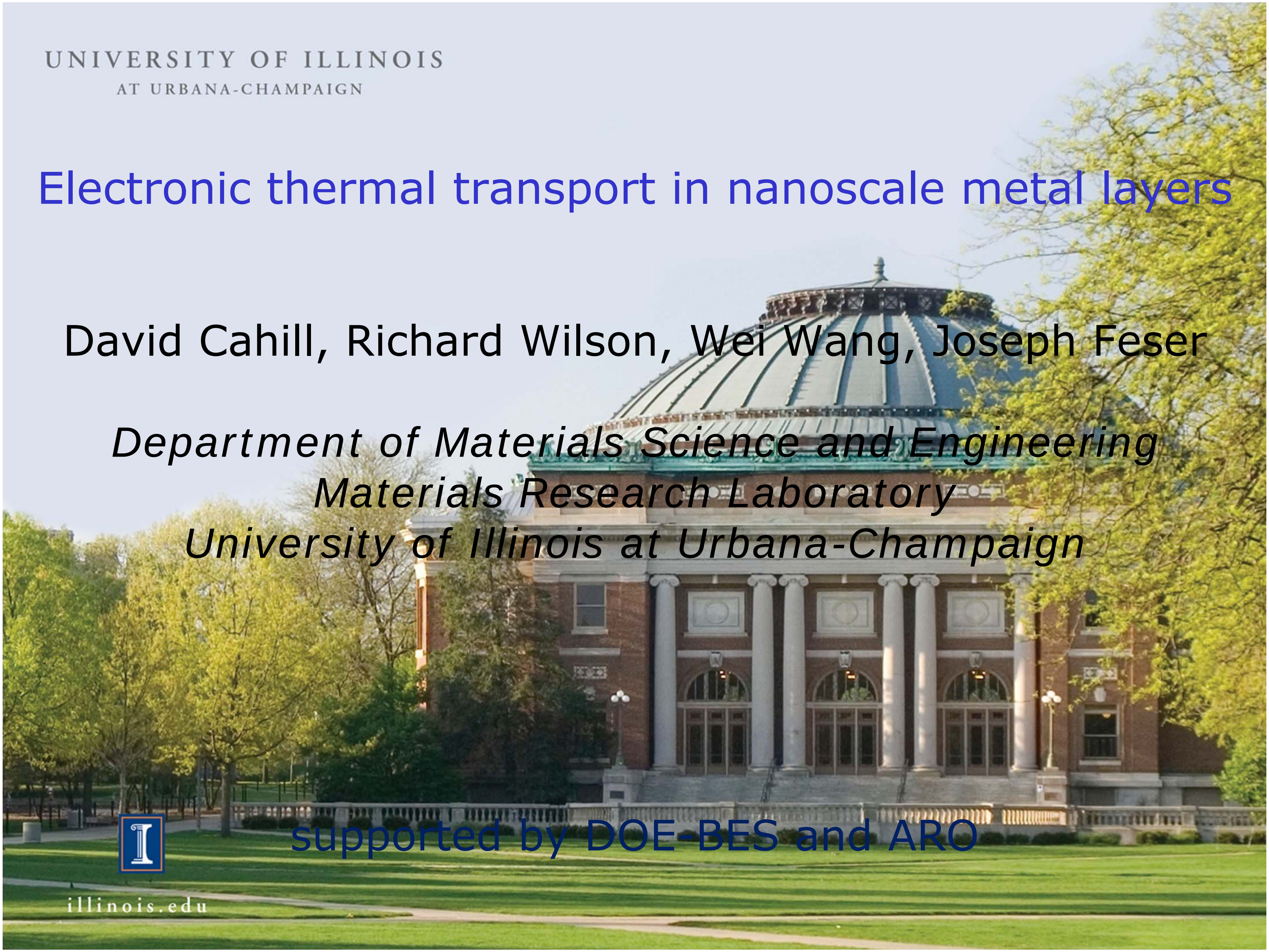

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Electronic thermal transport in nanoscale metal layers

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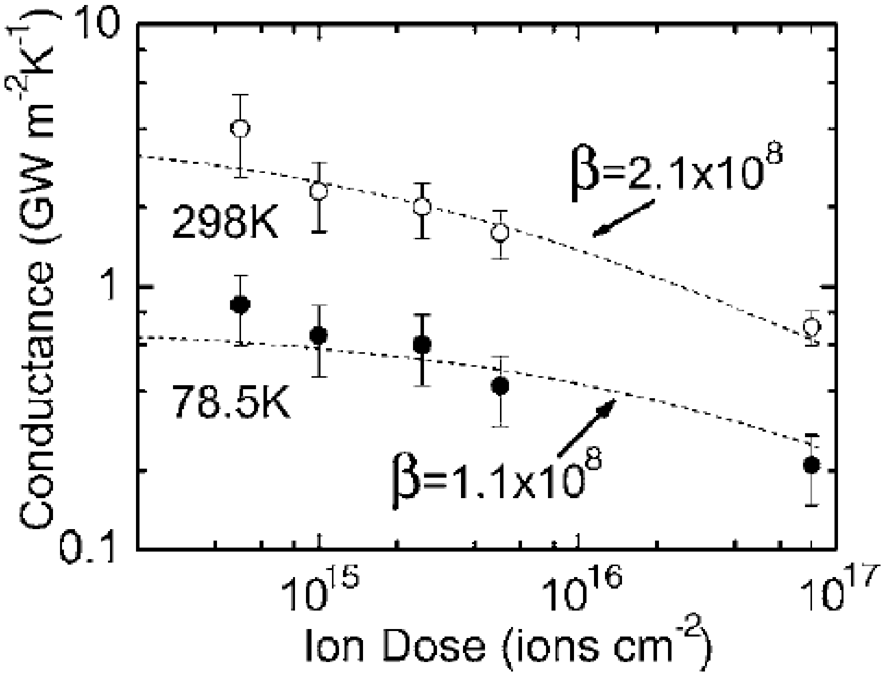
Outline

- Motivation
- Thermal conductance of metal-metal interfaces and the interfacial form of the Wiedemann-Franz law.
- Suppression of heat transport in metallic bilayers by weak-electron phonon coupling.
- In-plane thermal conductivity of thin metal layers using time-domain thermoreflectance (TDTR) with offset laser spots.

Motivation is mostly fundamental materials physics but there are "broader impacts"

- Thermal transport in phase change memory devices (liquid $Ge_2Sb_2Te_5$ is a metal).
- Emerging field of "spin caloritronics" (drive spin currents and magnetic polarization with a heat flux).
- Characterization of interfaces is always challenging. Thermal transport as an analytical tool.

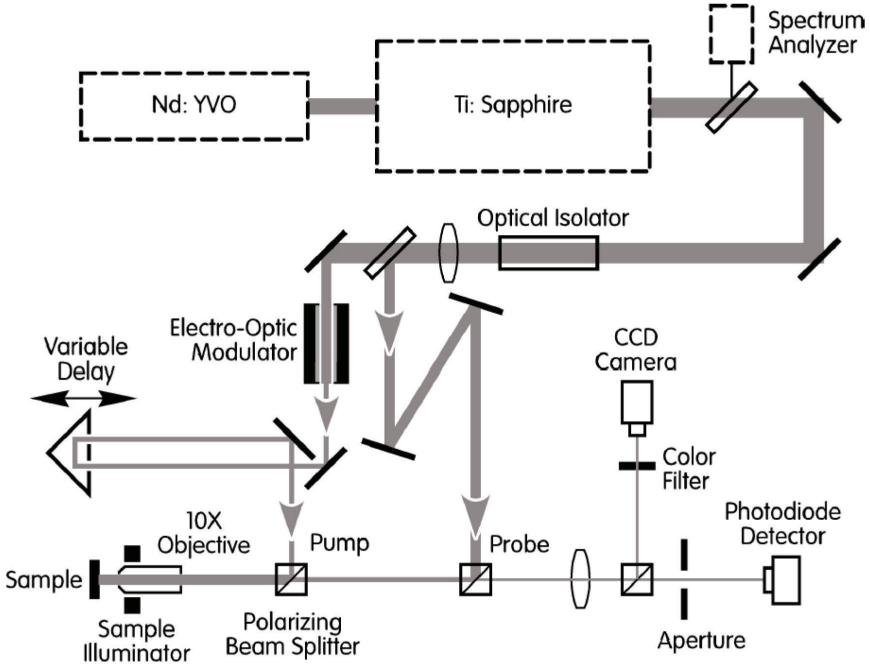
Thermal conductance of intermetallic layer formed by ion irradiation of a Al-Cu interfaces

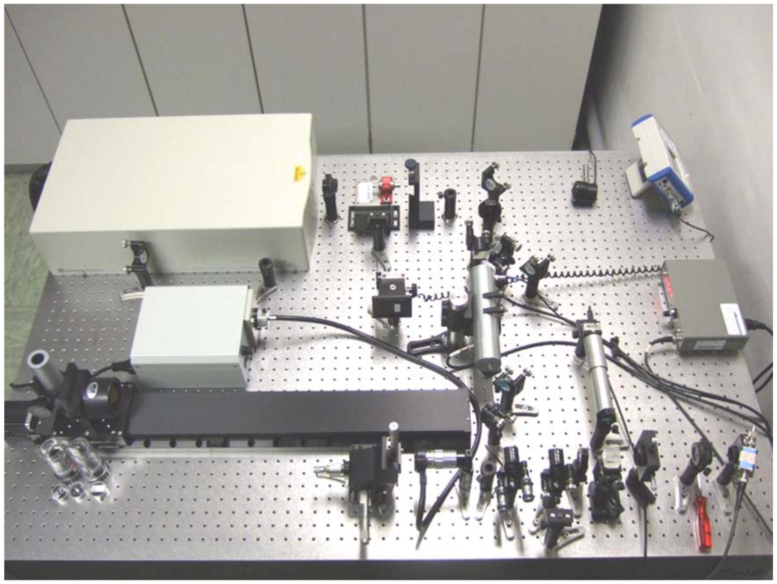


Gundrum *et al*. PRB (2005)

Time domain thermoreflectance since 2003

- Improved optical design
- Normalization by out-of-phase signal eliminates artifacts, increases dynamic range and improves sensitivity
- Exact analytical model for Gaussian beams and arbitrary layered geometries
- One-laser/two-color approach tolerates diffuse scattering





Clone built at Fraunhofer Institute for Physical Measurement, Jan. 7-8 2008

Are conductance of charge and heat at a metal-metal interface linearly related?

- Wiedemann-Franz law for interfaces

$$\frac{G(AR)}{T} = L$$

G = thermal conductance of the interface
AR = specific resistance of the interface
L = Lorenz number
(for degenerate electrons $L = L_0 = 24.5$ nΩ W K^{-2}

Typically, $G \sim 10$ GW m^{-2} K^{-1}
$AR \sim 1$ fΩ m^2

Prior work: Diffuse-mismatch model for electrons

- Transmission coefficient side 1→2

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

v= velocity, *D*=density of states

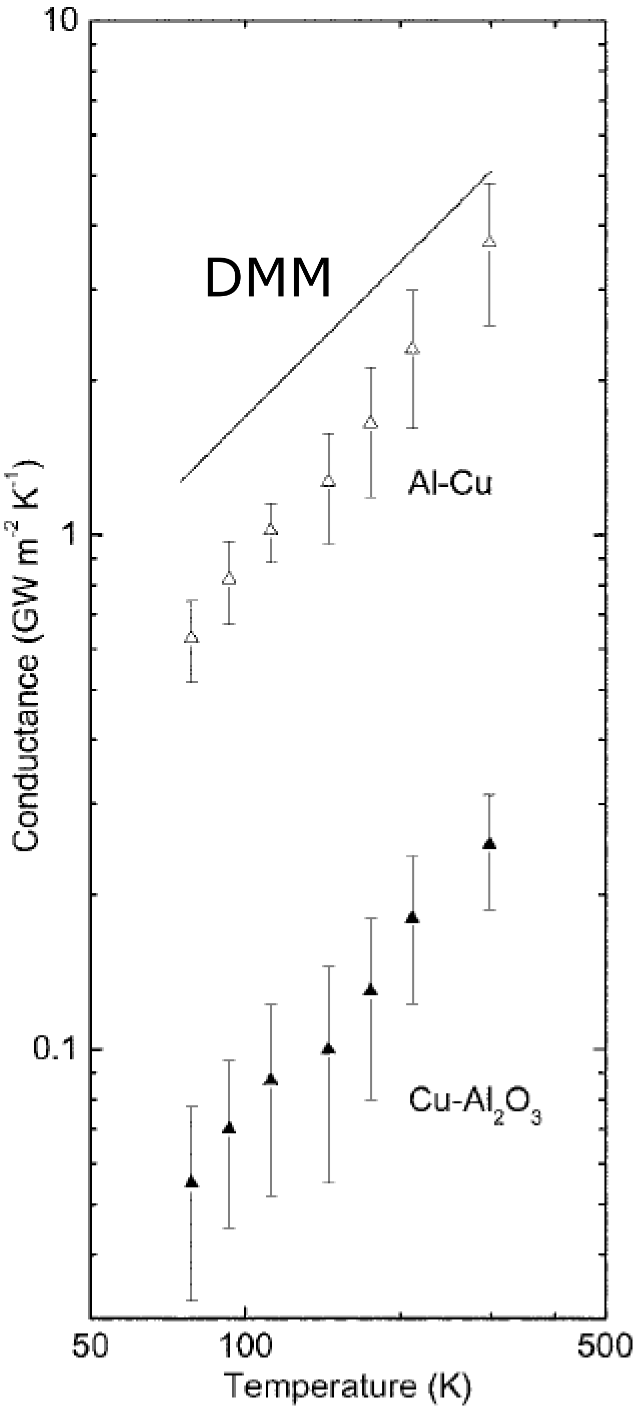
- For degenerate, isotropic Fermi surface

$$G = \frac{Z_1 Z_2}{4(Z_1 + Z_2)} \qquad Z = \gamma v_F T$$

γ = electronic heat capacity

v_F = Fermi velocity

Gundrum *et al*. PRB (2005)



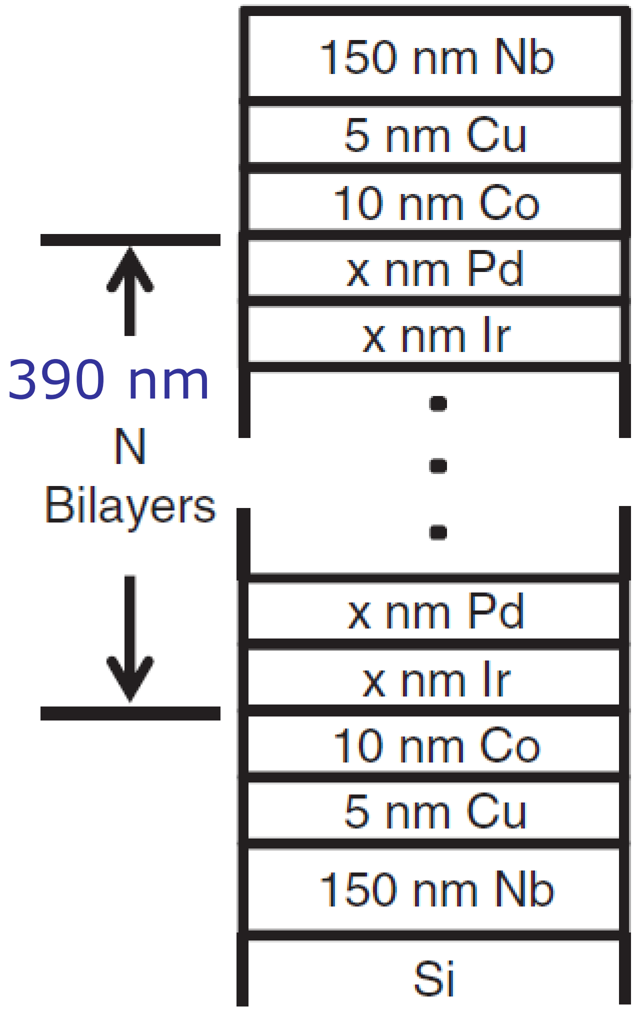
Hard to measure specific electrical resistance of a individual metal interface so use multilayers

- Samples prepared and electrical transport measurements by Jack Bass’s group at MSU.
    - Measurement at T=4 K using Nb superconducting leads

Acharyya *et al.*, APL (2009)

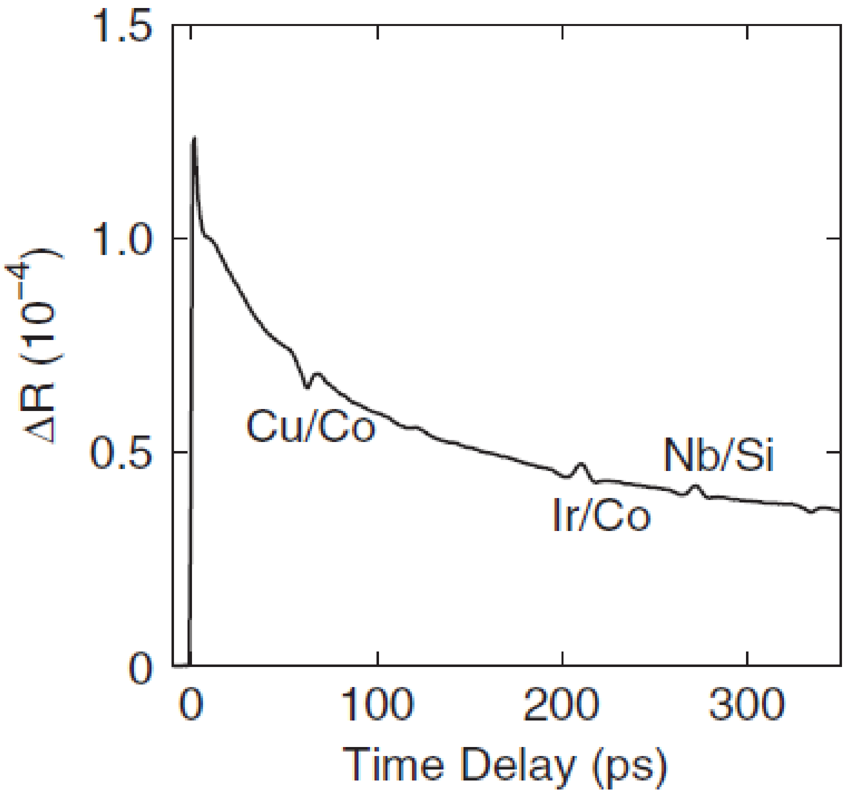
- TDTR measurement of thermal conductivity of the same samples at Illinois, 80 < T < 300 K

Wilson and Cahill, PRL (2012)

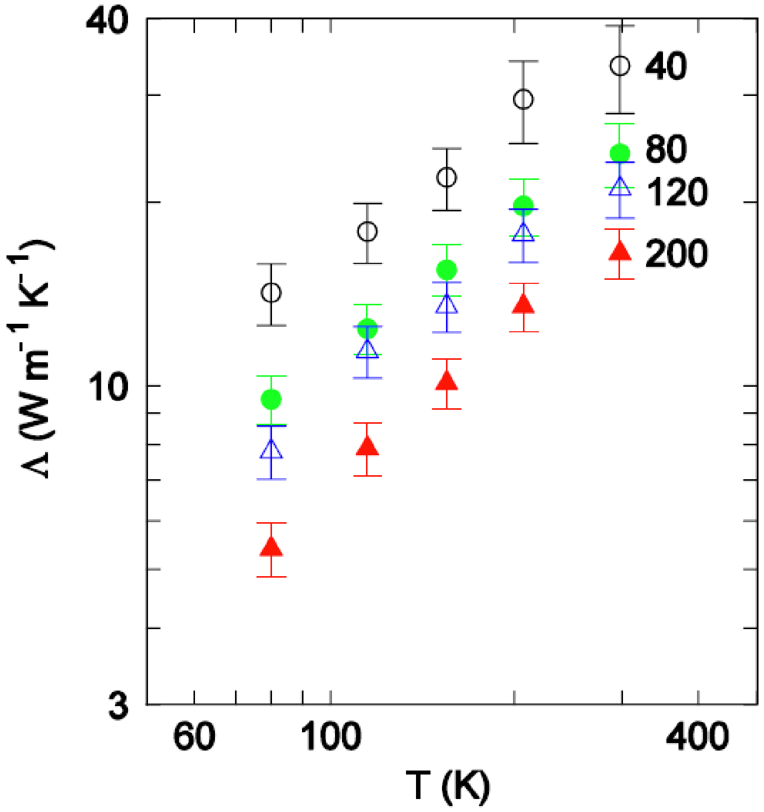


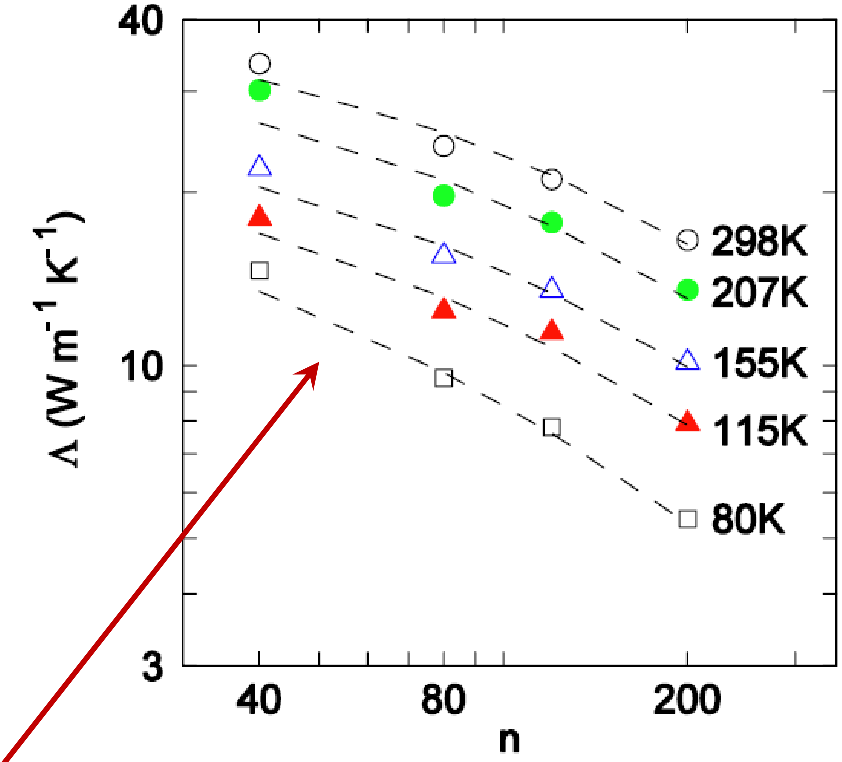
Error propagation from Nb thickness and thermal conductivity is not negligible

- Picosecond acoustics to measure thickness
- Estimate thermal conductivity of Nb from Wiedmann-Franz law and in-plane electrical conductivity
  - Uncertainty in the temperature dependence of the Lorenz number of an impure metal



Measure thermal conductivity as a function of *T* and the number of bilayers $40 < n < 200$



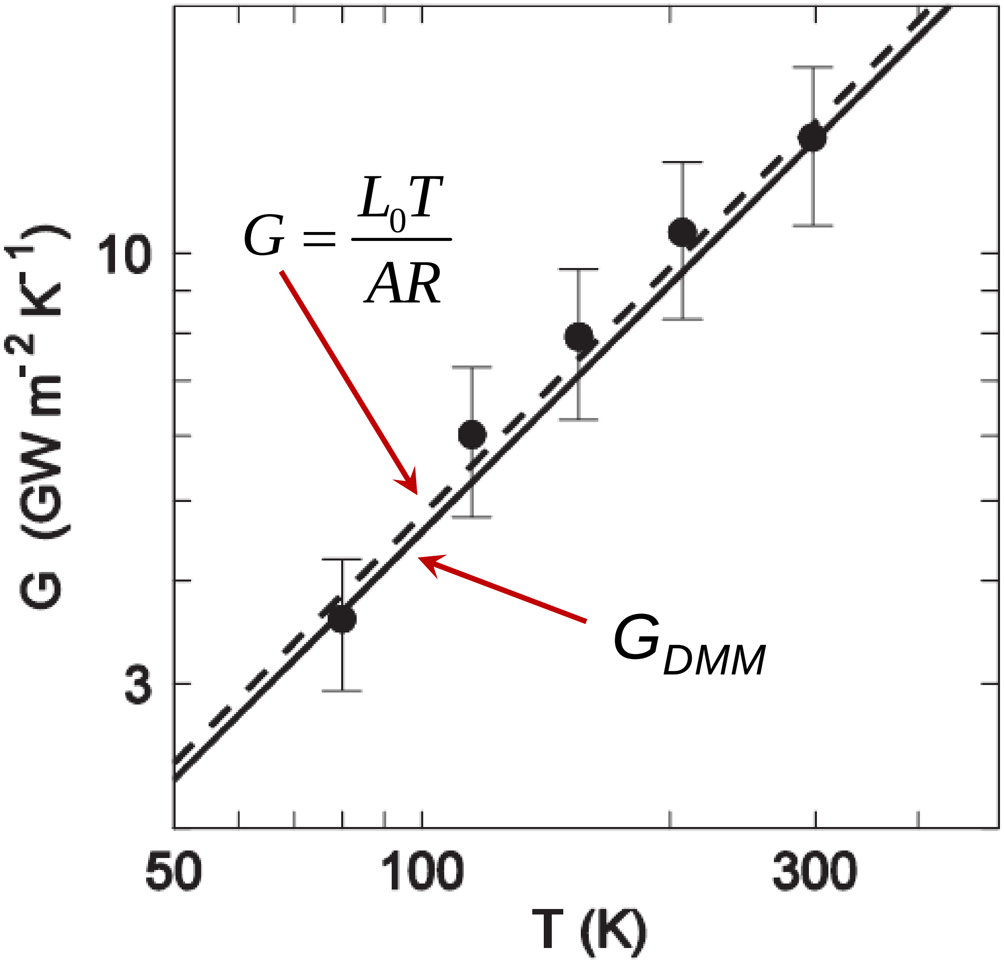


Dashed lines are series resistor models for *G* and thermal conductivity of the layers

Good agreement with both the Wiedmann-Franz law and DMM for electrons

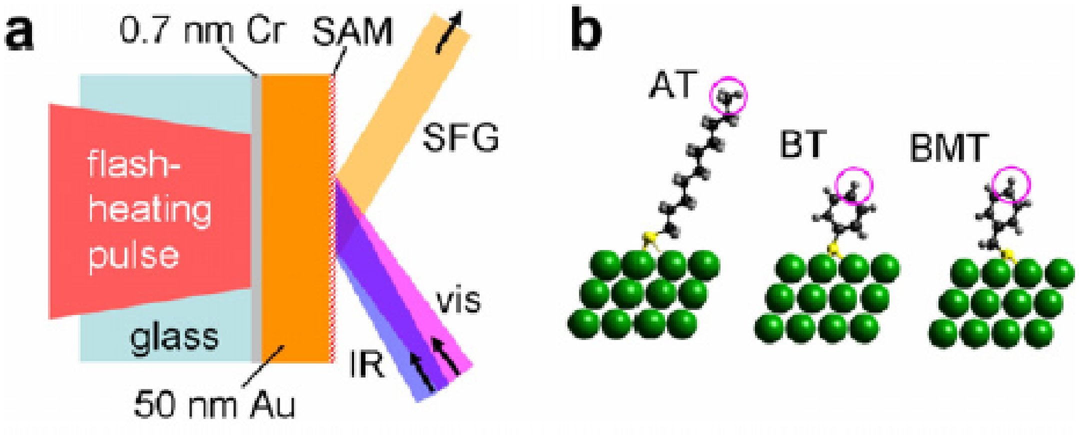
- Reasonable assumption: specific resistance *AR* measured at T=4 K is independent of temperature

  AR=0.51 fΩ m2



Wilson and Cahill, PRL (2012)

Can we indirectly heat a Au layer through contact with a transition metal on ultrafast time scales?



- Au provides well-defined chemistry for studies of molecular layers, however
  - Optical absorption is small so large temperature excursions are challenging using a laser oscillator.
  - hot-electron effects are a problem if we heat Au with amplified laser pulses.

Wang, Dlott *et al*., Chem. Phys. (2008)

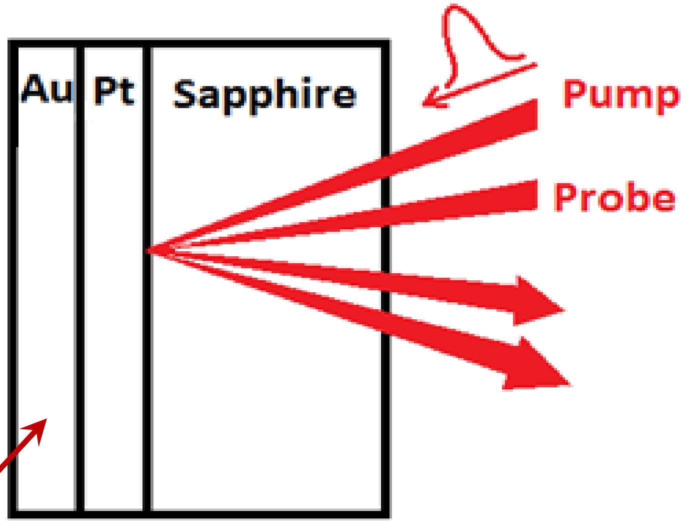
Unfortunately, heating of the Au layer is slow because of weak electron-phonon coupling in Au

- Electronic thermal conductance of Pt/Au interface is large
  - estimate $G \sim 10$ GW m^{-2} K^{-1} (experiment in progress)
- Effective conductance between Au electrons and Au phonons is not large.

g = electron-phonon coupling parameter (3×10^{16} W m^{-3} K^{-1})

h = Au thickness (20 nm)

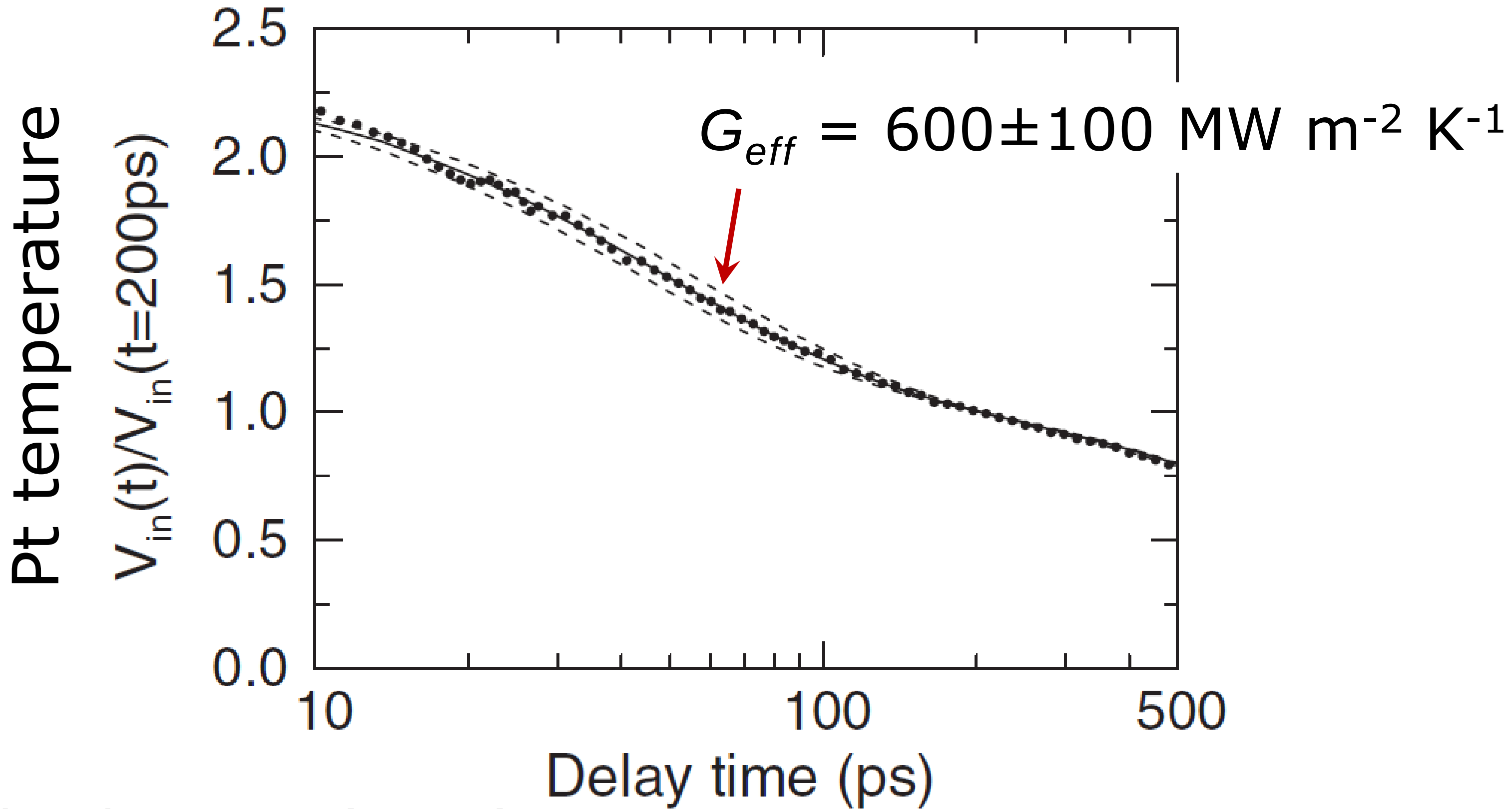
$$G_{eff} = gh = 600 \text{ MW m}^{-2} \text{ K}^{-1}$$



Unfortunately, heating of the Au layer is slow because of weak electron-phonon coupling in Au

- Characteristic time scale for the heating of the Au phonons

$$\tau = \frac{C}{g} \approx 80 \text{ ps}$$

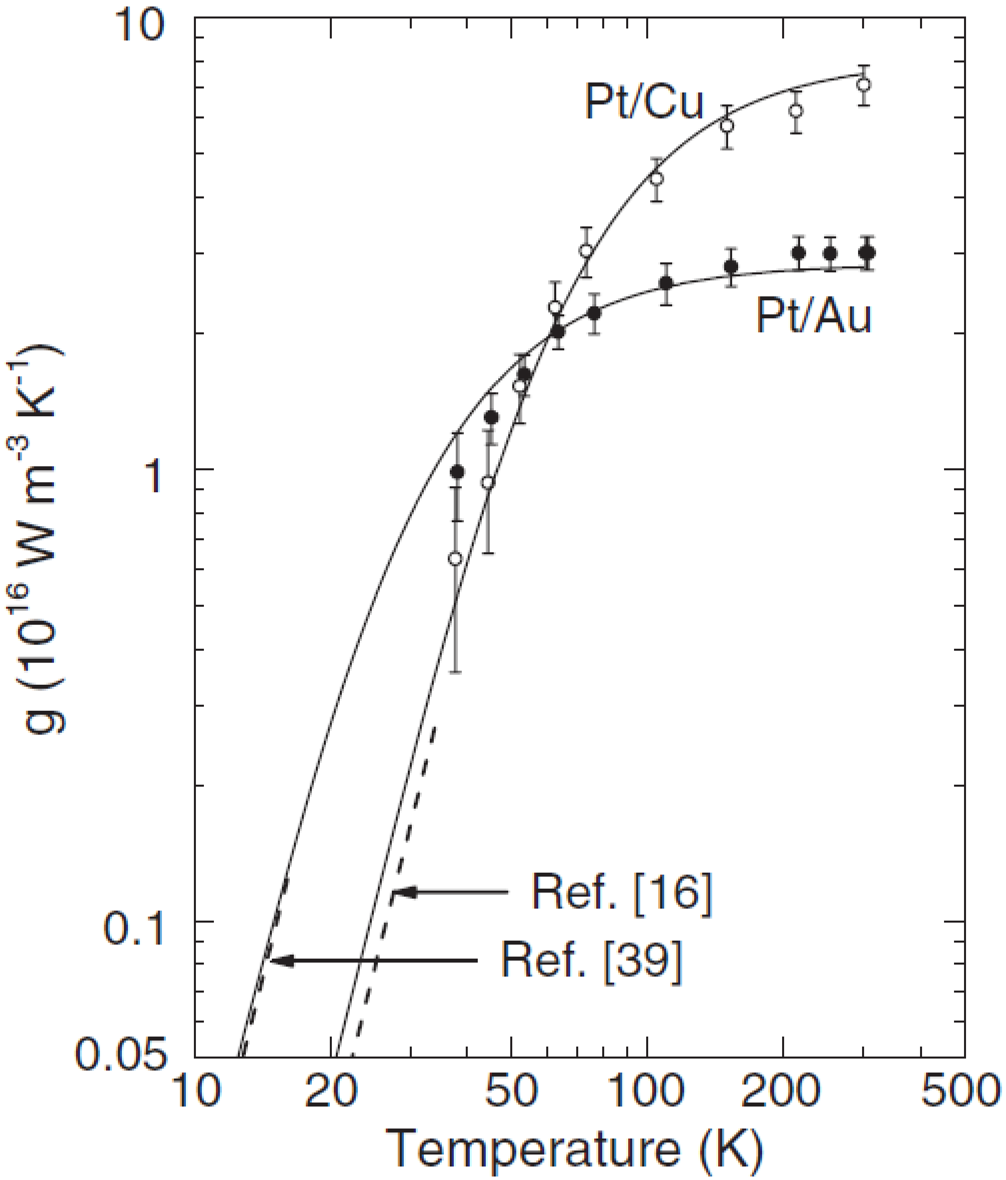


G_{eff} = 600±100 MW m^{-2} K^{-1}

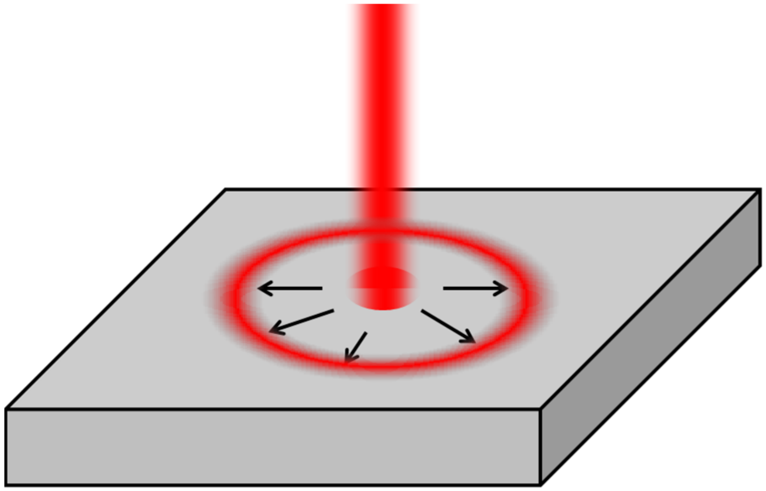
Wang and Cahill, PRL (2012)

Make lemonade out of lemons: lousy way to heat Au but excellent way to measure *g*

- Solid lines are the predictions of the original Kaganov “two-temperature” model of 1957
- Dashed lines are T^4 extrapolations of low temperature physics experiments.

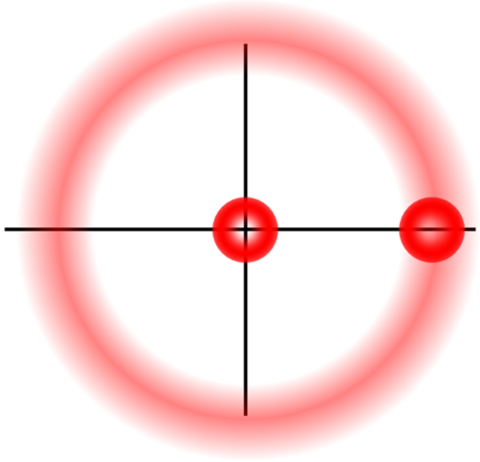


In-plane thermal conductivity using TDTR with offset laser beams



$$\Delta T(f)=\frac{2\pi}{A_S}\int_0^\infty G(f,k)P(k)\underset{\uparrow}{S(k)}k\,dk$$

The only difference from normal TDTR!

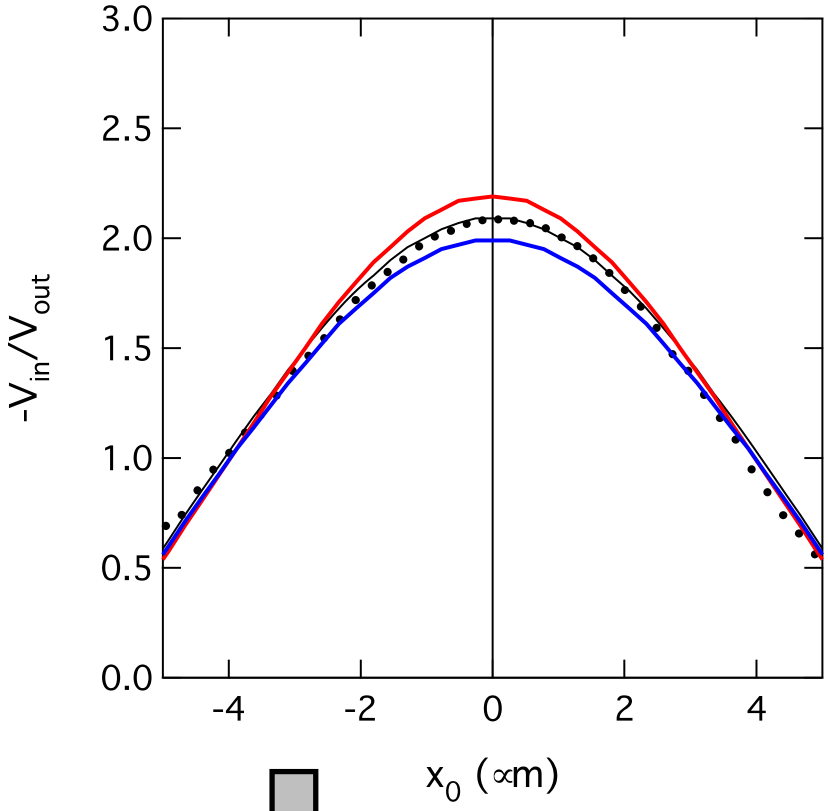


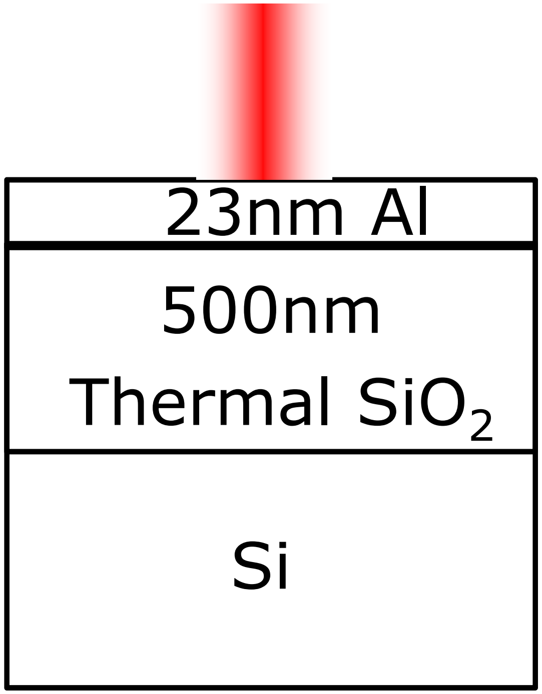
$$\underbrace{\frac{2A_S}{\pi w_S^2}\exp\left(-\frac{2\left[(x-x_0)^2+y^2\right]}{w_S^2}\right)}_{\text{Offset Gaussian Intensity}} \Leftrightarrow \underbrace{\left(\frac{2A_S}{\pi w_S^2}\right)\exp\left(-\frac{2(r^2+x_0^2)}{w_S^2}\right)I_0\left(\frac{4x_0 r}{w_S^2}\right)}_{\text{Distributed Ring Intensity}}$$

J.P. Feser, D.G. Cahill, Rev. Sci. Instr., 83, 104901 (2012).

For Al, electronic thermal conductivity should dominate; measured κ is in agreement with W-F

$f = 1.6$ MHz, $w_0 = 2.5$ μm, and $t = 1000$ ps





23nm Al

500nm Thermal SiO_2

Si

$$k_{\parallel} = 120 \text{ W/m-K } (k_{W-F} = 132 \text{ W/m-K})$$

Pushing the sensitivity requires smaller spot size and a low thermal conductivity substrate

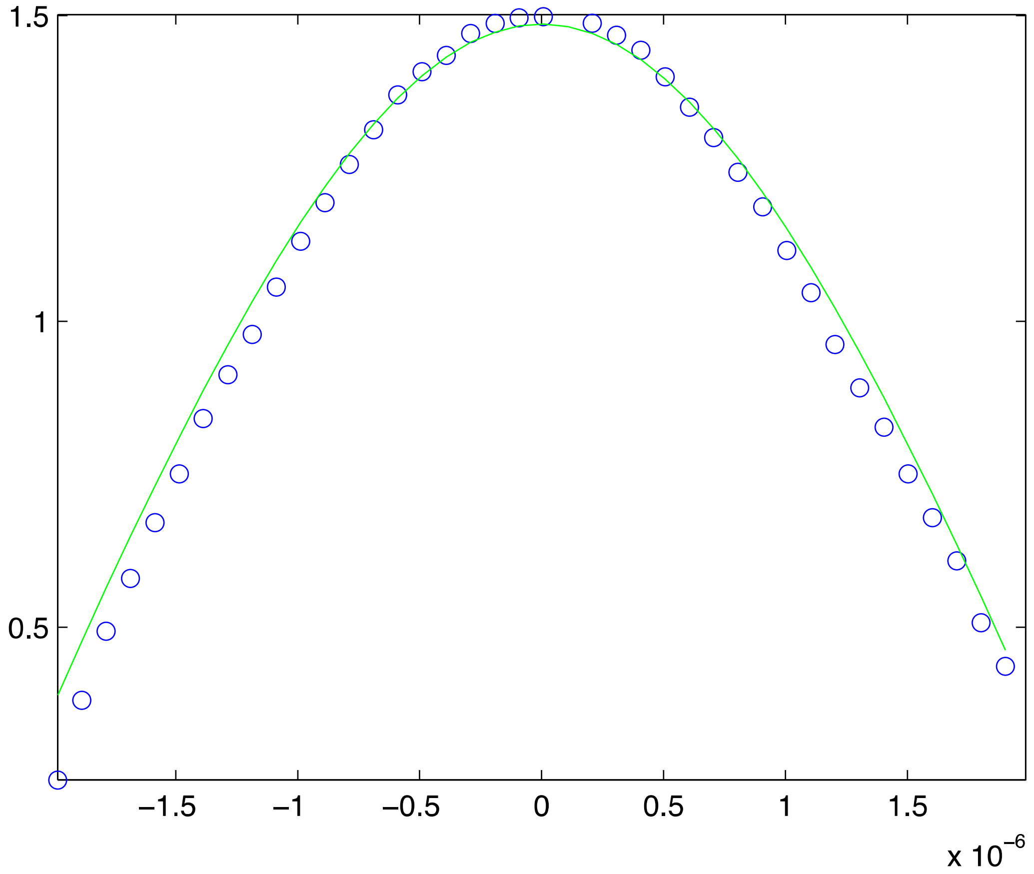
- In most cases, the relevant parameter is the lateral conductance of the film $h\kappa$, h layer thickness, κ in-plane thermal conductivity

V(32 nm)/BK7

w_0 = 1.05 μm

κ = 14.5 W m^{-1} K^{-1}

κ_{el}= 15.0 W m^{-1} K^{-1}



Conclusions

- Diffuse-mismatch model for electronic interface thermal conductance works well. Measurements of the thermal and electrical conductivity of the same multilayer shows confirm the validity of the interfacial form of the Wiedemann-Franz law.

- Thermal transport in a bilayer can be used to probe the strength of electron-phonon coupling. First measurement of the coupling parameter for Au and Cu over a wide temperature range agrees with the 1957 two-temperature model.

- In-plane thermal conductivity of a thin metal layer can be measured by TDTR using offset laser spots. Sensitivity to conductance is currently ~0.1 μW K^{-1}