Thermal Transport Across Interfaces (Part I)

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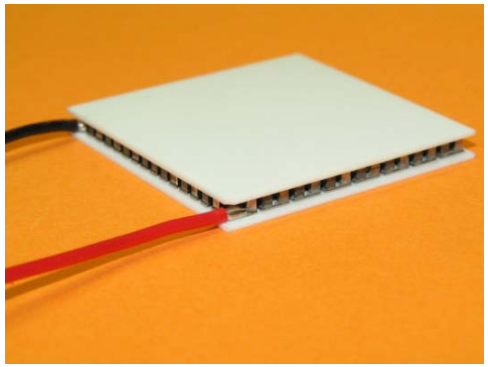
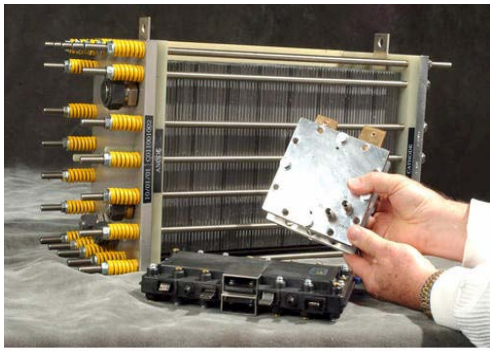
Outline

- Introduction
- Lattice vibrations and phonons
- The vibrating string
  - Interfaces between dissimilar strings: acoustic mismatch
  - Discrete masses and the vibrational eigenspectrum
- General thermal transport theory

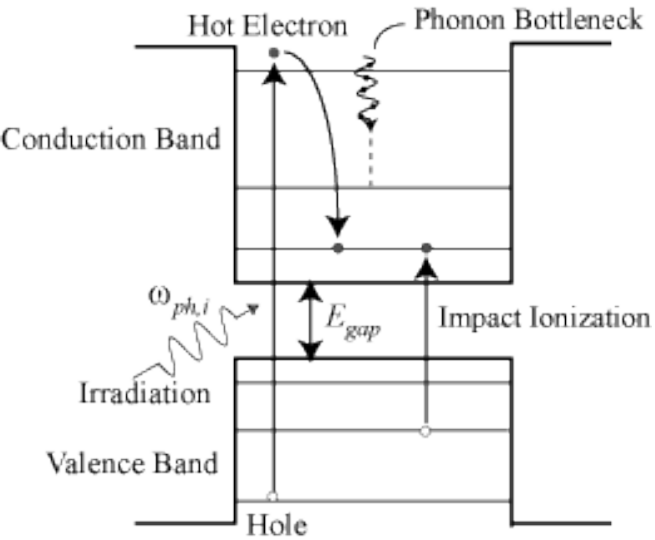
Applications of Nanoscale Energy Transport

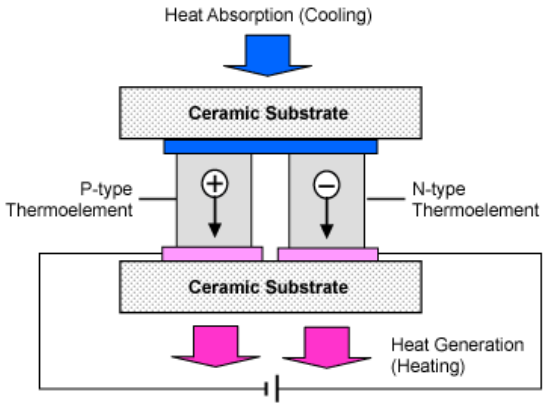
Energy conversion

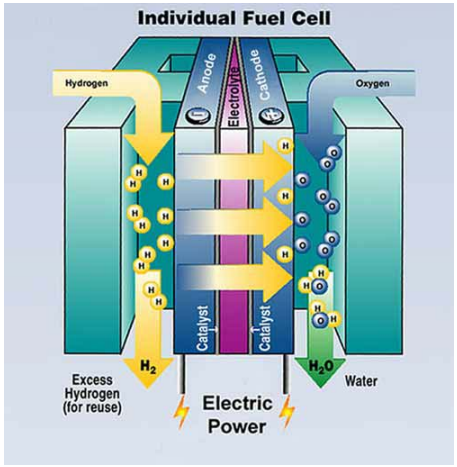




Courtesy of The National Renewable Energy Laboratory (NREL)







photovoltaics

absorptivity (photon-electron coupling), electron-phonon coupling

thermoelectrics

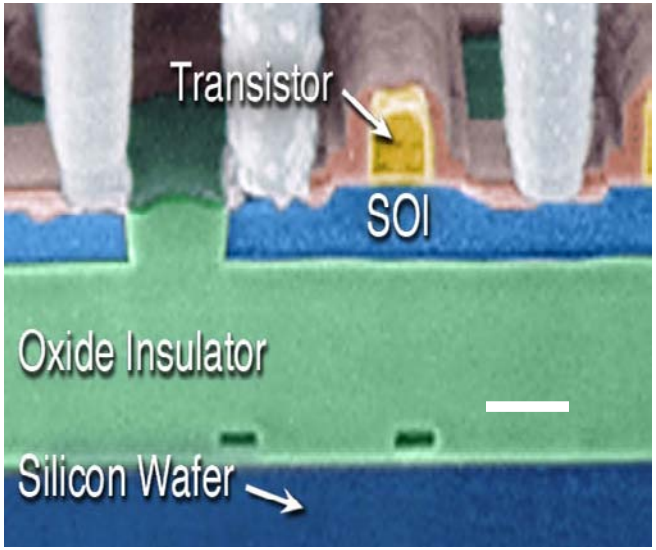
Seebeck coefficient, thermal conductivity, electrical conductivity

PEMFC fuel cell

H_2, O_2, and H_2O transport in nanopores

Applications of Nanoscale Energy Transport

- Cooling of electronics
- Laser manufacturing
- Bio systems
- Atmospheric heating



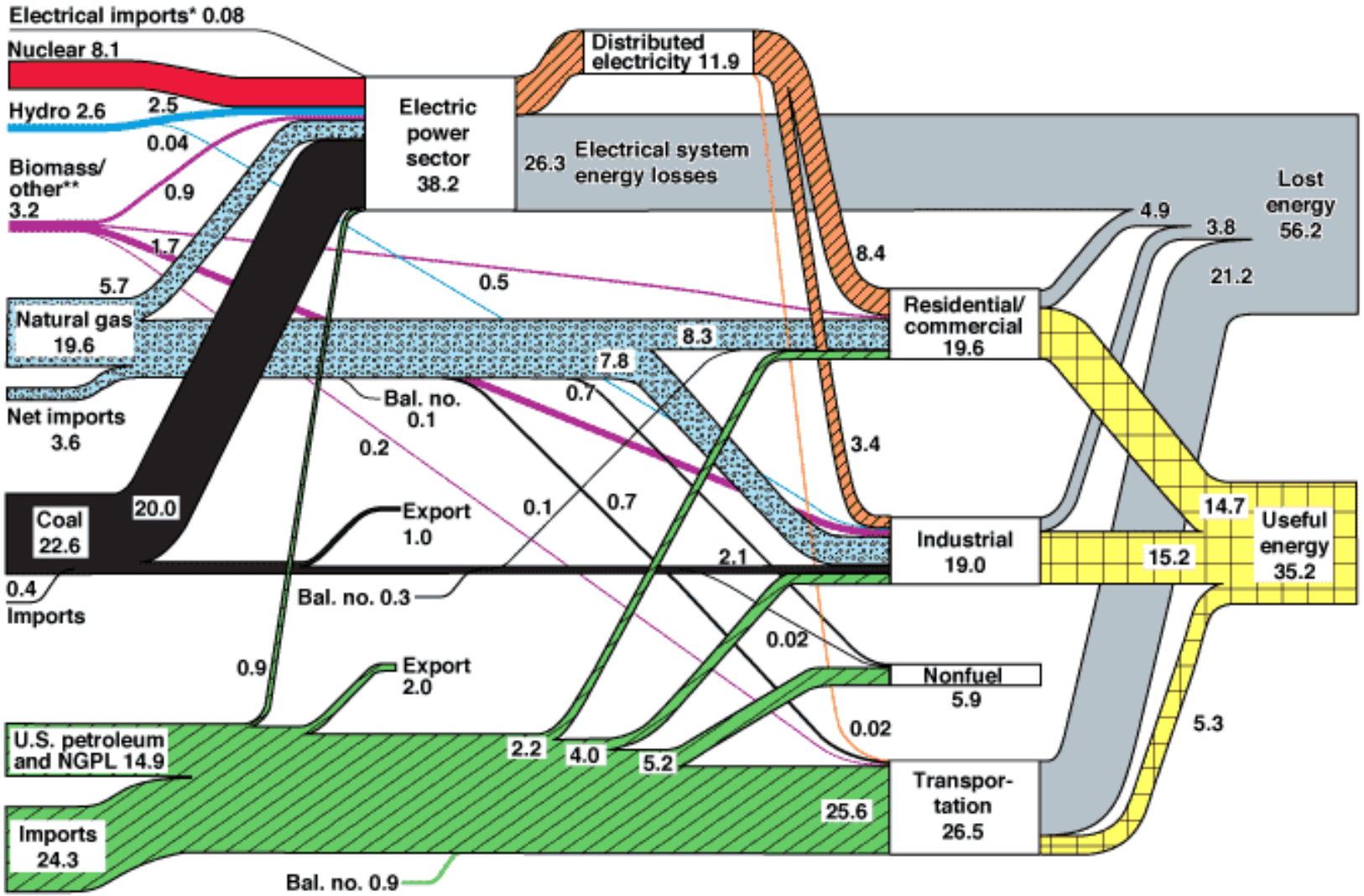
transistor

phonon-phonon scattering,
Phonon-boundary scattrering,
electron-phonon scattering

laser manufacturing

photon-electron interaction,
electron-phonon coupling

U.S. Energy Flow Trends – 2002
Net Primary Resource Consumption ~97 Quads



Source: Production and end-use data from Energy Information Administration, *Annual Energy Review 2002*.
*Net fossil-fuel electrical imports.
**Biomass/other includes wood, waste, alcohol, geothermal, solar, and wind.

June 2004
Lawrence Livermore National Laboratory
http://eed.llnl.gov/flow

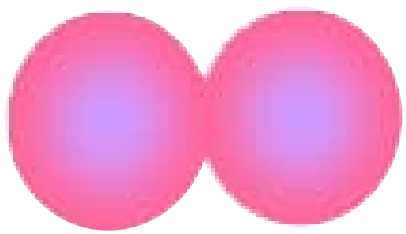
>90% of primary energy sources use thermal processes!

Principal Energy Carriers

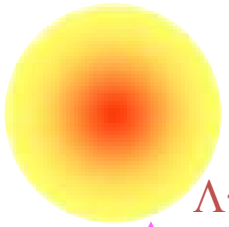
- Phonon: quantum of lattice vibrations
  - acoustic phonon: contributes to sound
  - optical phonon: infrared active or Raman active
- Electron: charged particle surrounding the nucleus
  - responsible for chemical bonding
- Fluid particle: single atoms or molecules in constant random motion
  - ideal gas: elastic collisions, forces of attraction are negligible
  - liquids: intermolecular forces
- Photon: quantum of electromagnetic fields

Nano for Energy

- Increased surface area
- Interface and size effects

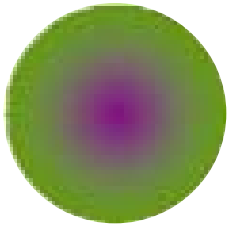


Molecules
Λ ~ 1–100 nm
λ~1 nm

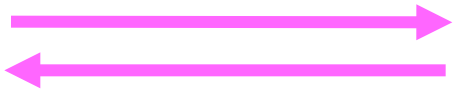


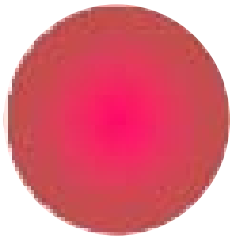
Photons
Λ > 10 nm
Λ~100-10,000 nm

Λ---Mean free path
λ---wavelength



Electrons
Λ~10-100 nm
λ~10-50 nm



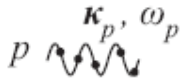
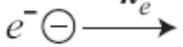
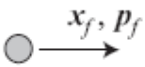
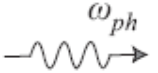


Phonons
Λ~10-100 nm
λ~1 nm

Slide courtesy of G. Chen

Carrier Distribution Functions (Occupation Number)

Table 1.2. Thermal equilibrium particle (energy occupancy) distribution (statistical) function $f_i^{\circ}(E_i)$, $i = p,\ e,\ f,\ ph$, and its temperature dependence for different energy carriers.

Attributes	Phonon	Electron (and Hole)	Fluid Particle	Photon
				
Energy presentation	wave vector κ_p or frequency ω_p, modes and polarization	wave vector κ_e, in conduction and valence bands, spins	momentum p_f, kinetic, potential, and electronic energy states	frequency ω_{ph}, polarization
Particle type	Bose-Einstein (Boson)	Fermi-Dirac (Fermion)	Maxwell-Boltzmann (M-B)	Bose-Einstein (Boson)
Nature of particle	particles are indistinguishable, integer spin (angular momentum), and any number of particles may occupy a given eigenstate	particles are indistinguishable, odd, half-integer spin (angular momentum) and obey the Pauli exclusion principle (only one particle may be found in a given eigenstate)	particles are distinguishable, and any number of particles may occupy a given eigenstate (classical particle or non-degenerate limit)	particles are indistinguishable, integer spin (angular momentum), and any number of particles may occupy a given eigenstate
Equilibrium distribution (occupancy) function, $f_i^{\circ}(E_i)$	$\frac{1}{\exp(\frac{E_p}{k_B T}) - 1}$	$\frac{1}{\exp(\frac{E_e - \mu}{k_B T}) + 1}$	$\frac{1}{\exp(\frac{E_{f,t}}{k_B T})}$	$\frac{1}{\exp(\frac{E_{ph}}{k_B T}) - 1}$

M. Kaviany, *Heat Transfer Physics*, 2008.

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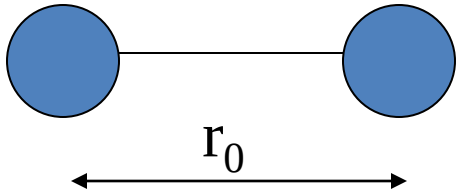
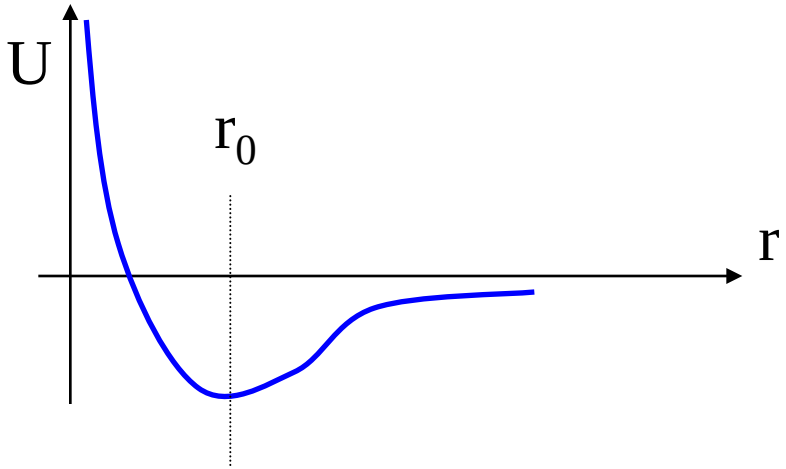
Lattice Vibrations

- 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…

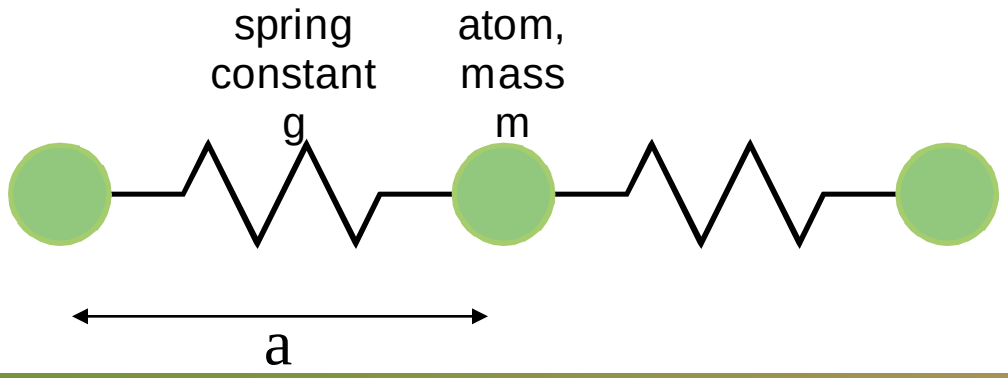


Lattice Vibrations, cont'd

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

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

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



Lattice Energy and Motion

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

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

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

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

- Simplified notation

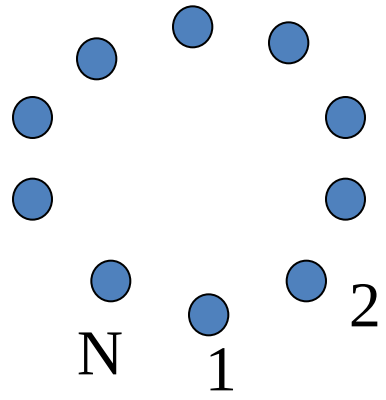
$$m\frac{d^2u_n}{dt^2} = -g\{2u_n - u_{n-1} - u_{n+1}\}$$

Lattice Motion, cont'd

- Seek solutions of the form

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

- Boundary conditions
  - Born-von Karman: assume that the ends of the chain are connected
    - $u_{N+1} = u_1$
    - $u_0 = u_N$



Lattice Motion, cont'd

- Then the boundary conditions become

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

$$u_1 \sim \exp\left\{i\left[Ka-\omega t\right]\right\}$$

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

where n is an integer

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

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

K = wave vector

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

Lattice Equations of Motion

- Substitute exponential solution into equation of motion

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

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

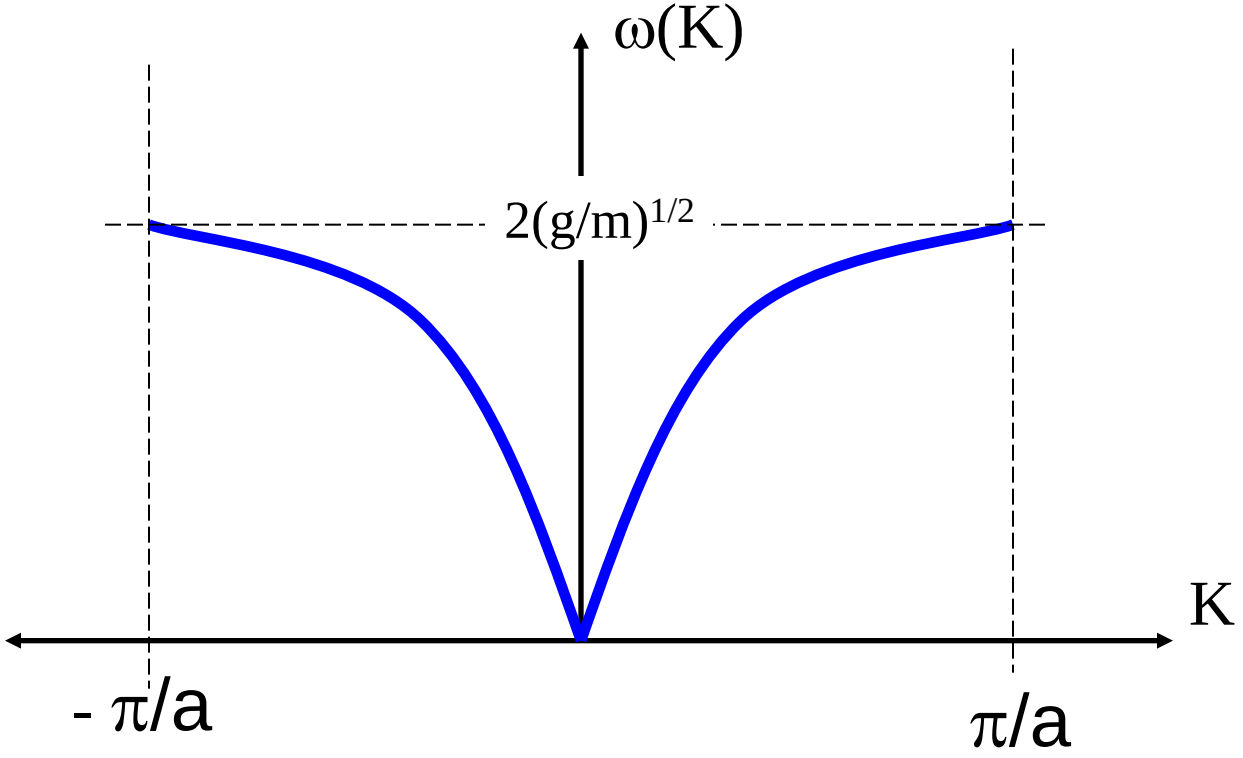
- Solve for ω

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

- 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 *u* 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? $\frac{u_{n+1}}{u_n} = \exp\{iKa\} = \exp\{i\pi\} = \cos\pi + i\sin\pi = -1$
  - 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

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Displacement of a Loaded String

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

- Consider a generic ODE of the form

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

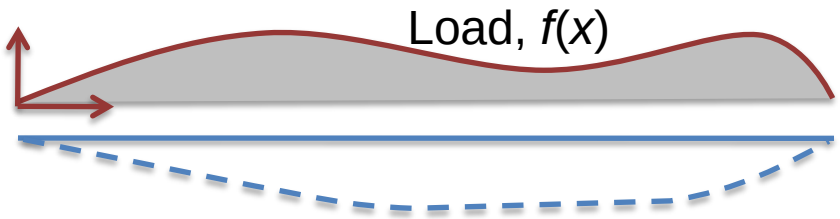
  - Where the prime (‘) denotes a derivative

- 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)

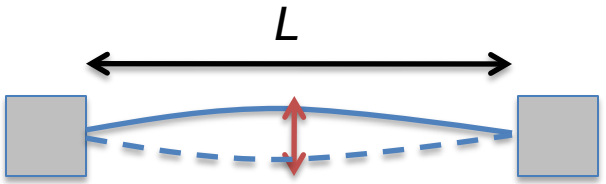


Note: in this case, *f* is the downward force per unit length per force of string tension

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

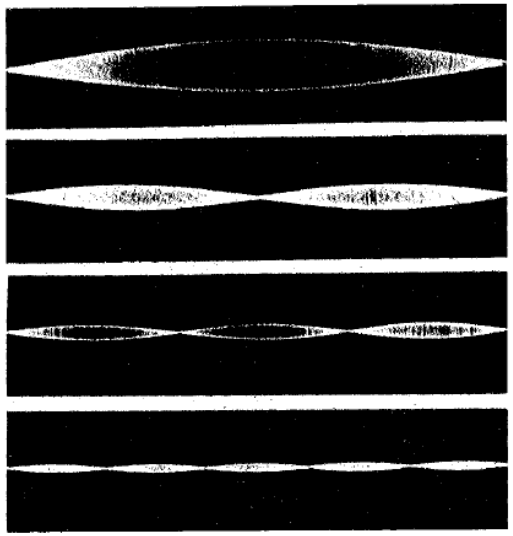


Fig. 6–1 Vibration of a string in various simple modes (n = 1, 2, 3, 5). (From D. C. Miller, The Science of Musical Sounds, Macmillan, New York, 1922.)

AP French, *Vibrations and Waves*, 1971

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

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

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 <u>wavevector</u>

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

Progressive Waves

- Consider the following trigonometric identity

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

- Applied to our previous displacement result…

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

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

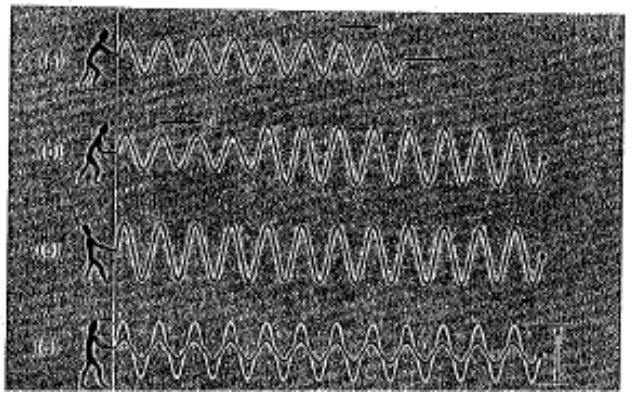
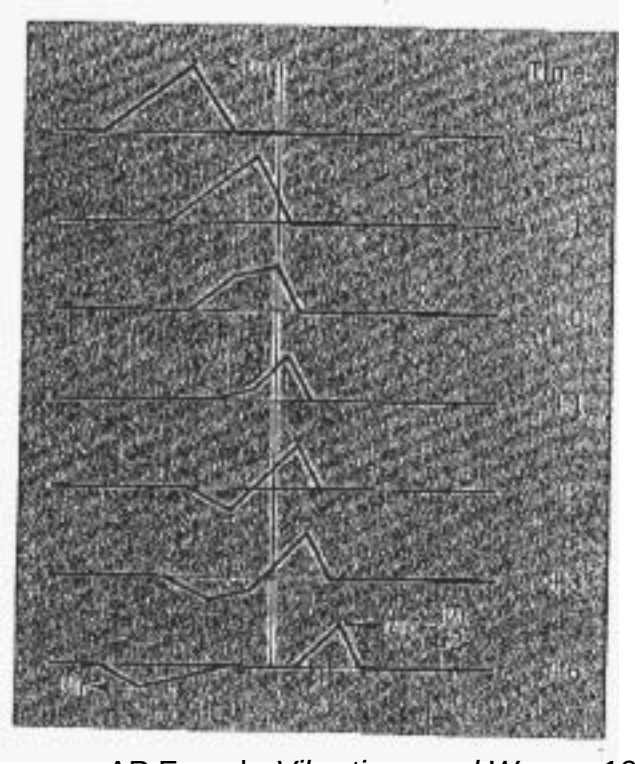


Fig. 7–1 (a) Traveling wave being generated. (b) Traveling wave plus reflected traveling wave. (c) Resultant standing wave (normal mode) at maximum amplitude. (d) Some standing wave as in (c) but at an instant when the displacements are much less than maximum.

AP French, *Vibrations and Waves*, 1971

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 reflect 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 moving wave $f_2(t - x/v_2)$



AP French, *Vibrations and Waves*, 19

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

$$f_2(t) = \frac{2v_2}{v_2 + v_1} f_1(t)$$

$$g(t) = \frac{v_2 - v_1}{v_2 + v_1} f_1(t)$$

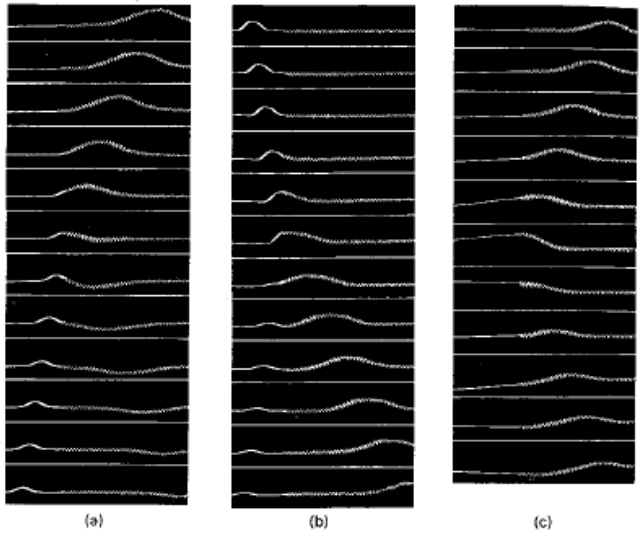


Fig. 8–4 Photographs of pulses encountering the boundary between two media. (a) Pulse passing from a light spring (right) to a heavy spring. At the junction the pulse is partially transmitted and partially reflected. You will note that the reflected pulse is upside down. (b) Pulse passing from a heavy 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.)

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

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 rate of energy flow

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

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

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

Interface Impedance and Transmission

- Most commonly, an acoustic impedance is defined, as

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

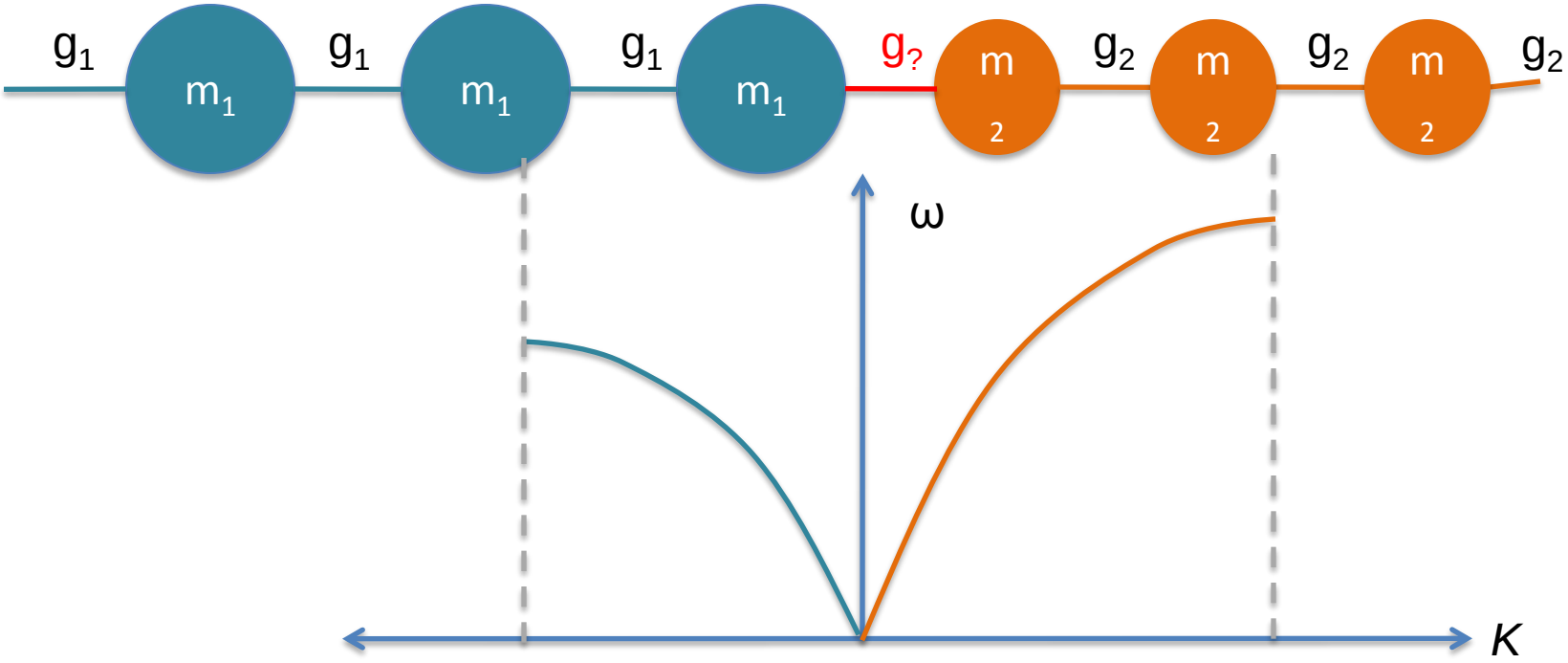
- Then, the normal-direction boundary transmittance $\tau_{b,1\text{-}2}$ becomes

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

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

Discrete Masses

- Now consider the problem of two different atomic chains joined at an interface



Dispersion for Point Masses

- Recall our earlier relation

$$\omega_{n,i}(K_{n,i}) = \sqrt{\frac{2g_i(1-\cos K_{n,i}a_i)}{m_i}} = 2\sqrt{\frac{g_i}{m_i}}\left|\sin(\tfrac{1}{2}K_{n,i}a_i)\right|$$

  - where $K_{n,i} = \dfrac{2\pi n}{a_i N}$

- Near the origin (K→0), the dispersion is linear (as for the continuous string→const velocity)
- Near the shortest wavelengths (longest wavevectors), the curve becomes flat (low velocity)
- This is the vibrational eigenspectrum

Acoustic Mismatch for Discrete Mass Chains

- The group velocity in a given chain portion *i* is

$$v_{g,i}(K_i;\omega) = \frac{\partial\omega}{\partial K} = a_i\sqrt{\frac{g_i}{4m_i}}\cos\left(\frac{K_i a_i}{2}\right)$$

- Then an acoustic impedance of each chain can be defined as

$$Z_i\left(K_i;\omega\right) = \frac{g_i}{v_{g,i}}$$

- And an acoustic boundary transmittance can be calculated as

$$\tau_b(\omega) = \frac{4Z_1Z_2}{\left(Z_1+Z_2\right)^2}$$

Outline

- Introduction
- Lattice vibrations and phonons
- The vibrating string
  - Interfaces between dissimilar strings: acoustic mismatch
  - Discrete masses and the vibrational eigenspectrum
- General thermal transport theory

General Analysis of Heat Flow

- Consider a perfect, free-standing quantum wire over which a small temperature difference (δT) is applied
- In the absence of scattering, the net heat flow through the wire would be

Sum over all polarizations or bands

$$J_{Qx,net} = \sum_{p} \int_{0}^{\infty} \frac{v_{g,p}\left[E_{i,p} - \mu\right]}{2\pi} \delta f_i^{o}(\delta T) dk_x$$

Carrier wavevector, $p_x/\hbar$

K-Space to Frequency Space

- For 1D problems, the conversion is very simple because $v_g = d\omega/dk$

$$J_{Qx,net} = \sum_p \int_0^\infty \frac{\left[E_{i,p} - \mu\right]}{2\pi} \delta f_i^o(\delta T) \frac{d\omega}{dk_x} dk_x$$

$$= \sum_p \int_0^\infty \frac{\left[E_{i,p} - \mu\right]}{2\pi} \delta f_i^o(\delta T) d\omega$$

- Convenient outcome: the flux term no longer contains velocity explicitly

Phonon Thermal Conductance and Conductivity

- The thermal conductance ($G_Q = J_{Q,net}/\delta T$) under perfect transmission is independent of the wire length L

$$G_Q(T) = \frac{k_B T}{2\pi\hbar} \sum_p \int_0^\infty f_{BE}^o(x)^2 e^x x^2 dx$$

$$= \sum_p \frac{k_B^2 \pi}{6\hbar} T$$

Bose-Einstein statistics for phonons

- Each active branch produces a 'quantum' of thermal conductance (Rego and Kirczenow, Phys Rev Lett **81** 232, 1998)
  - Measured by Schwab et al. (Nature **404** 974, 2000)
- …and the thermal conductivity is therefore length-dependent (and temperature-dependent)

$$\kappa = \frac{\text{rate of heat flow (W)}}{\text{area (m}^2\text{) x temperature gradient (K/m)}} = \frac{LG_Q(T)}{A}$$

  - Where A is the effective cross-sectional area

The Quantum of Thermal Conductance

- Consider only a single acoustic phonon polarization and perform the conductance integral (from $\omega = 0$ to ∞)
- Rego and Kirczenow [Phys Rev Lett 81 232 (1998)] showed that each such branch provides a fixed thermal conductance that depends only on the temperature T

$$G_Q^* = \frac{k_B^2 \pi}{6\hbar} T = T \times 9.462 \times 10^{-13} \ \frac{\mathrm{W}}{\mathrm{K}^2}$$

- This quantity represents the **maximum** thermal conductance that a given phonon branch can provide, even with none of the usual loss mechanisms (e.g., impurity scattering, umklapp scattering)

Inclusion of Scattering

- General expression for conductance

Scattering (transmission) function

$$G_Q(T) = \sum_p \int_0^\infty \frac{v_{g,p} k_B}{c_d} f_i^o(T)^2 e^{\frac{E_{i,p}(k)-\mu}{k_B T}} \left[\frac{E_{i,p}(k)-\mu}{k_B T}\right]^2 \Xi_p(k) k^{d-1} dk$$

- Some postulates concerning internal device (diffusive) scattering
  - Expect transmission function Ξ(k) to decrease with increasing device length *L* and with decreasing scattering length λ(k)
  - Rudimentary model:

$$\Xi(k) = \frac{\lambda(k)}{L}$$

Model Correction

- Problem: as L→0, rudimentary model predicts Ξ→∞ (not physical)
- Correction*:

$$\Xi(k) = \frac{\lambda(k)}{\lambda(k) + L}$$

- Corrected model satisfies quantum conductance criterion, Ξ→1 as L→0, and also reverts to the rudimentary model for large L

*see Datta, Electronic Transport in Mesoscopic Systems Cambridge University Press, Cambridge, 1995

Diffusive Heat Flow

- 3D thermal conductivity κ from kinetic theory:

$$\kappa = \frac{1}{3} C_v v_g \lambda$$

- Apply Landauer form of conductance with v_g=const and $\Xi=\lambda/(\lambda+L)\approx\lambda/L$ (assumed << 1) for phonons (μ=0)

$$\kappa = \frac{LG_Q}{A_d} = \frac{L}{A_d} v_g \frac{\lambda}{L} \underbrace{\sum_p \int \frac{E_{i,p}(k)}{(2\pi)^d} \frac{\partial f_i^o}{\partial T} d\mathbf{k}}_{C_v} = \frac{C_v v_g \lambda}{A_d}$$

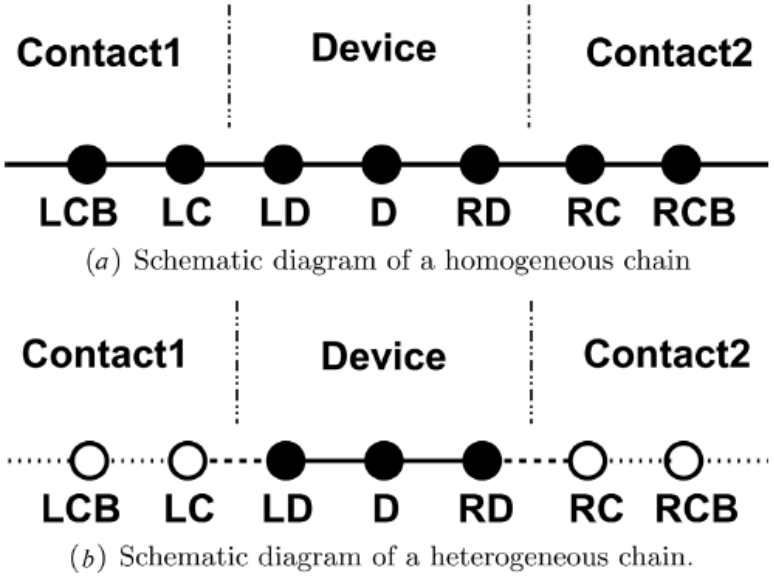
Constant that depends on dimensionality

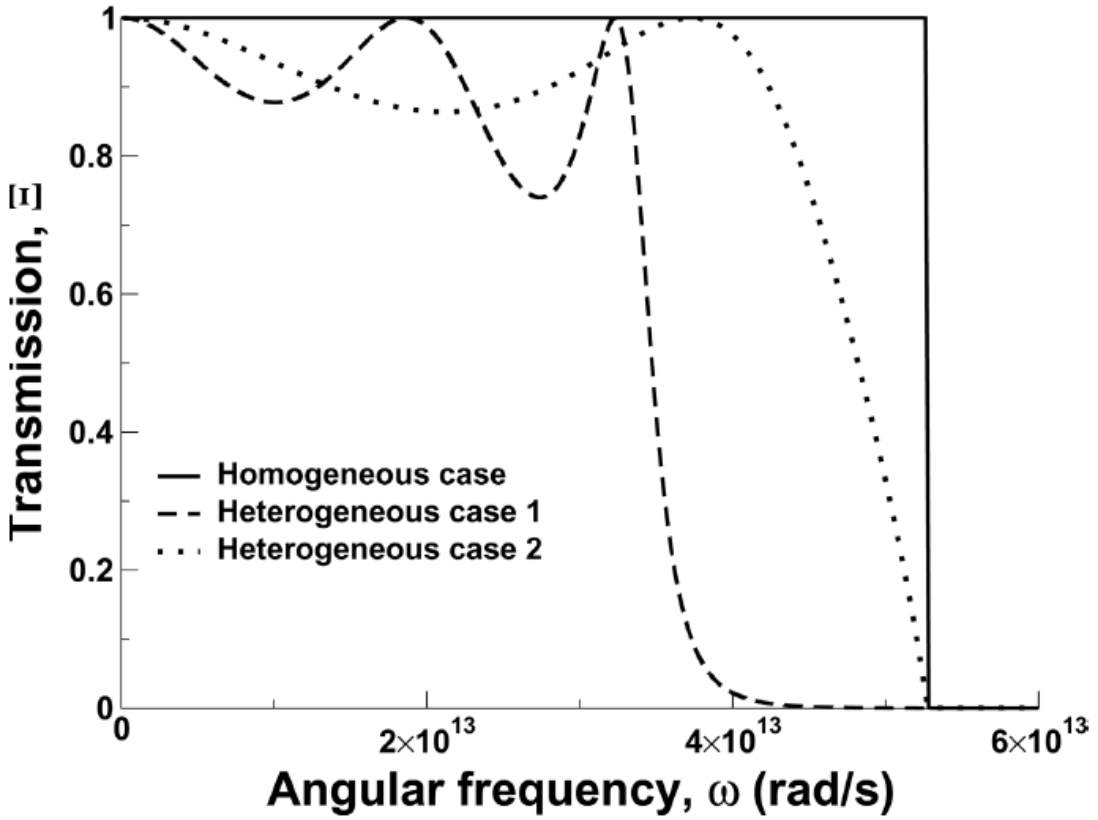
Example: Carbon Nanotube Thermal Conductivity

$$\kappa = \frac{1}{3} C_v v_g \lambda$$

- $v_g \approx 1 \times 10^4$ m/s (for Si, $v_g \approx 0.6 \times 10^4$ m/s)
- $\lambda > 1000$ nm at room temp (for Si, $\lambda \approx 50$ nm)
- $\kappa \approx 5000$ W/mK at room temp (for Si, $\kappa \approx 150$ W/mK)
- Largest factor is the increase in mean free path (λ) (Berber et al. *Phys. Rev. Lett.* 84 4613, 2000)

Conclusion: A 'Toy' Transmission Function

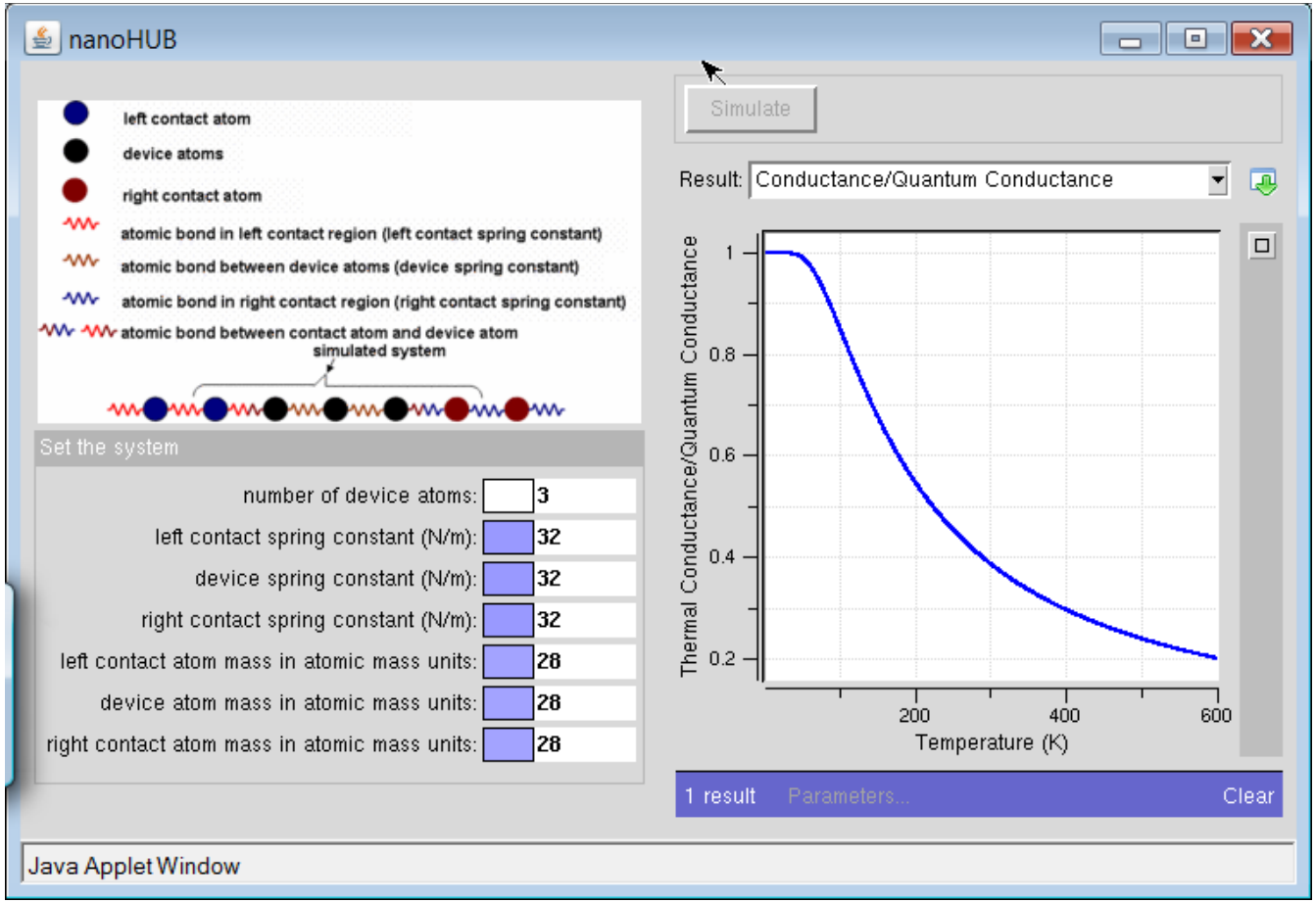




See https://www.nanohub.org/tools/greentherm

Zhang et al. Num. Heat Transfer B 51 333 (2007)

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



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