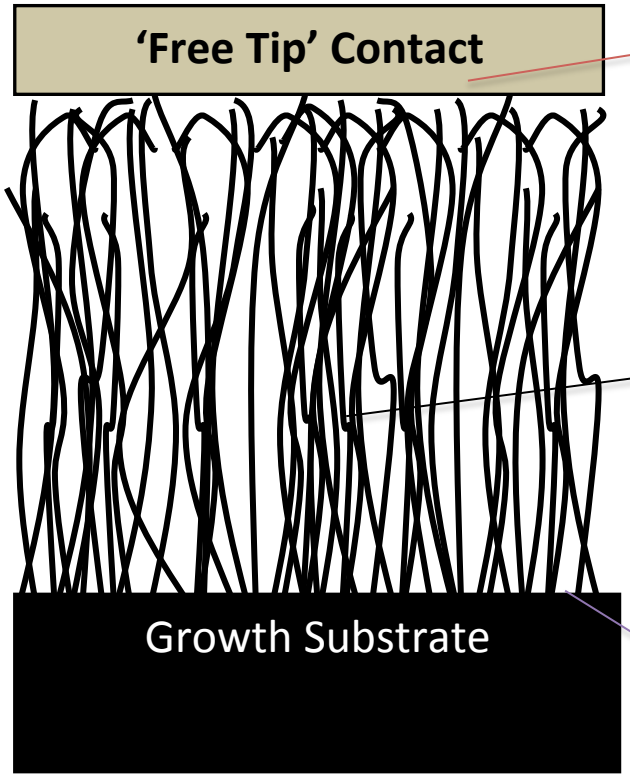






Smith, and Fisher, in review, *Int J Hyd En.*

Conclusion: Many Questions Remain for Real Interfaces

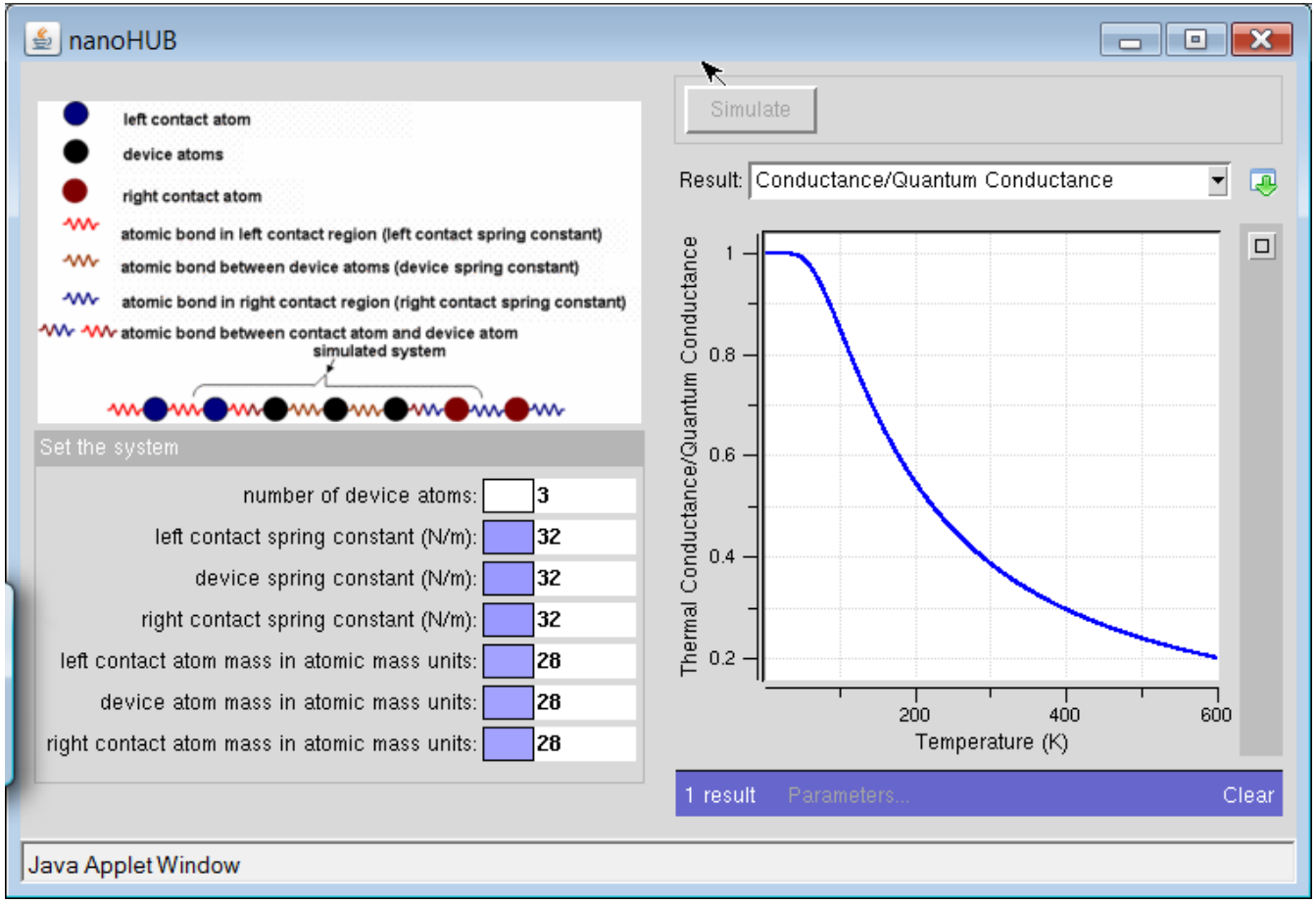


- Tip configurations at contact?
- Effects of clumping?
- Metalized tips?
- Chemical functionalization?

- Effective conductivity of CNT mat?
  - "Ultralow" conductivity?
- Effect of alignment?
- Effect of fillers. Wax? Phase change material?

- Covalent bonding at the growth base?
- Effect of synthesis technique? Thermal or plasma CVD? Post-treatment?
- Addition of 'sticky' metals (e.g., Ti)?
- Graphene-metal as a model?

nanoHUB Tool: Atomistic Green's Function 1D Atomic Chain Simulation



https://www.nanohub.org/tools/greentherm

Thank You
S4800 5.0kV x2.00k SE(M,LA0)
20.0um