Understanding Phonon Dynamics via 1D Atomic Chains

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Nanotechnology 501 Lecture Series

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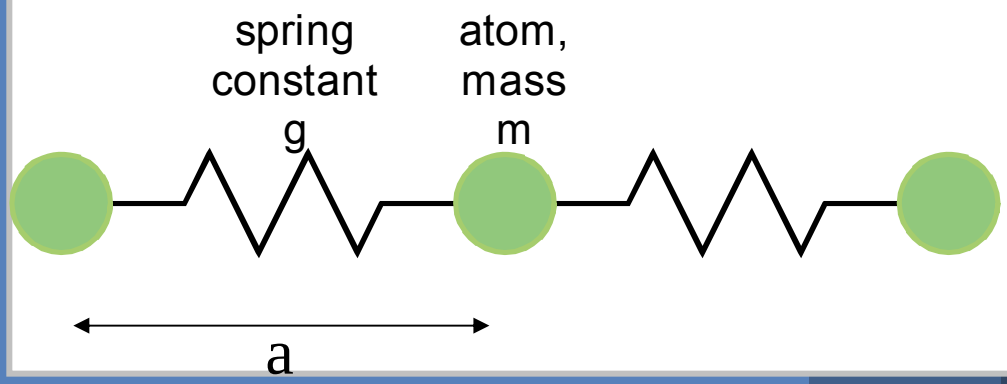
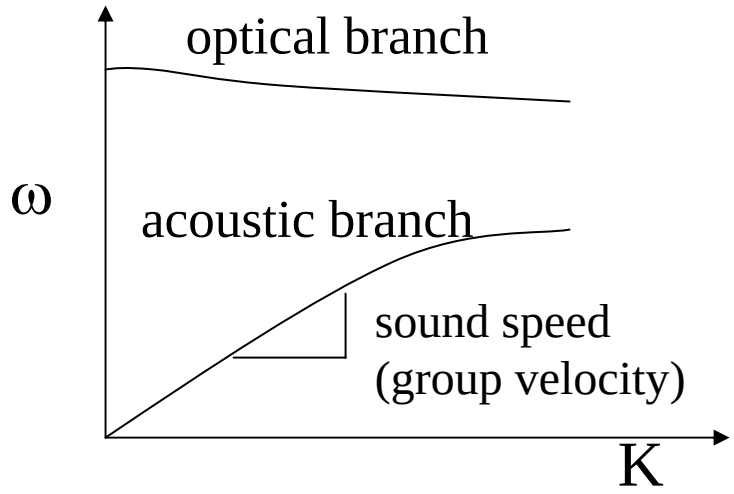
Phonon Heat Conduction

- Phonons are quantized lattice vibrations
- Govern thermal properties in electrical insulators and semiconductors
- Can be modeled to first order with spring-mass dynamics

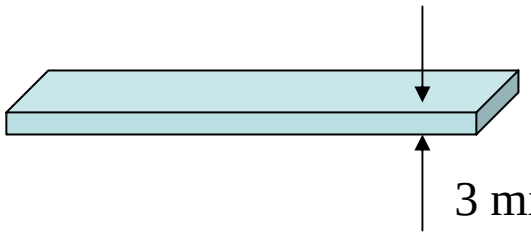


- Wave solutions
  - wave vector $K=2\pi/\lambda$
  - phonon energy=$\hbar\omega$
  - dispersion relations gives ω = fn(K)

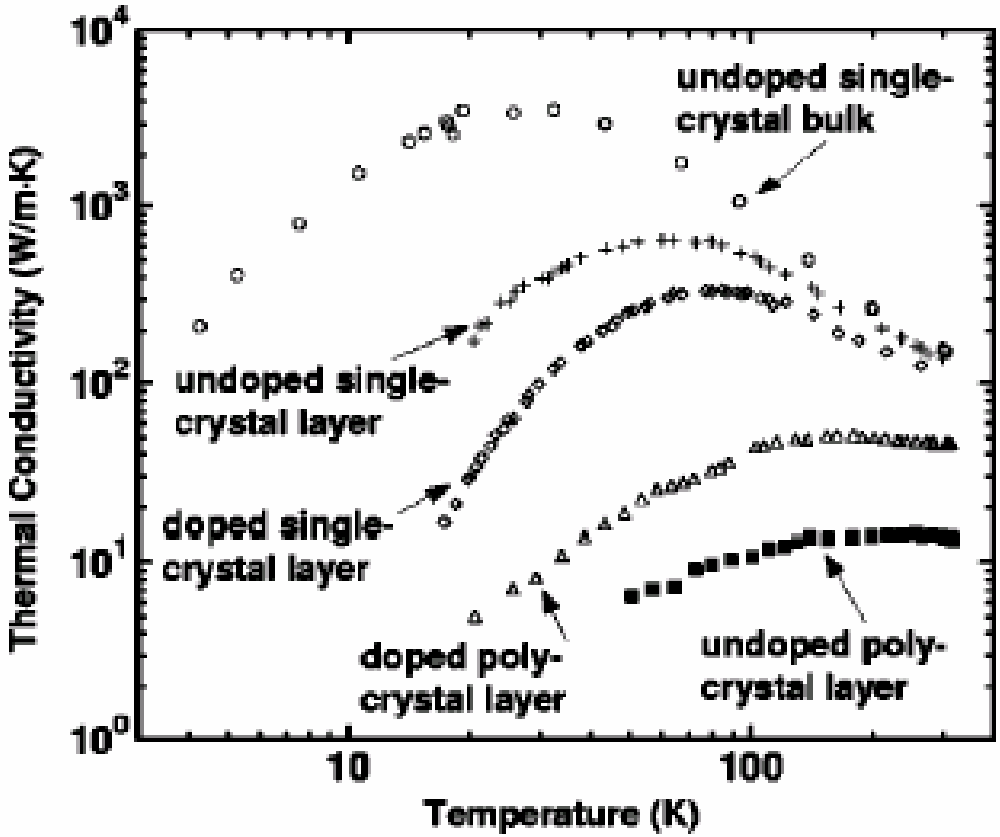


Heat Conduction Through Thin Films

- Experimental results for 3-micron silicon films



- Non-equilibrium scattering models work fairly well
- Crystalline structure has generally larger impact than film thickness

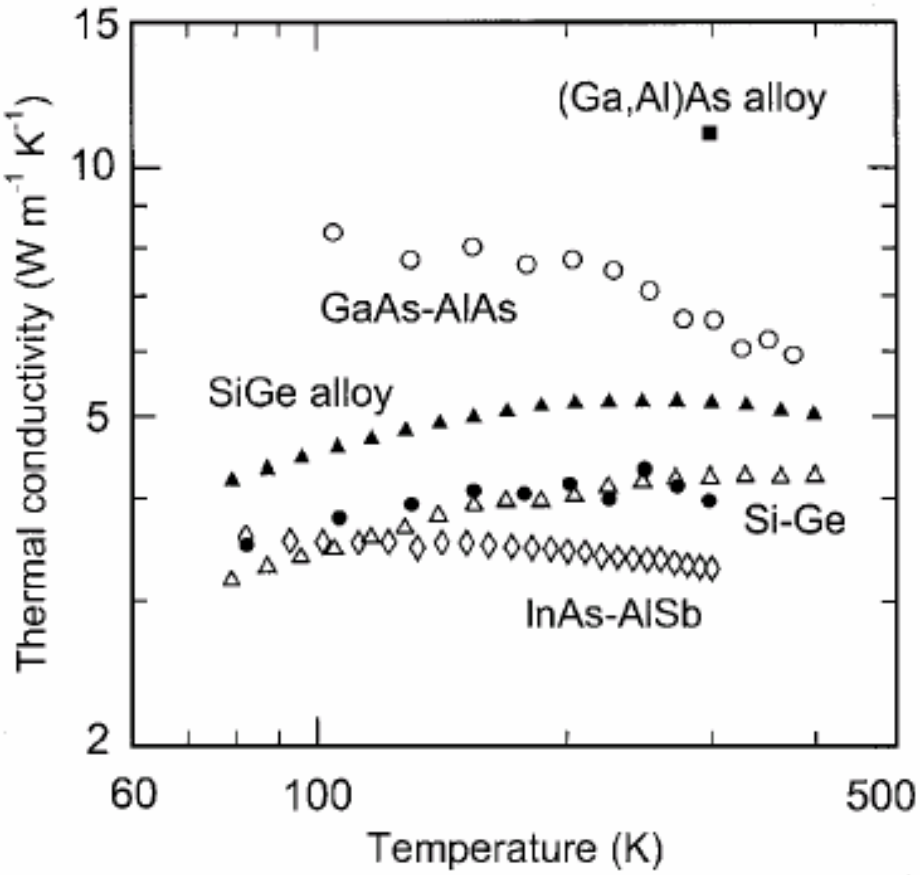


Asheghi et al., 1999

Heat Conduction Through Multiple Thin Films

- Fine-pitch 5 nm superlattices

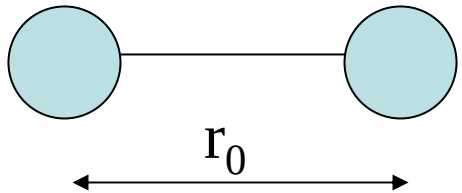
- Cross-thickness conductivity measurement
- Measured values are remarkably close to bulk alloy values (nearly within measurement error)
- Expected large reduction in conductivity not observed



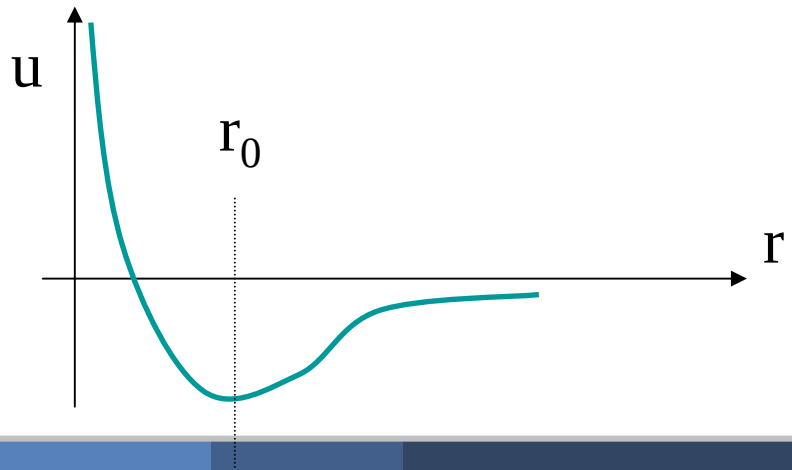
Cahill et al., 2003

Lattice Vibrations

- Consider two neighboring atoms that share a chemical bond



- The bond is not rigid, but rather like a spring with an energy relationship such as…

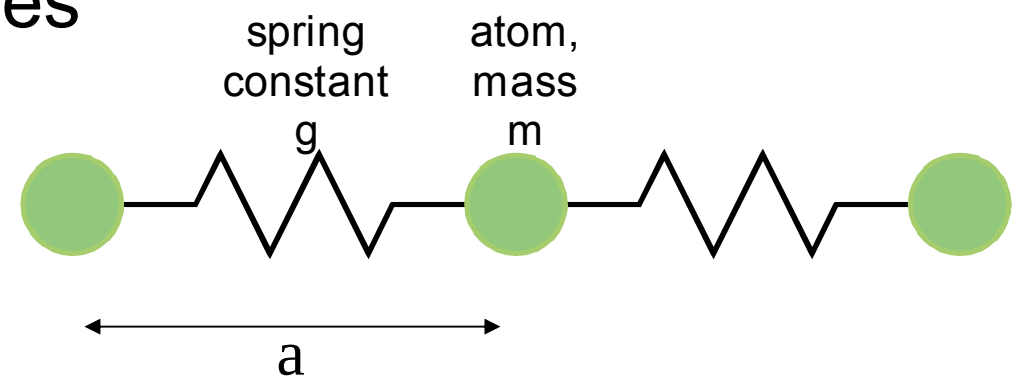


Lattice Vibrations, cont'd

- Near the minimum, the energy is well approximated by a parabola

$$u = \frac{1}{2} g x^2$$

  - $x = r - r_0$ and g = spring constant
- Now consider a one-dimensional chain of molecules



Lattice Energy and Motion

- Harmonic potential energy is the sum of potential energies over the lattice

$$U^{harm} = \frac{1}{2} g \sum_n \{x[na] - x[(n+1)a]\}^2$$

- Equation of motion of atom at location $x(na)$

$$F = m\frac{d^2x(na)}{dt^2} = -\frac{\partial U^{harm}}{\partial x(na)} = -g\{2x(na) - x[(n-1)a] - x[(n+1)a]\}$$

- Simplified notation

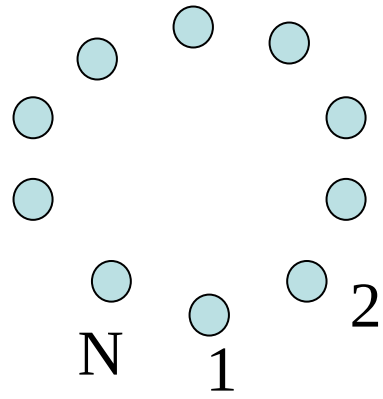
$$m\frac{d^2x_n}{dt^2} = -g\{2x_n - x_{n-1} - x_{n+1}\}$$

Lattice Motion, cont'd

- Seek solutions of the form

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

- Boundary conditions
  - Born-von Karman: assume that the ends of the chain are connected
    - $x_{N+1} = x_1$
    - $x_0 = x_N$



Lattice Motion, cont'd

- Then the boundary conditions become

$$x_{N+1} \sim \exp\left\{i\left[K\left(N+1\right)a-\omega t\right]\right\}$$

$$x_1 \sim \exp\left\{i\left[Ka-\omega t\right]\right\}$$

$$\rightarrow 1=\exp\left[iKNa\right]\rightarrow KNa=2\pi n,$$

where n is an integer

- Let λ be the vibration wavelength, $\lambda = aN/n$

$$K=\frac{2\pi n}{aN}=\frac{2\pi}{\lambda}$$

- Minimum wavelength, $\lambda_{min} = 2a = 2$(lattice spacing)

Solution to the Equations of Motion

- Substitute exponential solution into equation of motion

$$-m\omega^2 e^{i(Kna-\omega t)} = -g\left[2 - e^{-iKa} - e^{iKa}\right]e^{i(Kna-\omega t)}$$

$$= -2g\left(1-\cos Ka\right)e^{i(Kna-\omega t)}$$

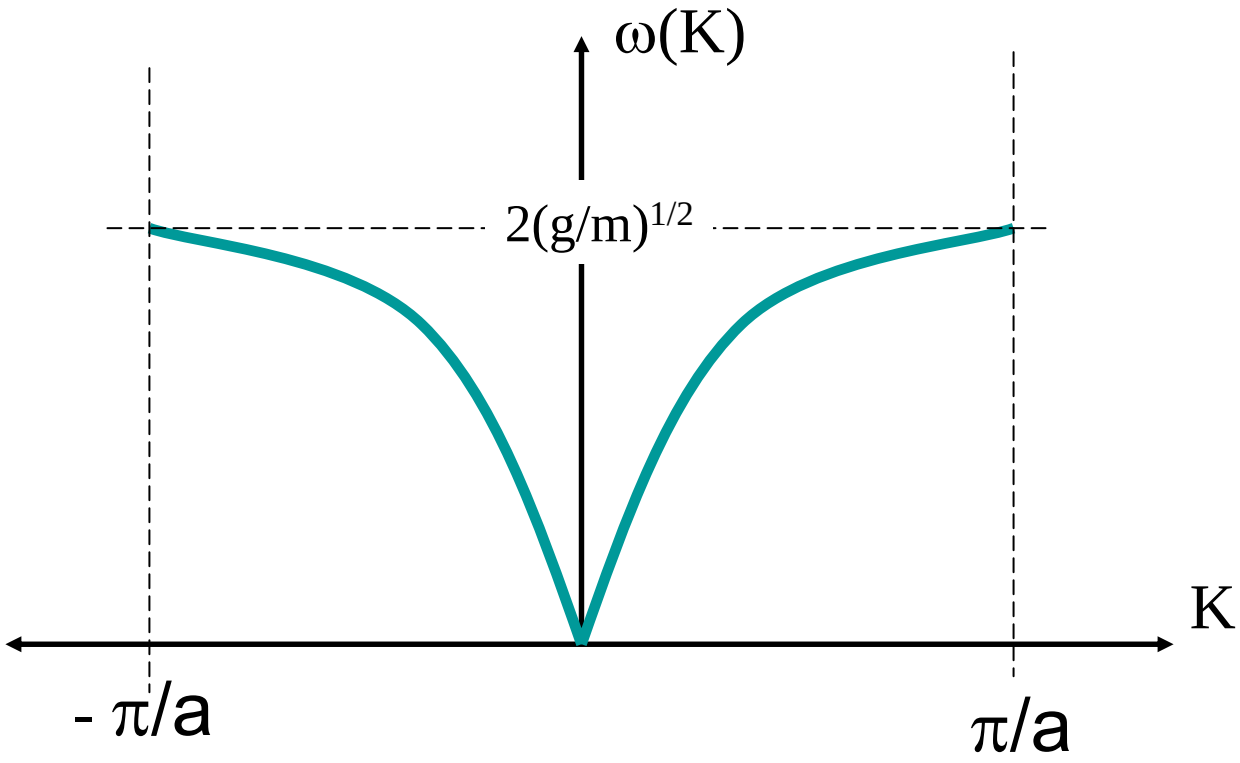
- Solve for ω

$$\omega(K) = \sqrt{\frac{2g(1-\cos Ka)}{m}} = 2\sqrt{\frac{g}{m}}\left|\sin(\tfrac{1}{2}Ka)\right|$$

- This is the dispersion relation for acoustic phonons
  - relates phonon frequency (energy) to wave vector (wavelength)

Dispersion Curve

- Changing *K* by $2\pi/a$ leaves *x* unaffected
  - Only *N* values of *K* are unique
  - We take them to lie in $-\pi/a < K < \pi/a$



Wave Velocities

- Phase velocity: $c = \omega/K$
- Group velocity: $v_g = \partial\omega/\partial K = a(g/m)^{1/2}\cos(Ka/2)$
- For small K:

$$\lim_{K\to 0} \omega = a\sqrt{\frac{g}{m}}|K|$$

$$\rightarrow \lim_{K\to 0} v_g = a\sqrt{\frac{g}{m}} = \left|\frac{\omega}{K}\right| = c$$

- Thus, for small K (large λ), group velocity equals phase velocity (and speed of sound)
- We call these acoustic vibration modes

Notes on Lattice Vibrations

- For $K = \pm\pi/a$, the group velocity is zero
  - why? $\frac{x_{n+1}}{x_n} = \exp\{iKa\} = \exp\{i\pi\} = \cos\pi + i\sin\pi = -1$
  - neighbors are 180 deg out of phase
- The region $-\pi/a < K < \pi/a$ is the first Brillouin zone of the 1D lattice
- We must extrapolate these results to three dimensions for bulk crystals

Density of Phonon States (Kittel, Ch5)

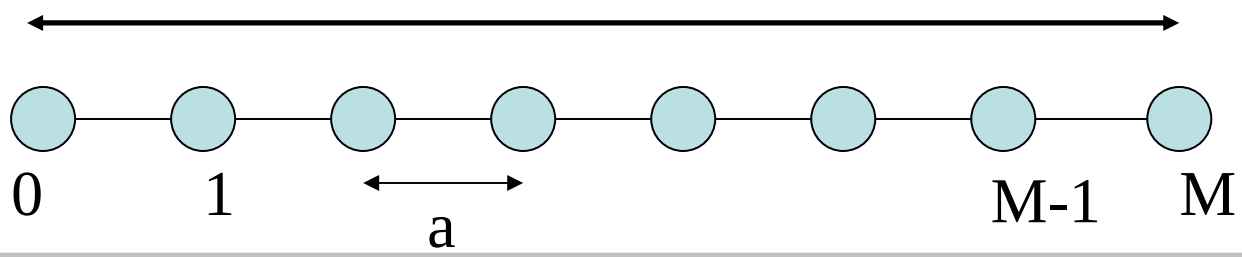
- Consider a 1D chain of total length *L* carrying *M*+1 particles (atoms) at a separation *a*
  - Fix the position of atoms 0 and *M*
  - Each normal vibrational mode of polarization p takes the form of a standing wave

$$x_n \sim \sin(nKa)\exp(-i\omega_{Kp}t)$$

  - Only certain wavelengths (wavevectors) are allowed $\lambda_{max}=2L$ ($K_{min}=\pi/L$), $\lambda_{min}=2a$ ($K_{max}=\pi/a=M\pi/L$)
  - In general, the allowed values of K are

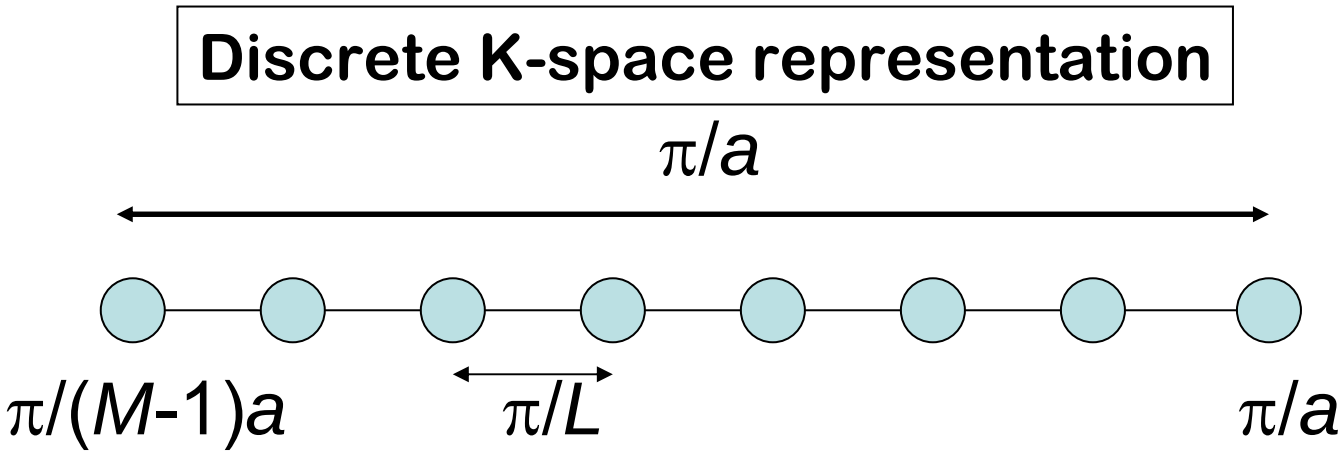
$$K = \frac{\pi}{L}, \frac{2\pi}{L}, \frac{3\pi}{L}, \ldots, \frac{(M-1)\pi}{L}$$

Note: $K=M\pi/L$ is not included because it implies no atomic motion, i.e., $\sin(nM\pi a/L)=\sin(n\pi)=0$.



Density of States, cont'd

- Thus, we have M-1 allowed, independent values of *K*
  - This is the same number of particles allowed to move
  - In *K*-space, we thus have *M*-1 allowable wavevectors
  - Each wavevector describes a single mode, and one mode exists in each distance π/L of *K*-space
  - Thus, $dK/dN = \pi/L$, where *N* is the number of modes



Density of States, cont'd

- The phonon density of states gives the number of modes per unit frequency per unit volume of real space

$$D(\omega) = \frac{1}{L^{\alpha=1}} \frac{dN}{d\omega} = \frac{1}{L} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{1}{\pi} \frac{1}{d\omega / dK}$$

  - The last denominator is simply the group velocity, derived from the dispersion relation

$$D(\omega) = \frac{1}{\pi v_g(\omega)} = \left[\pi a \sqrt{\frac{g}{m}} \cos\left(\frac{1}{2} K(\omega) a \right) \right]^{-1}$$

Note singularity for $K = \pi / a$

Periodic Boundary Conditions

- For more generality, apply periodic boundary conditions to the chain and find

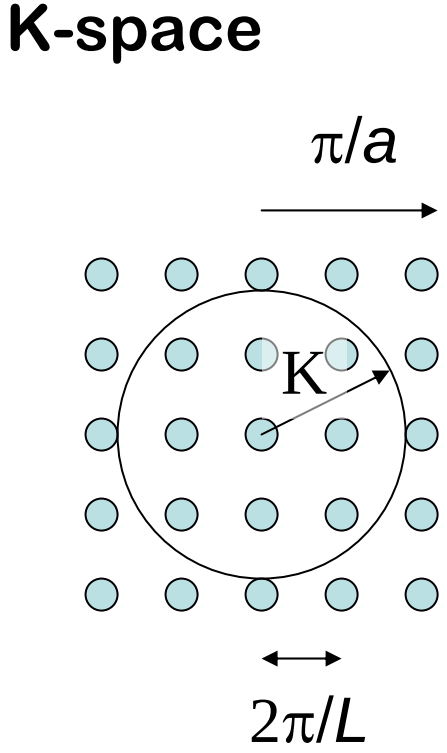
$$K = 0, \pm\frac{2\pi}{L}, \pm\frac{4\pi}{L}, \ldots, \frac{M\pi}{L}$$

  - Still gives same number of modes (one per particle that is allowed to move) as previous case, but now the allowed wavevectors are separated by $\Delta K = 2\pi/L$

  - Useful in the study of higher-dimension systems (2D and 3D)

2D Density of States

- Each allowable wavevector (mode) occupies a region of area $(2\pi/L)^2$
- Thus, within the circle of radius K, approximately $N=\pi K^2/(2\pi/L)^2$ allowed wavevectors exist
- Density of states



$$D(\omega) = \frac{1}{L^{\alpha=2}} \frac{dN}{d\omega} = \frac{1}{V} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{K(\omega)}{2\pi} \frac{1}{v_g(\omega)}$$

3D Density of States

- Using periodic boundary conditions in 3D, there is one allowed value of **K** per $(2\pi/L)^3$ volume of *K*-space
- The total number of modes with wavevectors of magnitude less than a given *K* is thus

$$N = \left(\frac{L}{2\pi}\right)^3 \left(\frac{4}{3}\pi K^3\right) = \frac{VK^3}{6\pi^2}$$

- The 3D density of states becomes

$$D(\omega) = \frac{1}{L^{\alpha=3}} \frac{dN}{d\omega} = \frac{1}{V} \frac{dN}{dK} \frac{dK}{d\omega} = \frac{K(\omega)^2}{2\pi^2} \frac{1}{v_g(\omega)}$$

Overview of Phonon Simulation Tools

- Boltzmann Transport Equation (BTE)
  - Requires boundary scattering models
  - Requires detailed understanding of phonon scattering and dispersion for rigorous inclusion of phonon physics
- Molecular Dynamics (MD)
  - Computationally expensive
  - Not strictly applicable at low temperatures
  - Handling of boundaries requires great care for links to larger scales and simulation of functional transport processes
- Atomistic Green's Function (AGF)
  - Efficient handling of boundary and interface scattering
  - Straightforward links to larger scales
  - Inclusion of anharmonic effects is difficult

Atomistic Green's Function (AGF) Modeling of Phonon Transport

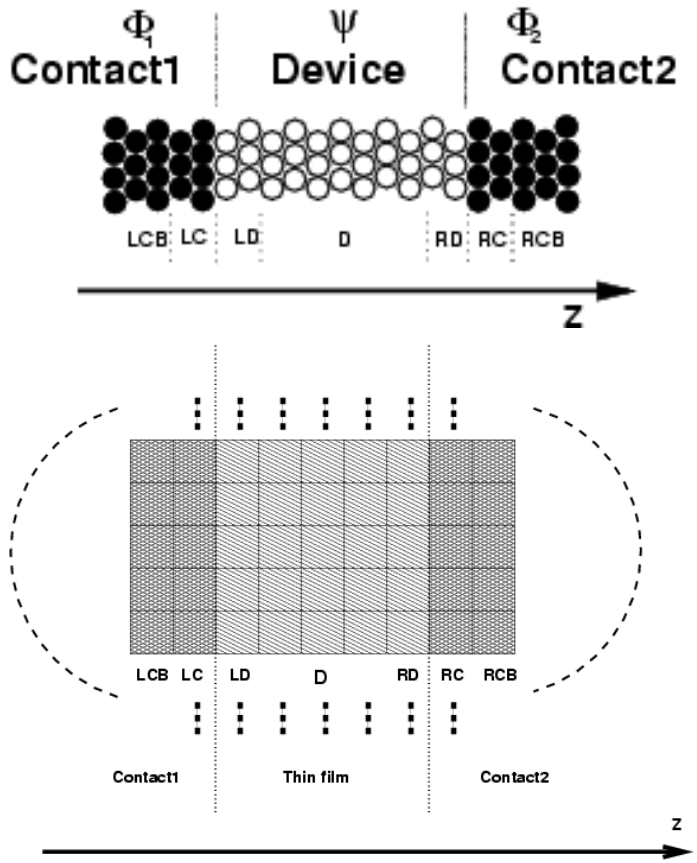
Based on Zhang et al. "The Atomistic Green's Function Method: An Efficient Simulation Approach for Nanoscale Phonon Transport," *Num Heat Trans-B*, in review.

Some Background

- Non-equilibrium Green's function method initially developed to simulate electron ballistic transport (see Datta, 1995)
- Very efficient in the ballistic regime but requires significant effort to implement scattering
- Recently applied to phonon transport (see Mingo, 2003; Zhang et al., in press)

Atomistic Green's Function

- 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



Recall 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 Matrix

- Define the **k** matrix as

$$k_{ij} = -\frac{\partial^2 U^{harm}}{\partial x_i \partial x_j}$$

$$\rightarrow k_{nn} = -2g$$

$$k_{n,n+1} = k_{n,n-1} = g$$

Here, *g* is the spring constant

- Then, define the harmonic matrix **H** as

$$H_{ij} = \frac{1}{\sqrt{M_i M_j}} k_{ij}, \text{ no index summation}$$

Harmonic 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

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

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

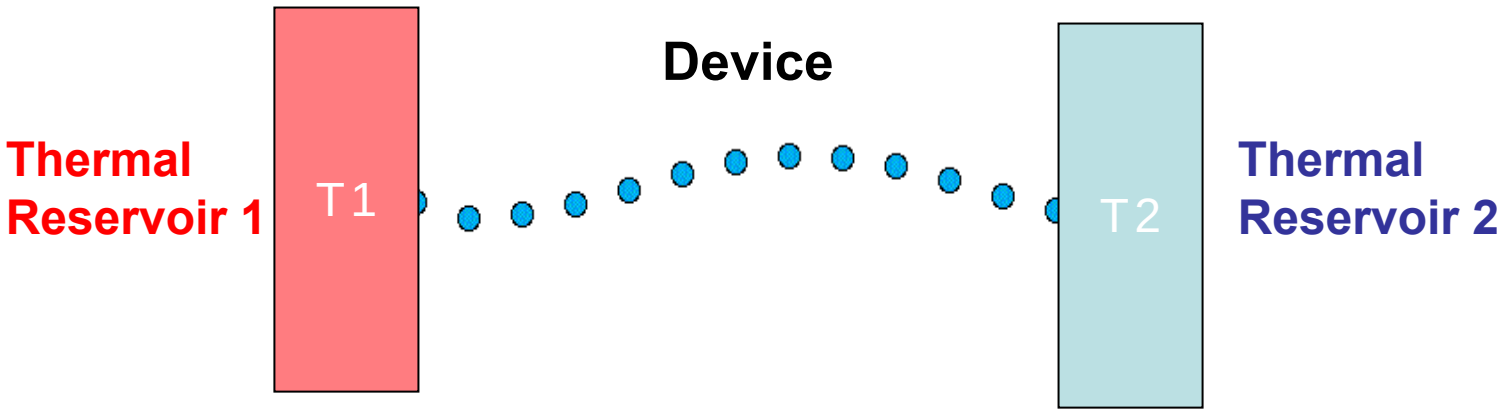
$$\mathbf{g} = \left[\left(\omega^2 + \delta i\right)\mathbf{I} - \mathbf{H}\right]^{-1}$$

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

Toward Realistic Problems

- So far, we have not made much progress in solving real problems

- To solve most practical problems, we need to incorporate different materials and interfaces



Why Green's Functions?

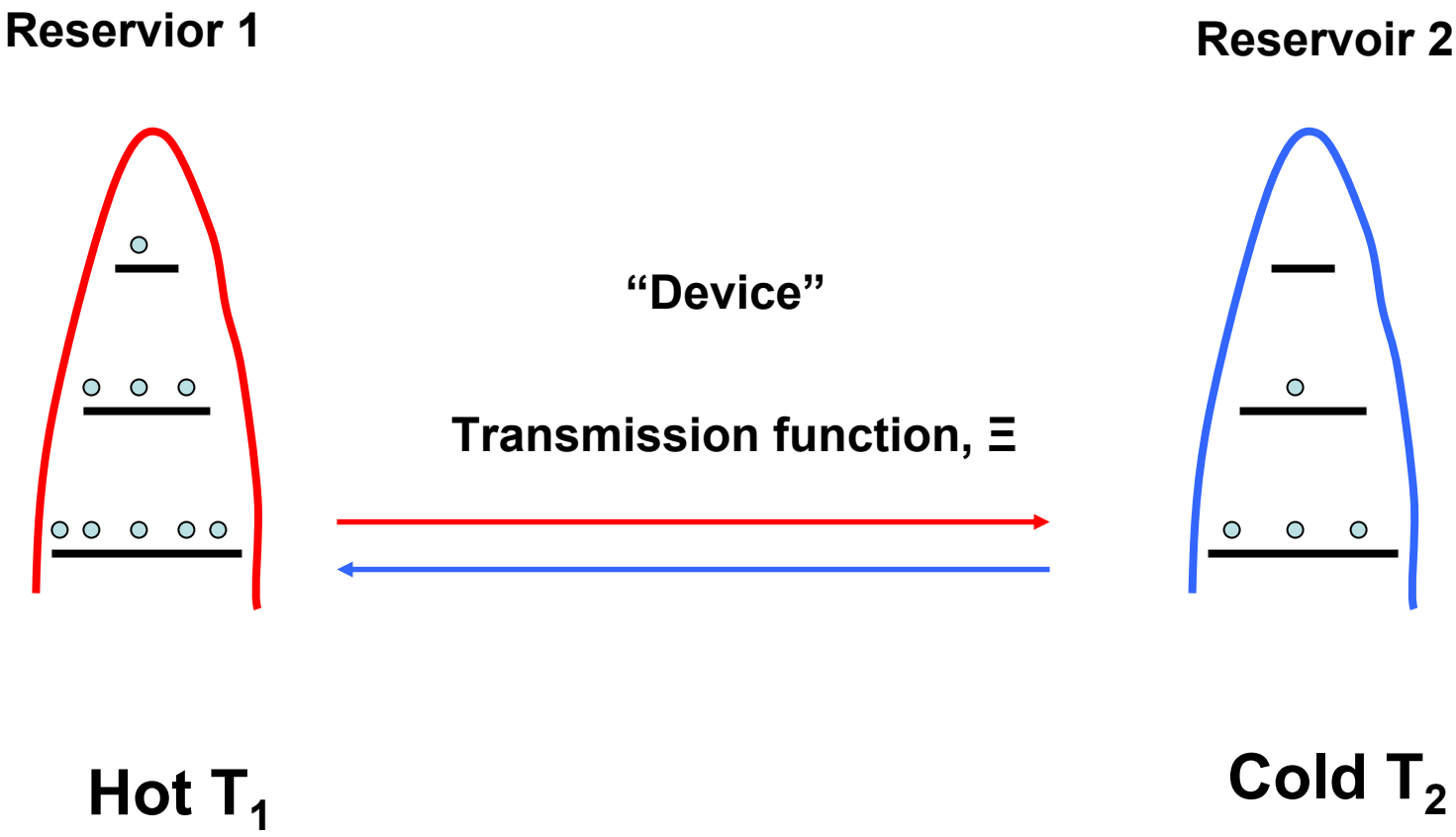
- We can model connections among different materials through the use of a different Green's function **G**

$$\mathbf{G} = \left[\omega^2 \mathbf{I} - \mathbf{H}_\mathbf{d} - \underbrace{\tau_1 \mathbf{g}_1 \tau_1^T}_{\Sigma_1} - \underbrace{\tau_2 \mathbf{g}_2 \tau_2^T}_{\Sigma_2} \right]^{-1}$$

superscript "*T*" = conjugate transpose

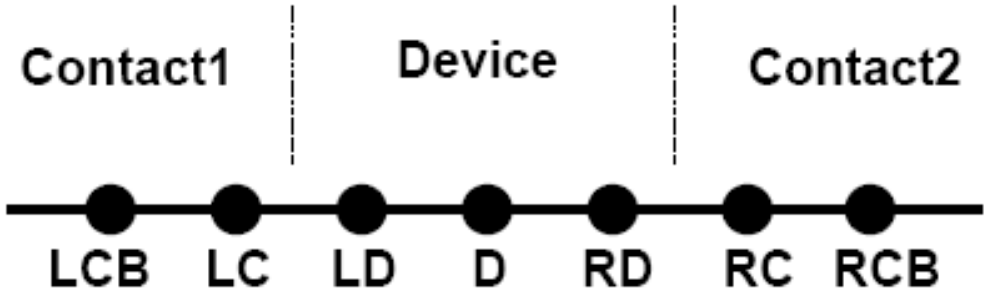
  - This matrix function includes self-energy matrices (Σ_1, Σ_2) that involve unperturbed Green's functions (**g**'s) associated with contacts (i.e., boundaries) in a transport problem
  - τ matrices handle connections between different system elements (materials, interfaces)
  - The full derivation is 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 scattering

Phonon Transport through a "Device" between Two Contacts

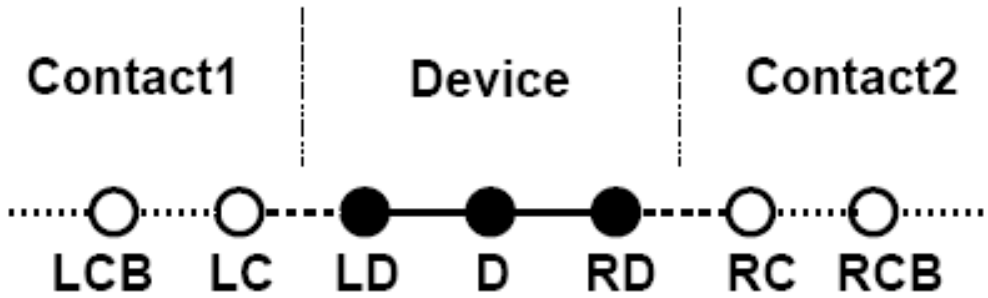


1D Atomic Chain

- 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.

Transmission and Heat Flux

$$J = \int_0^\infty \frac{\hbar\omega}{2\pi}\Delta N(\omega)\Xi(\omega)d\omega$$

(units = W)

Phonon energy

Phonon occupation difference

Transmission

$$\Delta N = N^+ - N^- \approx \frac{\hbar\omega}{k_B T^2}\frac{e^{\hbar\omega/k_B T}}{\left(e^{\hbar\omega/k_B T} - 1\right)^2}\Delta T$$

We need to evaluate transmission in order to calculate heat flux

Transmission and Green's Functions

- Some definitions of convenience

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

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

- The transmission function

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

The AGF Algorithm

Establish atomic positions and potential parameters ..... U and $\mathbf{x}_i$

↓

Assemble harmonic matrices (*H*) ..... $$\mathbf{H} = \{H_{ij}\} = \frac{1}{\sqrt{M_i M_j}} \begin{cases} \frac{\partial^2 U}{\partial u_i \partial u_j}, & \text{if i} \neq \text{j} \\ -\sum_{\text{m} \neq \text{j}} \frac{\partial^2 U}{\partial u_j \partial u_m}, & \text{if i} = \text{j} \end{cases}$$

↓

Calculate the Green's function (*g*) of uncoupled contacts ..... Uses decimation algorithm

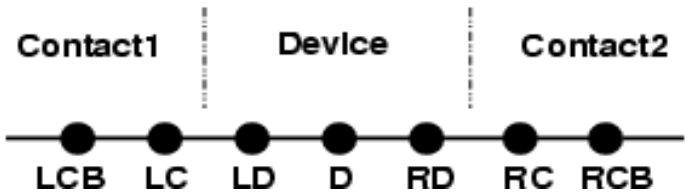
↓

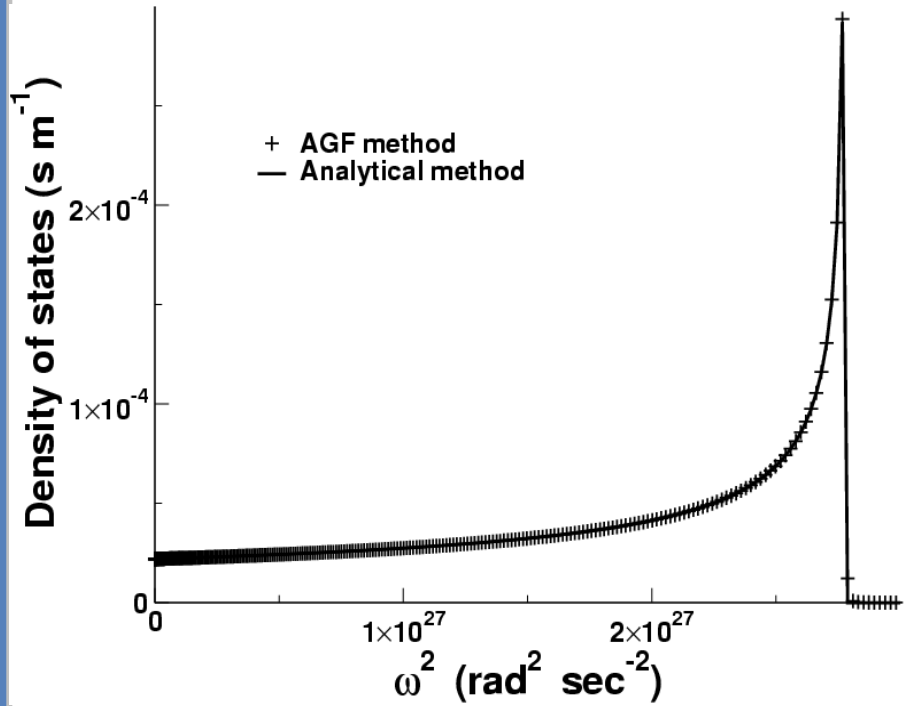
Calculate device **G** and phonon transmission (Ξ) ..... $$\Xi\left(\omega, \vec{\mathbf{k}}_{||}\right) = \text{Trace}\left[\mathbf{\Gamma}_L \mathbf{G}_{LD,RD} \mathbf{\Gamma}_R \mathbf{G}^{\dagger}_{LD,RD}\right]$$

↓

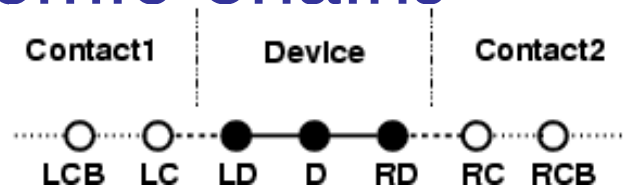
Integrate (Ξ) over phonon frequencies and $k_{||}$ to obtain the thermal conductance ..... $$J = \frac{1}{s} \int_0^{\infty} \int_{\vec{\mathbf{k}}_{||}} \frac{\hbar\omega}{2\pi} \Delta\tilde{N}(\omega) \Xi(\omega, \vec{\mathbf{k}}_{||}) \frac{d\vec{\mathbf{k}}_{||}}{(2\pi)^2} d\omega$$

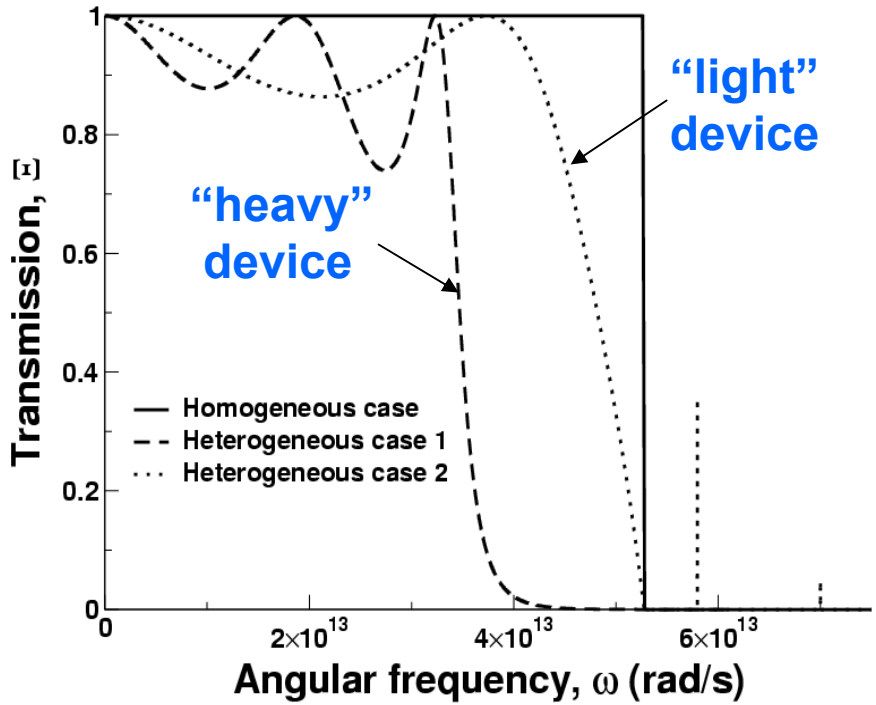
Results for Simple Atomic Chains





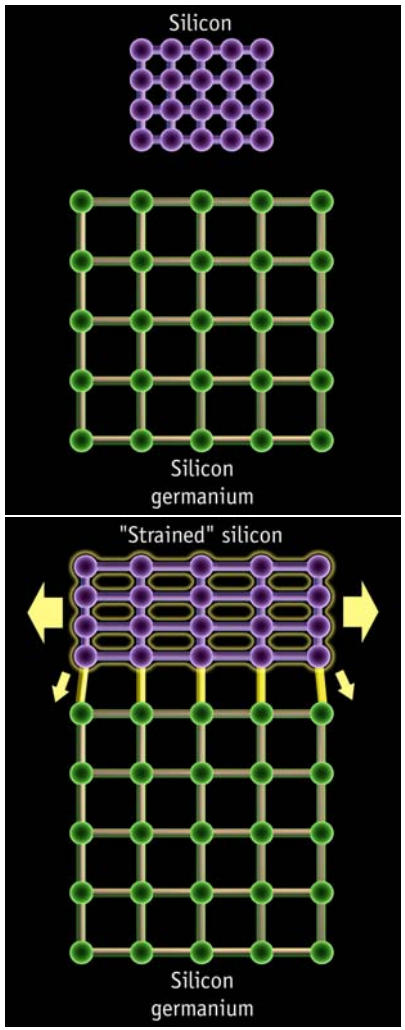
Homogeneous chain density of states





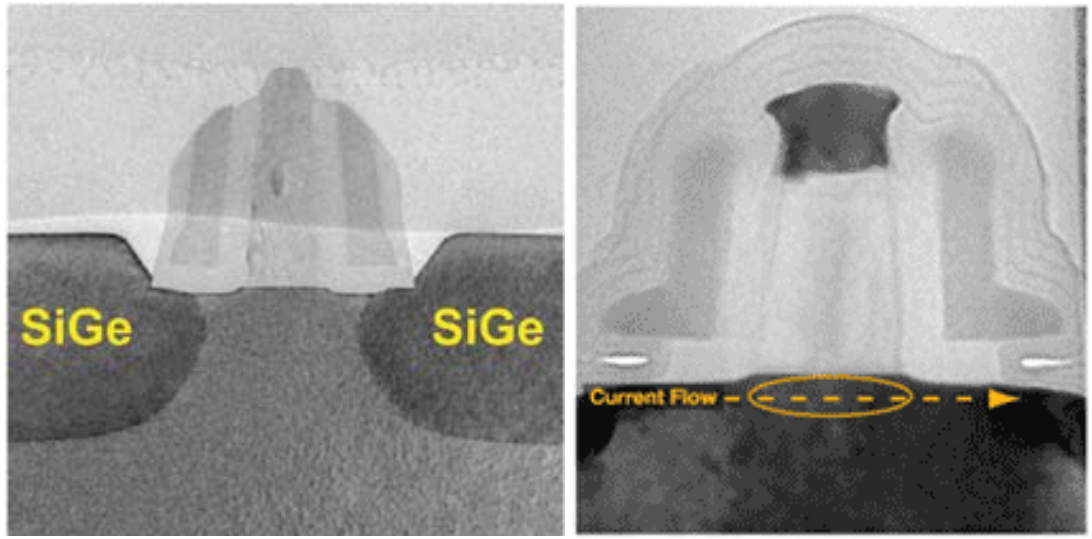
Homogeneous vs heterogeneous

Strained Silicon



T. Ghani, et al. at IEDM 2003

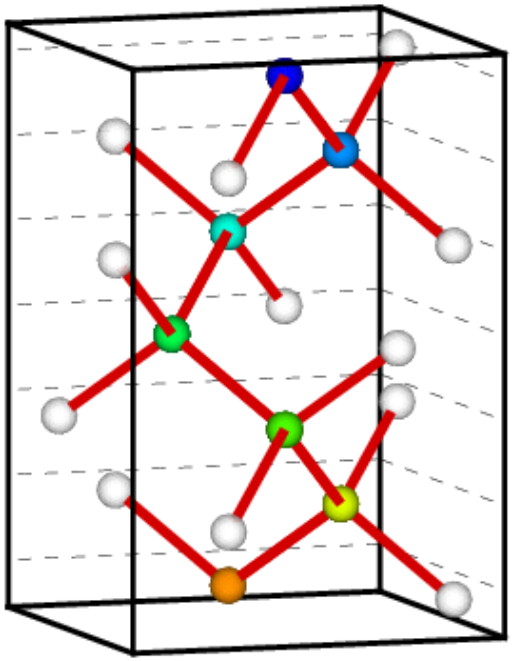
PMOS NMOS

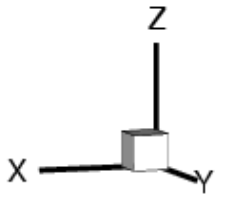


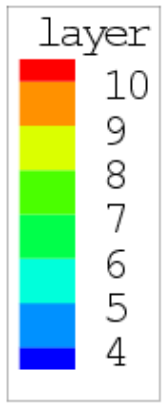
25 % drive current increase in PMOS;
10 % drive current increase in NMOS

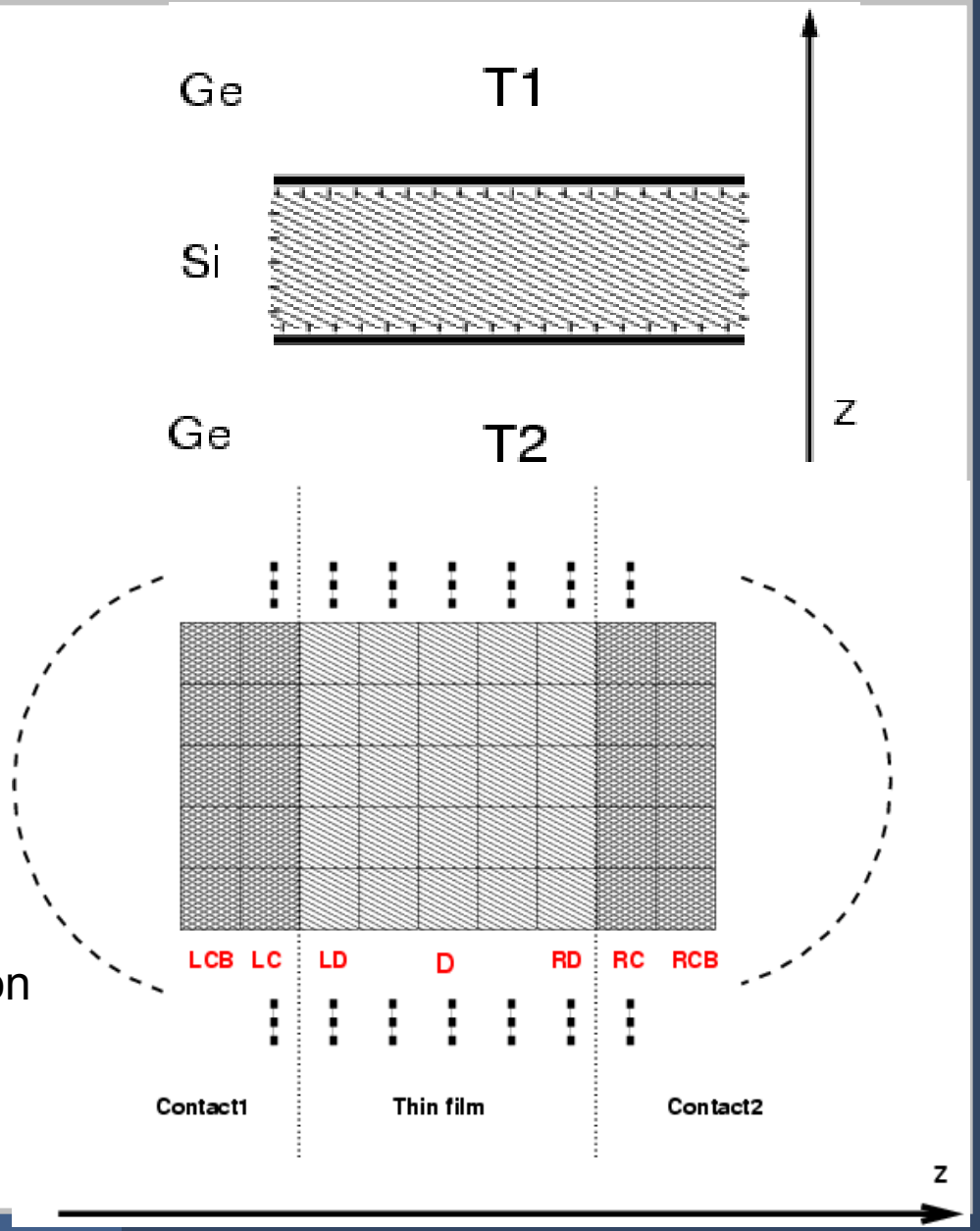
http://www.research.ibm.com/resources/press/strainedsilicon/

Thin Films





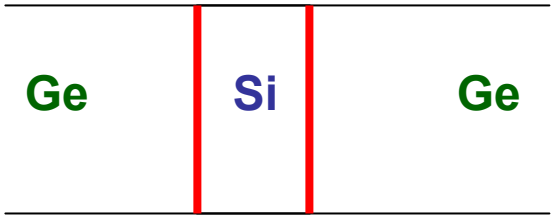




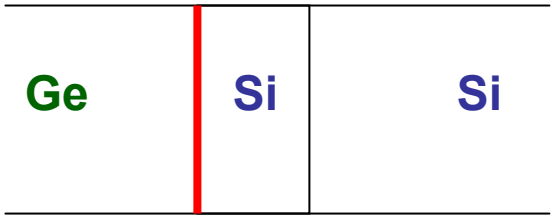
Transport along the (100) direction
(i.e., the *z* direction)

Model/Code Validation

Convert Ge/Si/Ge

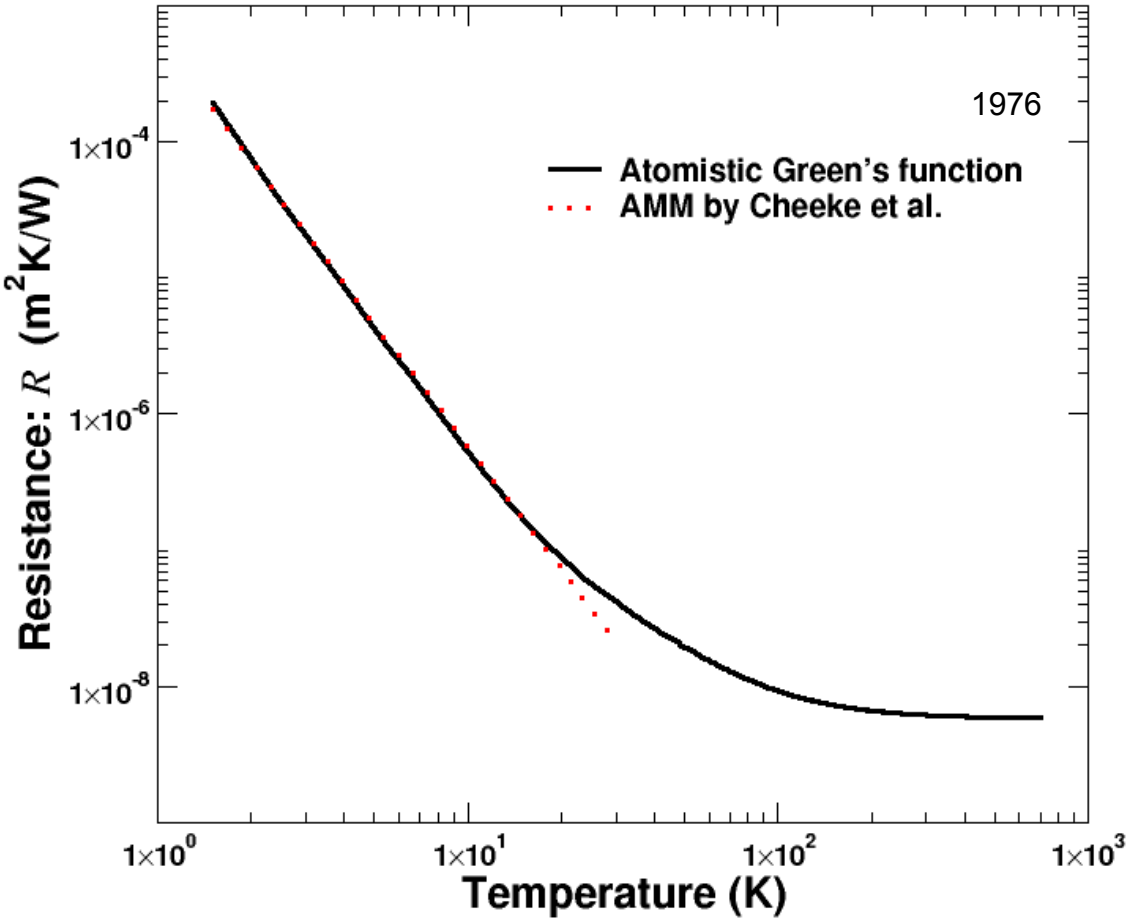


to Ge/Si/Si



to create a single interface.

Zhang et al., J. Heat Trans. in review

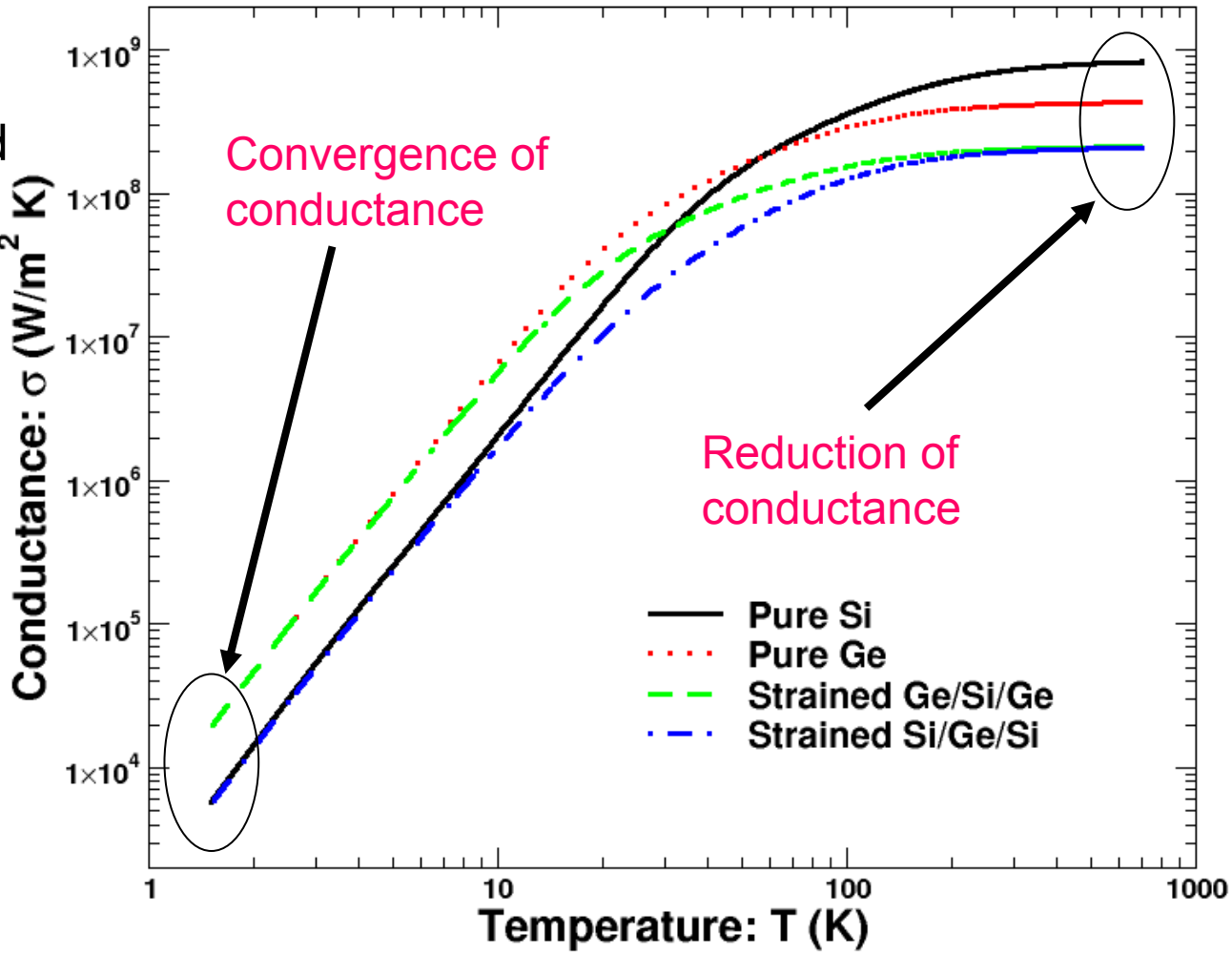


AMM is known to work well at low temperatures

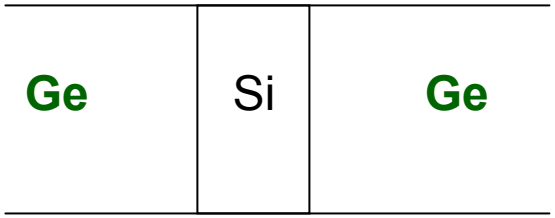
Thermal Conductance

Conductance reduced by 30 to 50% at room temperatures due to heterogeneous interfaces

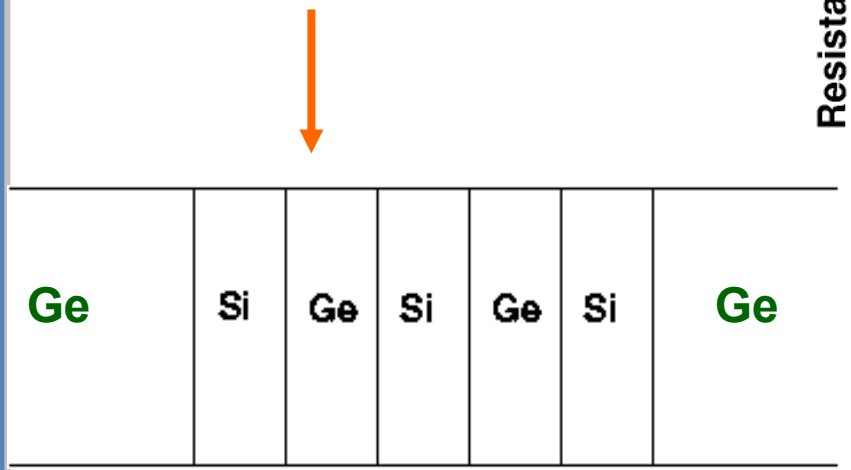
At low temperatures, conductance converges to that of the bulk contacts

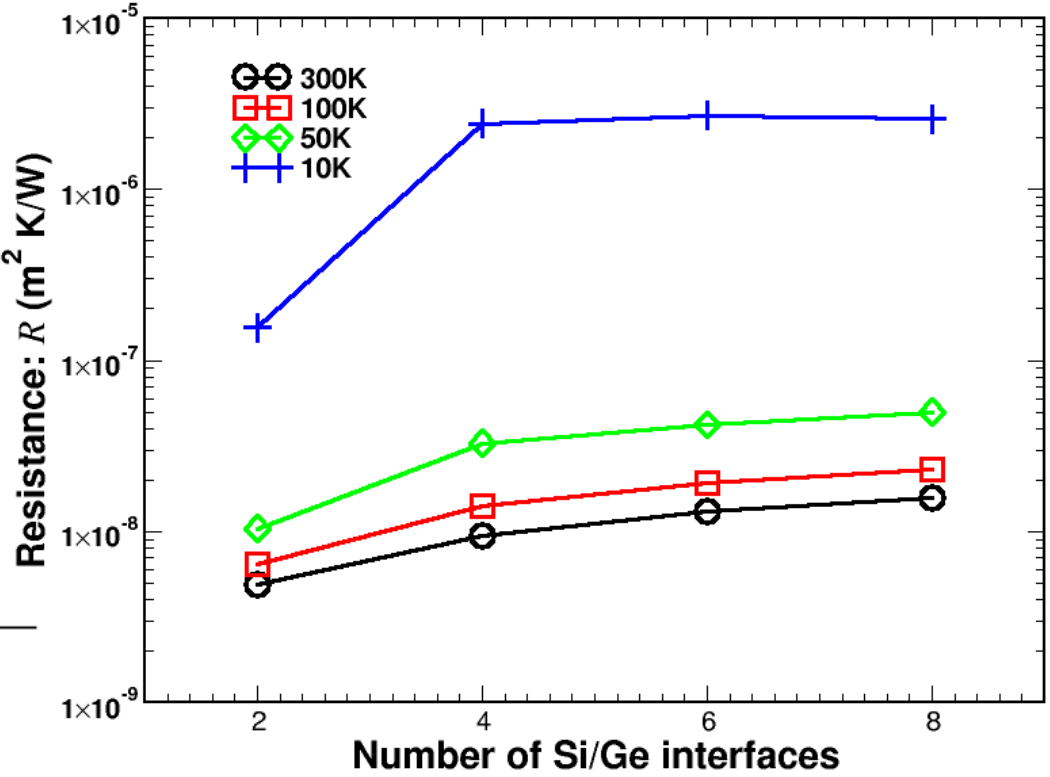


Multilayer Effects



Replace the Si device with a multi-layer structure





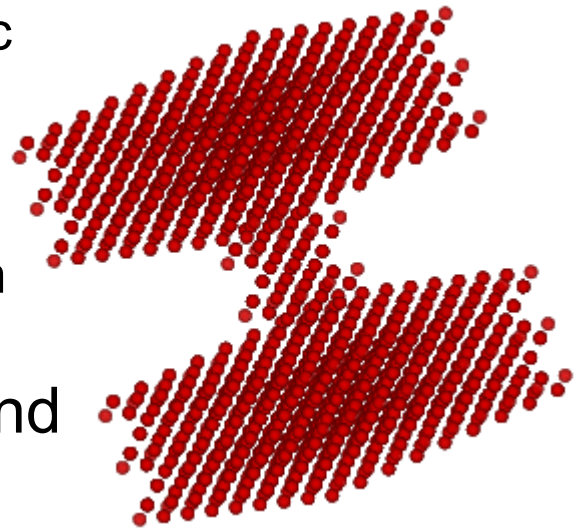
Asymptotic behavior is similar to that of radiation shields

Conclusions

- The AGF method is an effective tool in simulating ballistic phonon transport through relevant interfaces involving bulk and nanoscale materials
- Strain effects are small compared to heterogeneous-material effects
- A heterogeneous device layer reduces thermal conductance significantly at room temperature
- Increasing film thickness decreases thermal conductance
- The first few heterogeneous interfaces are most responsible for decreasing thermal conductance

Ongoing Work

- Numerical construction of harmonic matrices (*H*)
  - Enables the use of more complicated atomic configurations as well as advanced inter-atomic potentials
  - Implemented with EDIP potentials and benchmarked against bulk silicon dispersion curves
- Phonon transport through a nanowire and bulk contacts
  - The size mismatch between a nanowire and its bulk contact limits heat flow
  - (100) nanowire is being benchmarked



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Classes

- ME 597F: Micro and Nanoscale Energy Transfer Processes
- ME 595M: Computational Nanoscale Heat Transfer (with J. Murthy)