Thermal conductance of interfaces

David G. Cahill

_Frederick Seitz Materials Research Lab and Department of Materials Science_

_University of Illinois, Urbana_

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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 $\Lambda$ is a property of the continuum

\[
\vec{J} = -\Lambda \nabla T
\]

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

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

\[
\vec{J} = G \Delta T
\]

\[
G = \frac{1}{A k_B T^2} \int_0^\infty \langle q(t)q(0) \rangle \, dt
\]
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, $Z = \rho \nu$
  - 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$_2$O$_3$
- Au/water
- PMMA/Al$_2$O$_3$
- nanotube/alkane

**Modulated pump-probe apparatus**

- Nd:YVO
- Ti: Sapphire
- Optical Isolator
- F=10 MHz
- Electro-Optic Modulator
- 10X Objective
- Pump
- Probe
- Sample Illuminator
- Polarizing Beam Splitter
- CCD Camera
- Color Filter
- Photodiode Detector
- rf lock-in
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

- 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
  \[ g(r) = \frac{\exp(-qr)}{2\pi Ar} \quad q^2 = (i\omega/D) \]

- Hankel transform of surface temperature
  \[ G(k) = \frac{1}{\Lambda(4\pi^2 k^2 + q^2)^{1/2}} \]

- Multiply by transform of Gaussian heat source and take inverse transform
  \[ P(k) = A \exp(-\pi^2 k^2 w_0^2/2) \]
  \[ \theta(r) = 2\pi \int_0^\infty P(k)G(k)J_0(2\pi kr) \, k \, dk \]

- Gaussian-weighted surface temperature
  \[ \Delta T = 2\pi A \int_0^\infty G(k) \exp\left(-\pi^2 k^2 \left(w_0^2 + w_1^2\right)/2\right) \, k \, dk \]

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

\[
\begin{pmatrix}
B^+ \\
B^-
\end{pmatrix}_n = \frac{1}{2\gamma_n} \begin{pmatrix}
\exp(-u_nL_n) & 0 \\
0 & \exp(u_nL_n)
\end{pmatrix}
\times \begin{pmatrix}
\gamma_n + \gamma_{n+1} & \gamma_n - \gamma_{n+1} \\
\gamma_n - \gamma_{n+1} & \gamma_n + \gamma_{n+1}
\end{pmatrix}
\begin{pmatrix}
B^+ \\
B^-
\end{pmatrix}_{n+1}
\]

\[u_n = \left(4\pi^2k^2 + q_n^2\right)^{1/2} \quad q_n^2 = \frac{i\omega}{\Lambda_n} \quad \gamma_n = \Lambda_n u_n\]

\[G(k) = \left(\frac{B^+_1 + B^-_1}{B^-_1 - B^+_1}\right) \frac{1}{\gamma_1}\]

Einstein (1911)

- coupled the Einstein oscillators to 26 neighbors
- heat transport as a random walk of thermal energy between atoms; time scale of \(\frac{1}{2}\) vibrational period
- did not realize waves (phonons) are the normal modes of a crystal

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Einstein (1911)

Über die thermische Molekularbewegung in festen Körperm; von A. Einstein.

Works well for homogeneous disordered materials

- amorphous
- disordered crystal

W/Al₂O₃ 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

\[ G = \frac{\pi}{3} k_b \nu_{\text{max}}^3 \frac{v_D^2}{v_{\text{D}}} \]

\( \nu_{\text{max}} \) : vibrational cutoff frequency of material A

\( v_D \) : Debye velocity of material B

\( v_{\text{max}} = 1.8 \) THz for Bi, 2.23 THz for Pb


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$_2$O (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
  \[ G = 12 \text{ MW m}^{-2}\text{ K}^{-1} \]
- 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

- $G = 25 \text{ MW m}^{-2} \text{ K}^{-1}$

Constant heat flux $5 \times 10^6 \text{ W}$; heat sink from $L = 18$ to $L = 20 \text{ Å}$
Simulation: relaxation time

- Mimic the experiment: heat nanotube suddenly and let system equilibrate
- Use heat capacity to convert time constant to conductance. In the limit of long tubes:
\[ G = \frac{C}{\tau A} \]

\[ T(tube) - T(liquid) \ [K] \]

time [ps]

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

![Diagram showing Al, Cu, and Al₂O₃ layers](image)

Thermal Conductance of Al-Cu

- Metal-metal interface conductance is huge
  - 4 GW/m²-K at 298 K
- And increases linearly with temperature
Diffuse mismatch model for electrons

- Transmission coefficient $\Gamma$ for electron of energy $E$ from $1 \rightarrow 2$; $v(E)$ 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