Thermal conductance of interfaces

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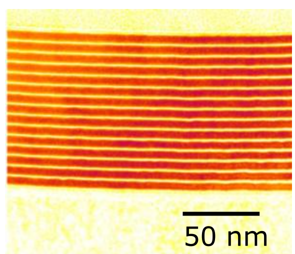
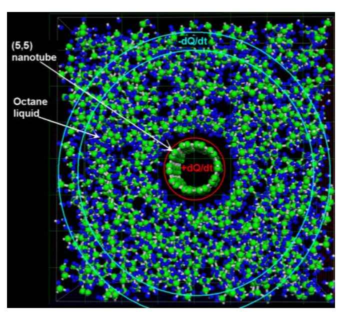
lecture for ECE 598EP, Hot Chips: Atoms to Heat Sinks

Interfaces are critical at the nanoscale

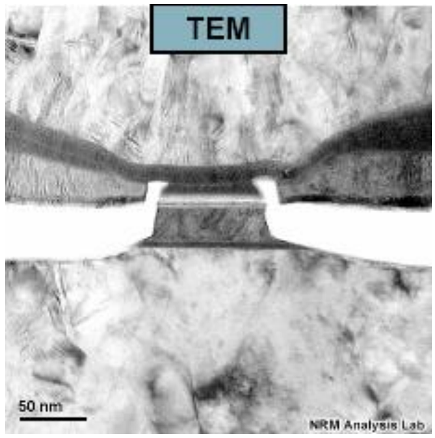
- Low thermal conductivity in nanostructured materials
  - improved thermoelectric energy conversion
  - improved thermal barriers
- High thermal conductivity composites and suspensions





Interfaces are critical at the nanoscale

- High power density devices
  - solid state lighting
  - high speed electronics
  - nanoscale sensors

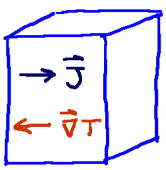


Micrograph of tunneling magnetoresistive sensor for 120 GB drives, M. Kautzky (Seagate)

Interface thermal conductance

- Thermal conductivity Λ is a property of the continuum

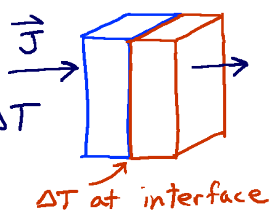
$\vec{J} = -\Lambda \vec{\nabla} T$



$$\Lambda = \frac{1}{3 V k_B T^2} \int_0^\infty \langle \vec{\jmath}(t) \cdot \vec{\jmath}(0) \rangle \, dt$$

- Thermal conductance (per unit area) *G* is a property of an interface

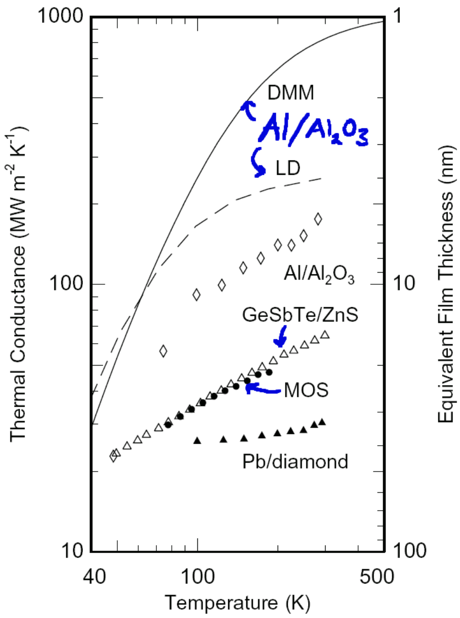
$J = G \Delta T$



$$G = \frac{1}{A k_B T^2} \int_0^\infty \langle q(t) q(0) \rangle \, dt$$

Interface thermal conductance (2001)

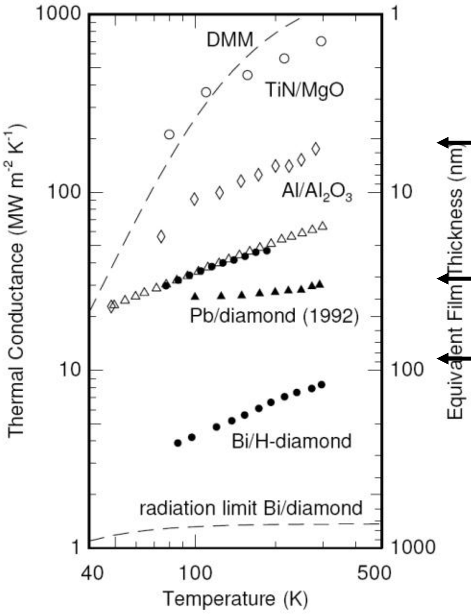
- Observations (2001) span a very limited range
  - Al/sapphire → Pb/diamond
  - no data for hard/soft
- lattice dynamics (LD) theory by Stoner and Maris (1993)
- Diffuse mismatch (DMM) theory by Swartz and Pohl (1987)



Acoustic and diffuse mismatch theory

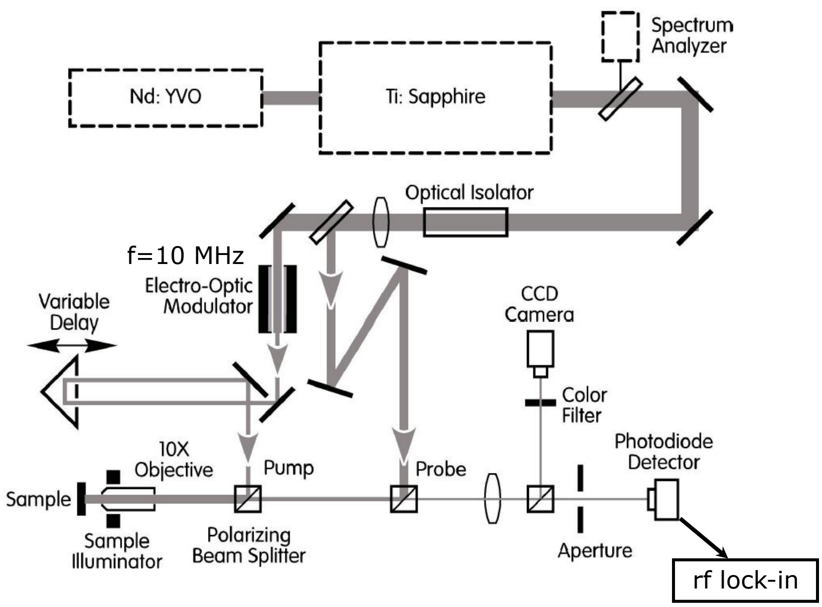
- Acoustic mismatch (AMM)
  - perfect interface: average transmission coefficient <*t*> given by differences in acoustic impedance, $Z=\rho v$
  - lattice dynamics (LD) incorporates microscopics
- Diffuse mismatch (DMM)
  - disordered interface: <*t*> given by differences in densities of vibrational states
- Predicted large range of *G* not observed (2001)
- For similar materials, scattering decreases *G*
- For dissimilar materials, scattering increases *G*

2005: Factor of 60 range at room temperature



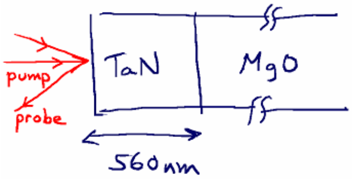
- W/Al_2O_3
- Au/water
- $PMMA/Al_2O_3$
- nanotube/alkane

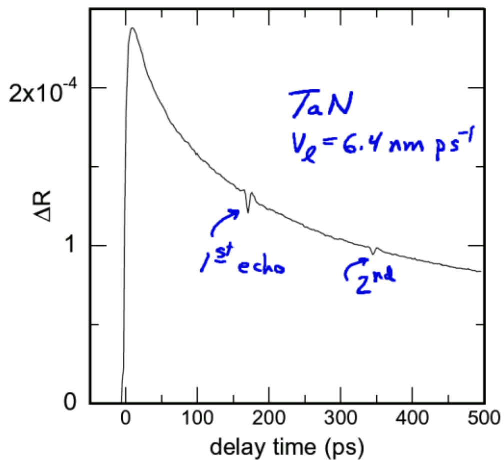
Modulated pump-probe apparatus



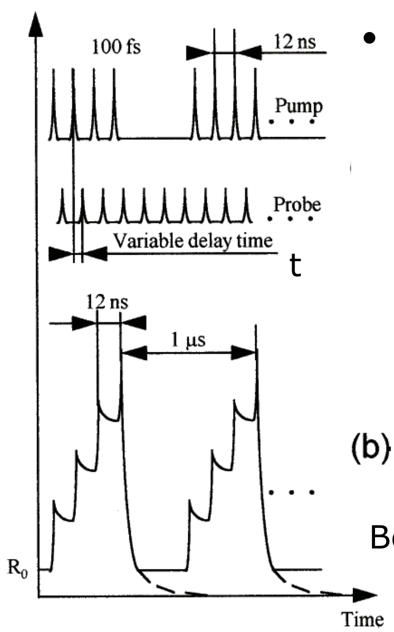
psec acoustics and time-domain thermoreflectance

- Optical constants and reflectivity depend on strain and temperature
- Strain echoes give acoustic properties or film thickness
- Thermoreflectance gives thermal properties





Modulated pump-probe



(b)

- four times scales:
  - pulse duration, 0.3 ps
  - pulse spacing, 12.5 ns
  - modulation period, 100 ns
  - time-delay, t

Bonello et al. (1998)

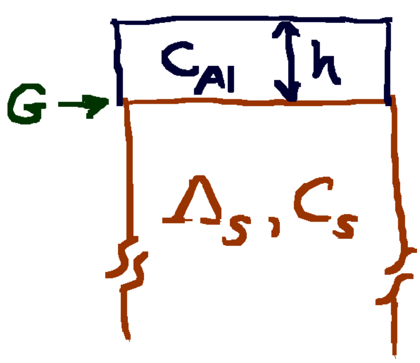
Analytical solution to 3D heat flow in an infinite half-space

- spherical thermal wave $$g(r) = \frac{\exp(-qr)}{2\pi\Lambda r} \qquad q^2 = (i\omega/D)$$
- Hankel transform of surface temperature $$G(k) = \frac{1}{\Lambda(4\pi^2k^2 + q^2)^{1/2}}$$
- Multiply by transform of Gaussian heat source and take inverse transform $$P(k) = A\exp(-\pi^2k^2w_0^2/2)$$ $$\theta(r) = 2\pi\int_0^\infty P(k)G(k)J_0(2\pi kr)\ k\ dk$$
- Gaussian-weighted surface temperature

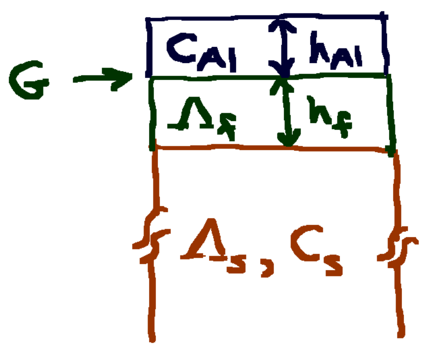
$$\Delta T = 2\pi A\int_0^\infty G(k)\exp\left(-\pi^2k^2\left(w_0^2 + w_1^2\right)/2\right)\ k\ dk$$

Two basic types of experiments

- thermal conductivity of bulk samples and thermal conductance of interfaces



- thermal conductivity of thin films



Iterative solution for layered geometries

$$\begin{pmatrix} B^+ \\ B^- \end{pmatrix}_n = \frac{1}{2\gamma_n} \begin{pmatrix} \exp(-u_n L_n) & 0 \\ 0 & \exp(u_n L_n) \end{pmatrix}$$

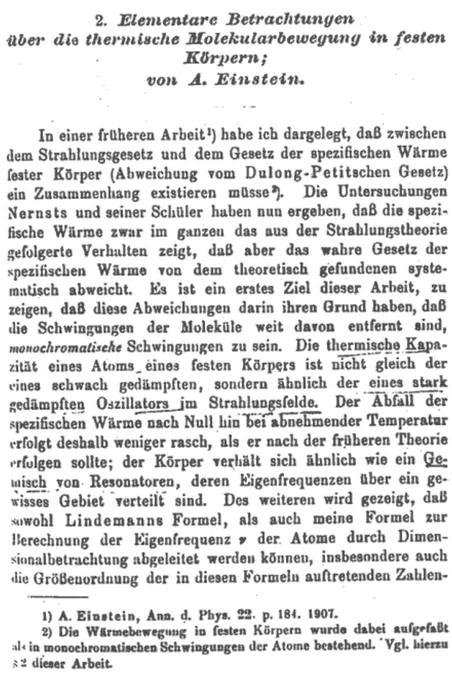
$$\times \begin{pmatrix} \gamma_n + \gamma_{n+1} & \gamma_n - \gamma_{n+1} \\ \gamma_n - \gamma_{n+1} & \gamma_n + \gamma_{n+1} \end{pmatrix} \begin{pmatrix} B^+ \\ B^- \end{pmatrix}_{n+1}$$

$$u_n = \left(4\pi^2 k^2 + q_n^2\right)^{1/2} \qquad q_n^2 = \frac{i\omega}{D_n} \qquad \gamma_n = \Lambda_n u_n$$

$$G(k) = \left(\frac{B_1^+ + B_1^-}{B_1^- - B_1^+}\right) \frac{1}{\gamma_1}$$

Einstein (1911)

- coupled the Einstein oscillators to 26 neighbors
- heat transport as a random walk of thermal energy between atoms; time scale of ½ vibrational period
- did not realize waves (phonons) are the normal modes of a crystal

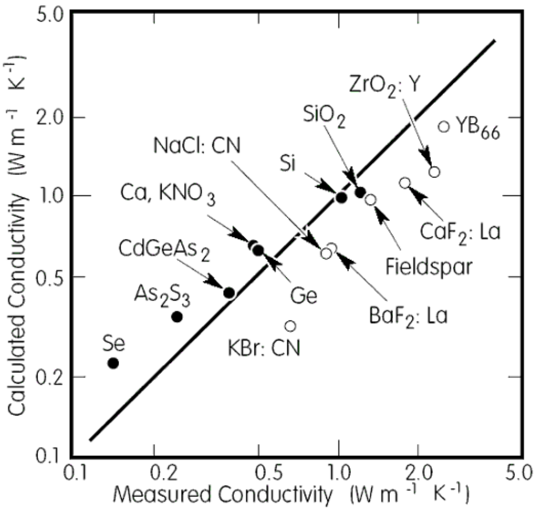


2. *Elementare Betrachtungen über die thermische Molekularbewegung in festen Körpern;*
von A. Einstein.

In einer früheren Arbeit[1]) habe ich dargelegt, daß zwischen dem Strahlungsgesetz und dem Gesetz der spezifischen Wärme fester Körper (Abweichung vom Dulong-Petitschen Gesetz) ein Zusammenhang existieren müsse[2]). Die Untersuchungen Nernsts und seiner Schüler haben nun ergeben, daß die spezifische Wärme zwar im ganzen das aus der Strahlungstheorie gefolgerte Verhalten zeigt, daß aber das wahre Gesetz der spezifischen Wärme von dem theoretisch gefundenen systematisch abweicht. Es ist ein erstes Ziel dieser Arbeit, zu zeigen, daß diese Abweichungen darin ihren Grund haben, daß die Schwingungen der Moleküle weit davon entfernt sind, *monochromatische* Schwingungen zu sein. Die thermische Kapazität eines Atoms eines festen Körpers ist nicht gleich der eines schwach gedämpften, sondern ähnlich der eines stark gedämpften Oszillators im Strahlungsfelde. Der Abfall der spezifischen Wärme nach Null hin bei abnehmender Temperatur erfolgt deshalb weniger rasch, als er nach der früheren Theorie erfolgen sollte; der Körper verhält sich ähnlich wie ein Gemisch von Resonatoren, deren Eigenfrequenzen über ein gewisses Gebiet verteilt sind. Des weiteren wird gezeigt, daß sowohl Lindemanns Formel, als auch meine Formel zur Berechnung der Eigenfrequenz ν der Atome durch Dimensionalbetrachtung abgeleitet werden können, insbesondere auch die Größenordnung der in diesen Formeln auftretenden Zahlen-

1) A. Einstein, Ann. d. Phys. 22. p. 184. 1907.
2) Die Wärmebewegung in festen Körpern wurde dabei aufgefaßt als in monochromatischen Schwingungen der Atome bestehend. Vgl. hierzu § 2 dieser Arbeit.

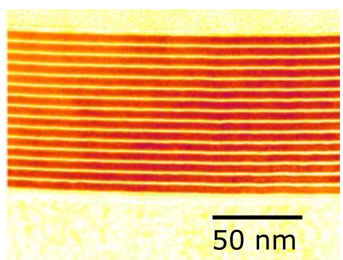
Works well for homogeneous disordered materials

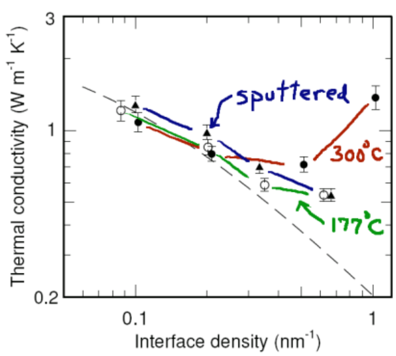


- ● amorphous
- ○ disordered crystal

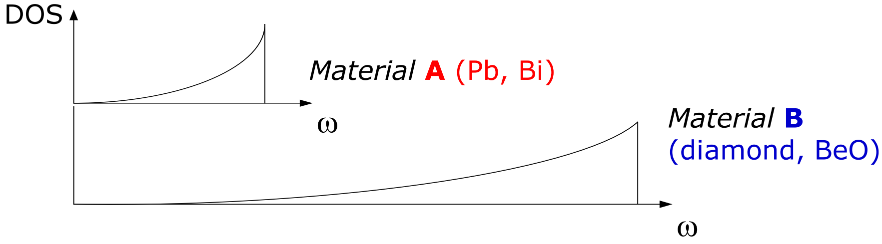
W/Al_2O_3 nanolaminates

- room temperature data
- sputtered in pure Ar
- atomic-layer deposition at 177 and 300 °C, S. George (U. Colorado)
- G = 220 MW m^{-2} K^{-1}





Interfaces between highly dissimilar materials



- high temperature limit of the radiation limit

$$G = \frac{\pi}{3} \frac{k_b \nu_{max}^3}{v_D^2}$$

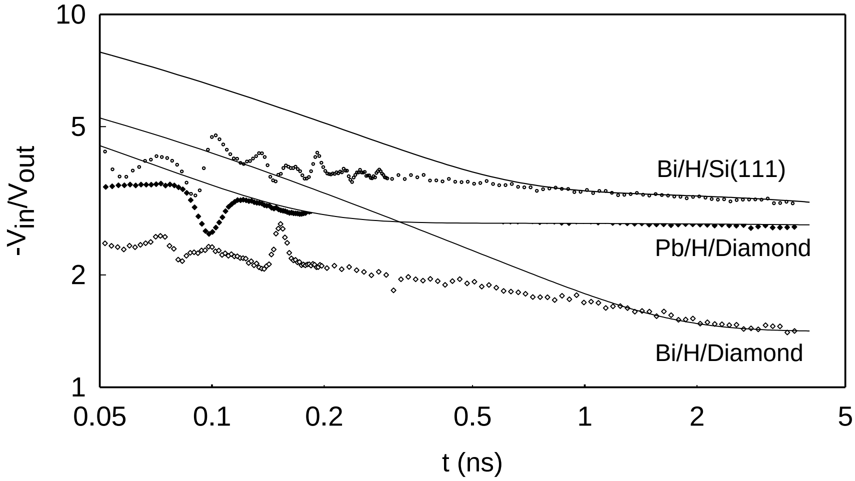
ν_{max} : vibrational cutoff frequency of material A

(ν_{max} = 1.8 THz for Bi, 2.23 THz for Pb)

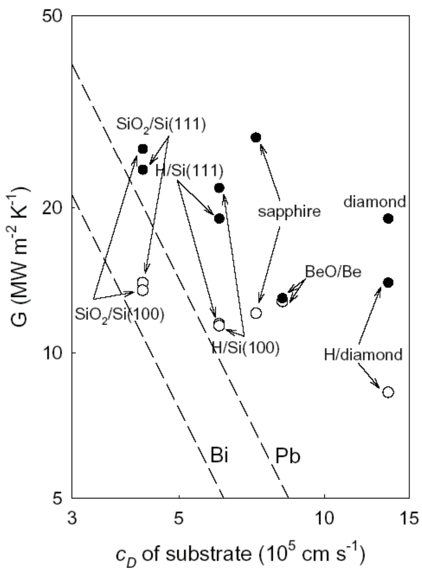
v_D : Debye velocity of material B

R. J. Stoner and H. J. Maris, *Phys.Rev.B* **48**, 22, 16373 (1993)

Thermoreflectance data for Bi and Pb interfaces

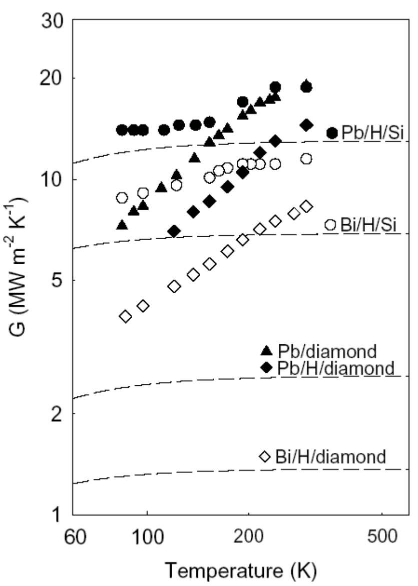


Room temperature thermal conductance



- Pb and Bi show similar behavior. Electron phonon coupling is not an important channel.
- Weak dependence on Debye velocity of the substrate.
- Pb/diamond a factor of two smaller than Stoner and Maris but still far too large for a purely elastic process.

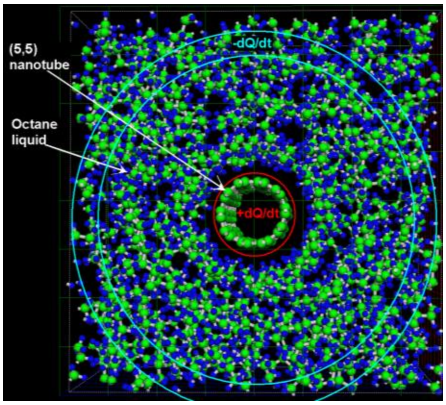
Temperature dependence of the conductance



- Excess conductance has a linear temperature dependence (not observed by Stoner and Maris).
- Suggests inelastic (3-phonon?) channel for heat transport

Carbon nanotube/alkane interfaces

- Experiment: nanotube suspension in surfactant (SDS) micelles in D_2O (with M. Strano)
- Computation by P. Keblinski: nanotube in octane

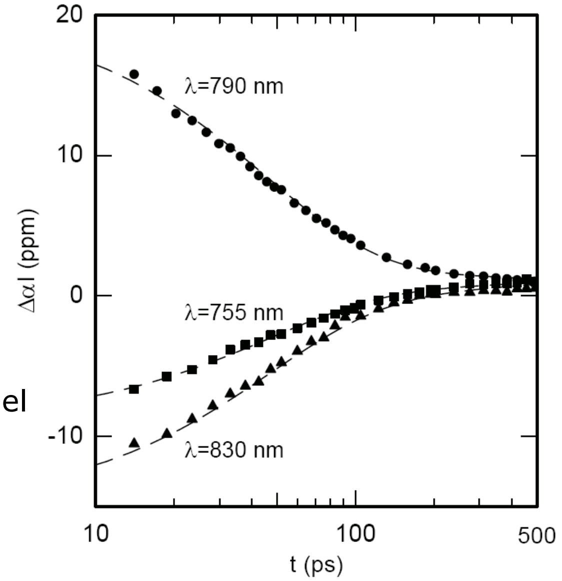


Transient absorption

- Optical absorption depends on temperature of the nanotube
- Cooling rate gives interface conductance

  G = 12 MW m^{-2} K^{-1}

- MD suggests channel is low frequency squeezing and bending modes strongly coupled to the fluid.



Critical aspect ratio for fiber composite

- Isotropic fiber composite with high conductivity fibers (and infinite interface conductance)

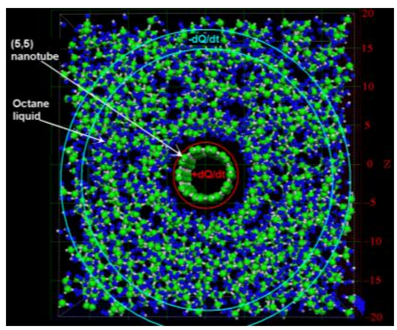
$$\Lambda_c = \frac{1}{3} V_f \Lambda_{NT}$$

- But this conductivity if obtained only if the aspect ratio of the fiber is high

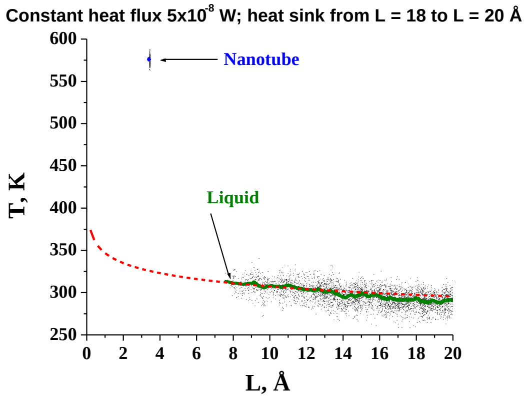
$$3\left(\frac{\Lambda_{NT}}{rG}\right)^{1/2} \approx 2000$$

Simulation: constant heat flux

- Pour heat into the tube and remove from the octane liquid
- G = 25 MW m^{-2} K^{-1}



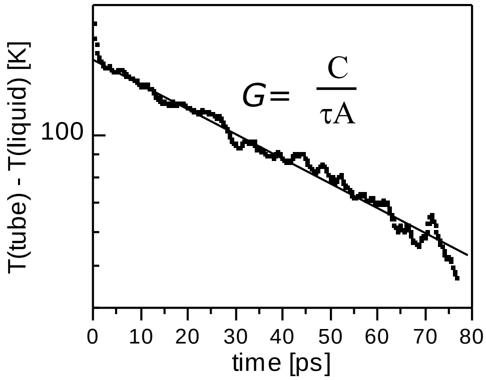
Constant heat flux 5×10^{-8} W; heat sink from L = 18 to L = 20 Å



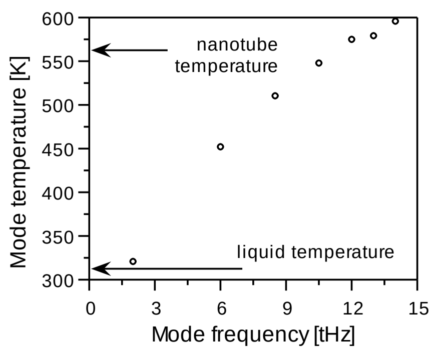
Simulation: relaxation time

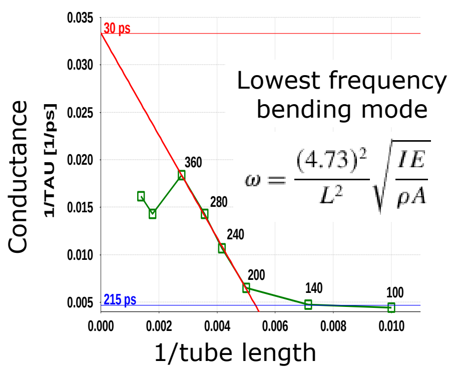
- Mimic the experiment: heat nanotube suddenly and let system equlibrate
- Use heat capacity to convert time constant to conductance. In the limit of long tubes:

  G = 22 MW m^{-2} K^{-1}



Simulation: Mechanisms for interface heat conduction

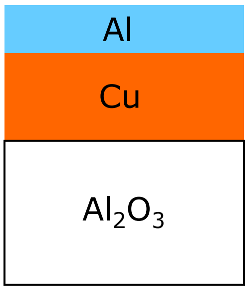


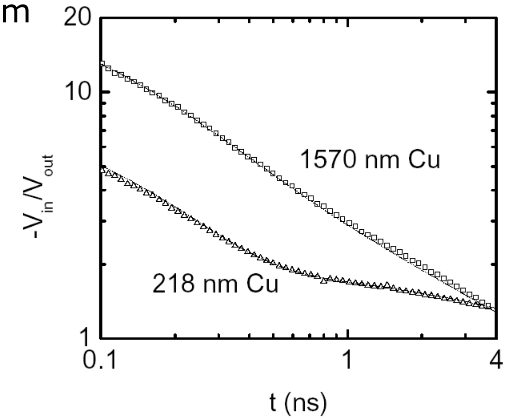


- Carbon nanotubes have a small number of low frequency modes associated with bending and squeezing. Only these modes can couple strongly with the liquid.

Metal-metal conductance: Al/Cu/sapphire samples and thermoreflectance data

- Two samples: Cu thicknesses of 218 nm and 1570 nm



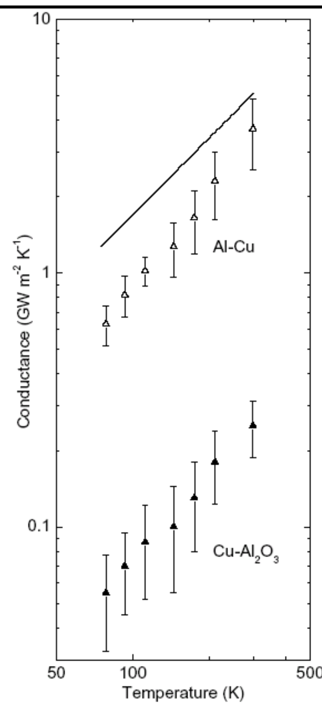


Thermal Conductance of Al-Cu

- Metal-metal interface conductance is huge

  4 GW/m^2-K at 298 K

- And increases linearly with temperature



Diffuse mismatch model for electrons

- Transmission coefficient Γ for electron of energy E from 1→2; v(E) is velocity; D(E) is density of states.

$$\Gamma_1(E) = \int_0^{\pi/2} \frac{v_2(E)D_2(E)}{v_1(E)D_1(E) + v_2(E)D_2(E)} \cos\theta \sin\theta d\theta$$

- Thermal conductance *G*; *N(E,T)* is occupation

$$G = \frac{1}{2} v_1(E_F)\Gamma_1(E_F) \int_0^{\infty} E \frac{dN_1(E,T)}{dT} dE$$

- Simplify, define Z = $\gamma v_F T$

$$G = \frac{Z_1 Z_2}{4(Z_1 + Z_2)}$$

Thermal Conductance of Al-Cu

- Diffuse mismatch model for electrons using

$$G = \frac{Z_1 Z_2}{4(Z_1 + Z_2)}$$

$$Z_{Al} = 0.18T, \; Z_{Cu} = 0.11T$$

in units of GW/m^2-K