

Electron-phonon coupling, the thermal conductance of interfaces, and limits to heat currents in metallic multilayers

David G. Cahill

Department of Materials Science and Engineering

Materials Research Laboratory

Department of Mechanical Sciences and Engineering

Department of Physics

University of Illinois at Urbana-Champaign

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Thermal conductance of interfaces

Costescu, Wall, and Cahill (2003); Majumdar and Reddy (2004)

Electron-phonon coupling from pump-probe optical spectroscopy

Allen (1987); Brorson et al. (1990)

Time-resolved, electronic heat transport in bilayers

Wang and Cahill (2006); Choi and Cahill (2010)

Limits to heat currents in metallic multilayers

Choi et al. (2015)

Out-of-equilibrium phonon-phonon and phonon-magnon transport near interfaces

Wilson, Feser, Hohensee, Cahill (2013)

Hohensee, Wilson, Feser, Cahill (2014)

Thermal transport coefficients in order of increasing exponent of the length scale in the denominator

- Conventional heat currents, e.g., heat diffusion equation, Fourier's law in steady-state governed by **thermal conductivity Λ**

$$J_Q = -\Lambda \nabla T \quad \Lambda \propto \text{W m}^{-1} \text{ K}^{-1}$$

- **Interface thermal conductance G** , ΔT =temperature across an interface

$$J_Q = G \Delta T \quad G \propto \text{W m}^{-2} \text{ K}^{-1}$$

- Volumetric heat currents exchanged between excitation, e.g., two-temperature model of electrons and phonons, **electron-phonon coupling parameter g**

$$j_Q = g(T_e - T_p) \quad g \propto \text{W m}^{-3} \text{ K}^{-1}$$

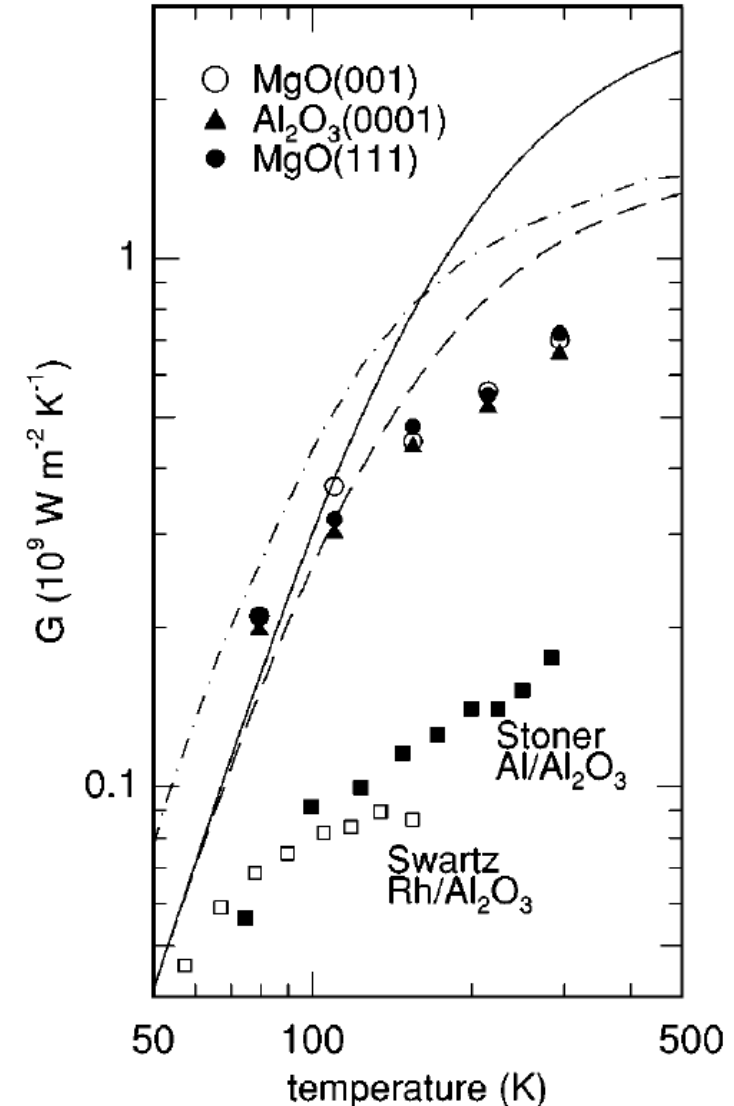
Thermal conductance of clean, strongly-bonded, abrupt interfaces is high but smaller than expected

UHV sputter deposition of TiN (metal) on MgO and Al₂O₃

TiN(001) layers grown on Mg(001) are essentially single crystals

Conductance is not sensitivity however to the details: same result for Mg(111) and Al₂O₃(0001)

Costescu, Wall, Cahill PRB (2003)



Enter Arun: Phonons are not the only important carriers of heat in this problem.

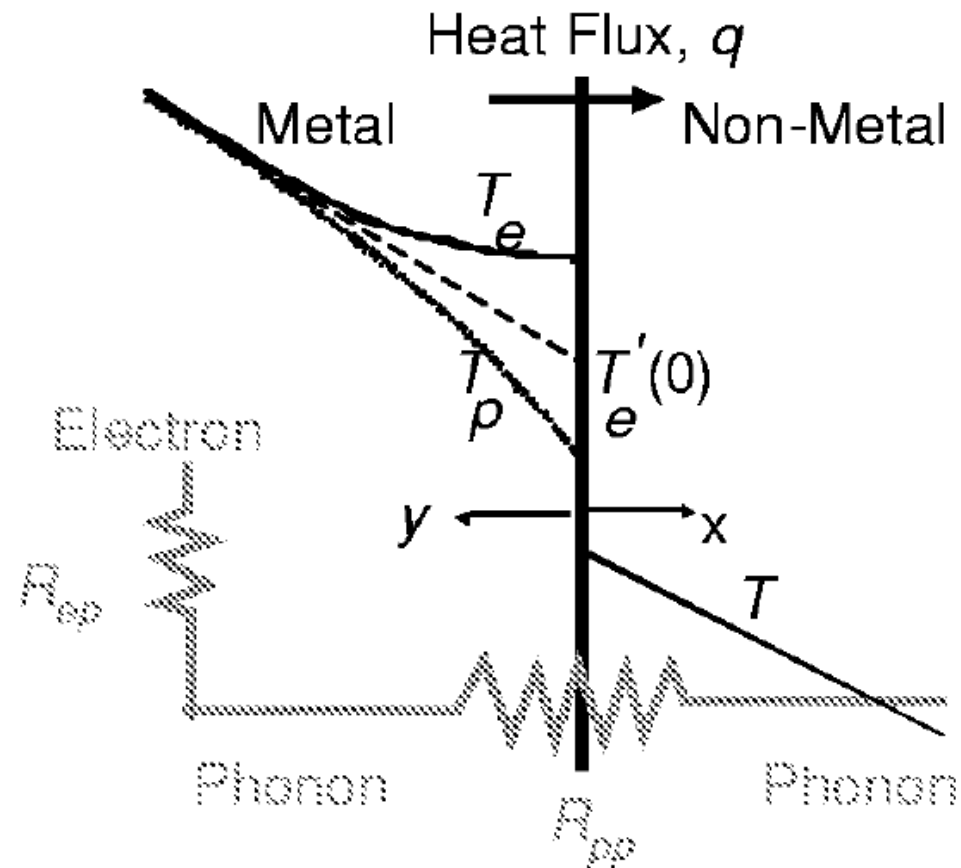
Heat current crossing a metal/dielectric interface creates a region where electrons and phonons are out of equilibrium

$$G_{ep} = \sqrt{g\Lambda_p}$$

$$2 \times 10^{16} < g < 10^{18} \text{ W m}^{-3} \text{ K}^{-1}$$

$$3 < \Lambda_p < 30 \text{ W m}^{-1} \text{ K}^{-1}$$

$$0.25 < G_{ep} < 5 \text{ GW m}^{-2} \text{ K}^{-1}$$



Majumdar and Reddy, APL (2004)

Measurements of electron-phonon coupling parameter were recently available from advances in theory and time-resolved experiment.

VOLUME 59, NUMBER 13

PHYSICAL REVIEW LETTERS

28 SEPTEMBER 1987

Theory of Thermal Relaxation of Electrons in Metals

Philip B. Allen^(a)

VOLUME 64, NUMBER 18

PHYSICAL REVIEW LETTERS

30 APRIL 1990

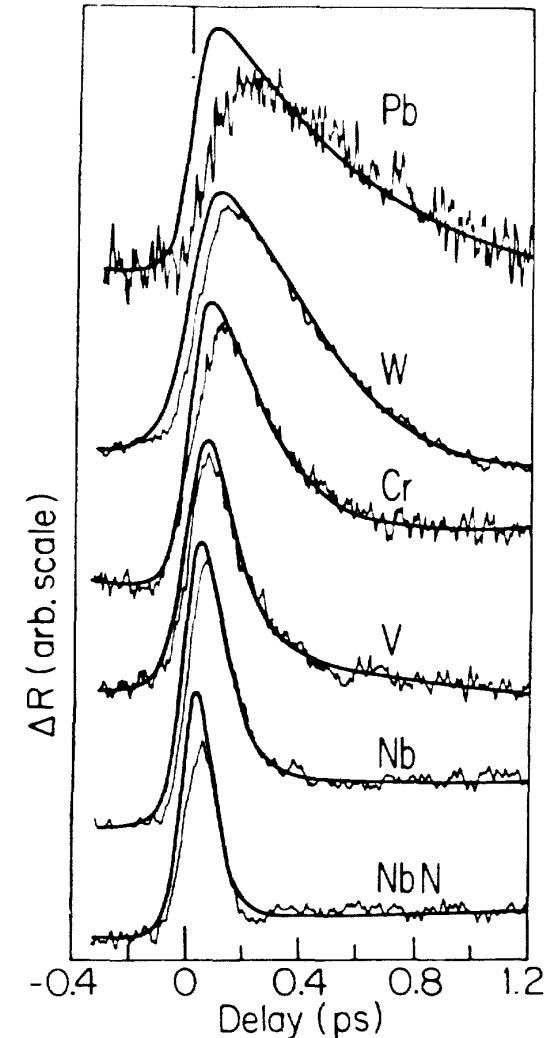
Femtosecond Room-Temperature Measurement of the Electron-Phonon Coupling Constant λ in Metallic Superconductors

S. D. Brorson,^(1,2) A. Kazeroonian,^(3,4) J. S. Moodera,⁽⁵⁾ D. W. Face,^{(3,4),(a)} T. K. Cheng,^(1,2)
E. P. Ippen,^(1,2) M. S. Dresselhaus,^(1,3,4) and G. Dresselhaus⁽⁵⁾

These measurements involve photon excitation energies that are huge compared to thermal energies

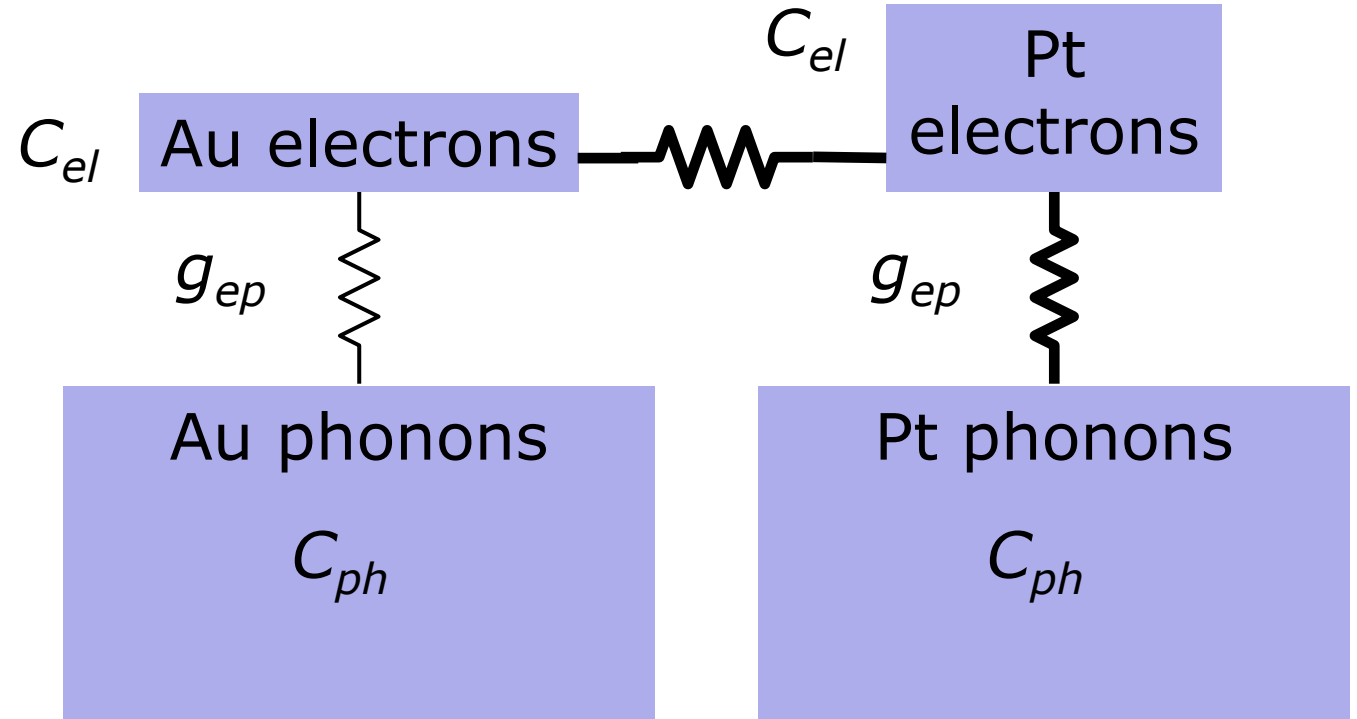
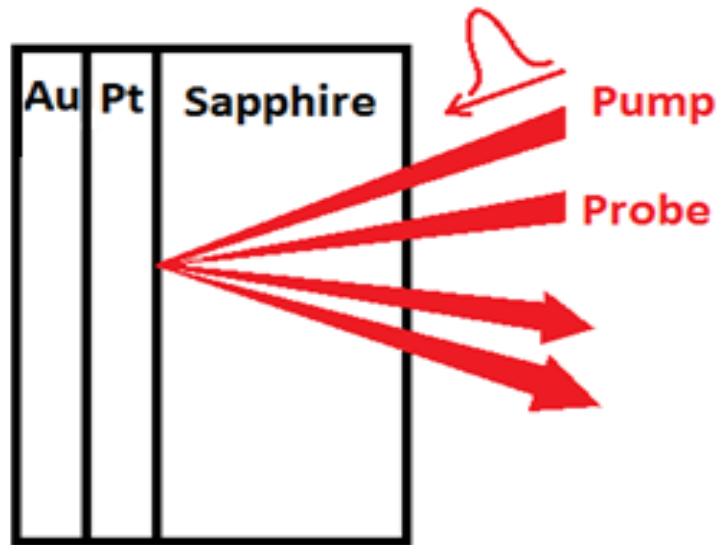
$$\hbar\omega \sim 50k_B T$$

Is this a reliable approach for predicting transport near equilibrium, i.e., is the same two-temperature model a good approximation for both a picosecond optical experiment and a transport measurement?



Brorson et al., PRL (1990)

Consider a metal bilayer that couples two metals, one with weak and one with strong electron-phonon coupling.



Heating of the Au layer is slow because electron-phonon coupling in Au is the smallest conductance in the problem

Characteristic time scale for the heating of the Au phonons

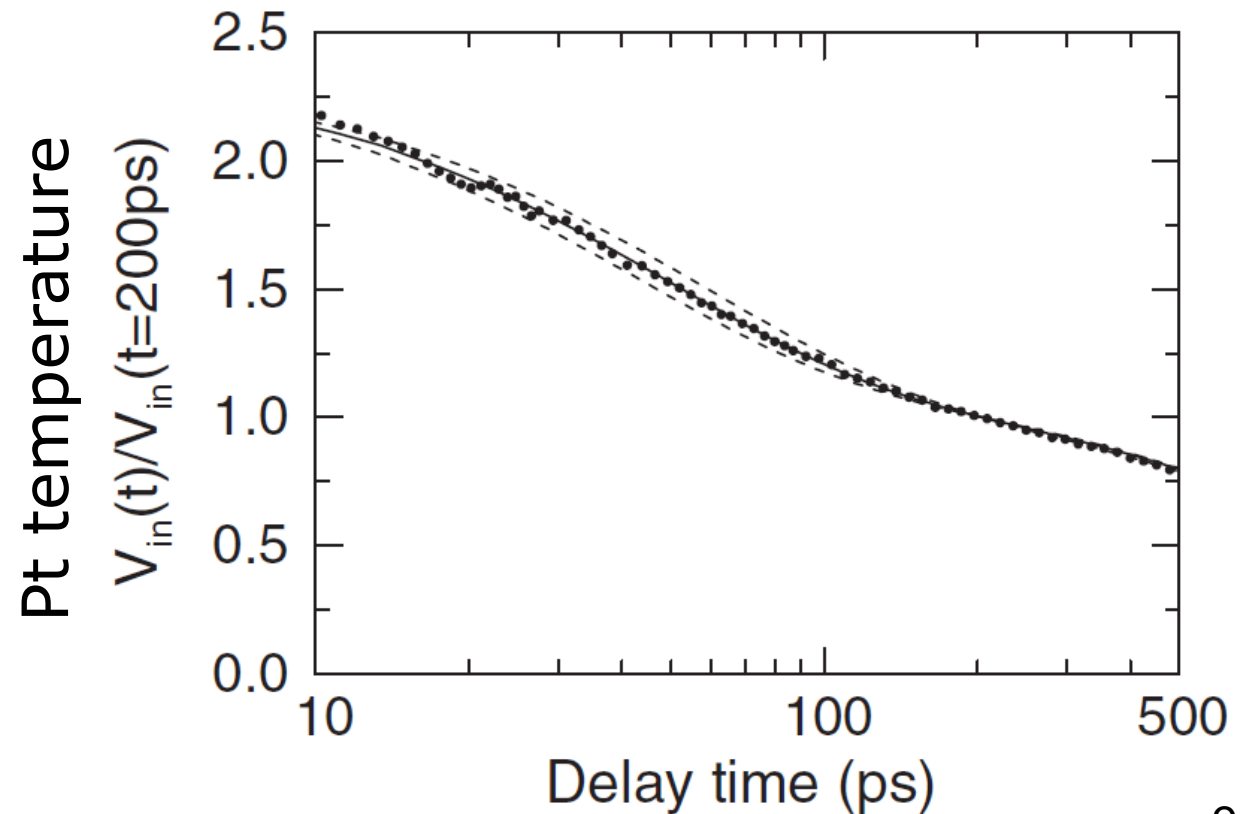
$$\tau = \frac{C_{Au,ph}}{g_{Au}} \approx 80 \text{ ps}$$

Data analyzed in terms of an effective G for the Au/Pt interface

$$G_{\text{eff}} = hg$$

$$g \approx \frac{600 \text{ MW m}^{-2} \text{ K}^{-1}}{20 \text{ nm}}$$

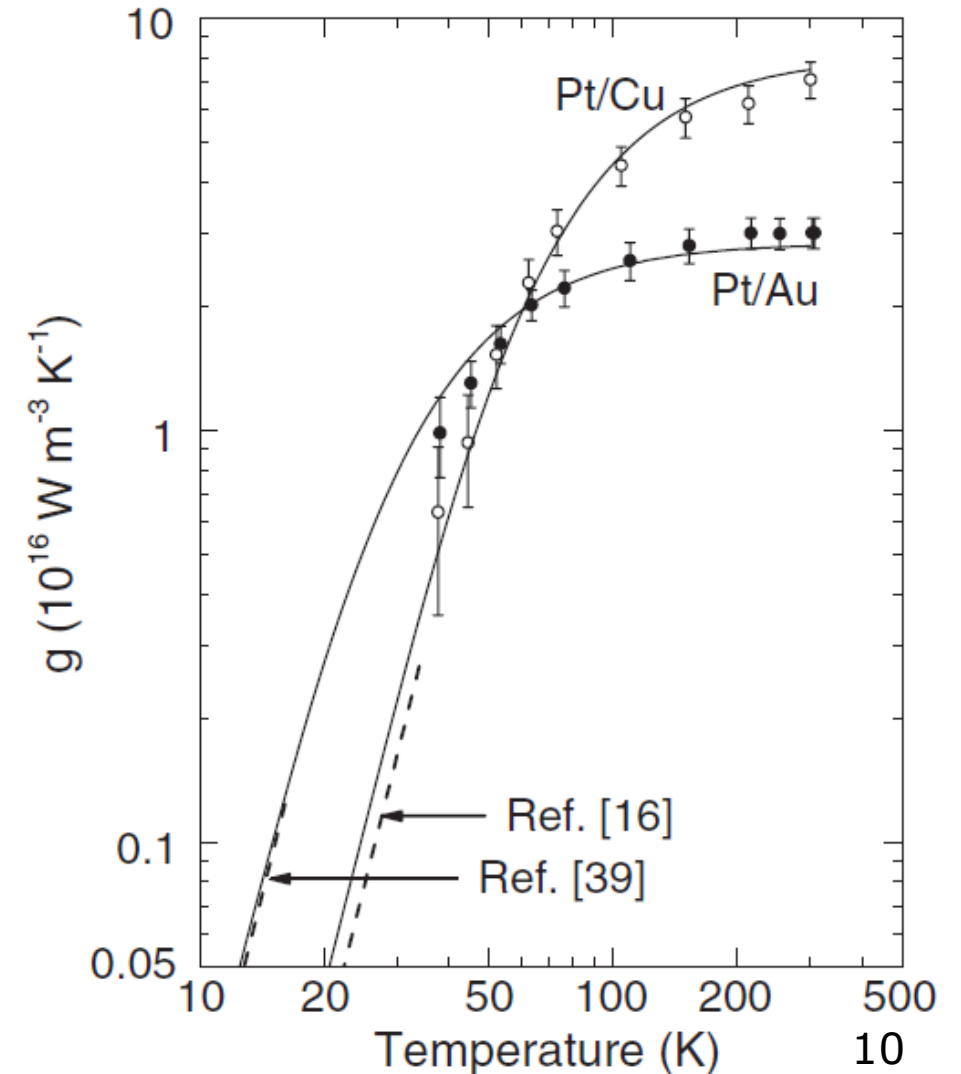
$$g \approx 3 \times 10^{16} \text{ W m}^{-3} \text{ K}^{-1}$$



We still can't say if the two-temperature model is a good approximation but this measurement is much closer to the transport problem that Arun studied

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.



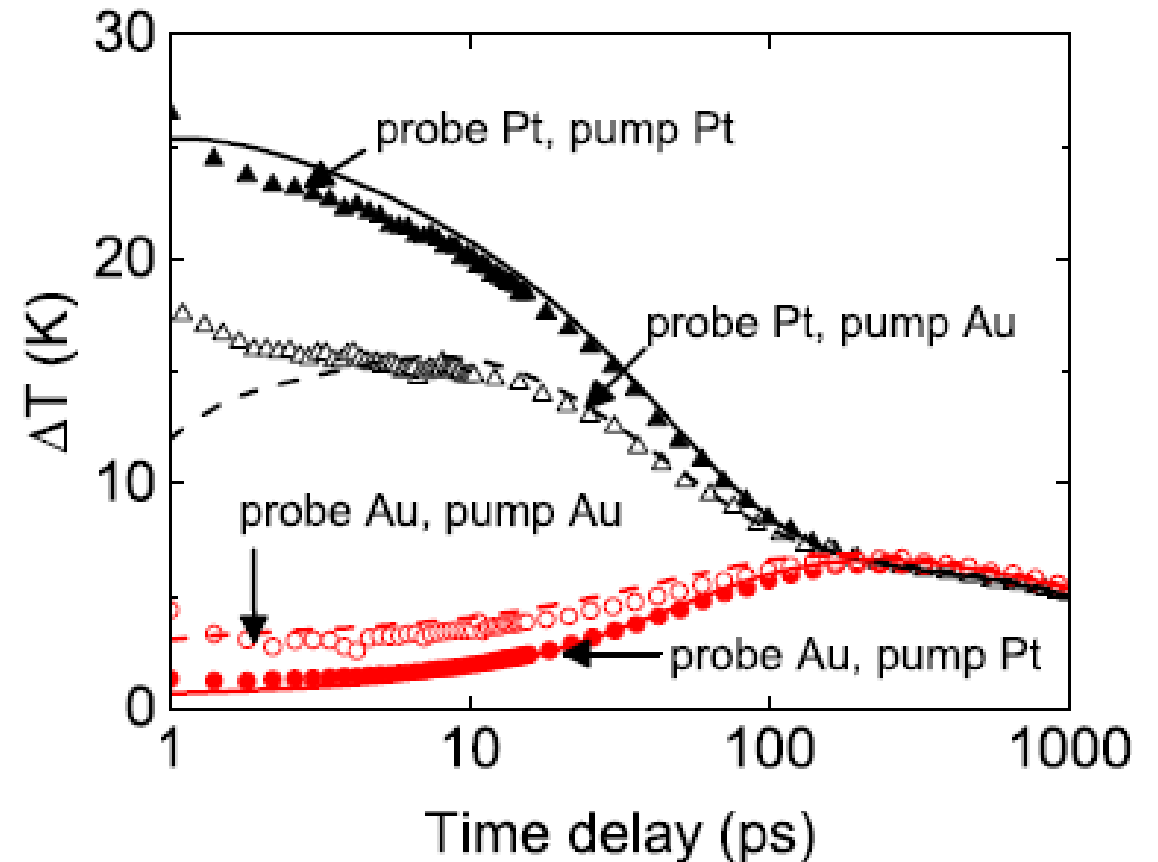
Wang and Cahill, PRL (2012)

Improve the sensitivity with a thicker Au layer and use all four configurations of pump and probe directions

More rigorous modeling of the experiment than previous study including finite optical absorption depth, and phonon and electron diffusion.

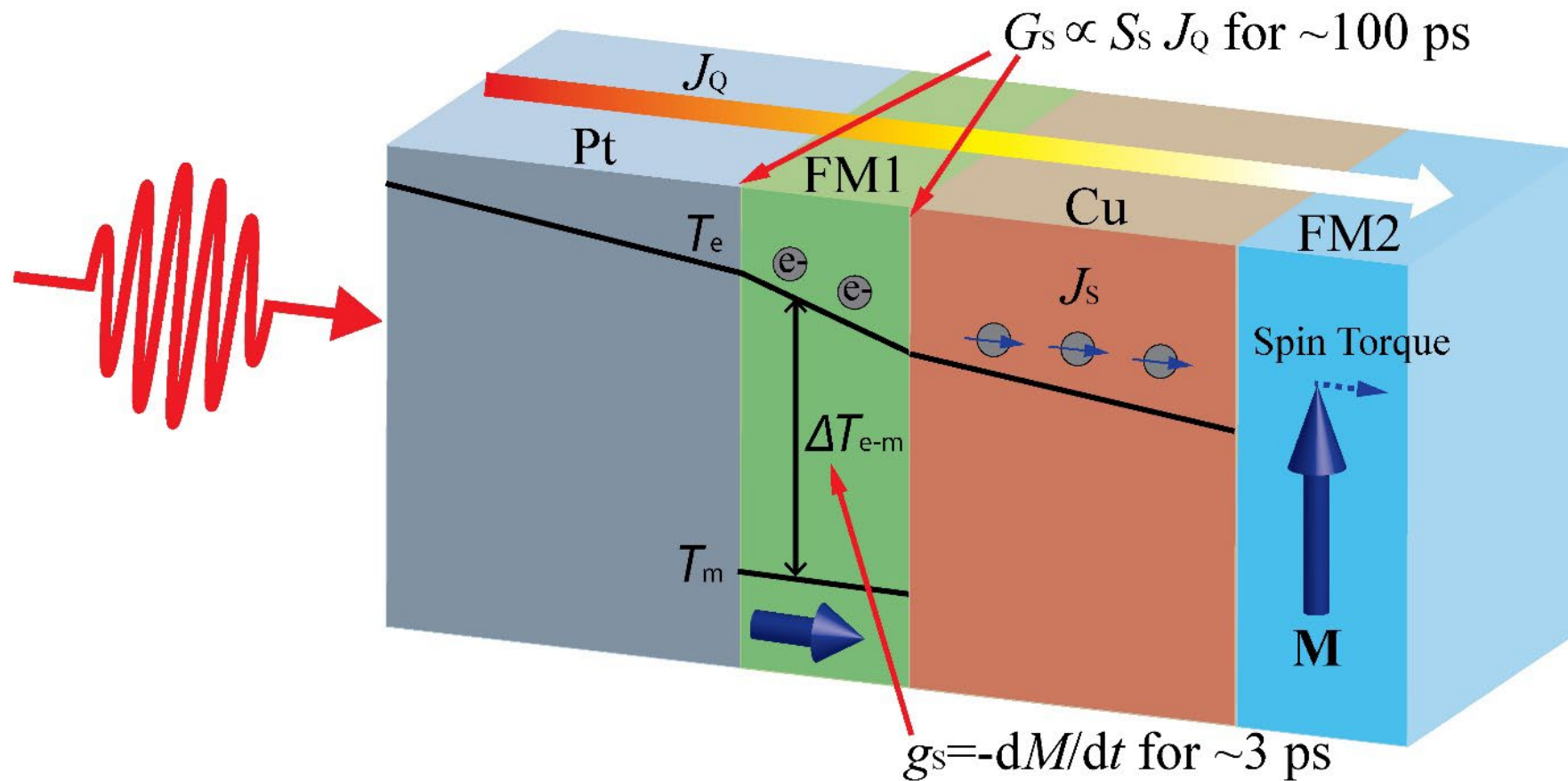
Counter intuitive result that a pump beam incident on the Au surfaces mostly heats the Pt.

$$g = 2.2 \times 10^{16} \text{ W m}^{-3} \text{ K}^{-1}$$



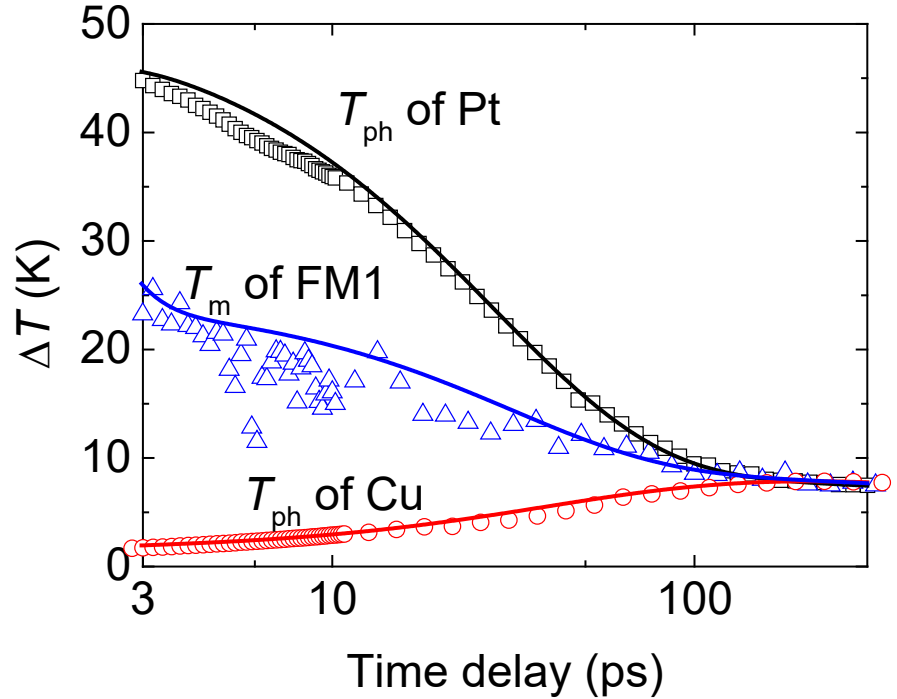
The electron-phonon coupling in Cu partially controls how well a thin Cu layer acts as a heat sink in an laser-driven magnetic device

Pt (20)/ [Co/Pt] or [Co/Ni] (3)/ Cu (100)/ CoFeB (2) (nm)

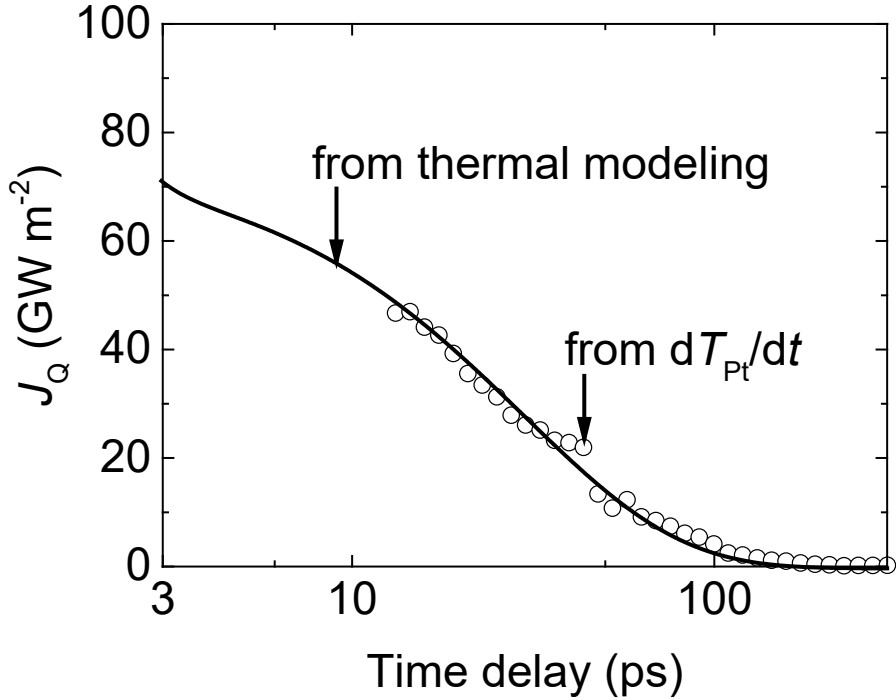


Heat flux of 70 GW m^{-2} for a temperature excursion of 45 K .
Effective interface conductance of $1.5 \text{ GW m}^{-2} \text{ K}^{-1}$

Temperature measurement



Heat current through FM1

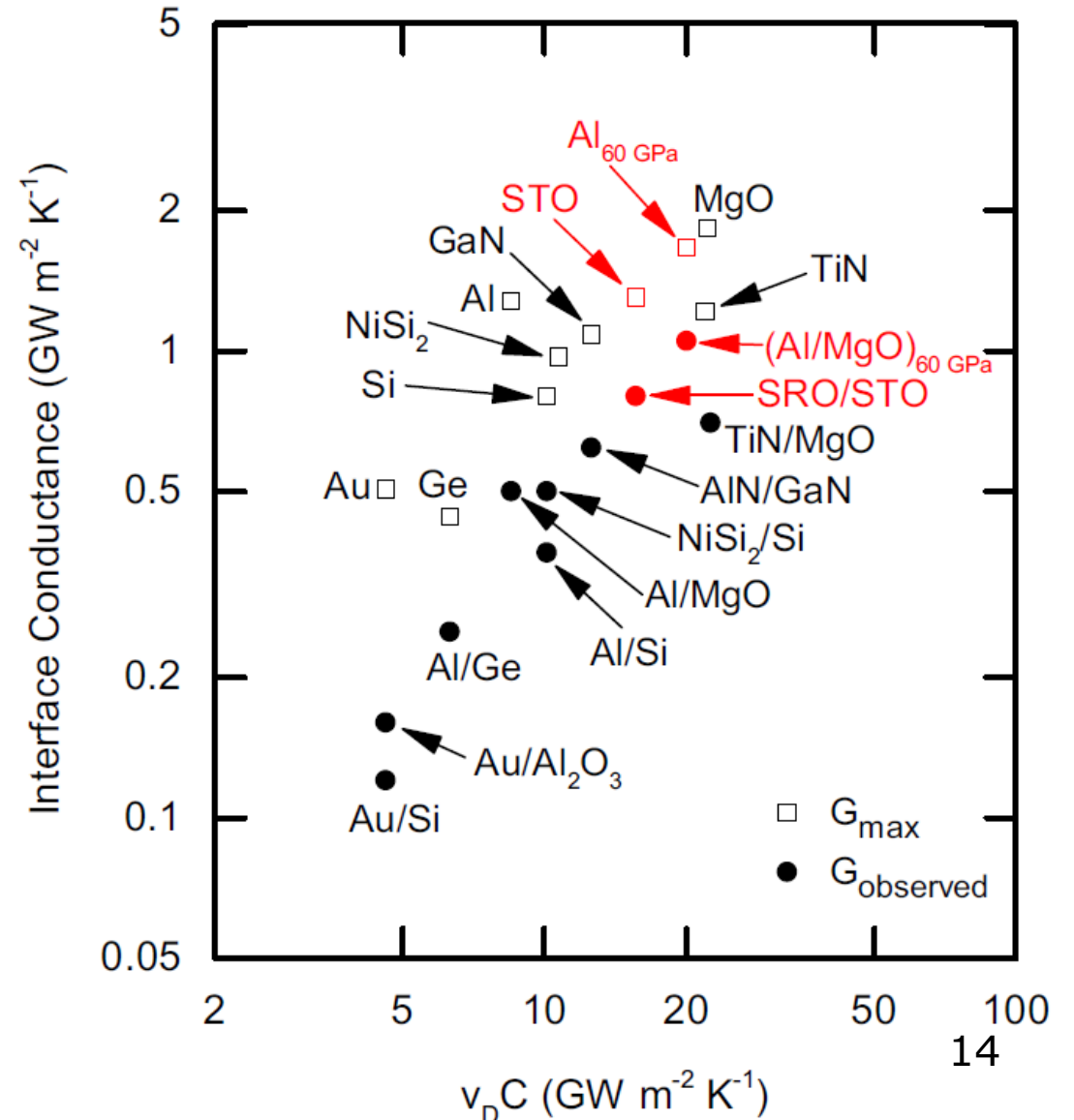


The J_Q is obtained from temperature measurement.
The J_Q persists for $\sim 100 \text{ ps}$.

Back to Arun's contribution. Can we observe the reduction of interface conductance created by out-of-equilibrium heat carriers at a metal/dielectric interface?

Maybe for Au, but the effect of finite electron-phonon coupling has not been isolated for a single interface.

The effect might be seen in nanoscale metal/dielectric multilayers. Difficult to separate from finite mean-free-path effects.



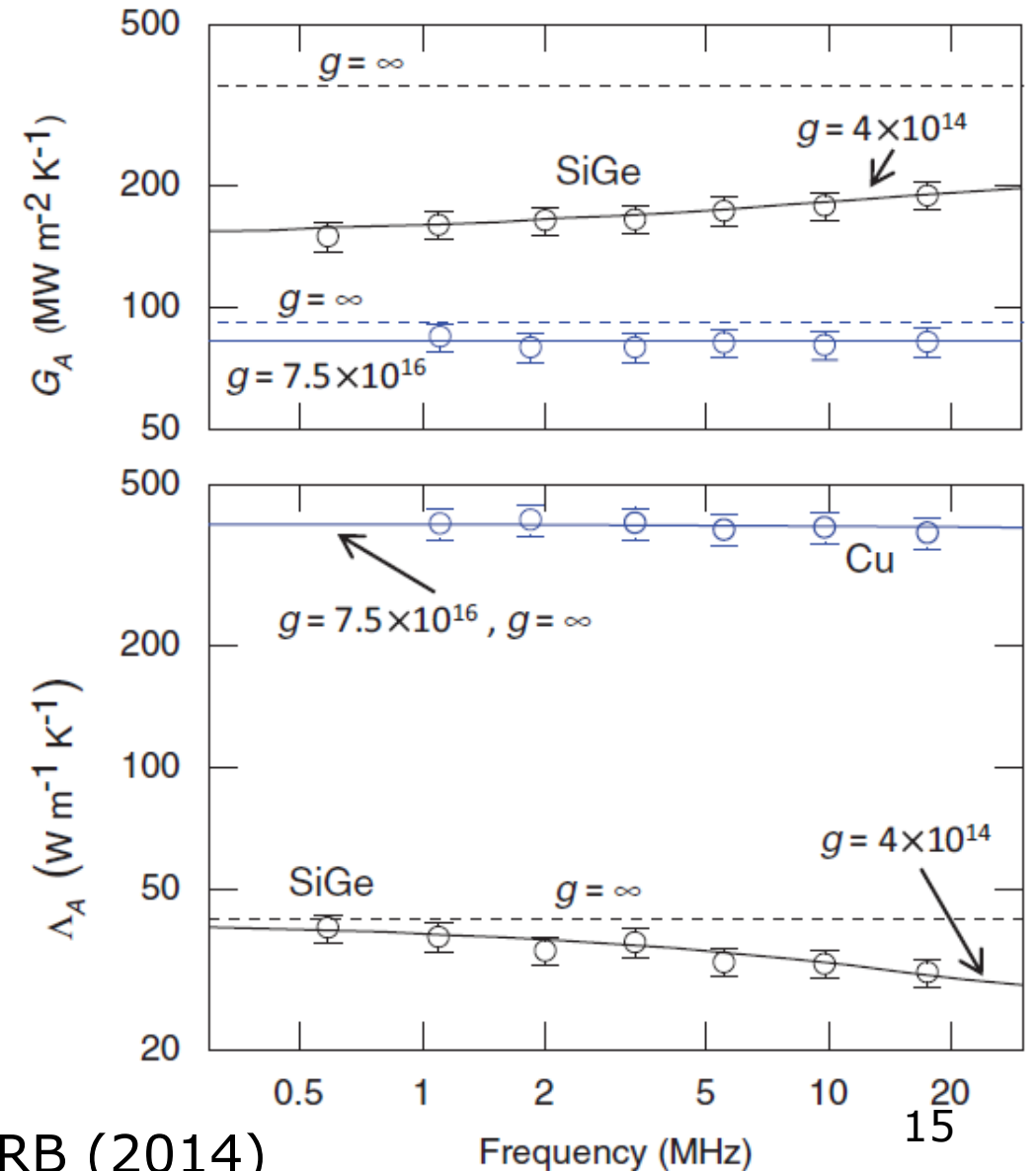
Strong effects of out-of-equilibrium phonon distributions at interfaces with semiconductor alloys.

Wilson's two-channel model of time-domain thermoreflectance (TDTR) measurements.

Describe transport at Al/SiGe interface with two channels: high and low frequency phonons.

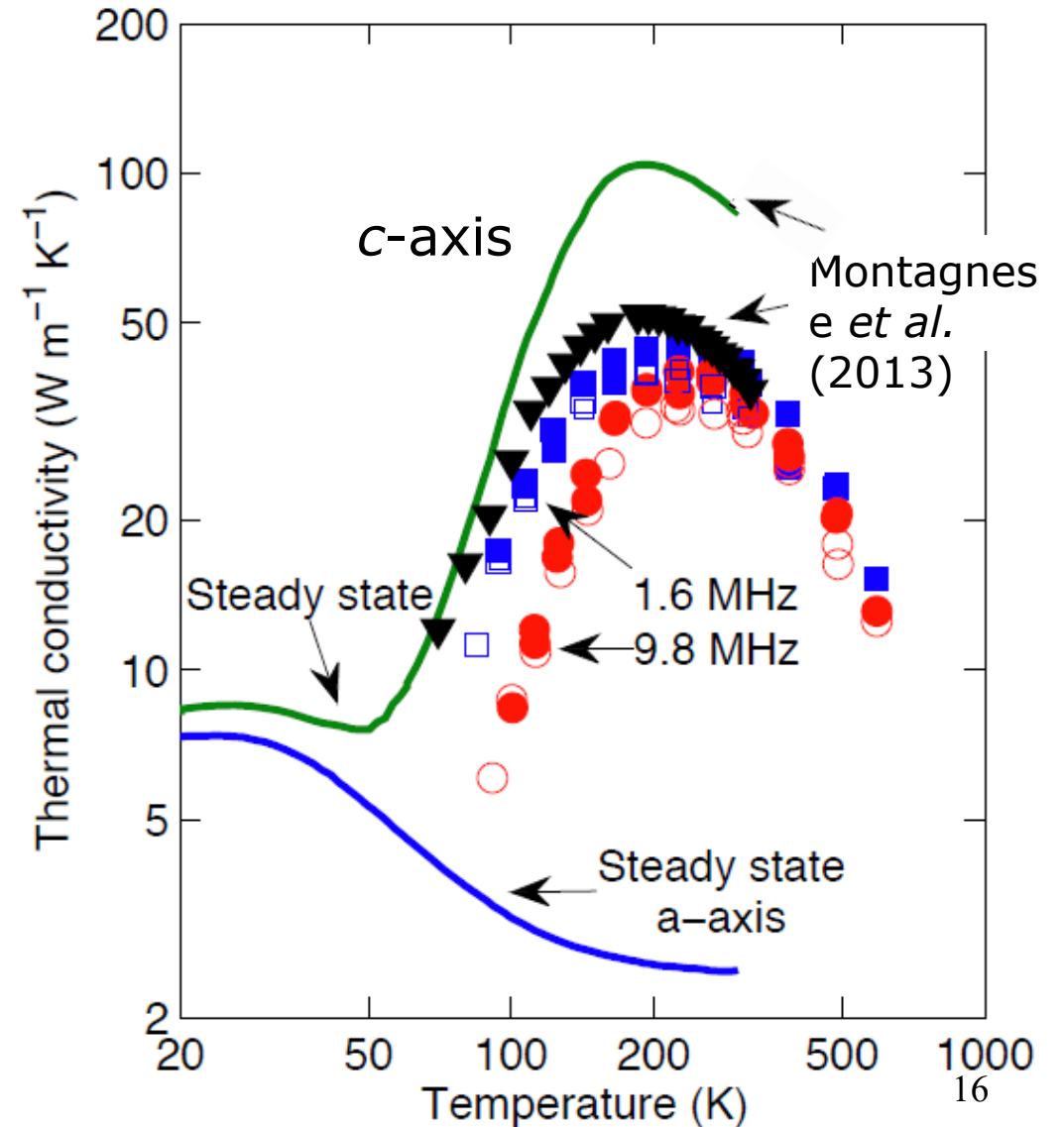
Apparent interface conductance G_A increases and apparent thermal conductivity Λ_A decreases with increasing frequency.

Explains Koh's frequency dependent thermal conductivity experiments.



Spin-wave thermal conductivity in $\text{Ca}_9\text{La}_5\text{Cu}_{24}\text{O}_4$ depends on the measurement frequency due to finite strength of magnon-phonon coupling

Magnon-phonon coupling becomes weaker at reduced temperatures and reduces the apparent thermal conductivity



Concluding comments

Thank you, Arun, for pointing out the importance of non-equilibrium between carriers that is created by a heat current crossing an interface.

The original proposal of a reduction in conductance of a metal-dielectric interface has been hard to observe.

Combinations of metals with high contrast in electronic heat capacity and electron-phonon coupling enables near-equilibrium measurements of electron-phonon coupling.

The same concept applied to out-of-equilibrium phonon-phonon and phonon-magnon distributions *is* observed and has important consequences for thermal transport in electronic materials.