Society of Vacuum Coaters Education Program in conjunction with the 66th Annual Technical Conference

Tutorial Course: M-230

Nanoscale Heat Transfer in Thin Films and Interfaces

Instructor: Patrick E. Hopkins
Chief Science Officer, Co-Founder, *Laser Thermal, Inc*.
patrick@laserthermal.com
https://laserthermal.com/
Whitney Stone Professor of Engineering, *University of Virginia*
phopkins@virginia.edu
https://patrickehopkins.com/

Monday, May 8, 2023

DISCLAIMER

These course materials are based upon sources believed to be reliable, but the Society of Vacuum Coaters and the author(s) disclaim any warrant or liability based on or relating to the contents of this publication, or the course presentation based upon this publication.

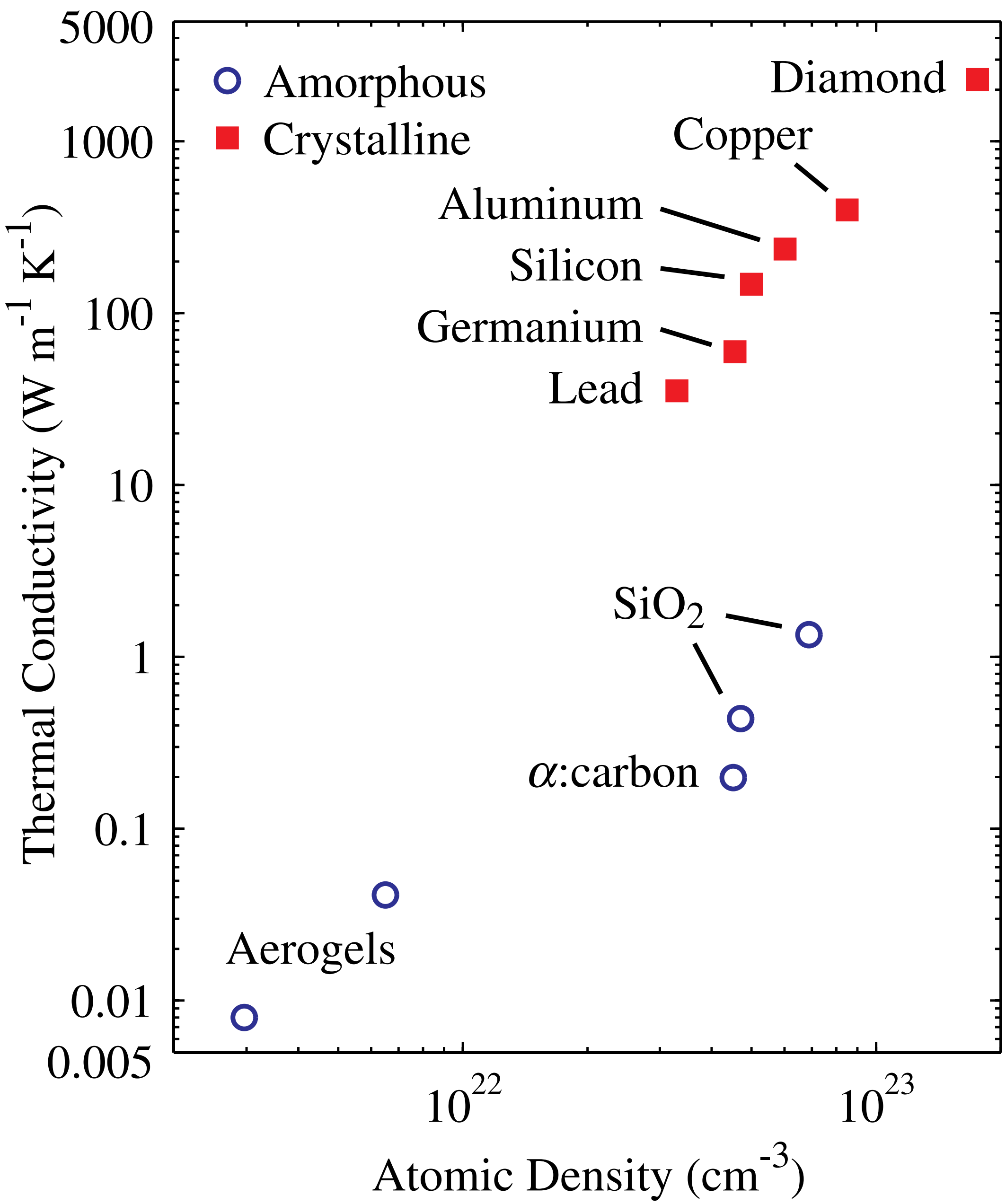
SURVEY

As an SVC tutorial attendee your feedback is *the* essential element that allows us to continuously improve and refine our educational offerings. Approximately one week after the course concludes you will receive an invitation to participate in a brief on-line survey. We would appreciate a few moments of your time to tell us how well we did and how we could do better going forward. All survey respondents will be entered into a raffle where the winner will receive a complimentary seat in any tutorial of their choice at the 2024 TechCon in Chicago, Illinois USA. Thank you!

Outline

1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

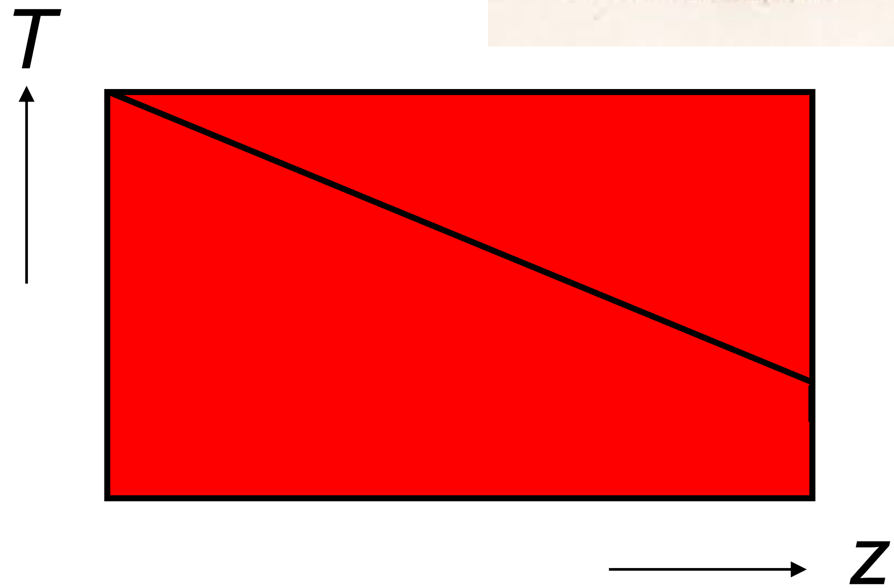
Thermal conductivity of materials – Macroscopic picture



PRL **110**, 015902 (2013)

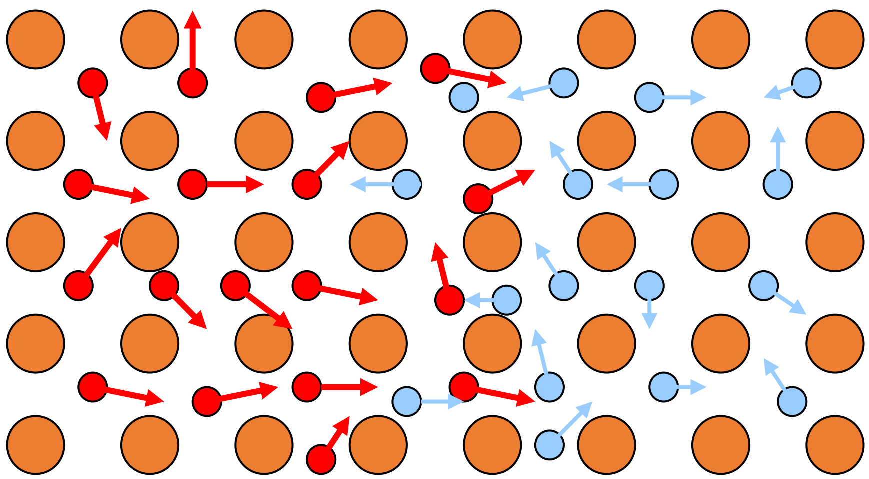
The Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$



Materials and Heat Transfer trends

Diffusion of "hot" electrons →



Metals:

Free electrons are the dominant energy carriers in metals, ballistic velocity ~10^6 m/s

Electron carrier density:

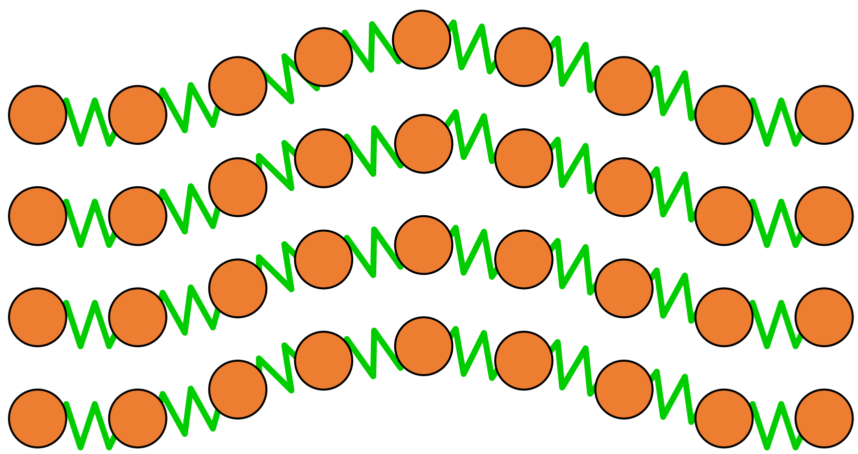
in metals ~10^{23} cm^{-3}

in semiconductors ~10^{18} cm^{-3}

atom

"hot" free electron

"cold" free electron



Phonon propagation →

Semiconductors:

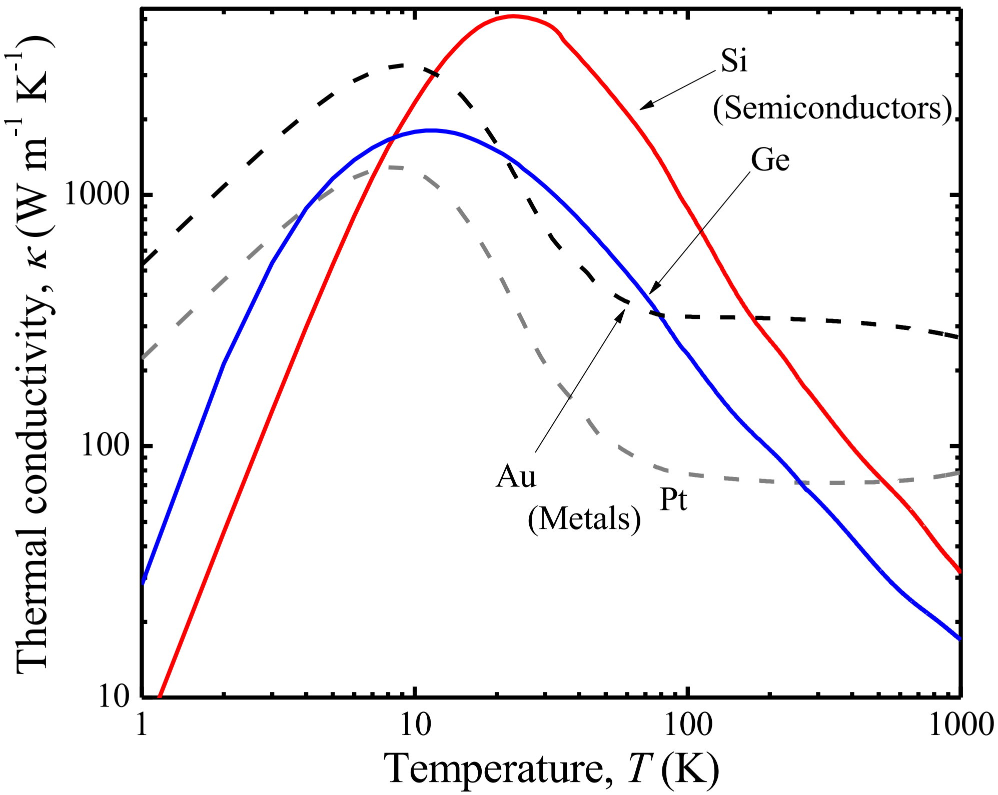
Phonons (lattice vibrations) are the dominant energy carriers in semiconductors, velocity ~10^3 m/s

$$\kappa = \frac{1}{3}Cv\lambda = \frac{1}{3}Cv_g^2\tau$$

Thermal conductivity of bulk materials

Thermal conductivity

$$q = -\kappa \frac{\partial T}{\partial z}$$



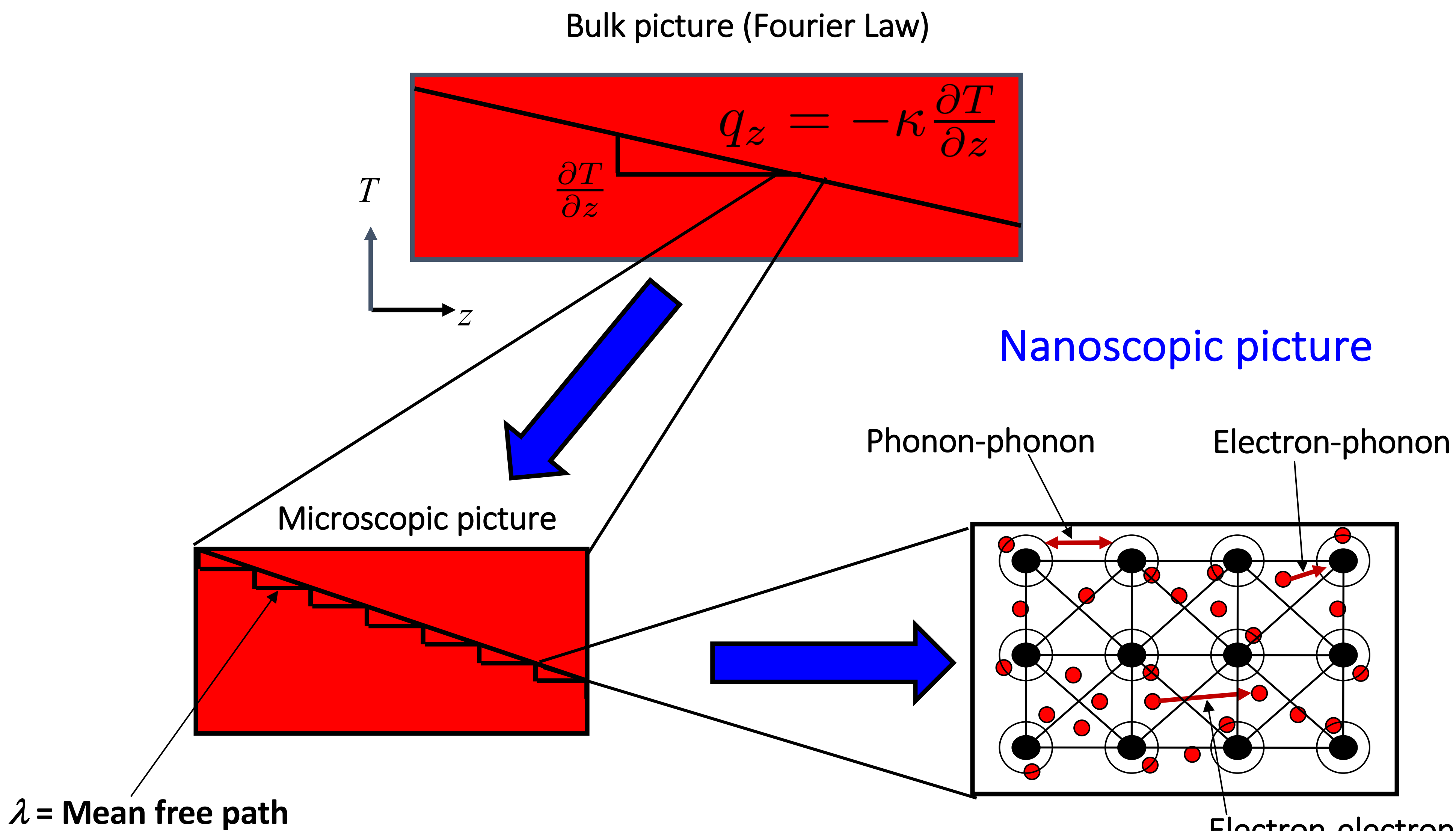
Temperature trends in κ related to energy carrier scattering in solids

Notice different trends between κ in metals and κ in semiconductors

Higher thermal conductivity = larger heat flux removed

Tabulated data from: Ho, Powell, and Liley, "Thermal conductivity of the elements," Journal of Physical and Chemical Reference Data, **1**, 279 (1972).

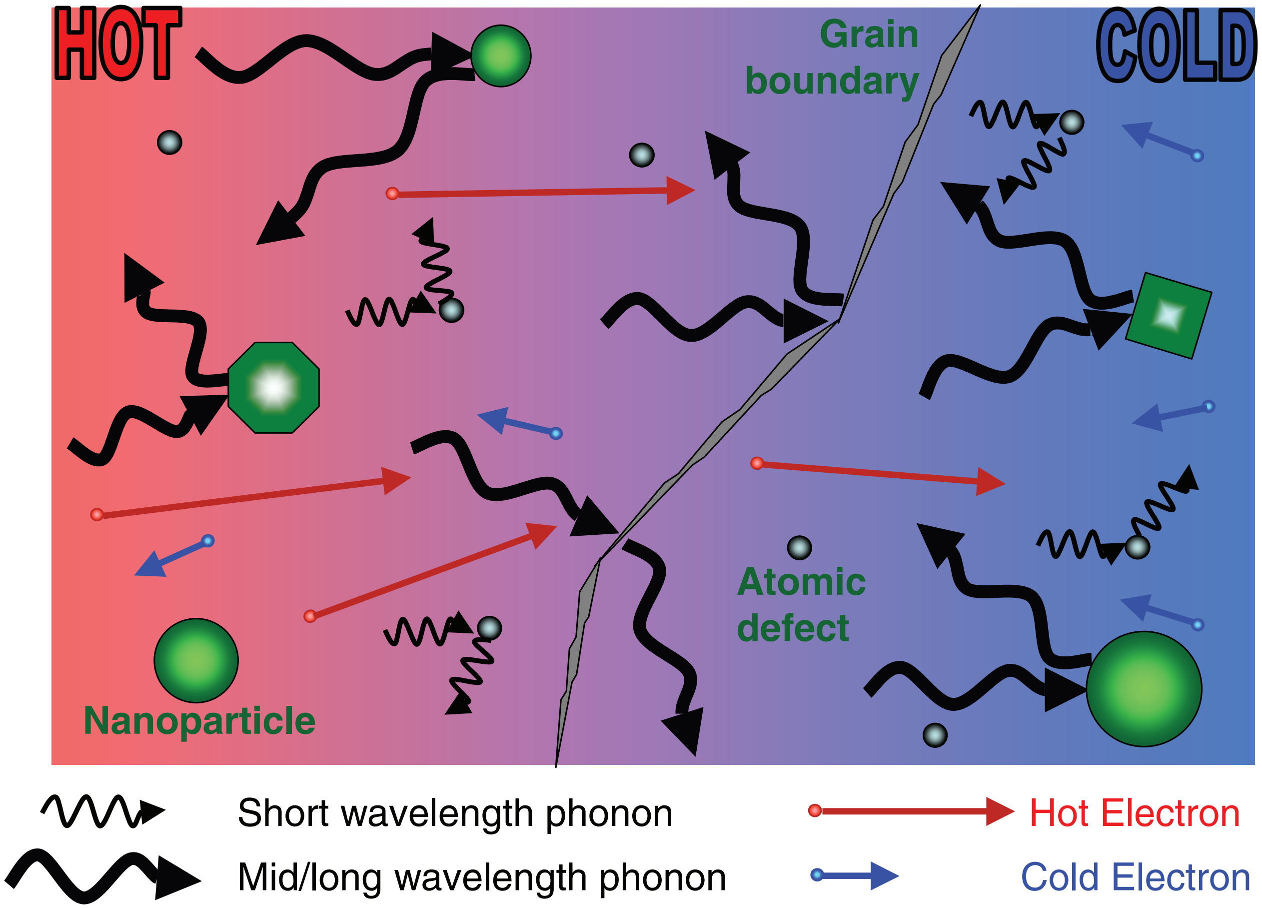
Thermophysics on the nanoscale



$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v^2 \tau$$

A nanoscopic view with Kinetic Theory

$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$



Adv. Mat. **22**, 3970

C: Heat capacity
"How much energy electrons/phonons store"

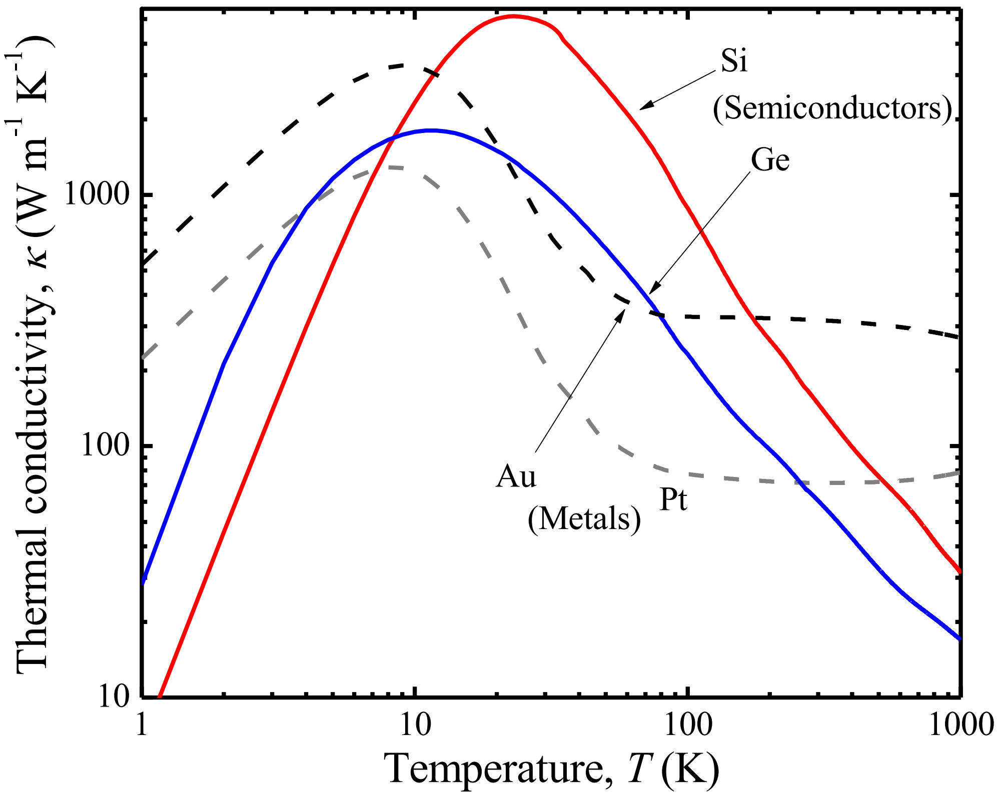
v: Velocity
"How fast the electrons/phonons move"

λ = Mean free path
"How far they move before losing energy/momentum"

Thermal conductivity of bulk materials

Thermal conductivity

$$q = -\kappa \frac{\partial T}{\partial z}$$



$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$

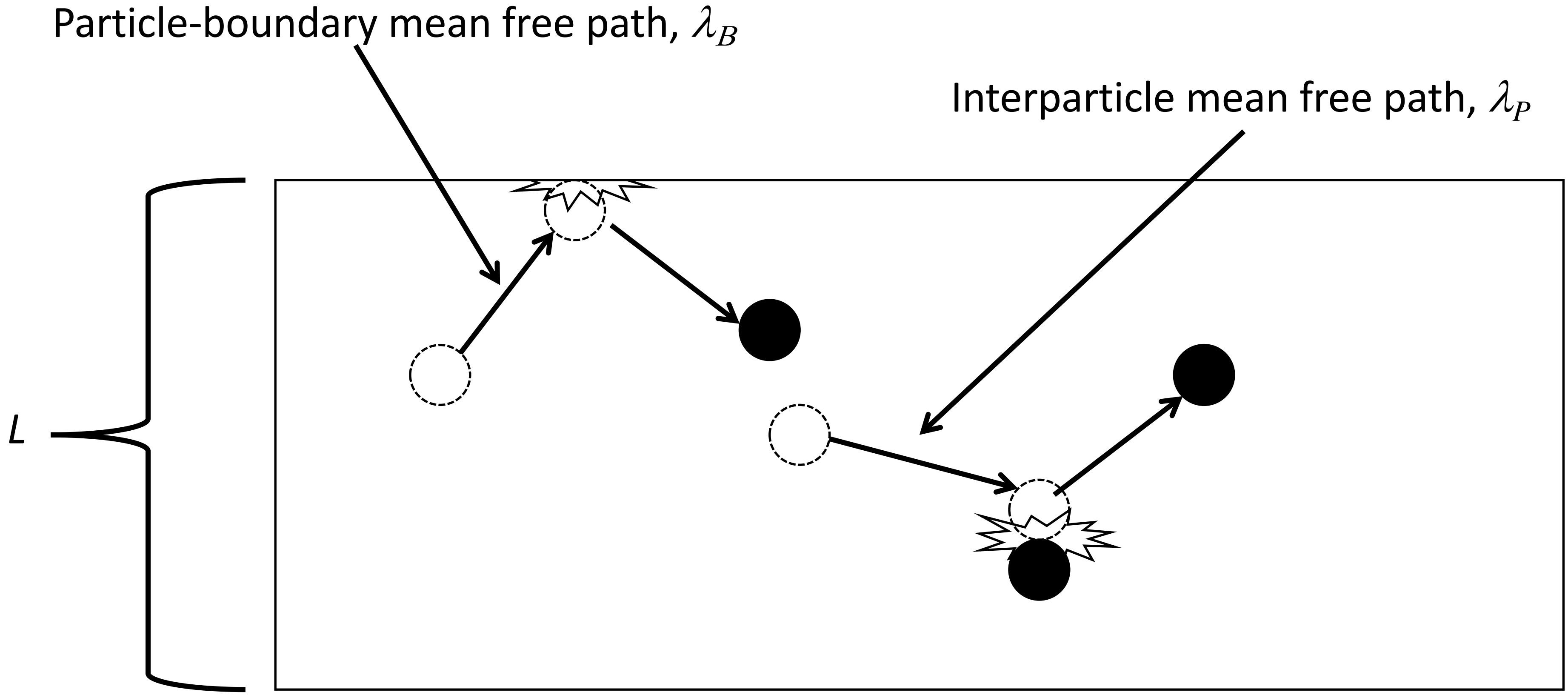
Different temperature dependencies for metals and non-metals (like semiconductors)

Why????

Electrons in metal (intrinsic scattering from electron-electron and electron-phonon)

Phonons in non-metal (intrinsic scattering from phonon-phonon)

Mean free path



$$\lambda_P = v\,\tau_P - \text{Material dependent}$$

$$\lambda_B = v\,\tau_B - \text{Geometry dependent} - L/2$$

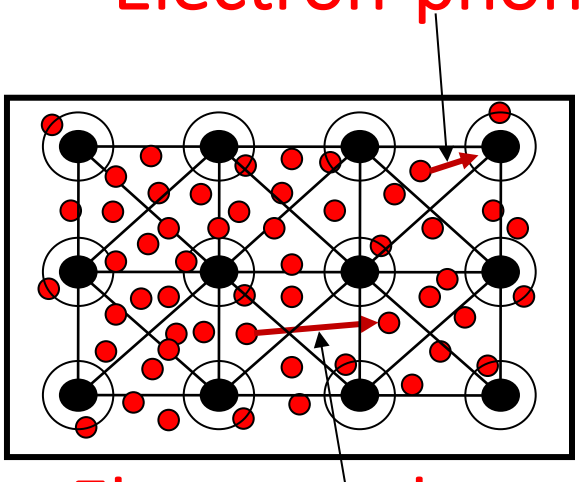
Mean free path in crystals

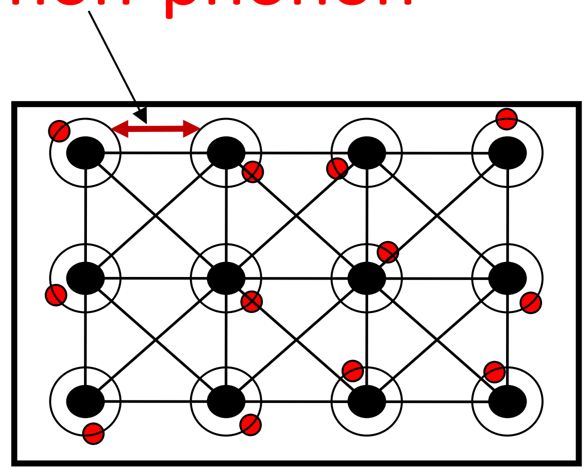
Metals

Electrons are the dominant carriers

Semiconductors

Phonons are the dominant carriers





$$\frac{1}{\tau_{metal}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_D}$$

$$\frac{1}{\tau_{semi}} = \frac{1}{\tau_{pp}} + \frac{1}{\tau_D}$$

Defect and impurity scattering

Thermal conductivity of metals

$$\frac{1}{\tau_{metal}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_D}$$

Electron-electron scattering

$$\frac{1}{\tau_{ee}} = AT_e^2$$

Electron-phonon scattering

$$\frac{1}{\tau_{ep}} = BT_p$$

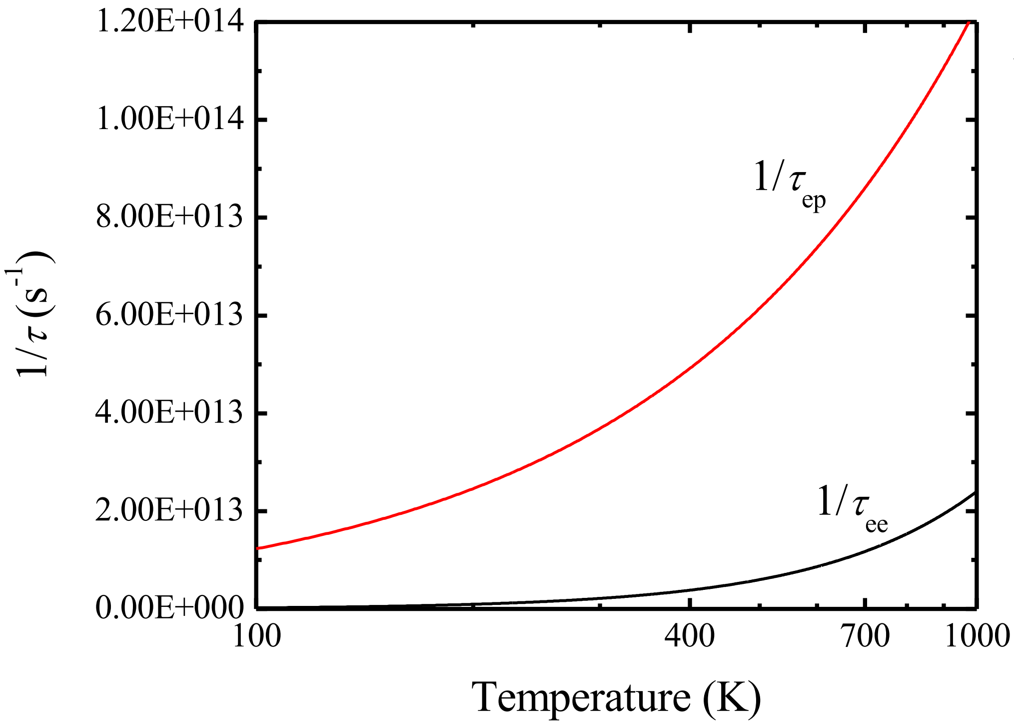
Electron-defect scattering

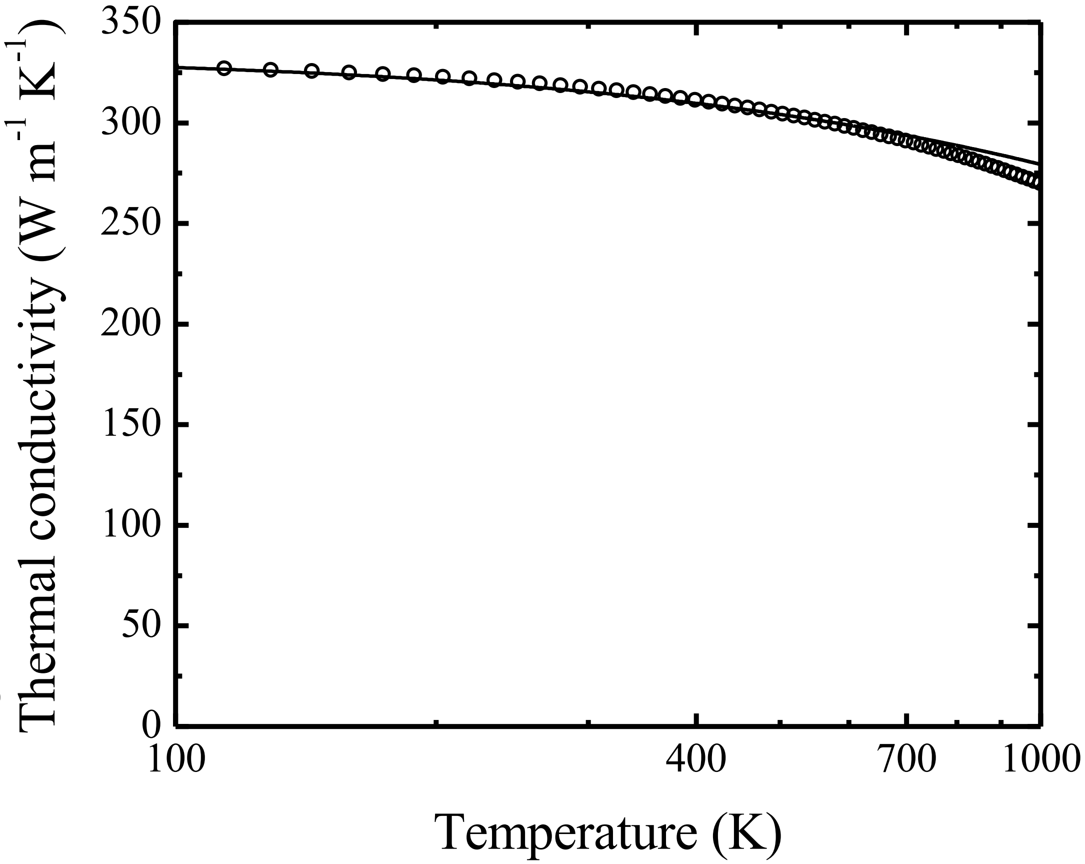
$$\frac{1}{\tau_D} \neq f(T_e, T_p)$$

Thermal conductivity of metals

Electron-electron and electron-phonon scattering rates in Au

$$T_e = T_p$$





$$\kappa = \frac{1}{3} C v_F^2 \tau_{metal}$$

$$\kappa = \frac{1}{3} \gamma T v_F^2 \tau_{metal}$$

$$\frac{1}{\tau_{metal}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_D}$$

Electron transport: WIEDEMANN-FRANZ LAW

Thermal conductivity

$$\frac{\kappa}{\sigma} = LT$$

Electrical conductivity

Temperature

Lorentz number

$$L = \frac{\kappa}{\sigma T} = \frac{\pi^2}{3}\left(\frac{k_B}{e}\right)^2 = 2.44 \times 10^{-8}\,\mathrm{W\,\Omega\,K^{-2}}.$$

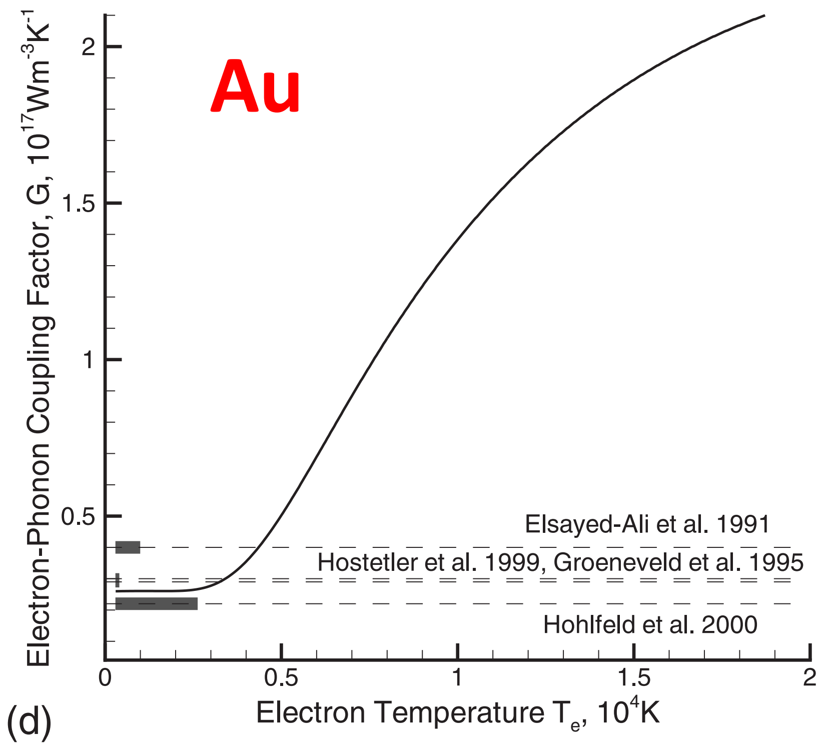
What does this mean????

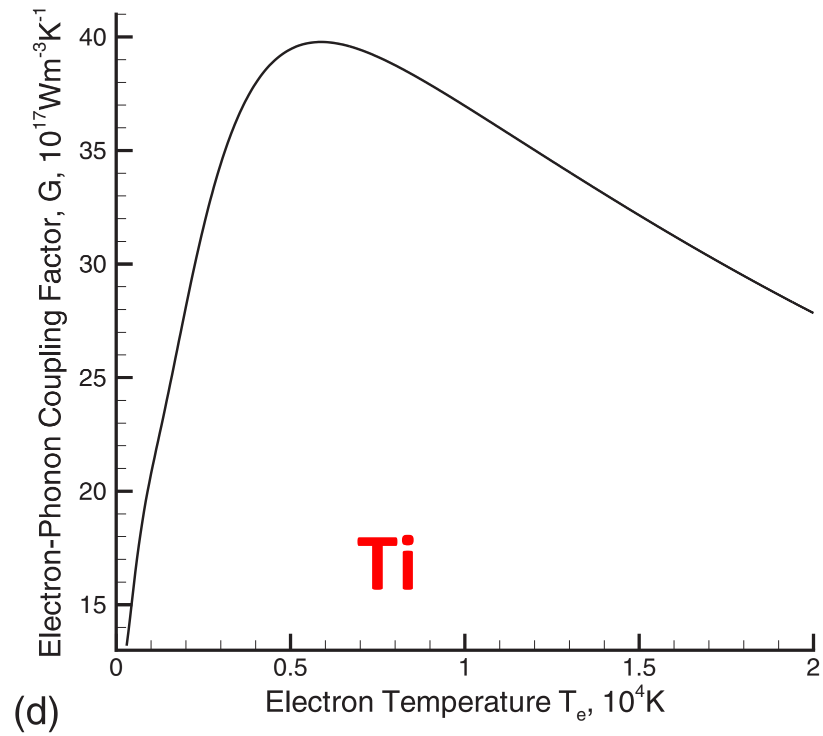
A metal with a high thermal conductivity also has a high electrical conductivity (low electrical resistivity)

A metal with a high thermal conductivity also has a high electrical conductivity (low electrical resistivity)

Material	ρ (Ω·m) at 20 °C	σ (S/m) at 20 °C	k (W/(m K))	k/σ (W Ω/K)
Silver	1.59×10^{-8}	6.30×10^{7}	429	6.81E-06
Copper	1.68×10^{-8}	5.96×10^{7}	401	6.73E-06
Gold	2.44×10^{-8}	4.10×10^{7}	318	7.76E-06
Aluminium	2.82×10^{-8}	3.50×10^{7}	230	6.57E-06
Calcium	3.36×10^{-8}	2.98×10^{7}	201	6.74E-06
Tungsten	5.60×10^{-8}	1.79×10^{7}	173	9.66E-06
Zinc	5.90×10^{-8}	1.69×10^{7}	116	6.86E-06
Nickel	6.99×10^{-8}	1.43×10^{7}	91	6.36E-06
Lithium	9.28×10^{-8}	1.08×10^{7}	85	7.87E-06
Iron	1.00×10^{-7}	1.00×10^{7}	80	8.00E-06
Platinum	1.06×10^{-7}	9.43×10^{6}	72	7.64E-07
Tin	1.09×10^{-7}	9.17×10^{6}	67	7.31E-07
Lead	2.20×10^{-7}	4.55×10^{6}	35	7.69E-06
Titanium	4.20×10^{-7}	2.38×10^{6}	22	9.24E-07

In general: larger electron-phonon scattering rates (EP coupling factor), lower k in metals





PHYSICAL REVIEW B **77**, 075133 (2008)

Electron-phonon coupling and electron heat capacity of metals under conditions of strong electron-phonon nonequilibrium

Zhibin Lin and Leonid V. Zhigilei*

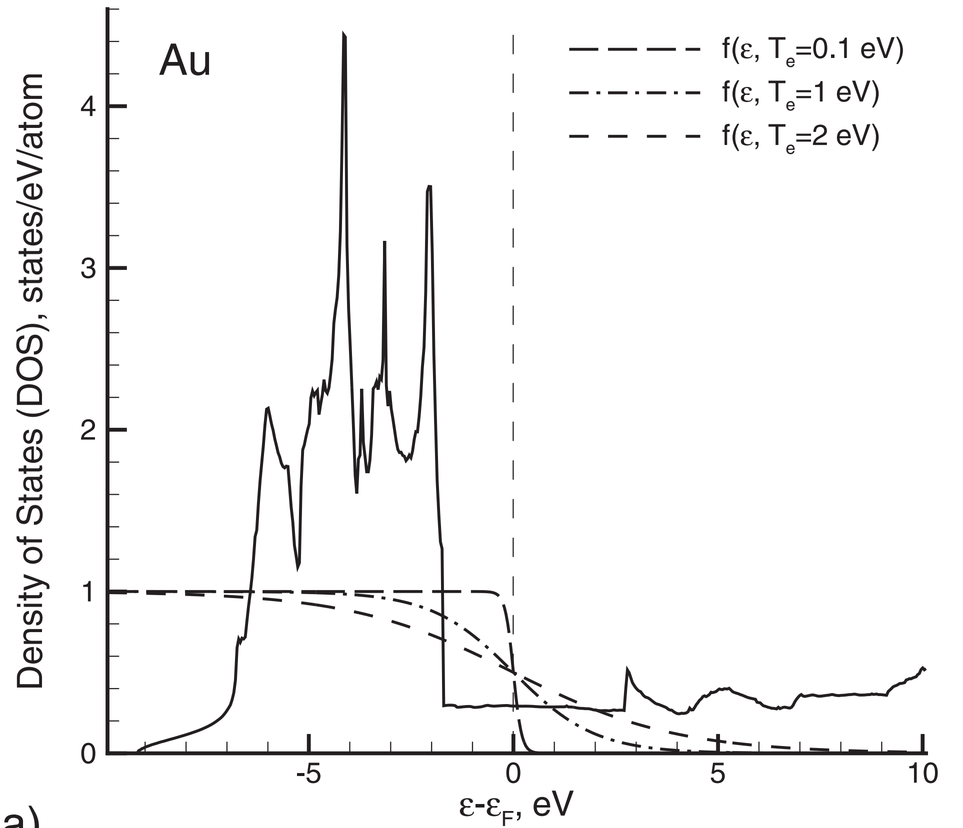
Department of Materials Science and Engineering, University of Virginia, 395 McCormick Road, Charlottesville, Virginia 22904-4745, USA

Vittorio Celli

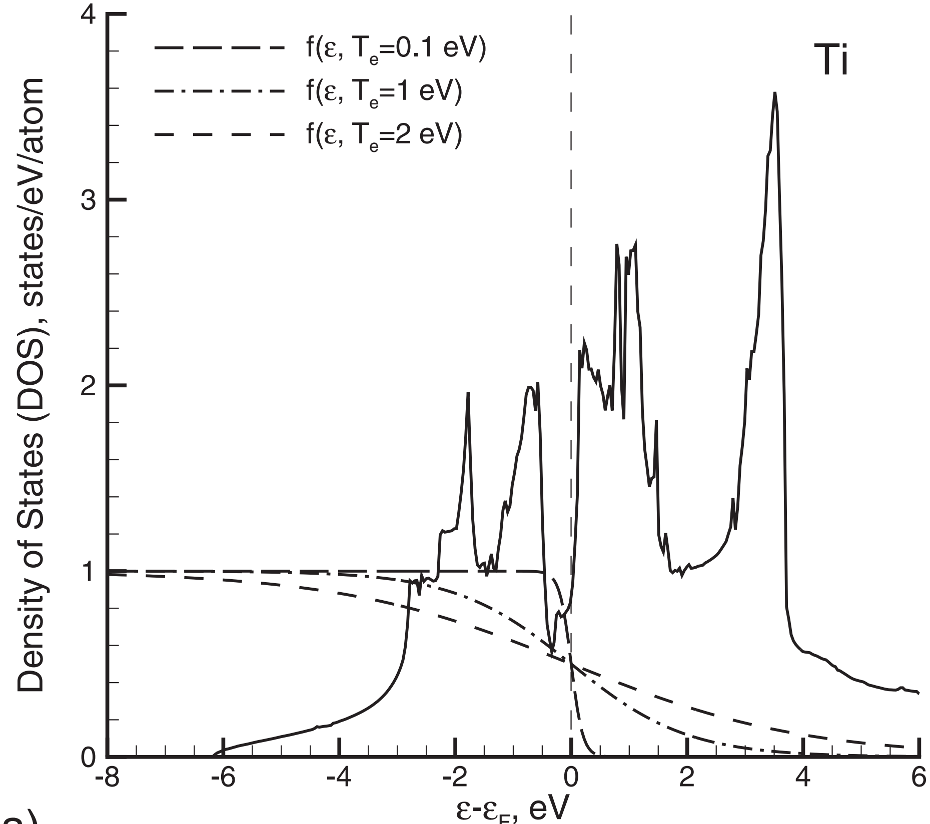
Department of Physics, University of Virginia, 382 McCormick Road, Charlottesville, Virginia 22904-4714, USA

(Received 23 August 2007; revised manuscript received 22 December 2007; published 28 February 2008)

Band-structure/DOS determines EP coupling and k



(a)



(a)

PHYSICAL REVIEW B **77**, 075133 (2008)

Electron-phonon coupling and electron heat capacity of metals under conditions of strong electron-phonon nonequilibrium

Zhibin Lin and Leonid V. Zhigilei*
Department of Materials Science and Engineering, University of Virginia, 395 McCormick Road, Charlottesville, Virginia 22904-4745, USA

Vittorio Celli
Department of Physics, University of Virginia, 382 McCormick Road, Charlottesville, Virginia 22904-4714, USA
(Received 23 August 2007; revised manuscript received 22 December 2007; published 28 February 2008)

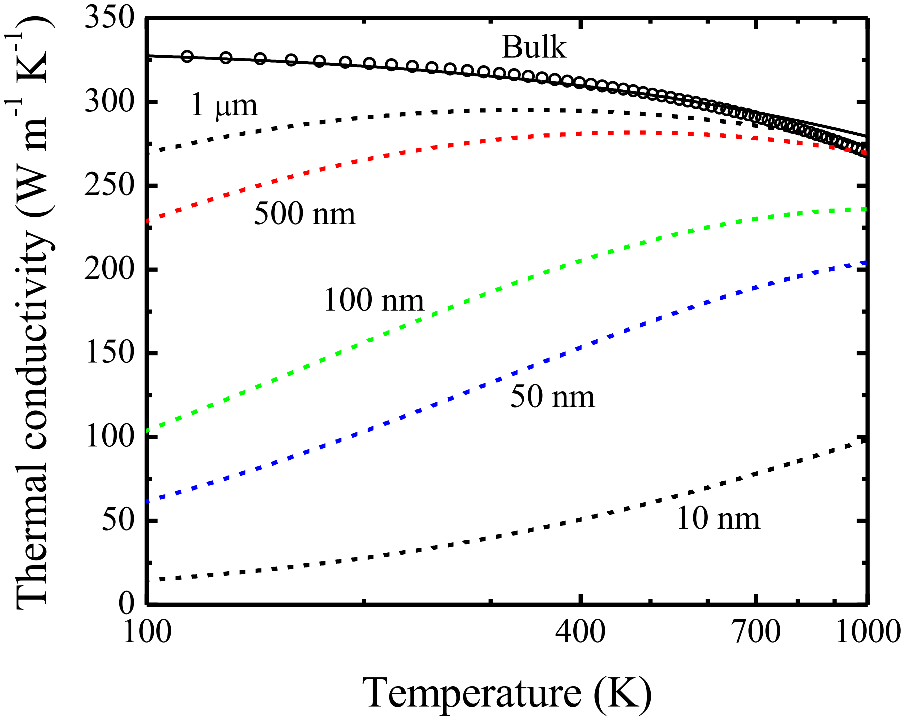
Band-structure/DOS determines EP coupling and k

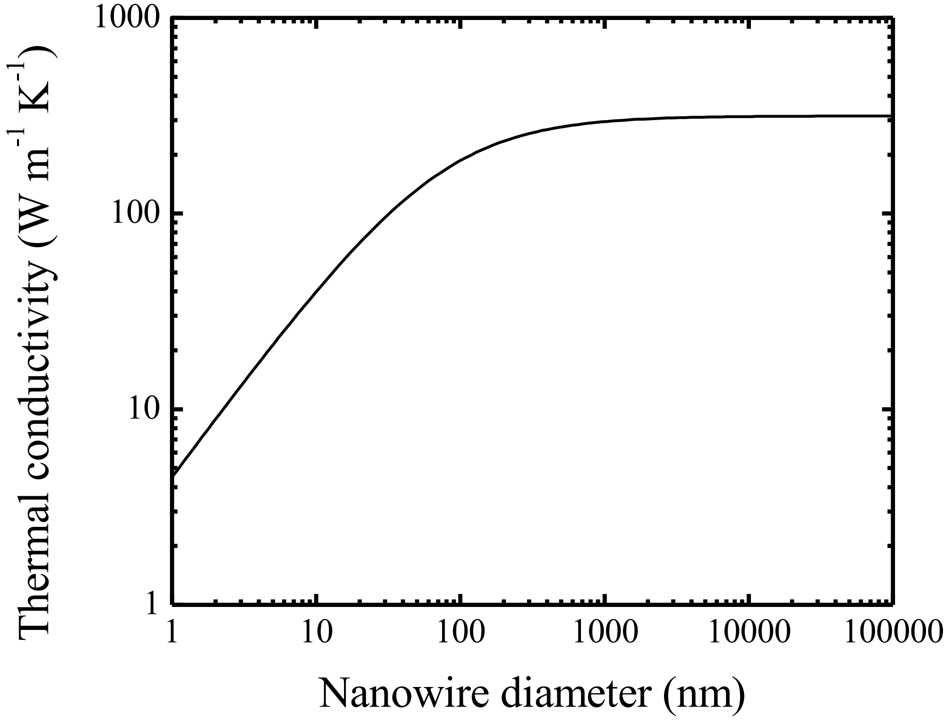
Metal	D-band to Fermi surface separation (eV)	κ at T = 300 K (W m^{-1} K^{-1})
Ag	4	429
Cu	2.15	401
Au	2.4	310
W	0.85	178
Cr	0.8	94
Ni	0.25	91

Thermal conductivity of metals

Size effects

$$\frac{1}{\tau_{metal,nano}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_D} + \frac{1}{\tau_B}$$

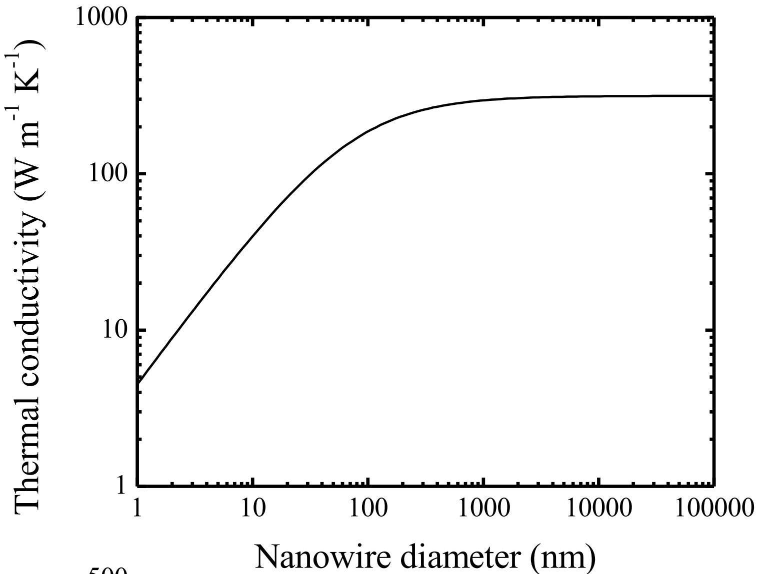


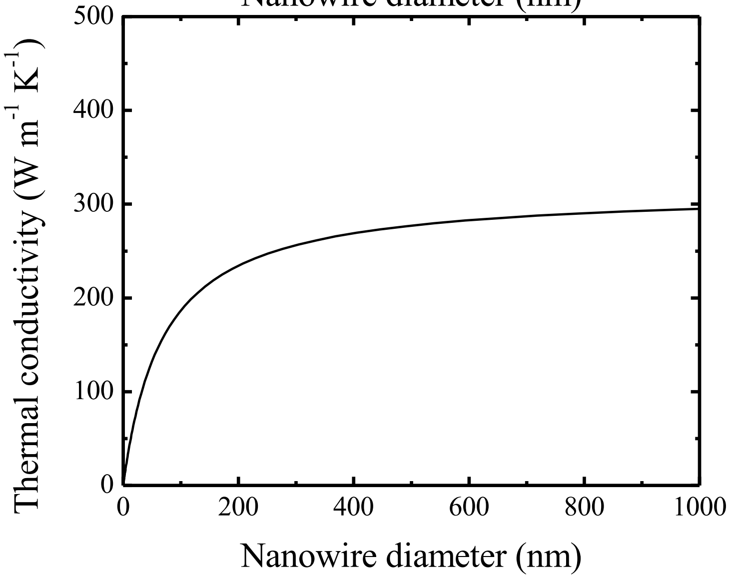


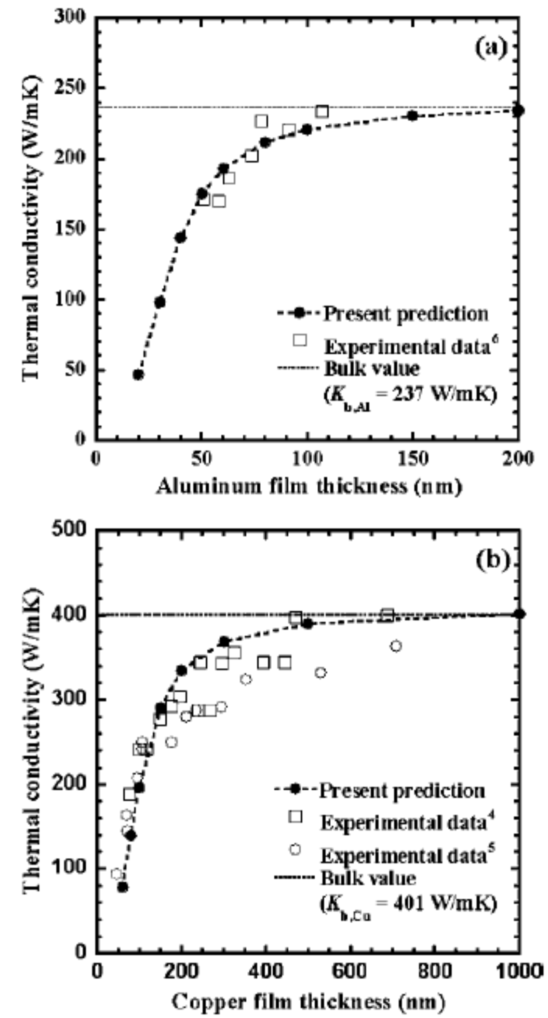
Thermal conductivity of metals

Size effects

$$\frac{1}{\tau_{metal,nano}} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_D} + \frac{1}{\tau_B}$$

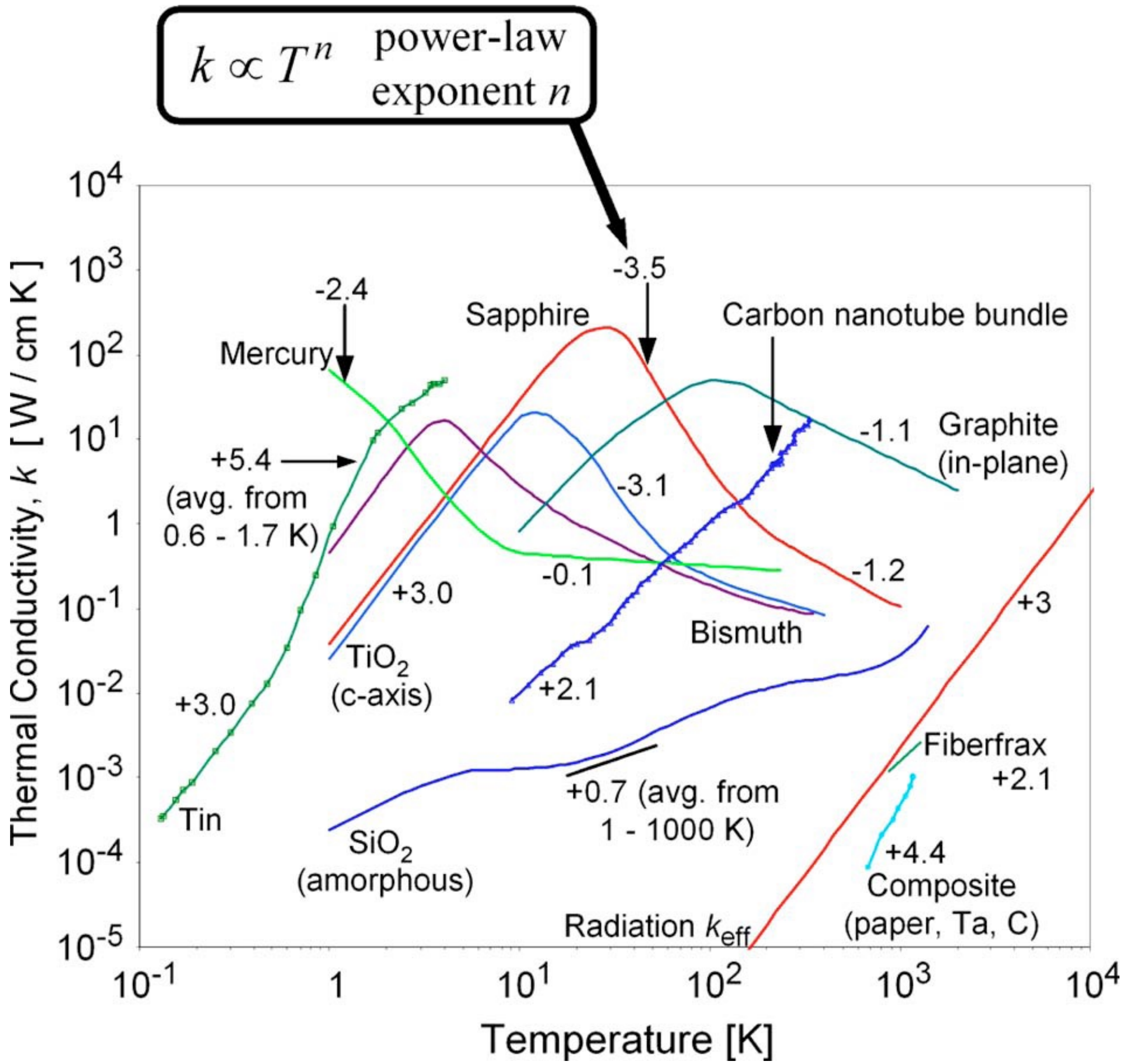






Jin *et al*, *APL* **92** 171910 (2008)

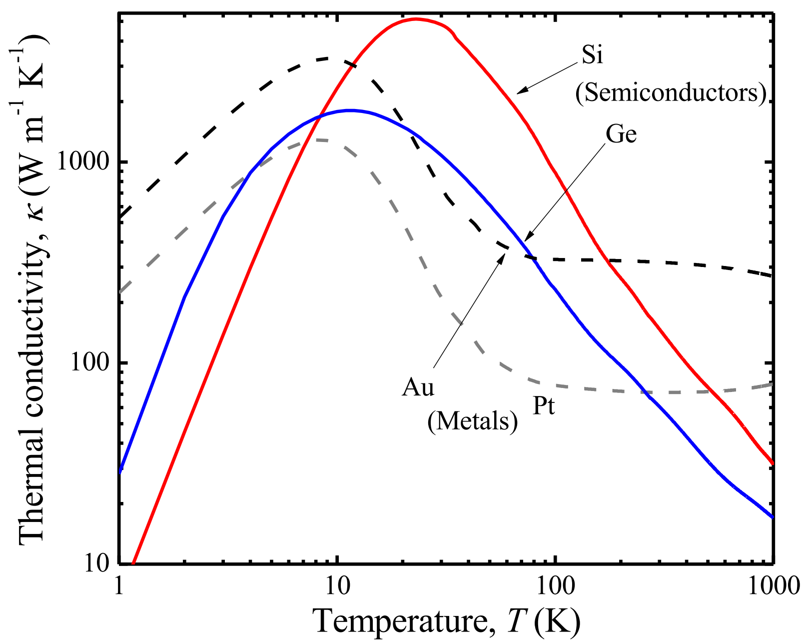
Some commonly used nonmetals

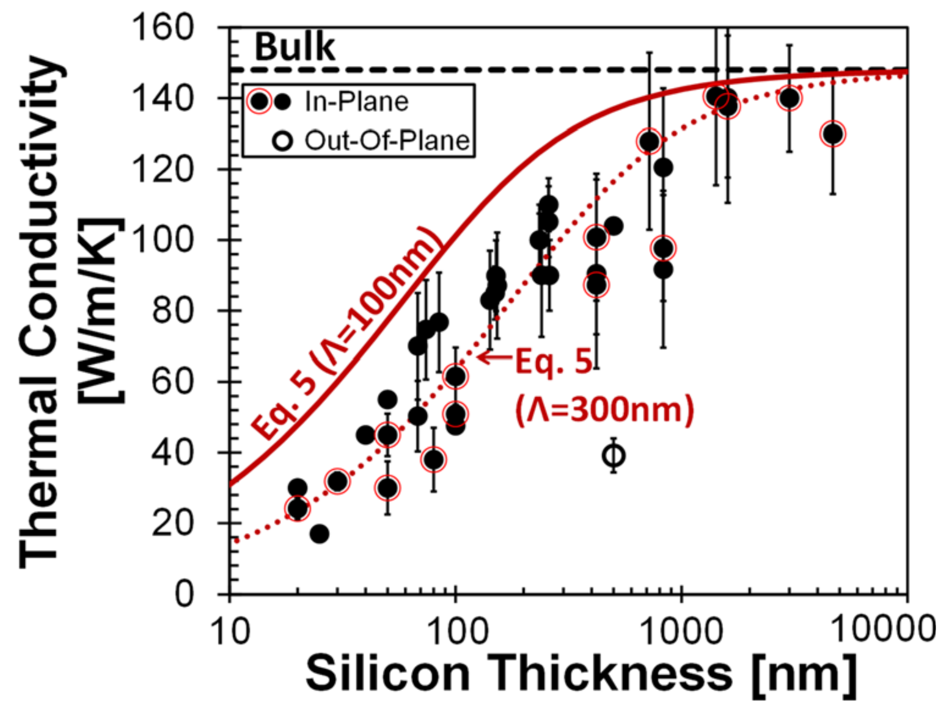


Dames, “Solid-state thermal rectification with existing bulk materials," Journal of Heat Transfer, **131**, 061301 (2009).

κ in Si nanosystems

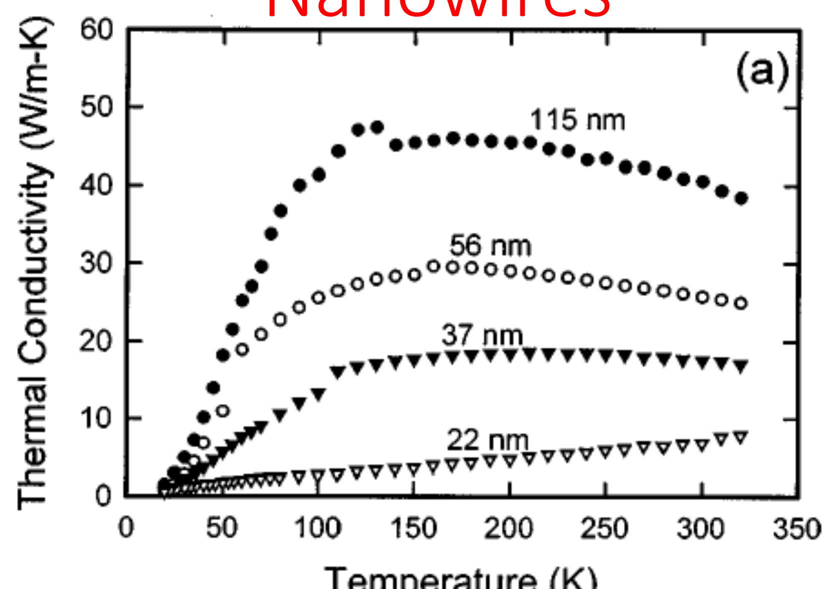
Bulk thermal conductivity





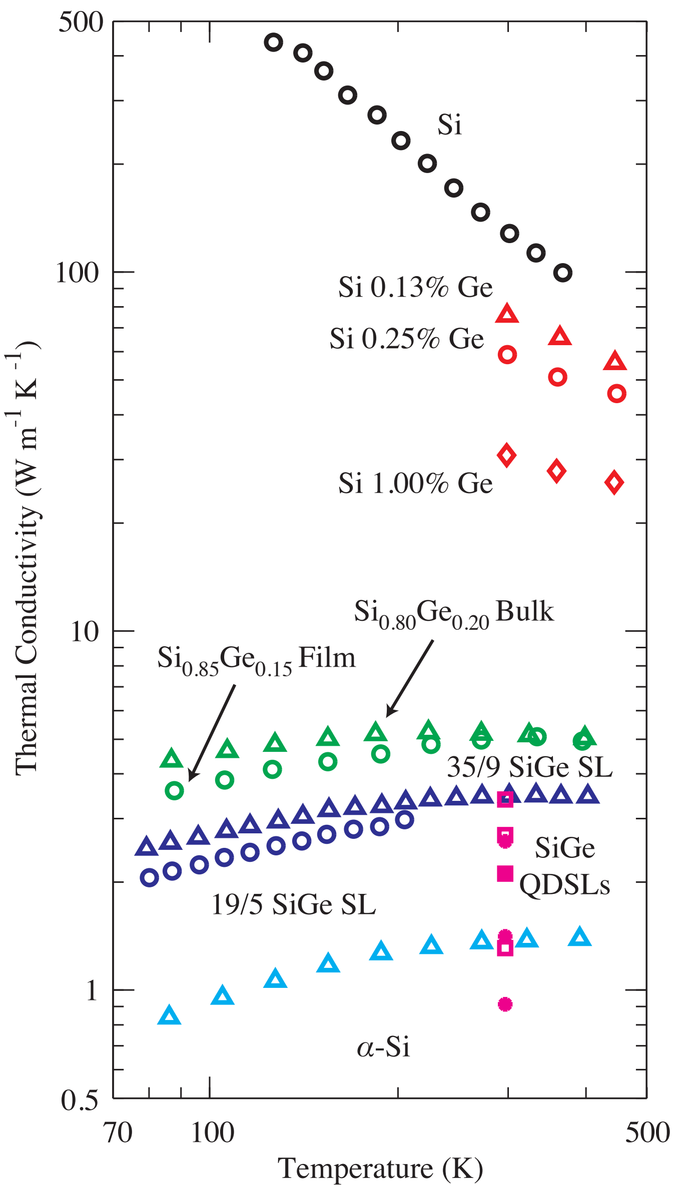
Marconnet, *JHT* **135**, 061601 (2013).

Nanowires



Li *et al*, *APL* **83**, 2934 (2003).

κ in Si: other "nano" resistances



Dilute alloy

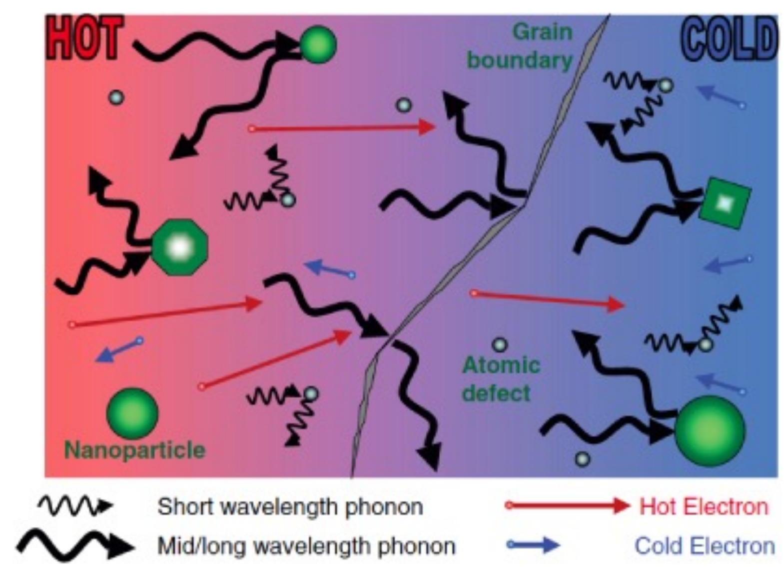
Bulk alloy

Interfaces

Complete disorder (amorphous)

$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$

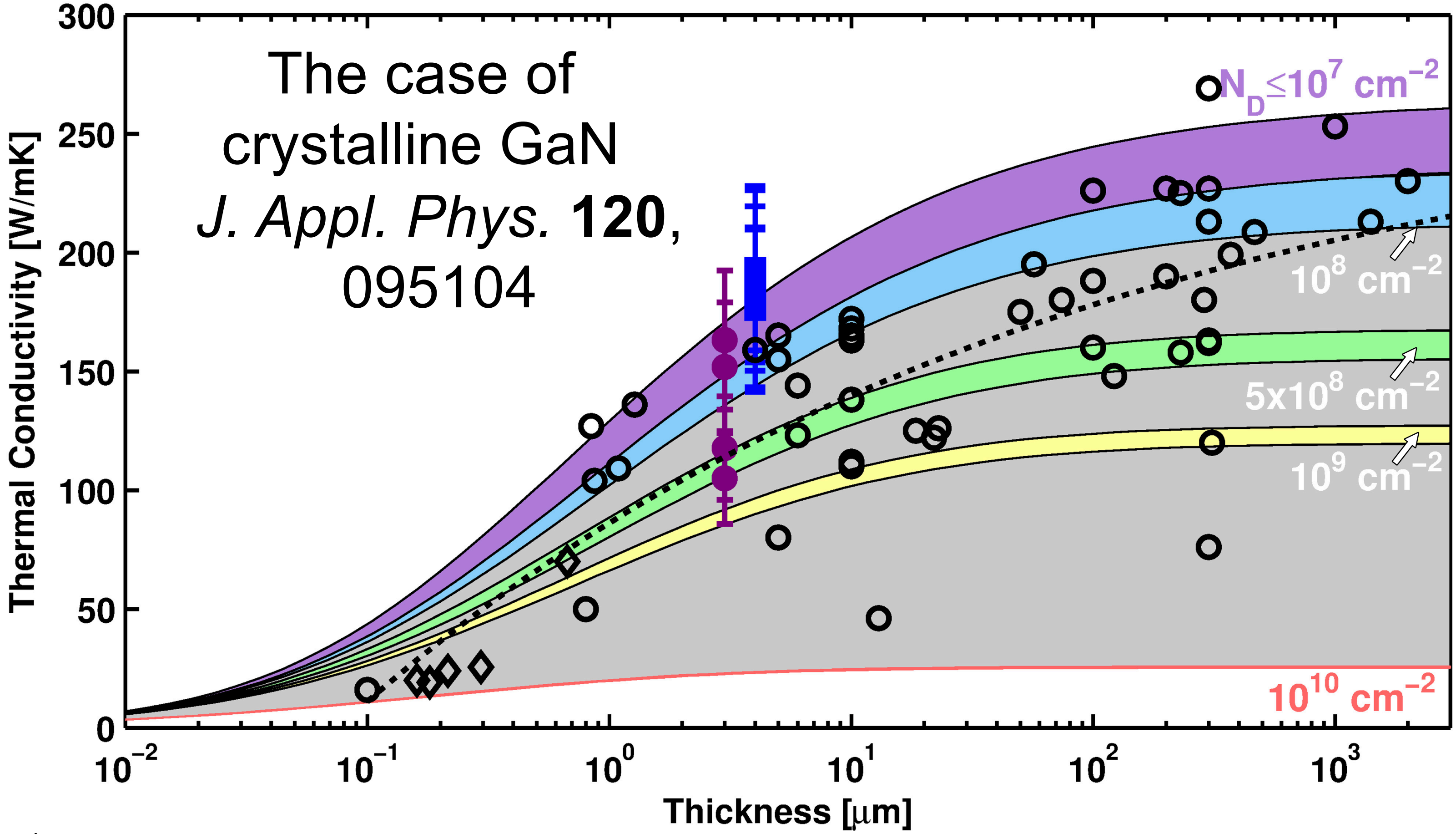
$$\tau = f\left(\tau_{\text{intrinsic}}, \tau_{\text{impurity}}, \tau_{\text{boundary}}\right)$$



Adv. Mat. **22**, 3970 (2010)

Thermal conductivity of materials – Nanoscale behavior

Well controlled and prescribed inclusions, defects, or interfaces change thermal conductivity based on manipulating the behavior of electrons and phonons



$$\kappa = \frac{1}{3} C v \lambda$$

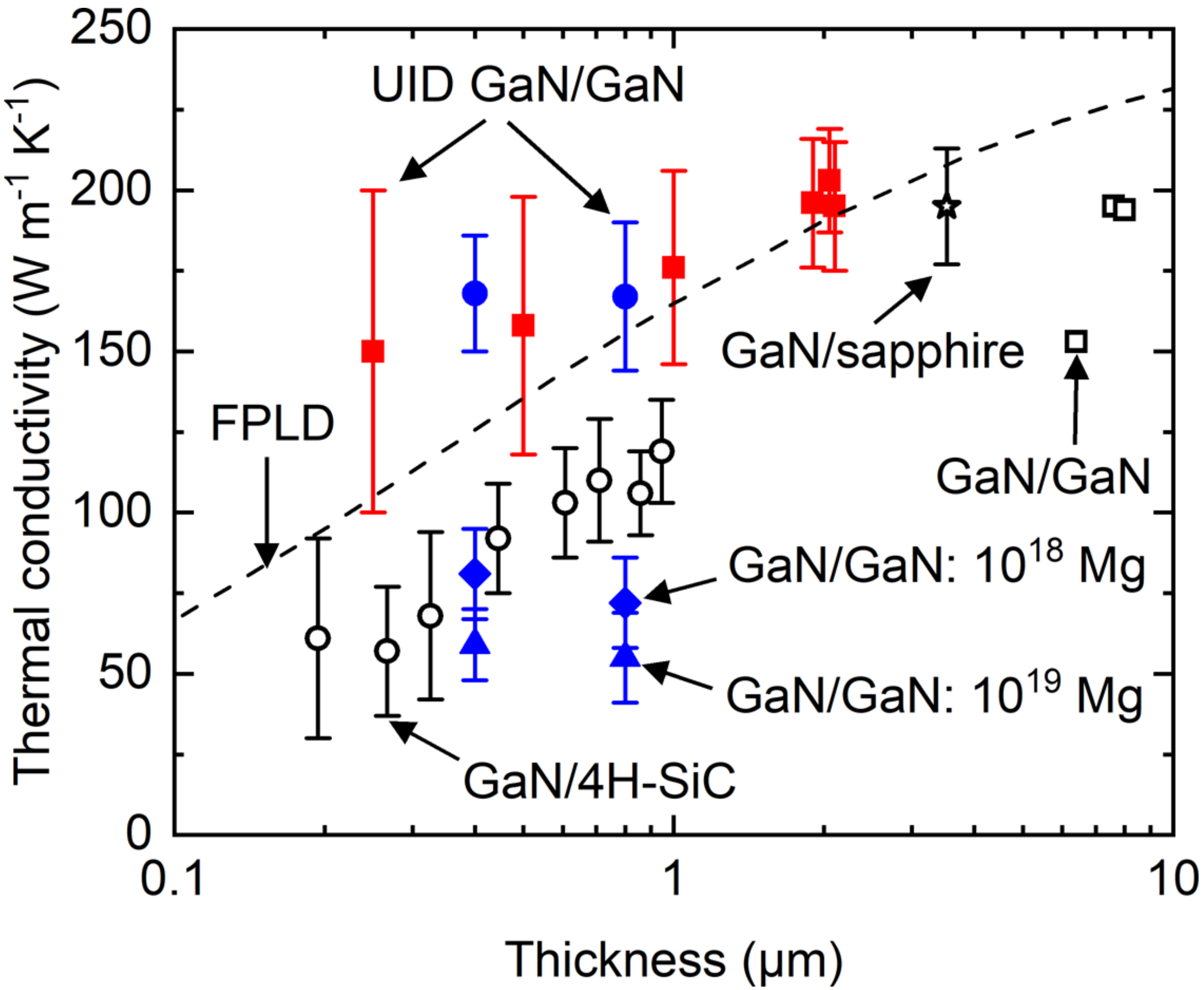
Interfaces critical when thickness < λ

Dislocations critical when spacing < λ

Thermal conductivity of materials – Nanoscale behavior

Well controlled and prescribed inclusions, defects, or interfaces change thermal conductivity based on manipulating the behavior of electrons and phonons

The case of crystalline GaN
Phys. Rev. Mat. **5**, 104604



$$\kappa = \frac{1}{3} C v \lambda$$

Dopant scattering critical when $(n_{density})^{-1/3} < \lambda$

Thermal conductivity of non-metals (phonons)

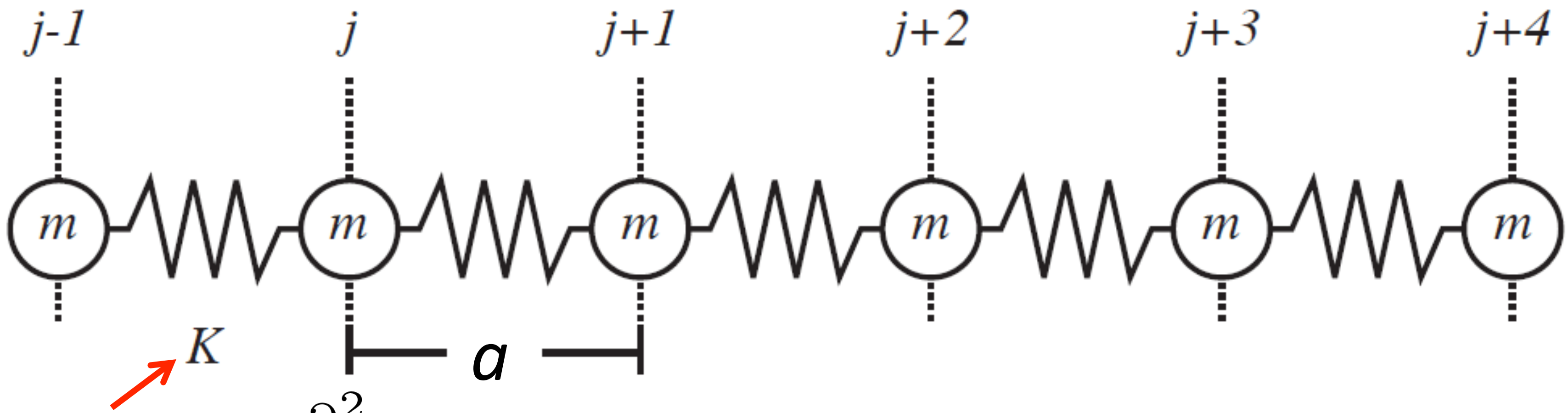
$$\frac{1}{\tau_{semi}} = \frac{1}{\tau_{pp}} + \frac{1}{\tau_D} + \frac{1}{\tau_B}$$

Phonon-phonon scattering $$\frac{1}{\tau_{pp}} = AT\omega^2 \exp\left[-\frac{B}{T}\right]$$

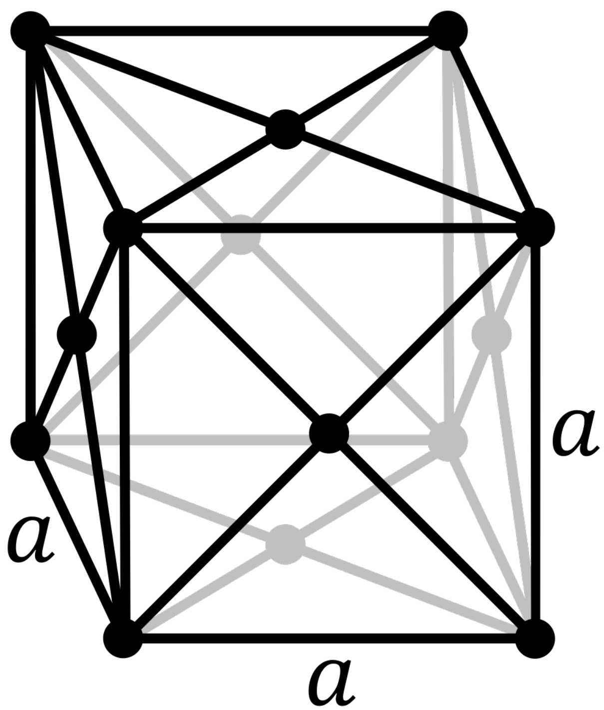
Phonon-defect scattering $$\frac{1}{\tau_D} = C\omega^4$$

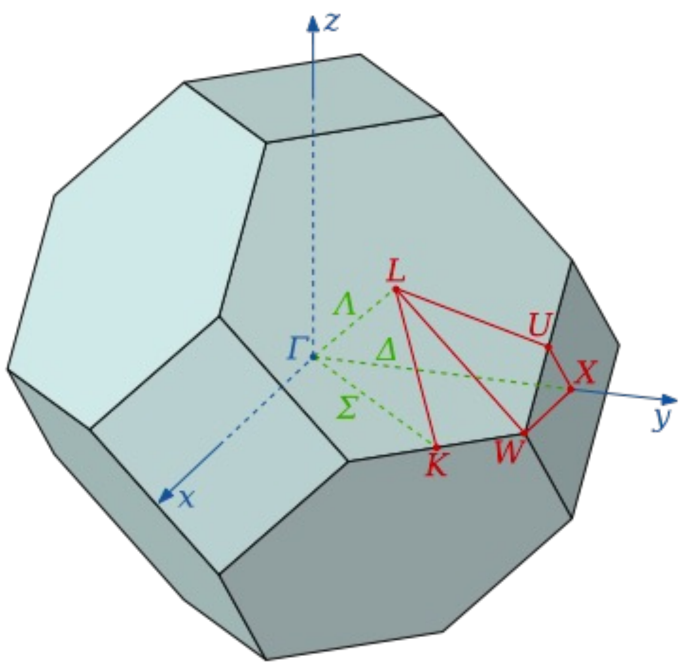
Phonon-boundary scattering $$\frac{1}{\tau_B} = \frac{2v}{L}$$

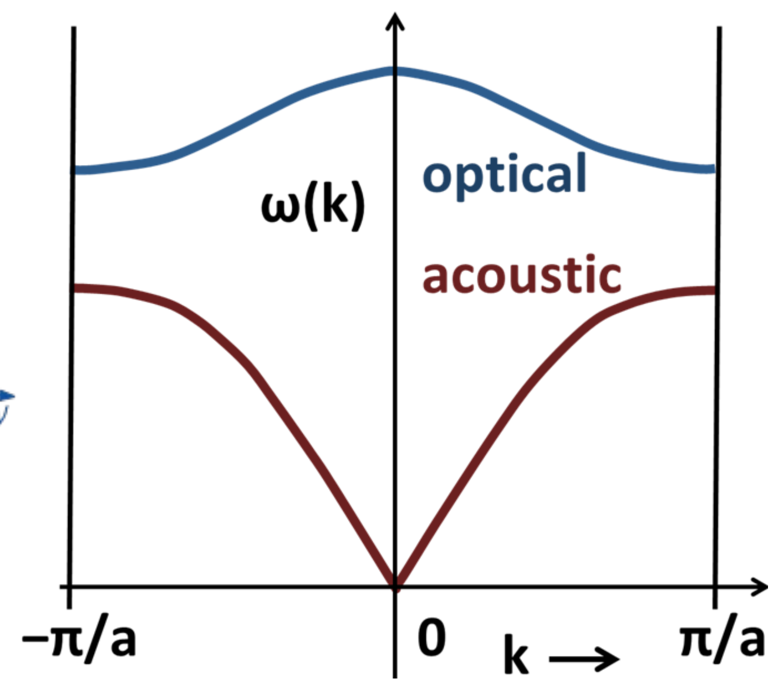
The spectrum of phonons



$$K = -\frac{\partial^2 u}{\partial x^2}$$





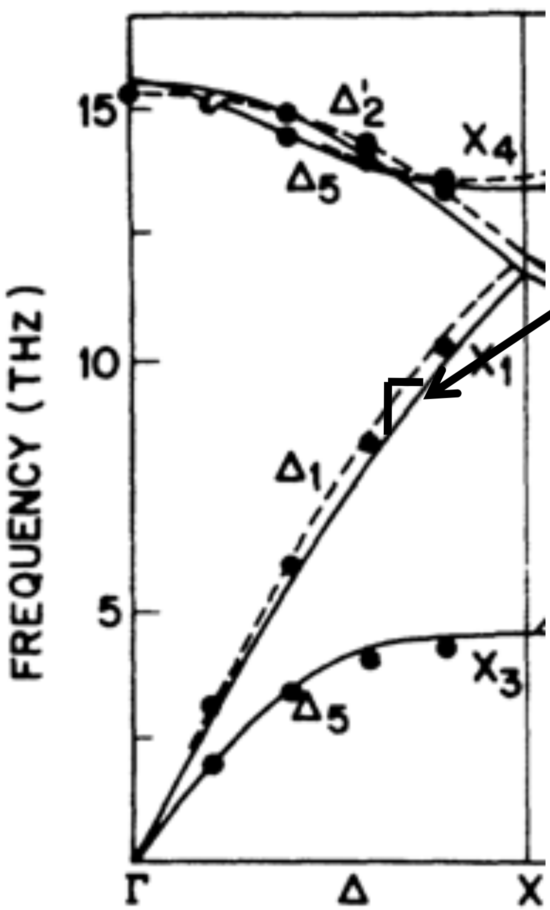


Wikipedia

Phonon dispersion and velocities

$$\Lambda = \frac{2\pi}{k}$$

@ k = 0, Λ = infinity so phonons are affected by the physical boundaries of the samples being measured

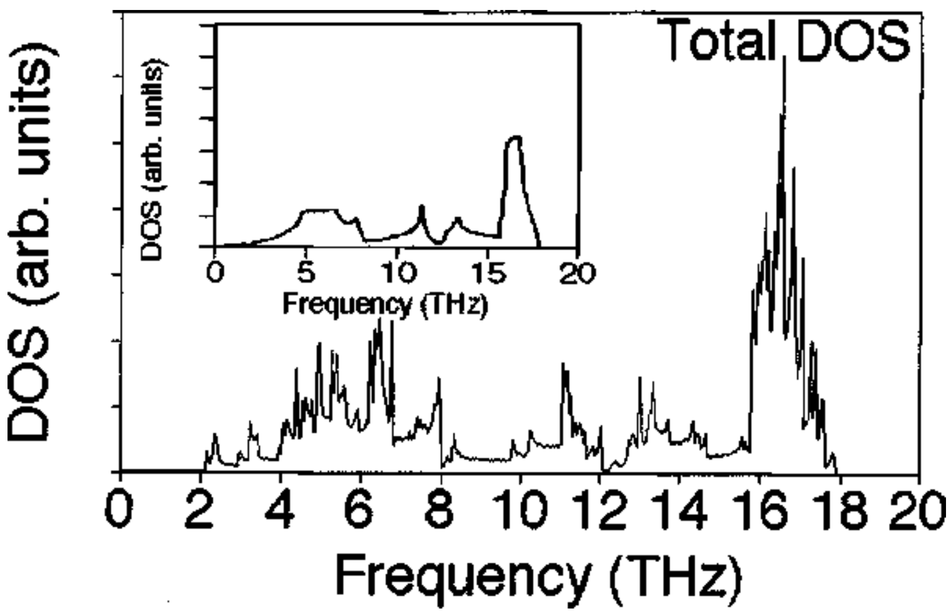


$$v(\omega) = \frac{\partial \omega}{\partial k}$$

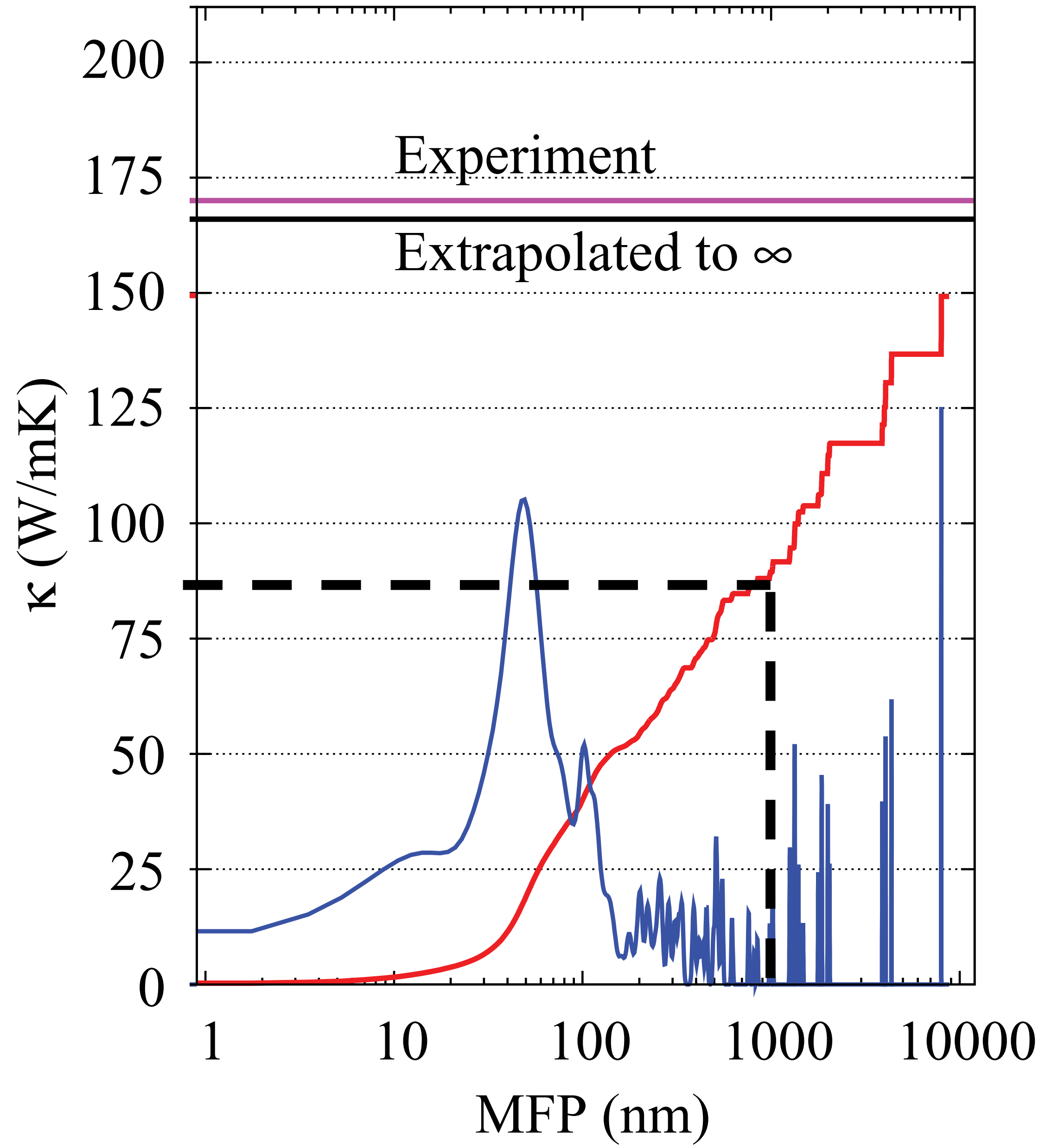
The "bandwidth" of phonons

$$\kappa = \int_\omega C_\omega v_\omega \lambda_\omega \, d\omega$$

50% of heat in Si carried by phonons with MFP's ~1 μm and less



Schelling, Phillpot, Keblinski
J. Appl. Phys. **95**, 6082

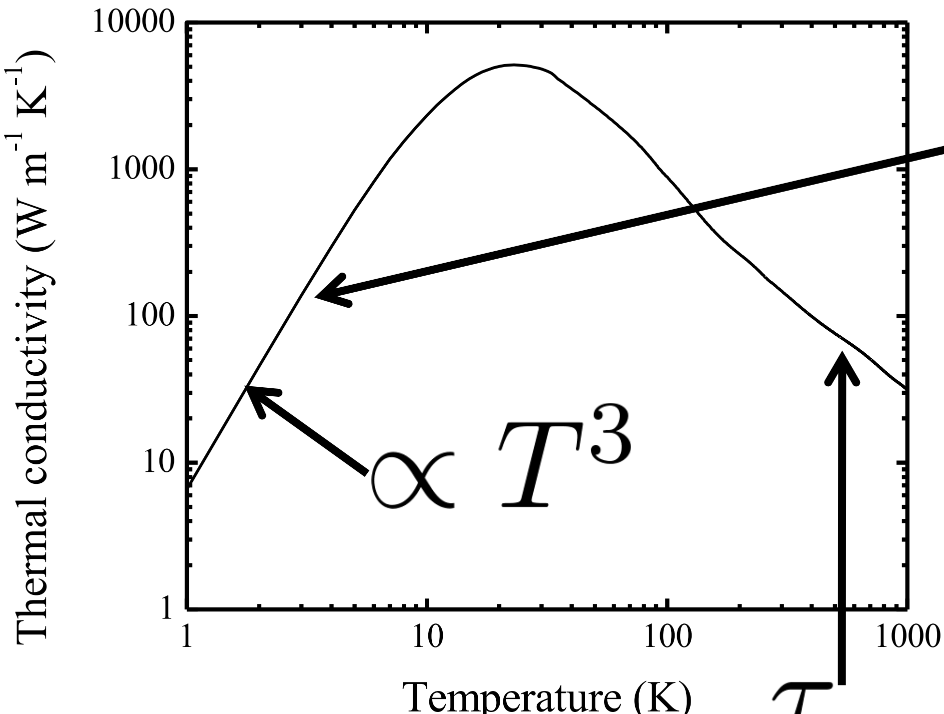


Phys. Rev. B **84**, 085204

Thermal conductivity of semiconductors

$$\kappa = \frac{1}{3} C v^2 \tau_{semi} \qquad v\tau = \lambda$$

$$\kappa = \frac{1}{3} \sum_j \int_0^{\omega_{max,j}} \hbar \omega D(\omega) \frac{\partial f}{\partial T} v_j^2(\omega) \tau_{semi} \, \mathrm{d}\omega$$



Follows heat capacity trend but reduced by boundary and impurity scattering

$$\frac{1}{\tau_{pp}} = AT\omega^2 \exp\left[-\frac{B}{T}\right]$$

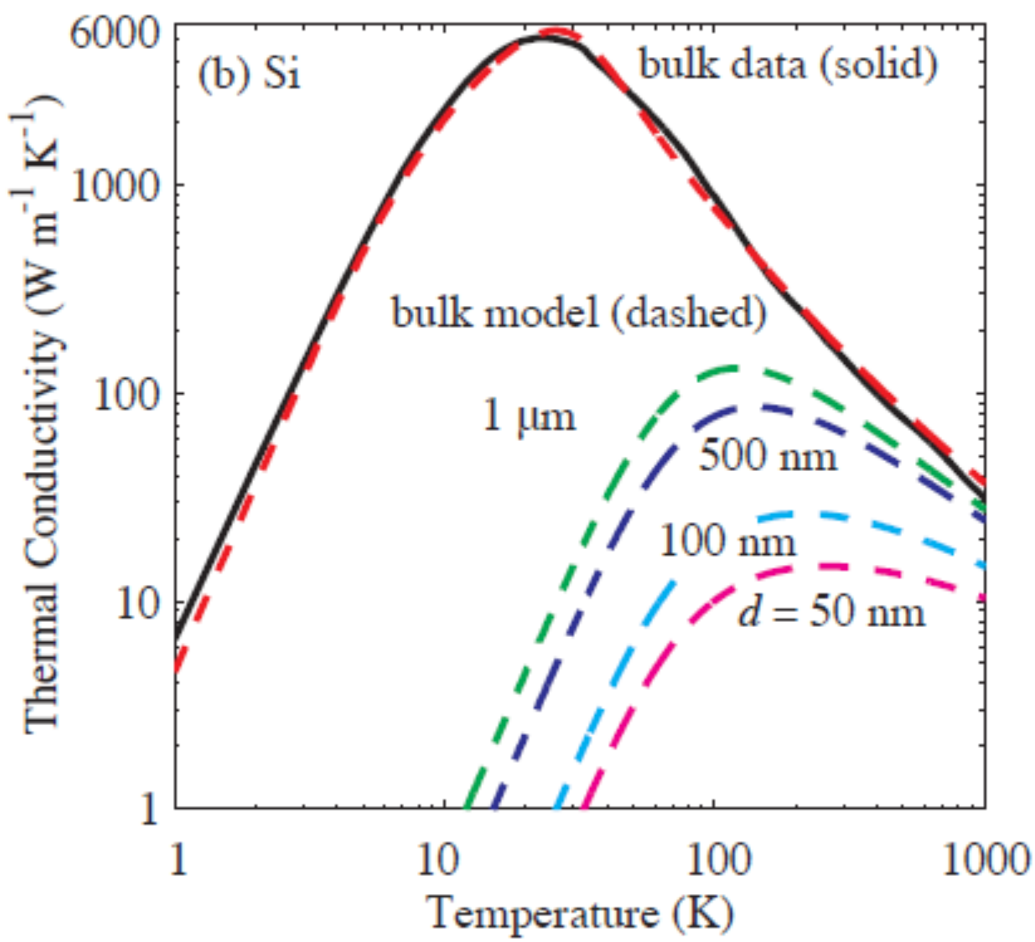
$$\tau_{pp} \propto \lambda_{pp} \propto T^{-1}$$

Thermal conductivity of semiconductors

Size effects

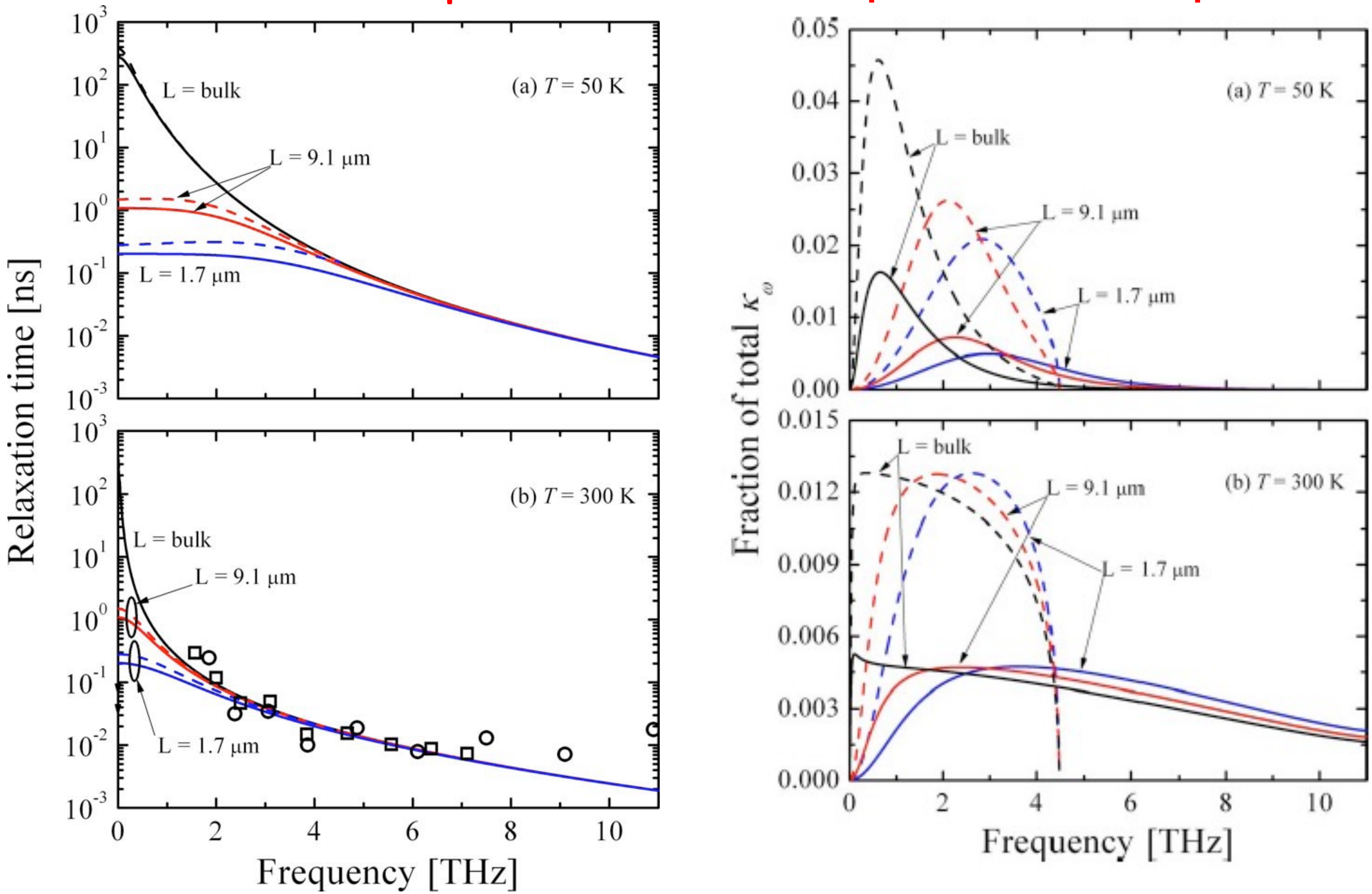
$$\kappa = \frac{1}{3}\sum_j \int_0^{\omega_{max,j}} \hbar\omega D(\omega)\frac{\partial f}{\partial T} v_j^2(\omega)\tau_{semi}\,\mathrm{d}\omega$$

$$\frac{1}{\tau_B} = \frac{2v}{L}$$



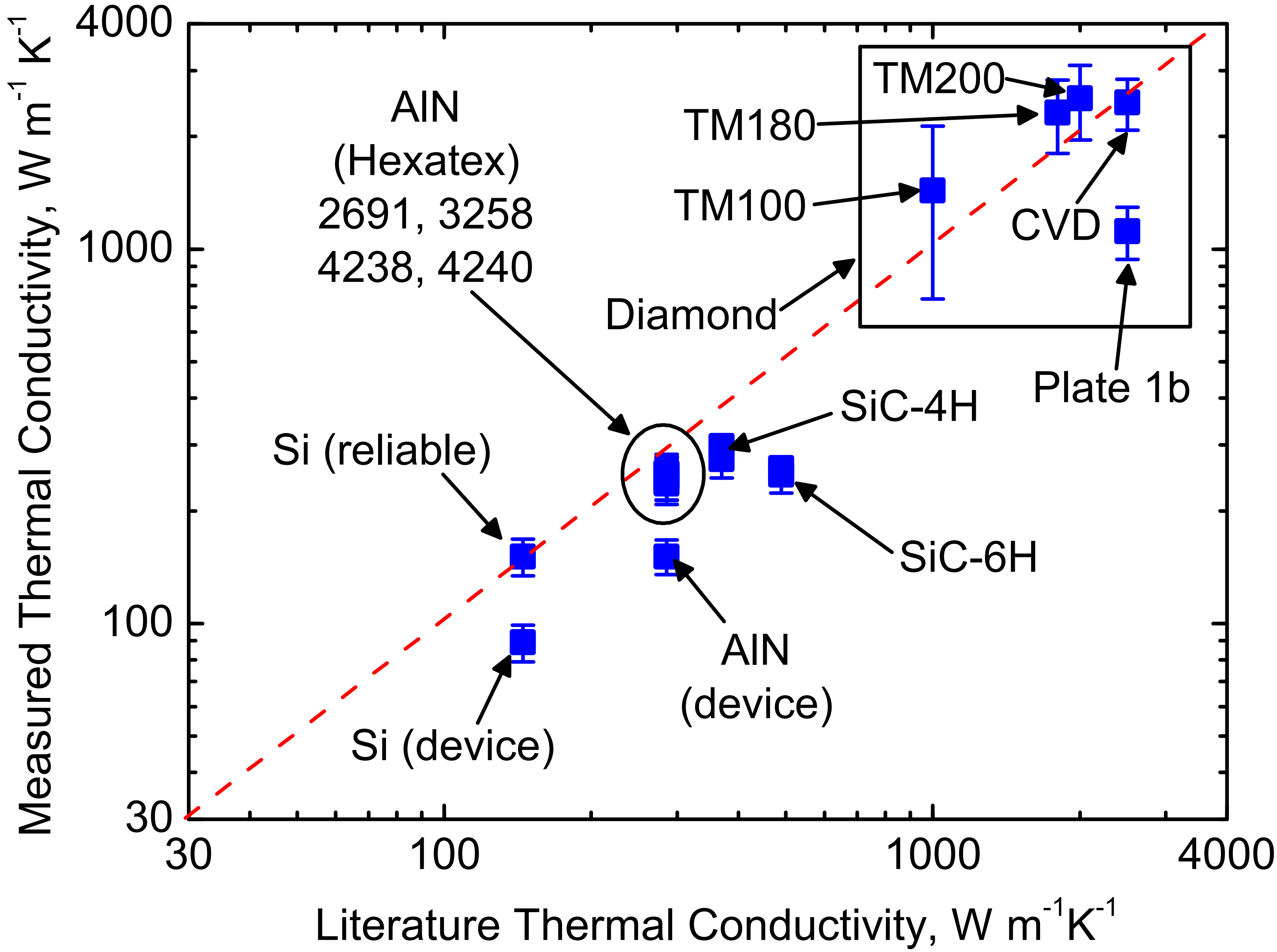
Thermal conductivity of semiconductors

Size effects: spectral nature of phonon transport



Hopkins *et al*., Appl. Phys. Lett **95**, 161902 (2009).

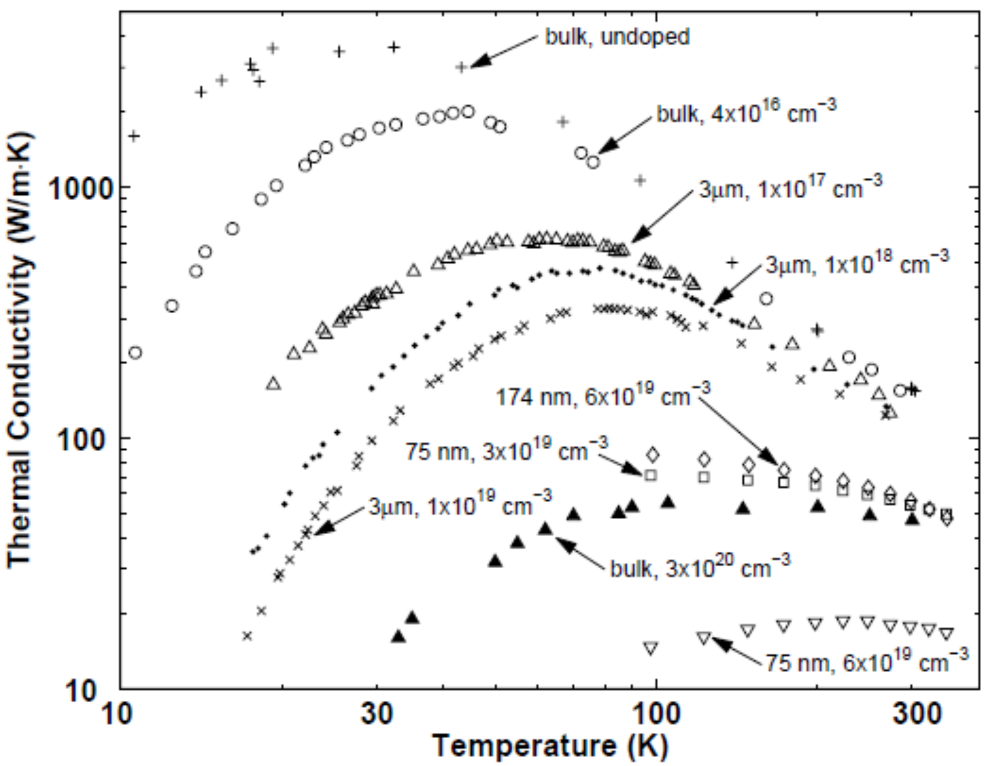
Point defects are a big concern

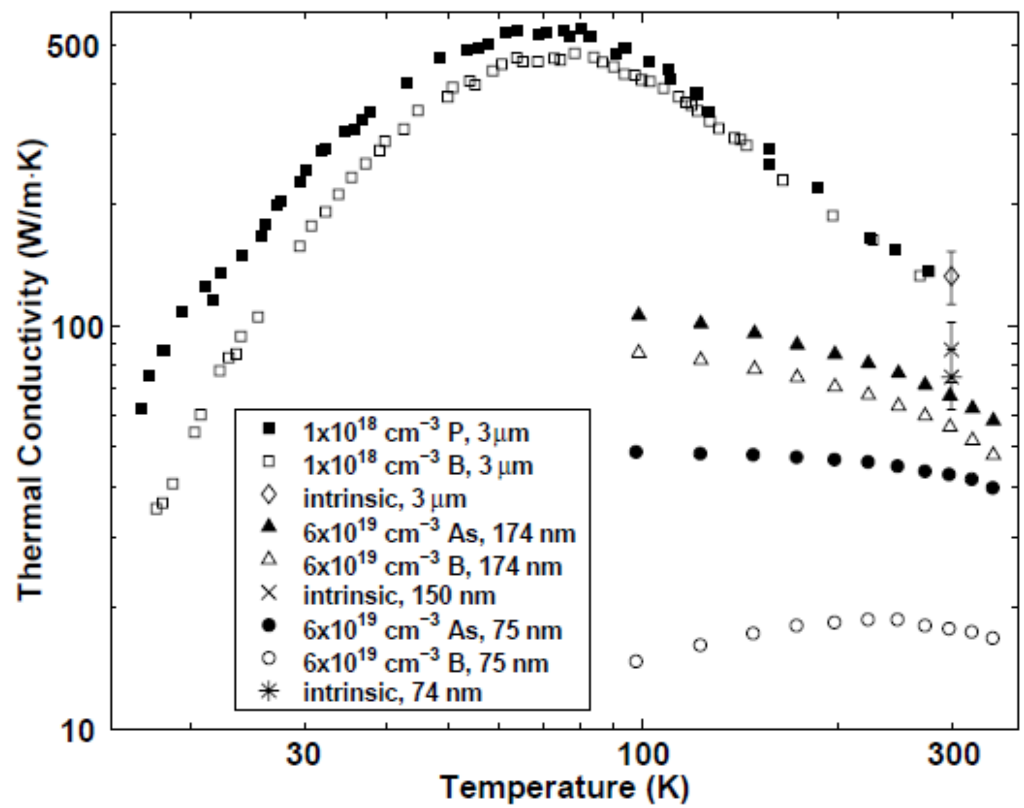


Other phonon scattering mechanisms

$$\frac{1}{\tau_{semi}} = \sum_j \frac{1}{\tau_j}$$

Dopant scatering





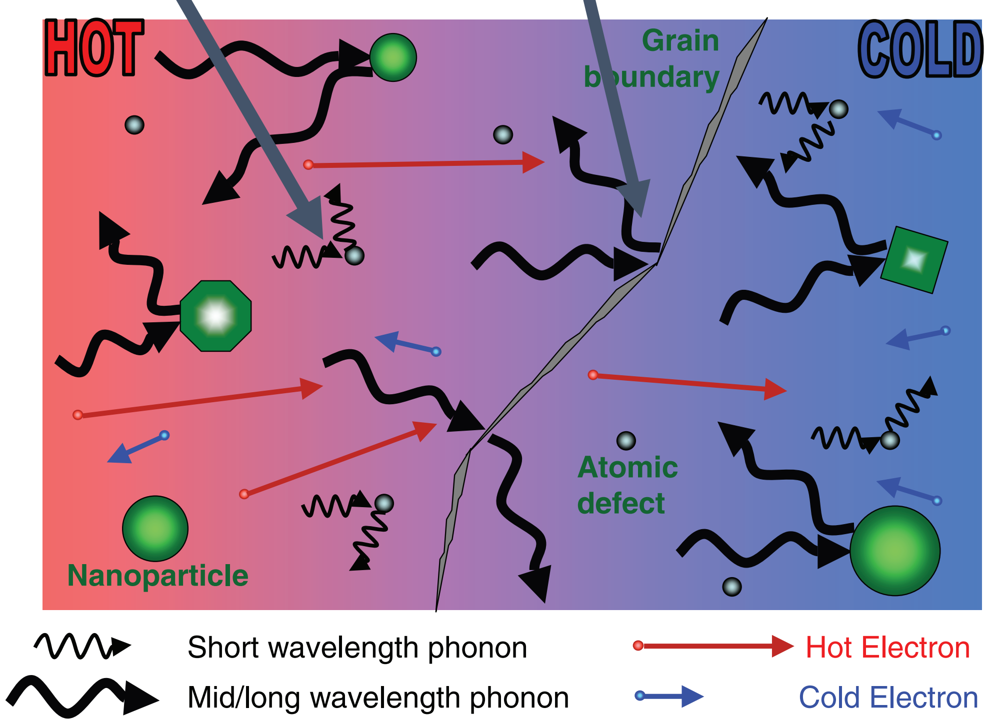
McConnell and Goodson, *Annual Review of Heat Transfer* **14**, 129 (2005).

Spectral phonon transport – The "bandwidth" of phonons

Thermal conductivity of alloys

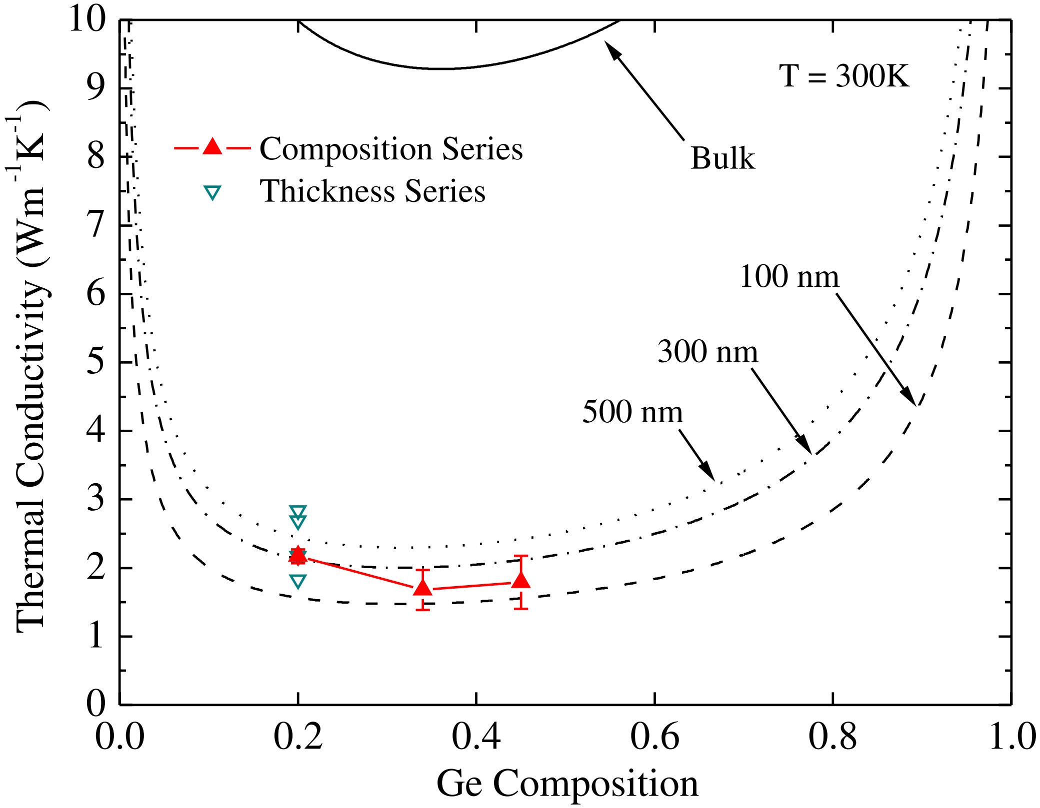
Short wave
point defects

Long wave
Boundaries



Adv. Mat. **22**, 3970

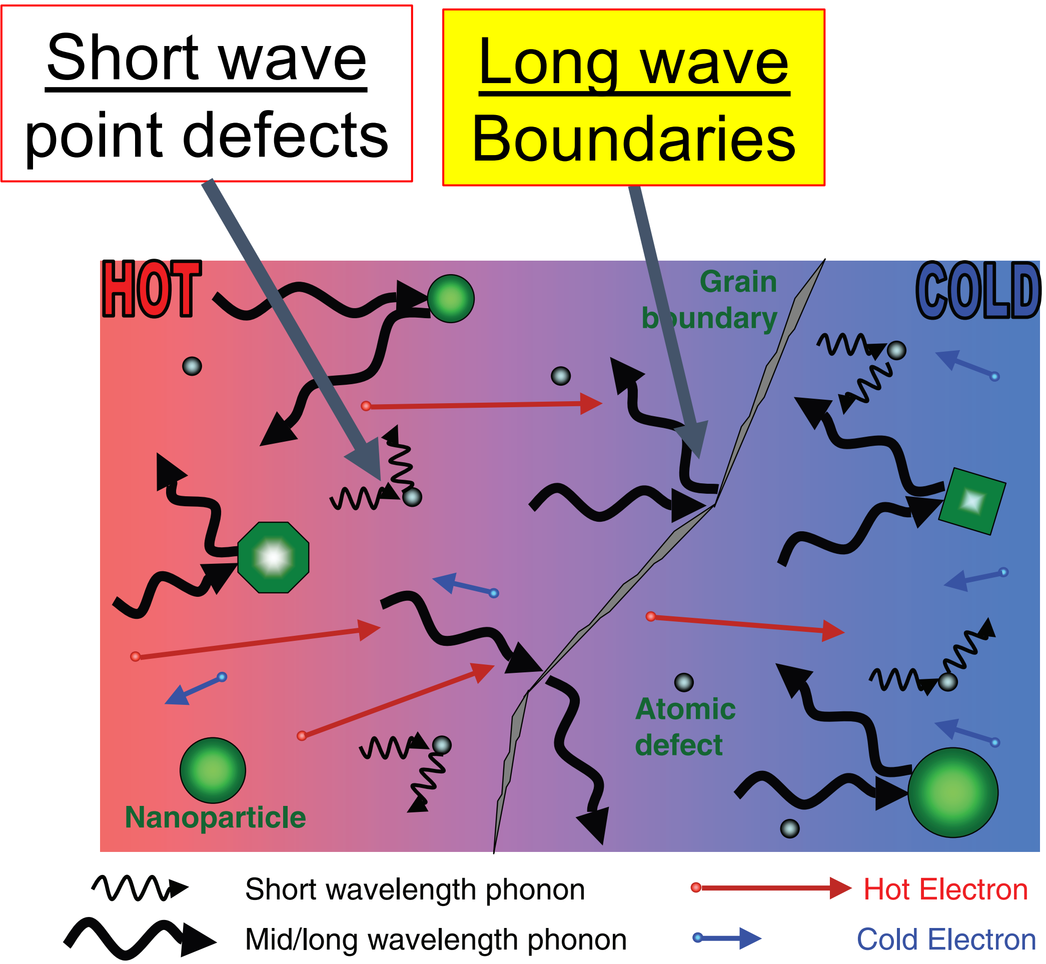
Thermal conductivity $Si_{1-x}Ge_x$ alloys



Phys. Rev. Lett. **109**, 195901

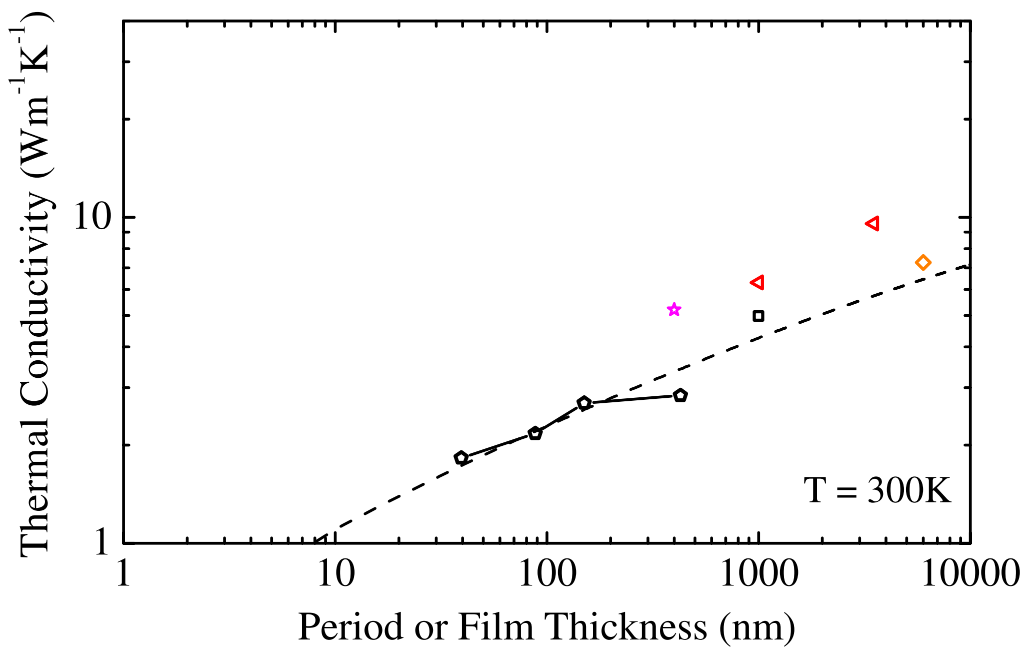
Spectral phonon transport – The “bandwidth” of phonons

Thermal conductivity of alloy thin films



Adv. Mat. **22**, 3970

Thermal conductivity $Si_{1-x}Ge_x$ alloys



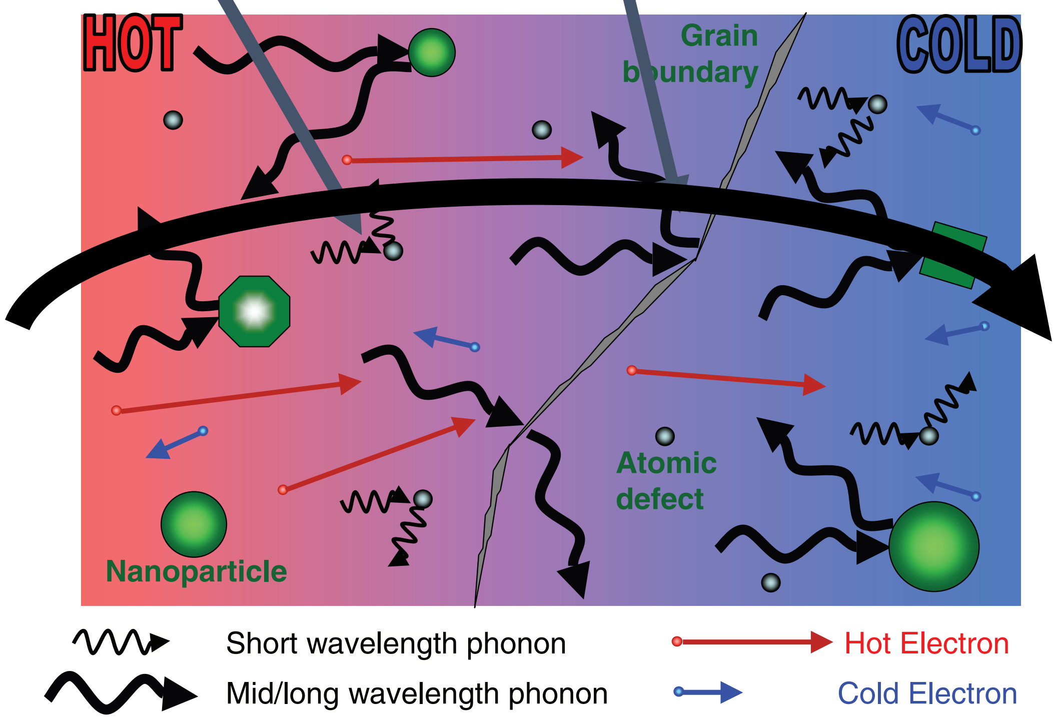
Phys. Rev. Lett. **109**, 195901

Spectral phonon transport – The “bandwidth” of phonons

Spectral thermal conductivity of alloys

<u>Short wave</u>
point defects

<u>Long wave</u>
Boundaries



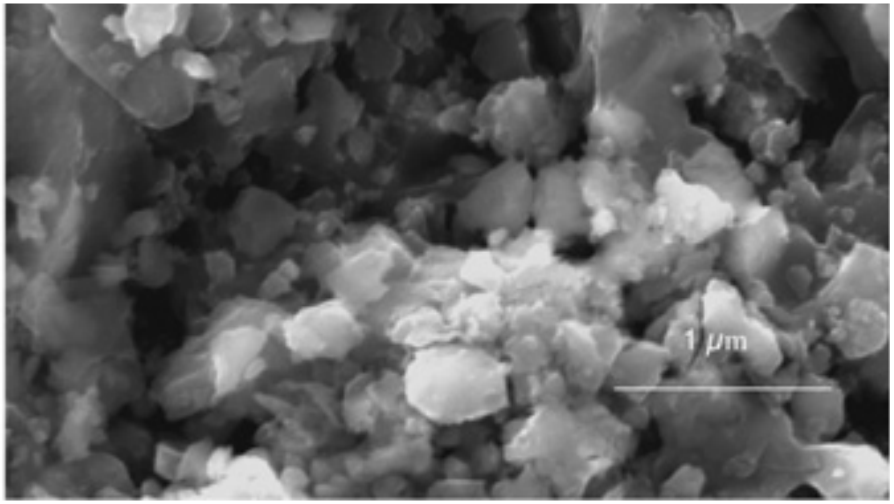
Adv. Mat. **22**, 3970

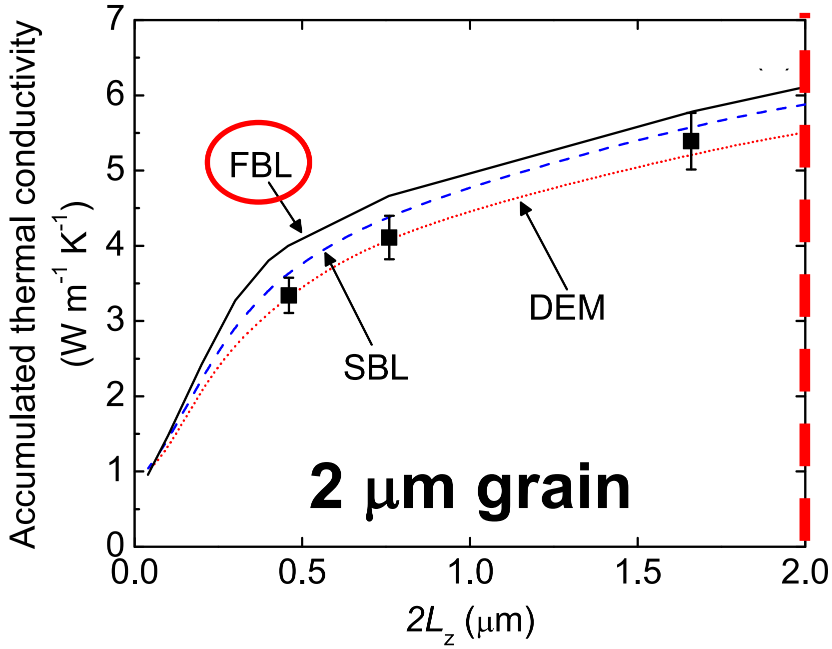
How about long, long wavelength phonons??

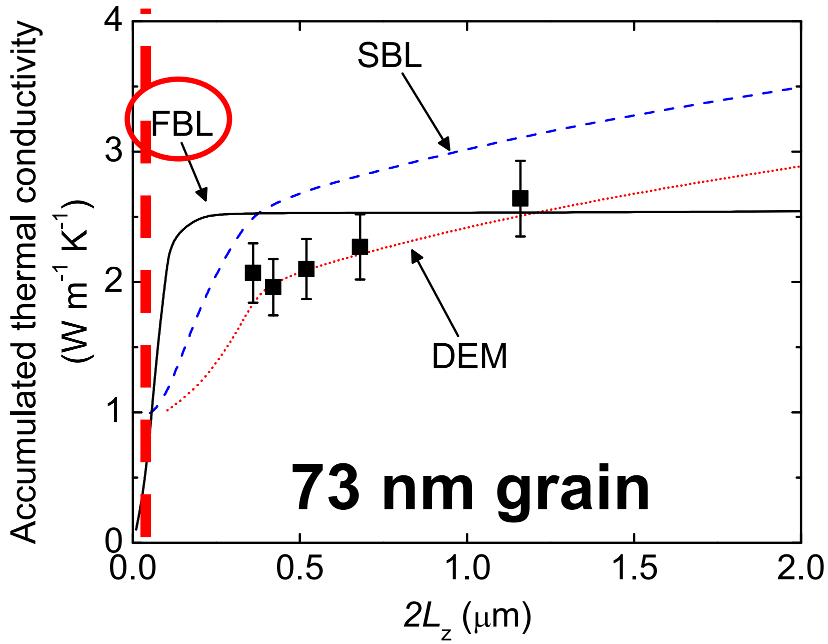
What happens when phonon wavelengths are much greater than boundaries/interfaces?

Spectral phonon transport – Nanograined alloys

Nanograined $Si_{80}Ge_{20}$



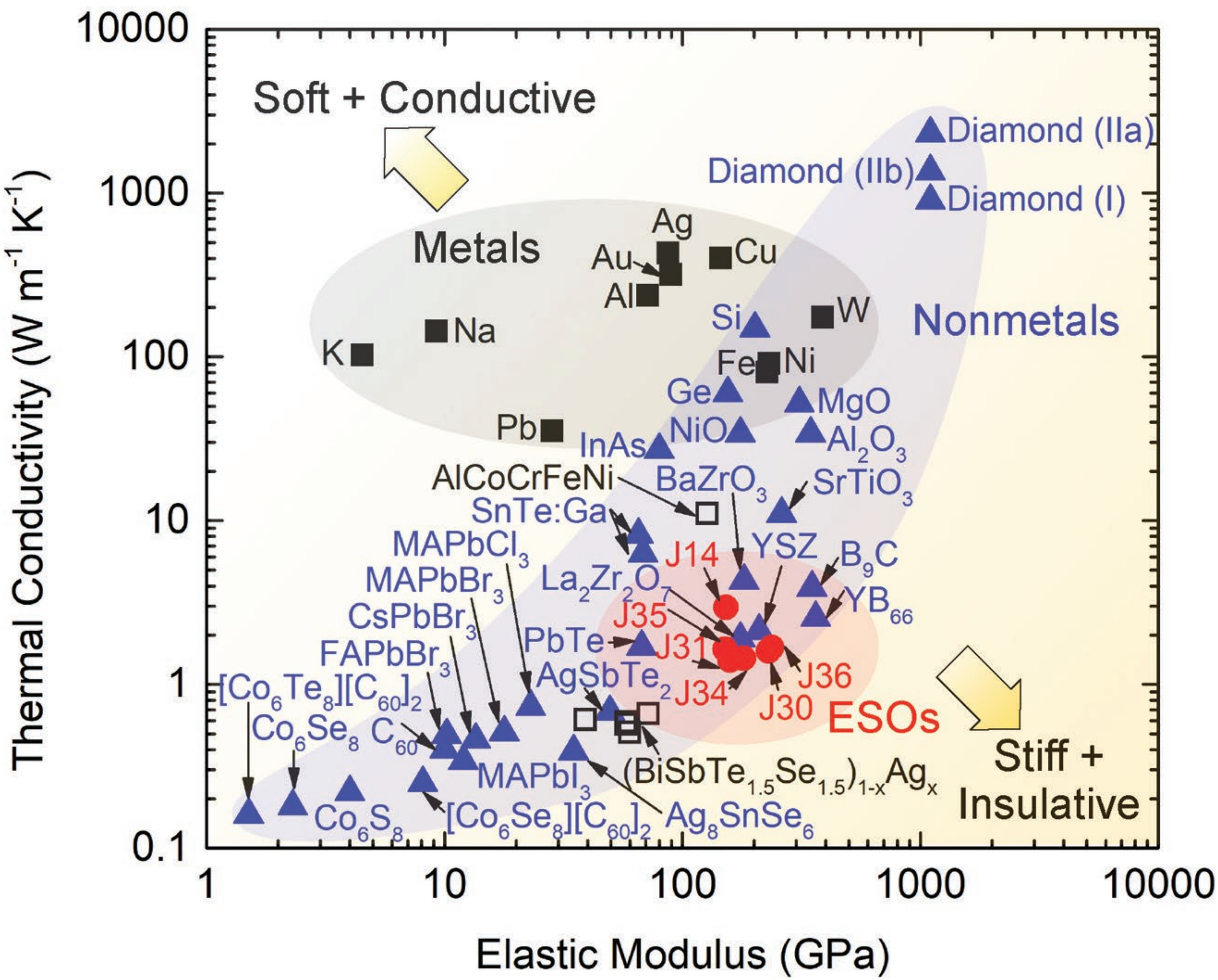




Phonons with wavelengths much larger than grain size do not scatter at grain boundaries

Appl. Phys. Lett. **11**, 131902

Thermal conductivity of crystals



Adv. Mat. **30**, 1805004 (2018)

Leibfried-Schlomann Eq.

V=volume of unit cell

constant

Debye frequency – maximum frequency of vibration in a solid

$$\kappa = A\frac{V^{1/3}\omega_D^3}{\gamma^2 T},$$

Temperature

γ=Gruneisen parameter
(related to anharmonicity of bonds)

$$\omega_D \propto \sqrt{\frac{K}{m}}$$

$$\omega_D \propto V^{1/6}\sqrt{B}$$

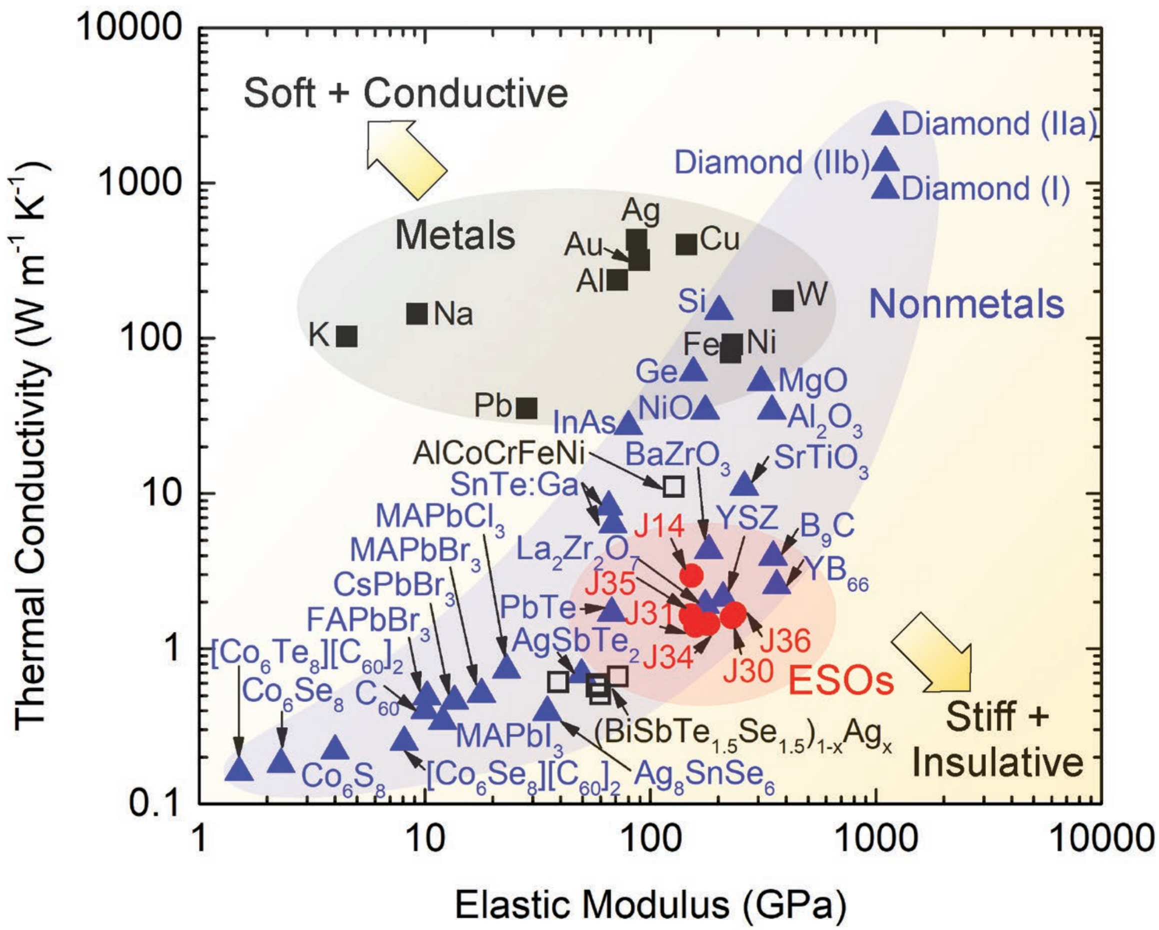
Bulk modulus

[21] G. Leibfried and E. Schlömann, Nach. Akad. Wiss. Gottingen, Math. Phyz. Klasse **4**, 71 (1954).

[22] M. Roufosse and P. G. Klemens, Phys. Rev. B **7**, 5379 (1973).

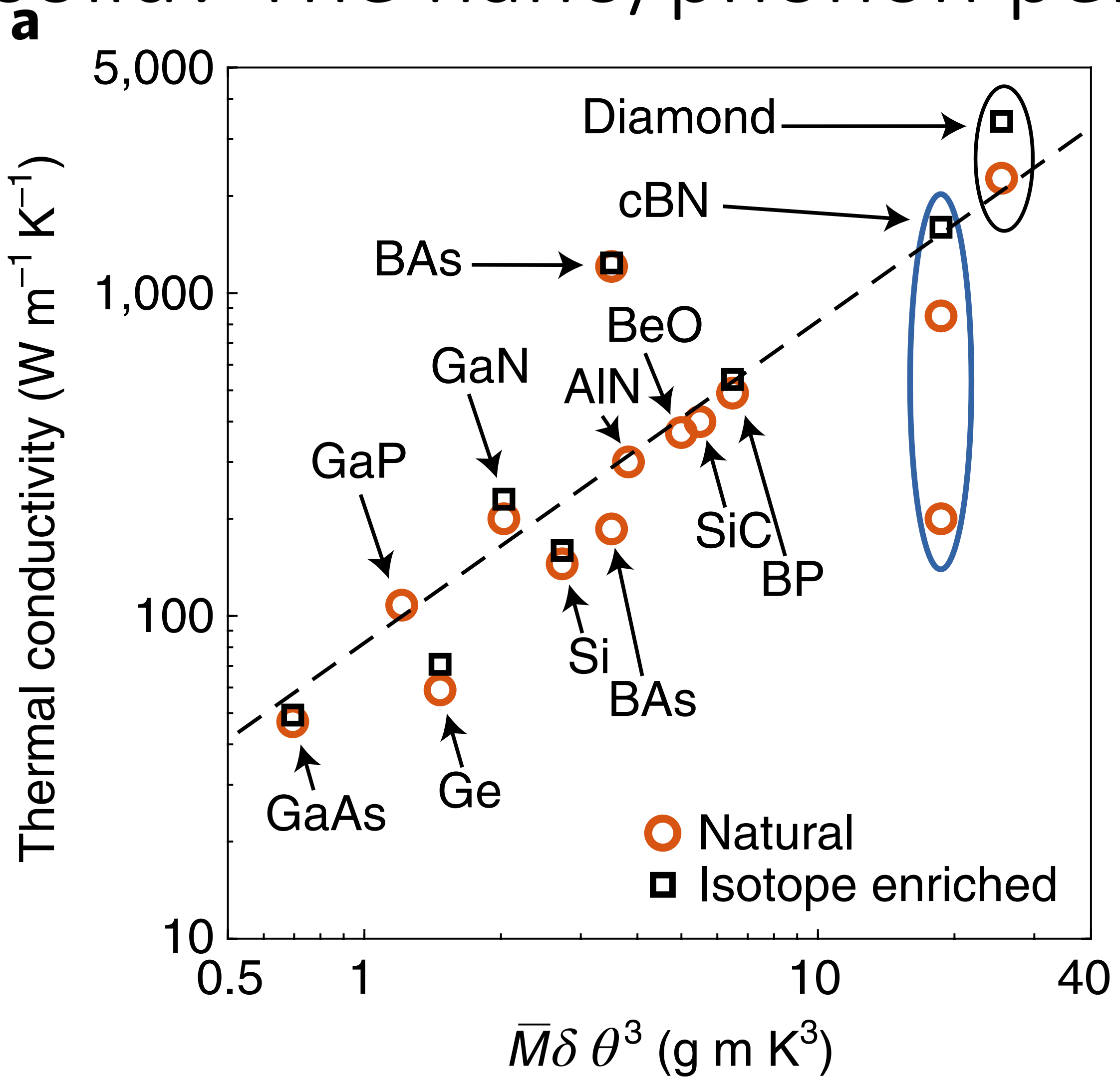
So how do you make a high thermal conductivity solid? The L-S perspective

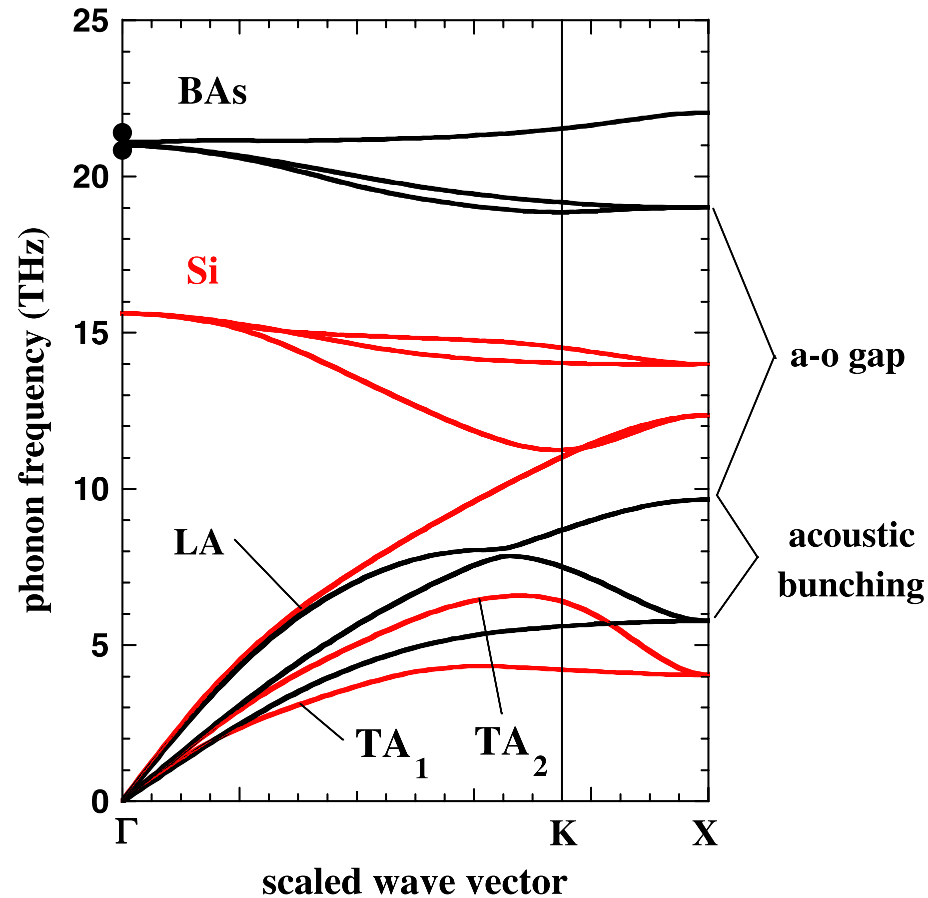
$$\kappa \propto K^{3/2} \propto B^{3/2}$$



Adv. Mat. **30**, 1805004 (2018)

So how do you make a high thermal conductivity solid? The nano/phonon perspective





THERMAL CONDUCTIVITY

Achieving a better heat conductor

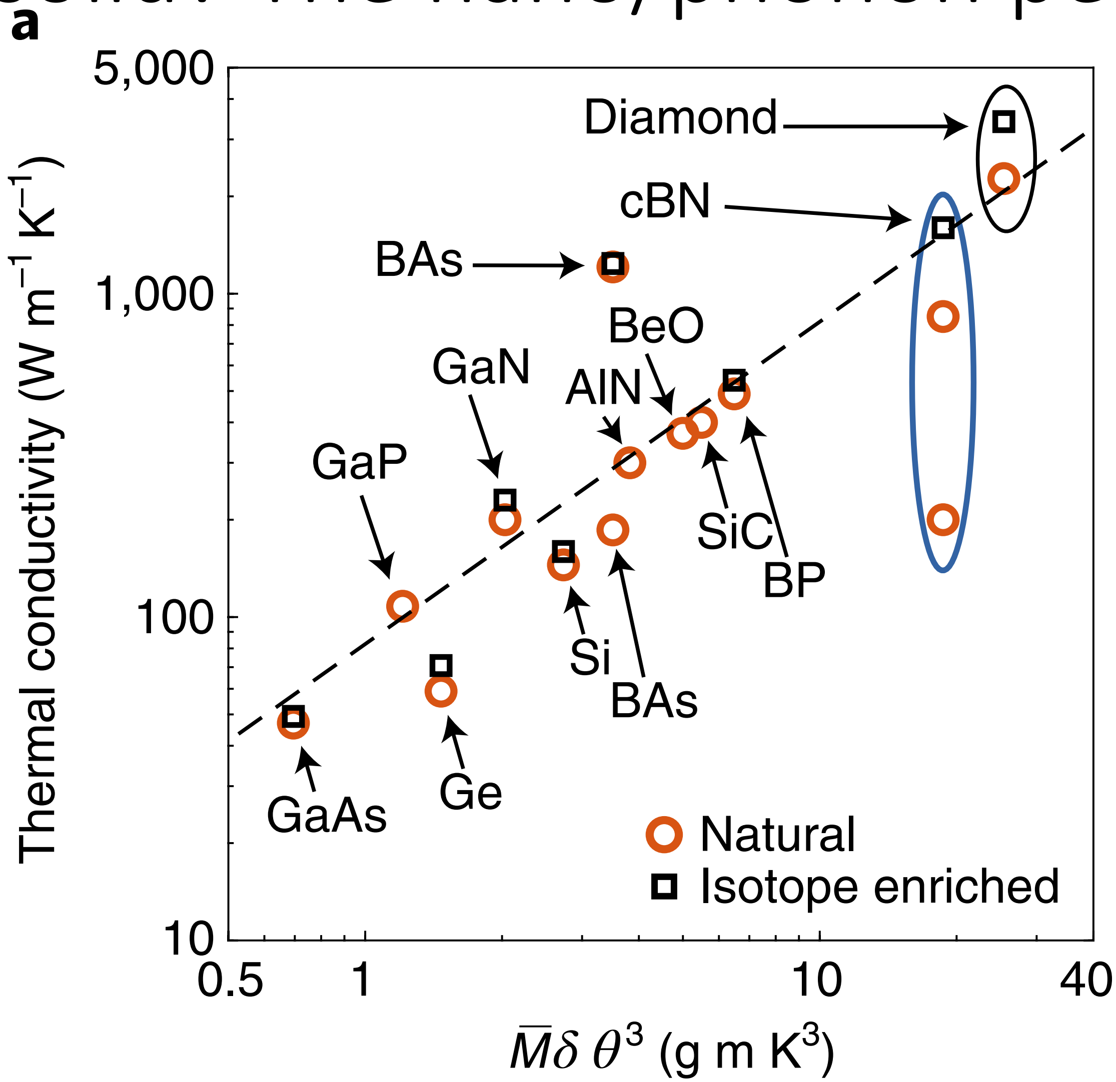
Finding a competitor for diamond as a good heat conductor remains challenging. Measurements on crystals of cubic boron nitride demonstrate a thermal conductivity of 1,600 W m^{-1} K^{-1} at room temperature, rivalling diamond.

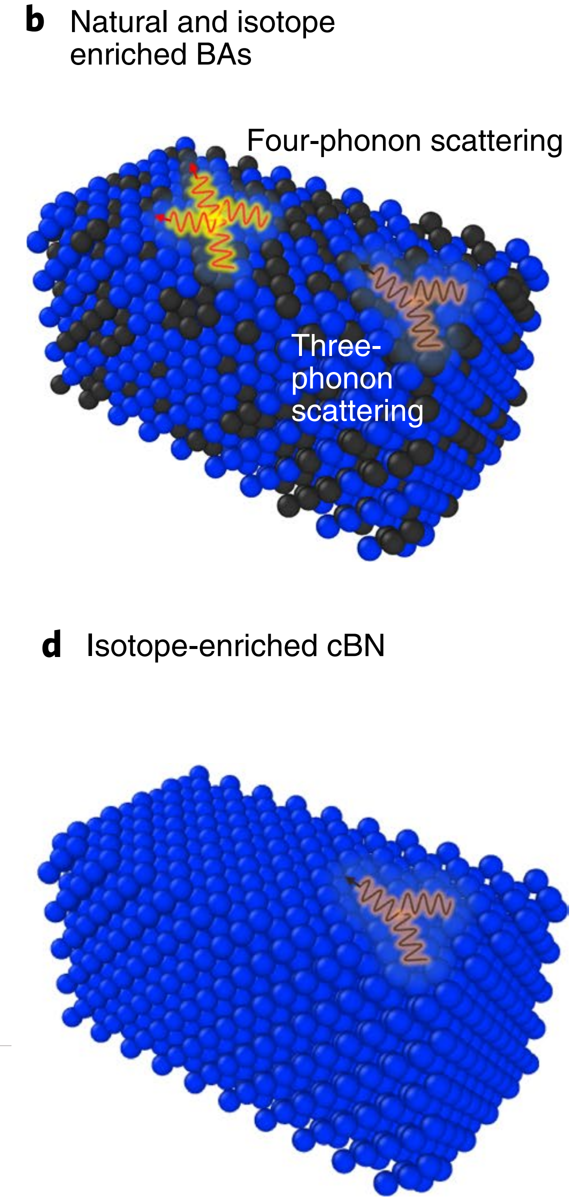
Ashutosh Giri and Patrick E. Hopkins

NATURE MATERIALS | VOL 19 | MAY 2020 | **482**

PRL **111**, 025901 (2013)

So how do you make a high thermal conductivity solid? The nano/phonon perspective





THERMAL CONDUCTIVITY

Achieving a better heat conductor

Finding a competitor for diamond as a good heat conductor remains challenging. Measurements on crystals of cubic boron nitride demonstrate a thermal conductivity of 1,600 W m^{-1} K^{-1} at room temperature, rivalling diamond.

Ashutosh Giri and Patrick E. Hopkins

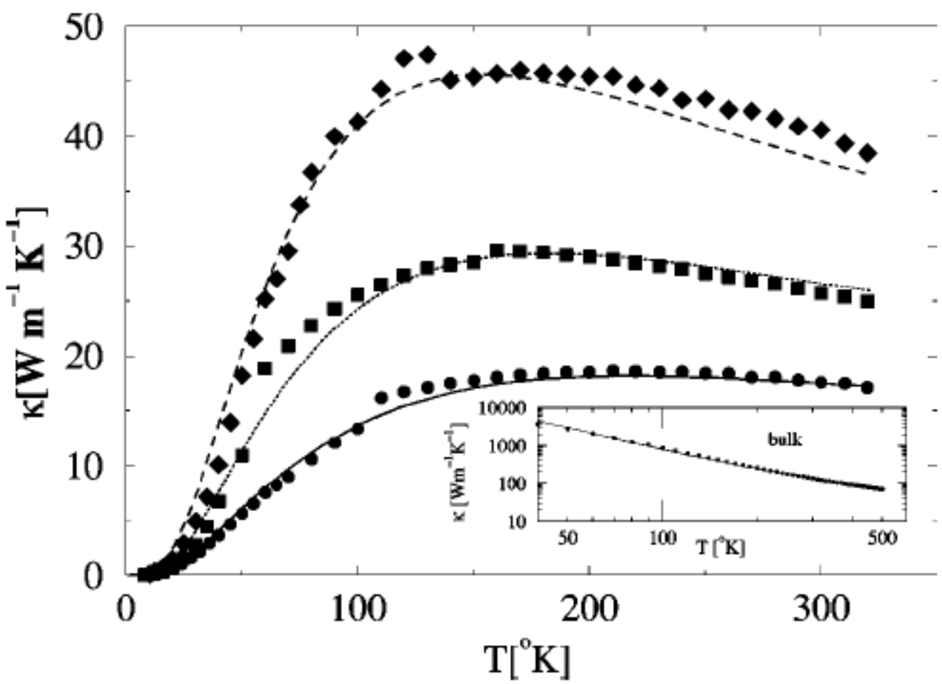
NATURE MATERIALS | VOL 19 | MAY 2020 | 482

Si nanowire κ

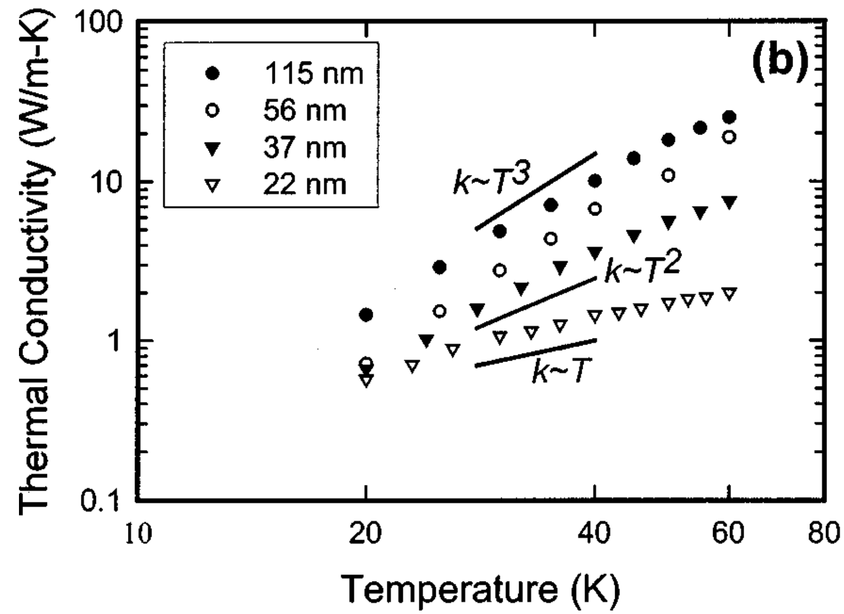
Now we know that the major reduction in κ in nanowires is boundary scattering. But what about the trends???

$$\kappa = \frac{1}{3}\sum_j \int_0^{\omega_{max,j}} \hbar\omega D(\omega)\frac{\partial f}{\partial T} v_j^2(\omega)\tau_{semi}\,\mathrm{d}\omega$$

$$\frac{1}{\tau_B} = \frac{2v}{L}$$



Mingo, *Phys. Rev. B* **68**, 113308 (2003).



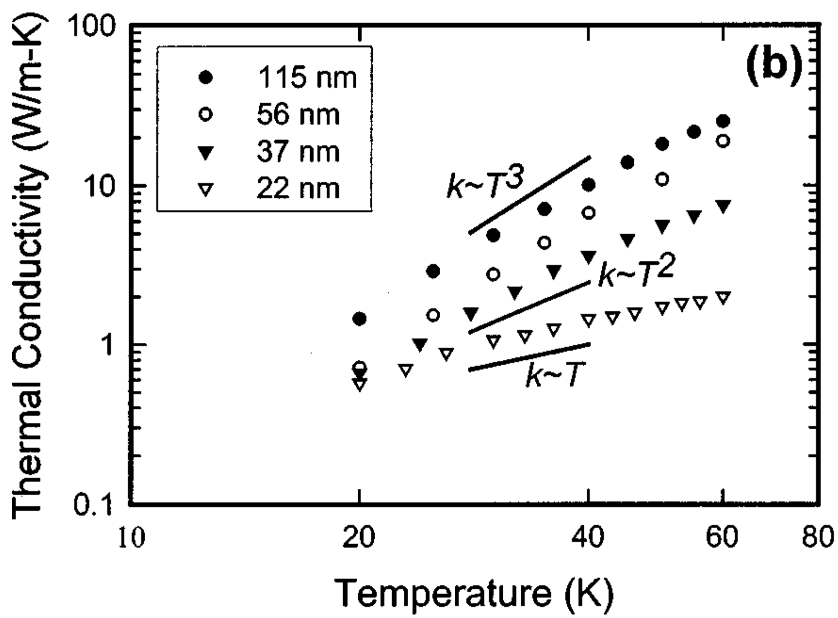
Recall: can be explained from *C* arguments

Li *et al.*, Appl. Phys. Lett **83**, 2934 (2003).

Si nanowire κ

Now we know that the major reduction in κ in nanowires is boundary scattering. But what about the trends???

Li *et al.*, Appl. Phys. Lett **83**, 2934 (2003).

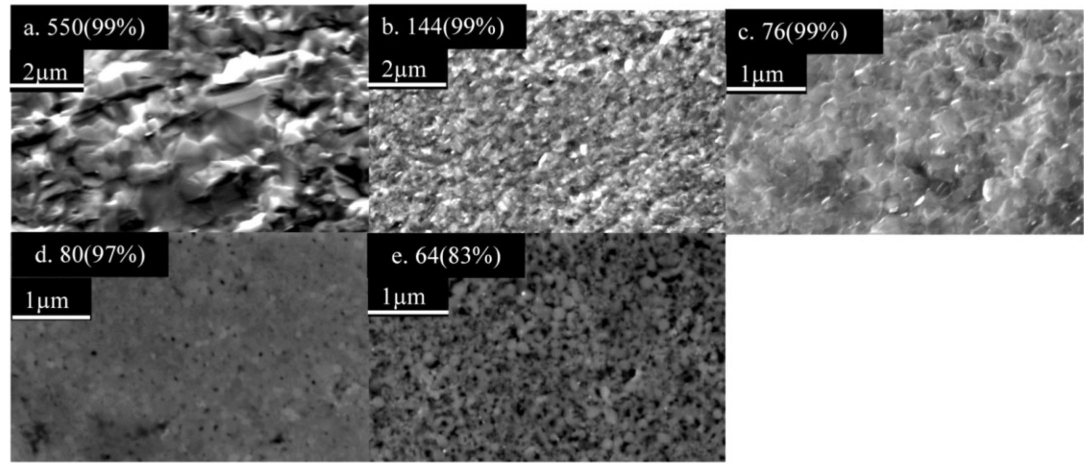


$$\kappa_{p,2D}(T) = \frac{k_B^3}{2\pi\hbar^2} T^2 \sum_j \frac{1}{v_j^2} \int_0^{x_D} x^3 \frac{\exp[x]}{(-1+\exp[x])^2} v_j^2 \tau \, \mathrm{d}x$$

This T^2 trend assumes τ is independent of ω

Nanograined Si thermal conductivity

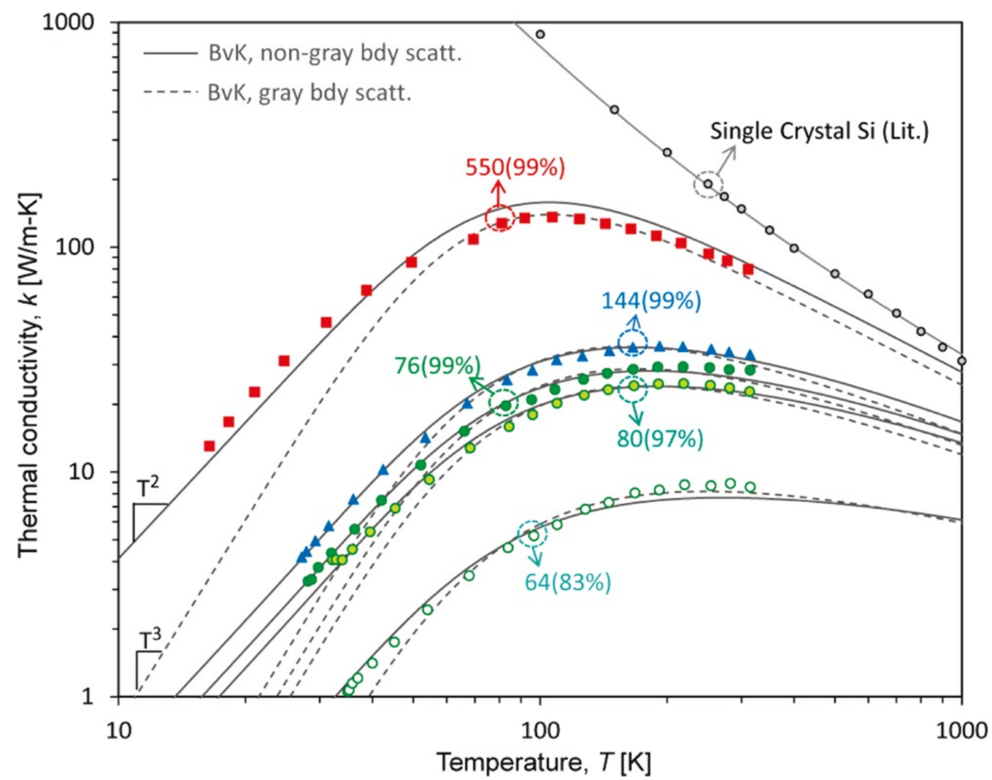
Now we know that the major reduction in κ in nanowires is boundary scattering. But what about the trends???



Typical assumption

$$\frac{1}{\tau_B} = \frac{2v}{L}$$

But T^2 trend still exists in BULK samples with nanograin boundaries (i.e., there are not any COHERENT mechanisms at play!!)



Wang *et al*. *Nano Lett* **11** 2206 (2011)

Minimum limit to thermal conductivity

$$\frac{1}{\tau_{semi}} = \sum_j \frac{1}{\tau_j}$$

Maximum scattering rate!!!

Einstein limit

Einstein, *Ann. Phys.* (1911)

$$\lambda_{\min} = a \approx n^{-1/3}$$

$$\tau_{\min} = \frac{\pi}{\omega}$$

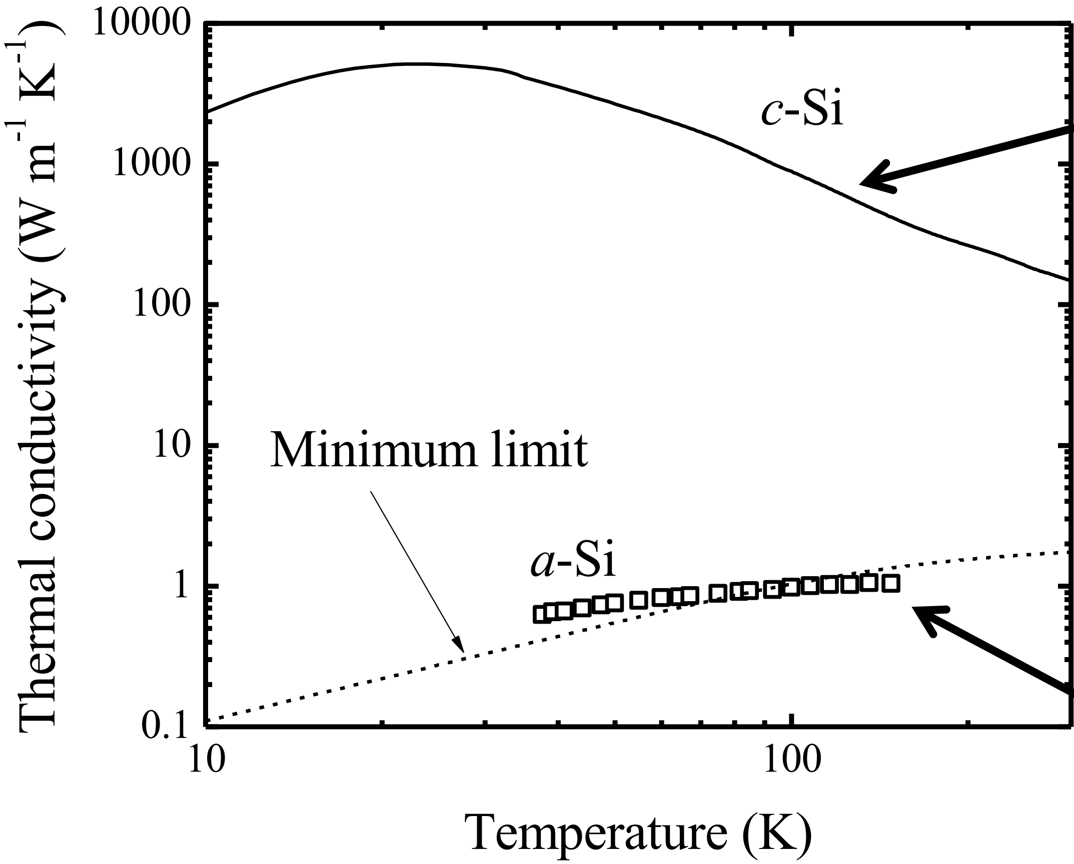
$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$

Cahill, Watson, Pohl limit
"coupled oscillators"
Phys. Rev. B. 46, 6131

$$\kappa_{min}(T) = \frac{1}{6\pi} \sum_j \frac{1}{v_j} \int_0^{\omega_{max,j}} \frac{\hbar^2 \omega^3}{k_B T^2} \frac{\exp\left[\frac{\hbar\omega}{k_B T}\right]}{\left(-1+\exp\left[\frac{\hbar\omega}{k_B T}\right]\right)^2} \mathrm{d}\omega$$

Minimum limit to thermal conductivity

Capture amorphous thermal conductivity trends well, as lack of periodicity causes phonon scattering at $\lambda = a$

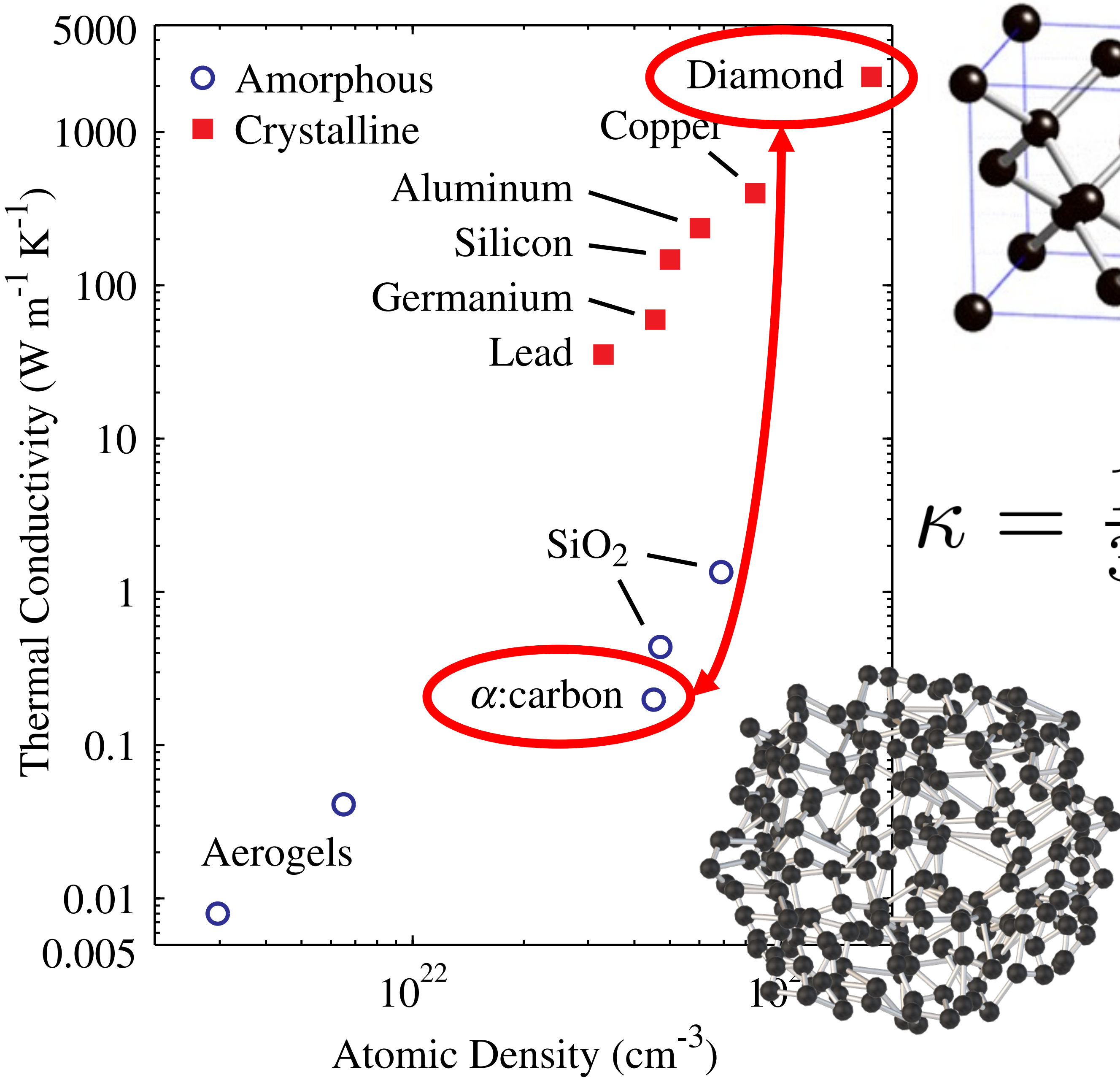


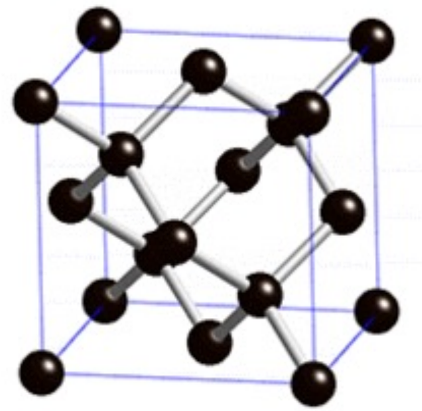
T^{-1} trend in crystalline material due to phonon-phonon scattering

Amorphous solid trends follow heat capacity since scattering is related to *a* and not temperature dependent

Hopkins and Beechem, *Nanoscale and Microscale Thermophysical Engineering* **14**, 51 (2010)

Amorphous materials have low thermal conductivities





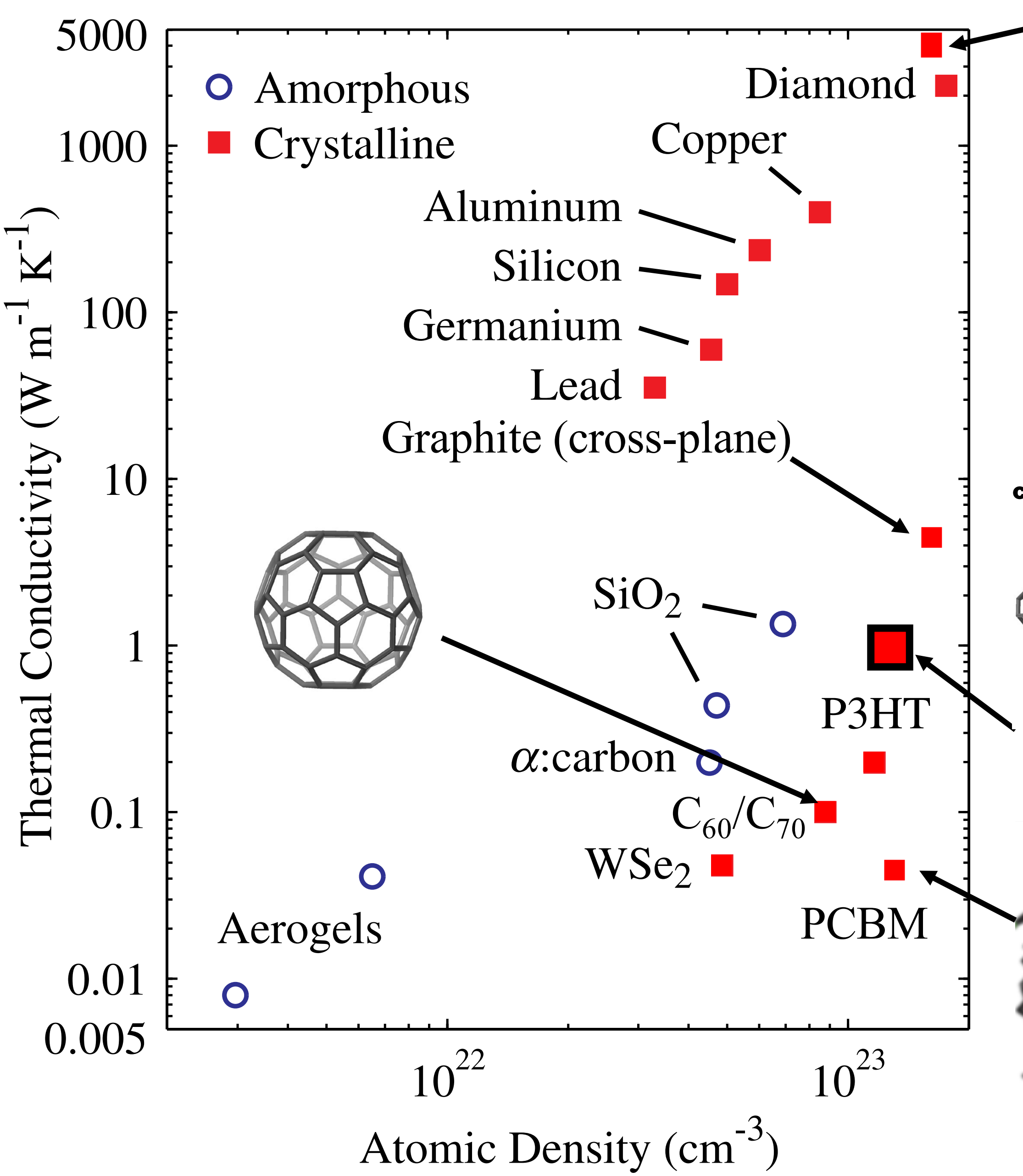
$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$

$$\lambda_{\min} = a \approx n^{-1/3}$$

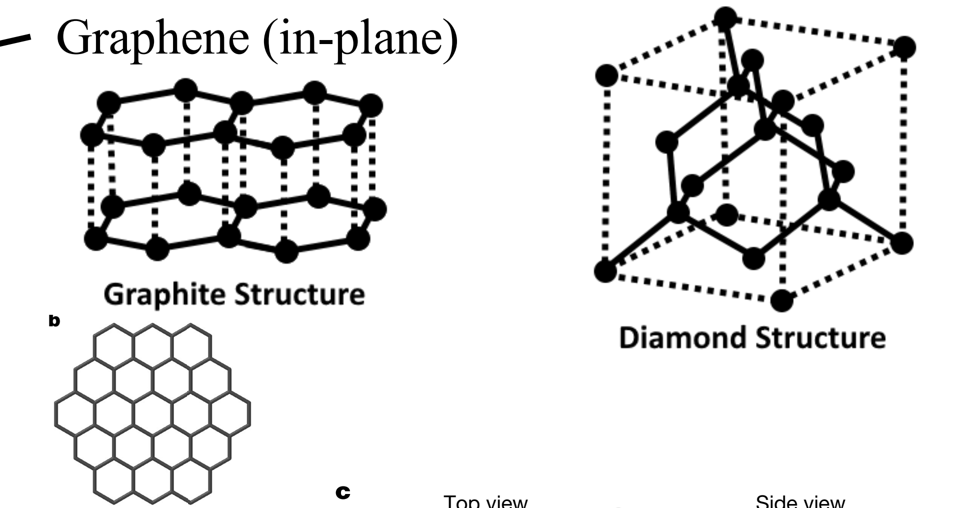
$$\tau_{\min} = \frac{\pi}{\omega}$$

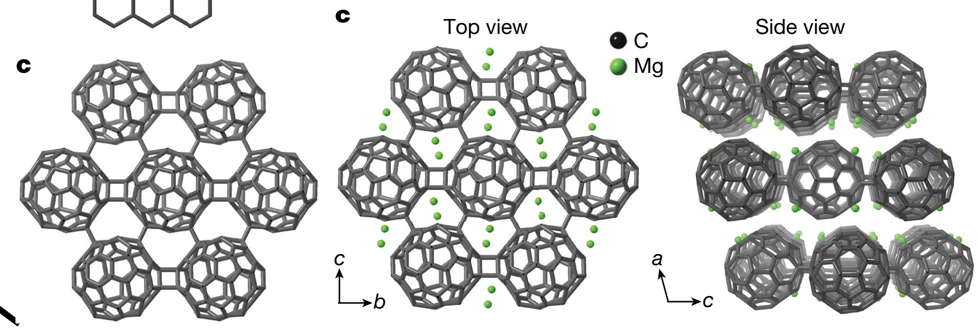
PRL **110**, 015902 (2013)

Making a low thermal conductivity material

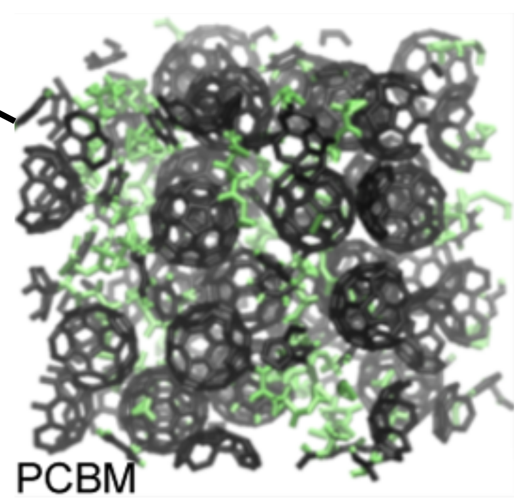


PRL **110**, 015902 (2013)





Graphullerne *Nature* 613, 71 (2023)



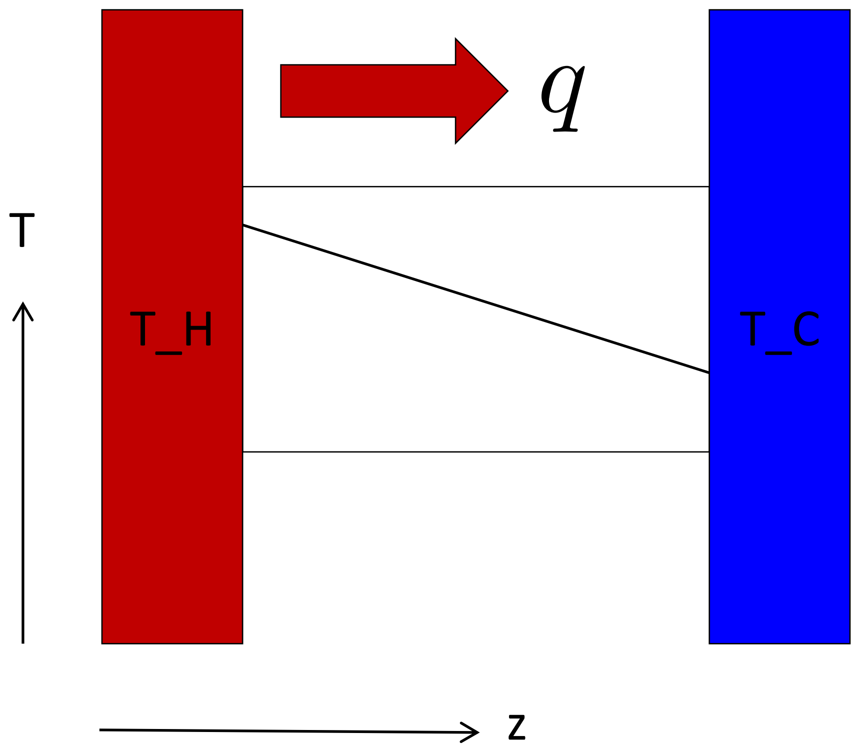
Outline

1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

Heat Transfer

Steady state = The Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$



Transient = The Heat Equation

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial z^2} + q(t)$$

$$q = \text{Flux}$$
$$\rho = \text{mass density}$$
$$\kappa = \text{Thermal conductivity}$$
$$T = \text{temperature}$$
$$C = \text{Heat Capacity}$$

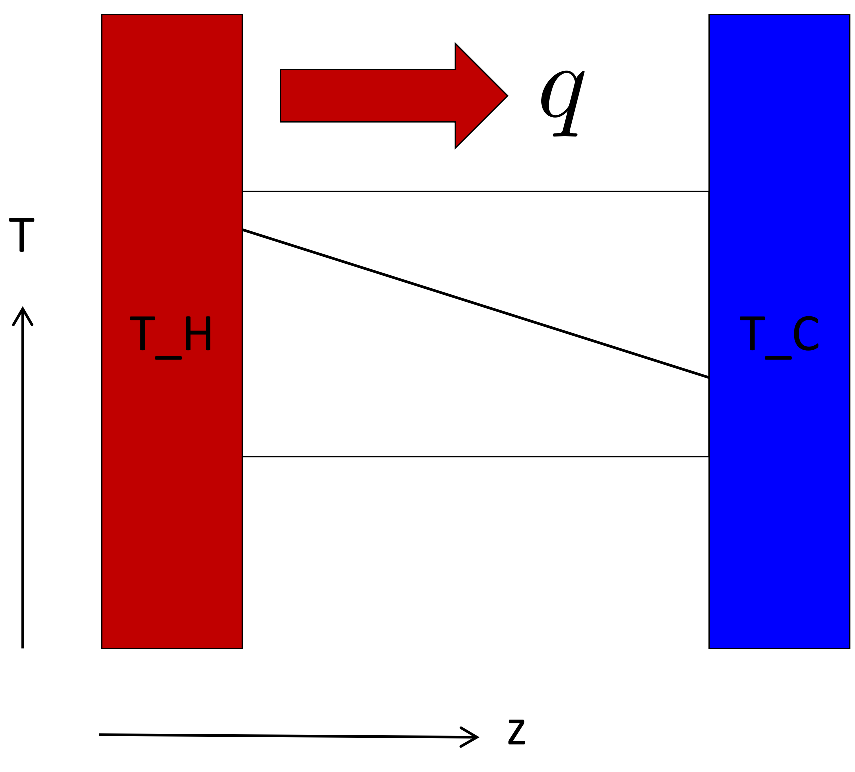
Outline

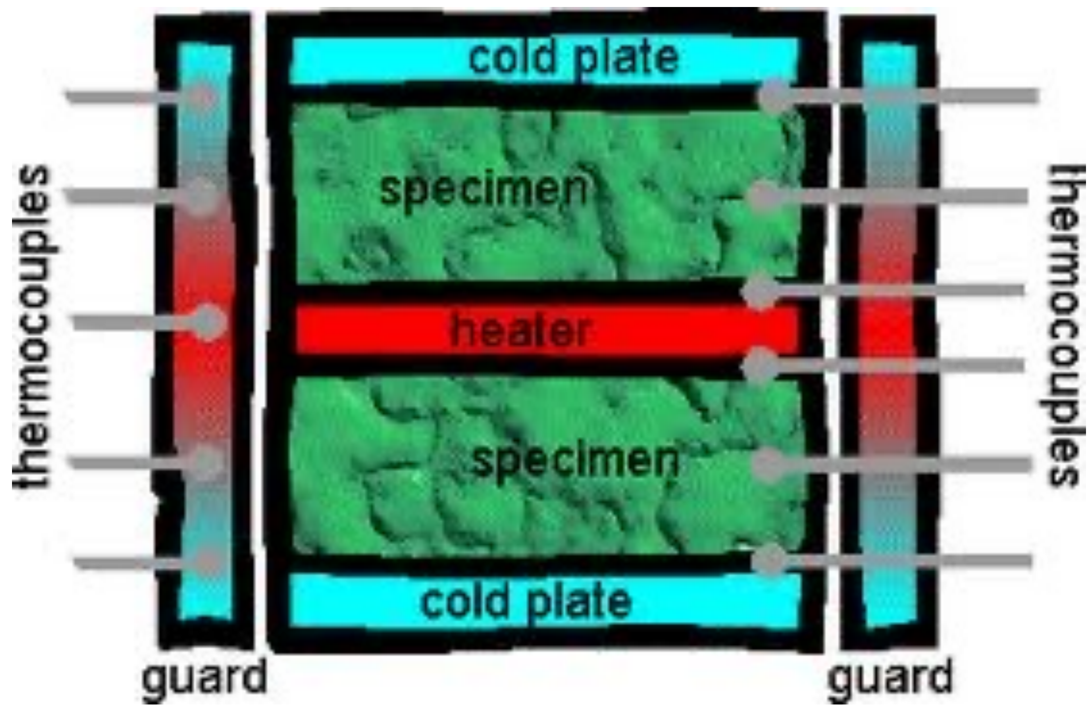
- Steady state electrical resistivity
- Transient techniques
  - “RC”
  - Heat capacity
  - Laser flash
  - Transient Electro-thermal
  - ns pulse
  - fs pulse
- Periodic techniques
  - Angstrom method
  - 3ω
  - FDTR
  - TDTR

Steady state measurements - bulk

"Guarded hot plate"

$$q = -\kappa \frac{\partial T}{\partial z}$$





Steady state measurements - nano

Electrical resistivity

Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$

$$\kappa V \frac{\partial^2 T}{\partial x^2} + I^2 R(T) = 0$$

Joule heating

$$q = I^2 R$$

$$\overline{R} = R_0 \left[\frac{2}{L\sqrt{\frac{I^2 R_0 \alpha}{w\, d\, L\, \kappa}}} \tan\left(\frac{L\sqrt{\frac{I^2 R_0 \alpha}{w\, d\, L\, \kappa}}}{2} \right) \right]$$

Measured resistance is related to thermal conductivity (and a lot of other known quantities)

Y. C. Tai, C. H. Mastrangelo, and R. S. Muller. Thermal conductivity of heavily doped low-pressure chemical vapor deposited polycrystalline silicon films. Journal of Applied Physics, 63:1442–1447, 1988.

Steady state measurements - nano

$$\overline{R} = R_0 \left[\frac{2}{L\sqrt{\frac{I^2 R_0 \alpha}{w d L \kappa}}} \tan\left(\frac{L\sqrt{\frac{I^2 R_0 \alpha}{w d L \kappa}}}{2} \right) \right]$$

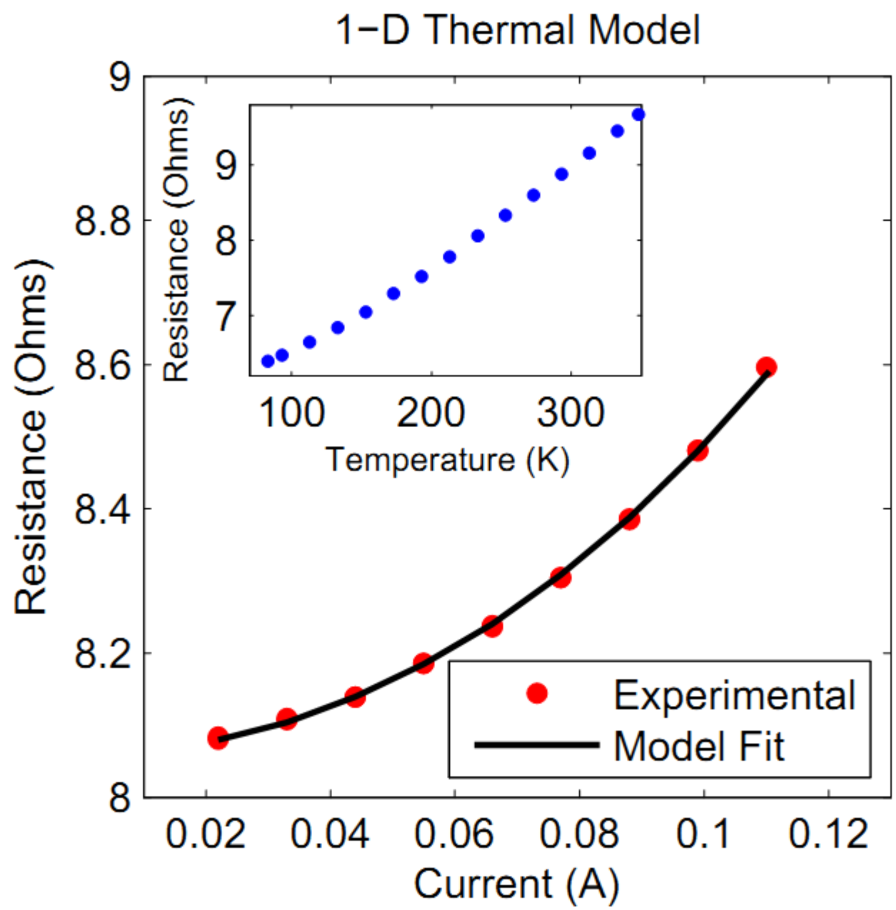


Fig. from: English, Phinney, Hopkins, and Serrano, "Thermal conductivity of single crystal silicon microbridges measured by electrical resistance thermometry and time domain thermoreflectance," *J. Heat Trans*. vol. **135**, 091103 (2013)

Steady state measurements - nano

Electrical resistivity - assumptions

$$\overline{R} = R_0 \left[\frac{2}{L\sqrt{\frac{I^2 R_0 \alpha}{w\, d\, L\, \kappa}}} \tan\left(\frac{L\sqrt{\frac{I^2 R_0 \alpha}{w\, d\, L\, \kappa}}}{2} \right) \right]$$

Current

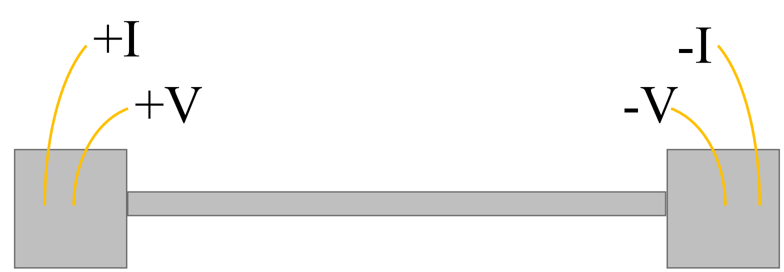
Equilibrium resistance

TCR

What you measure

Sample geometry

NEED TO KNOW SAMPLE GEOMETRY!!!

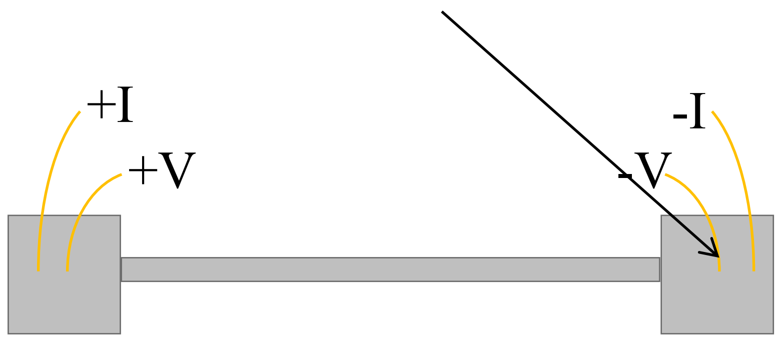


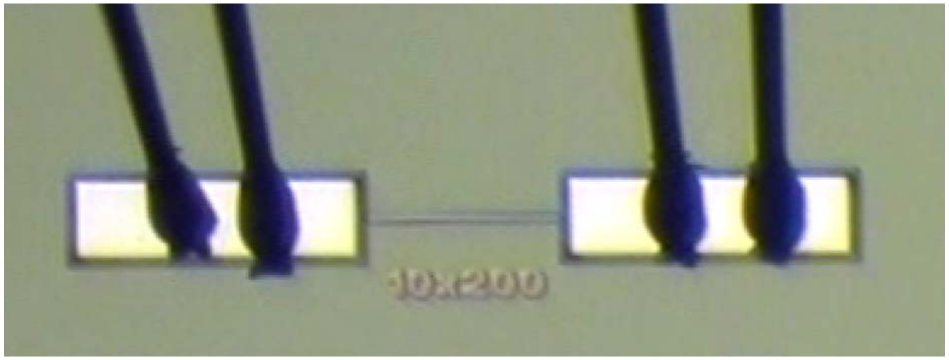
Y. C. Tai, C. H. Mastrangelo, and R. S. Muller. Thermal conductivity of heavily doped low-pressure chemical vapor deposited polycrystalline silicon films. Journal of Applied Physics, 63:1442–1447, 1988.

Fig. from: English, Phinney, Hopkins, and Serrano, “Thermal conductivity of single crystal silicon microbridges measured by electrical resistance thermometry and time domain thermoreflectance,” under review.

Steady state measurements - nano

Electrical/thermal contact resistances are inherently present in measurements





How do you make these contacts in a nanosystem??

When would these contact resistances matter in terms of sample geometry???

P. E. Hopkins and L. M. Phinney. Thermal conductivity measurements on polycrystalline silicon micro- bridges using the 3w technique.

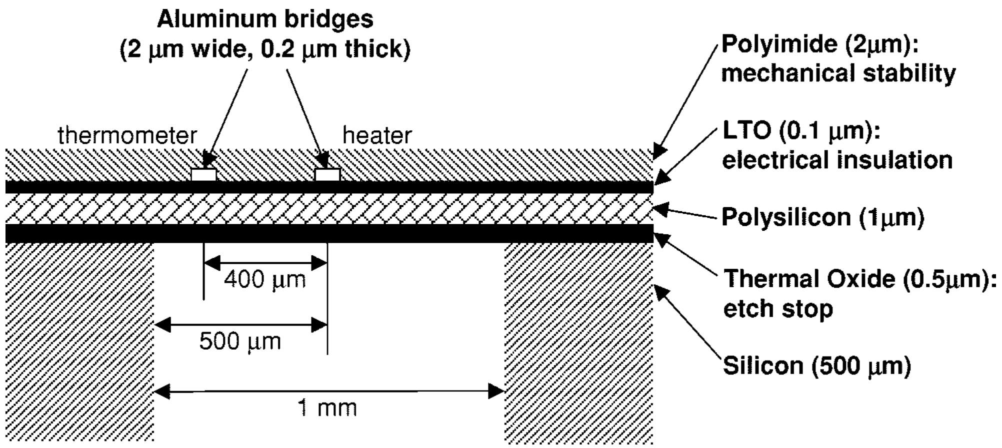
Journal of Heat Transfer, 131:043201, 2009.

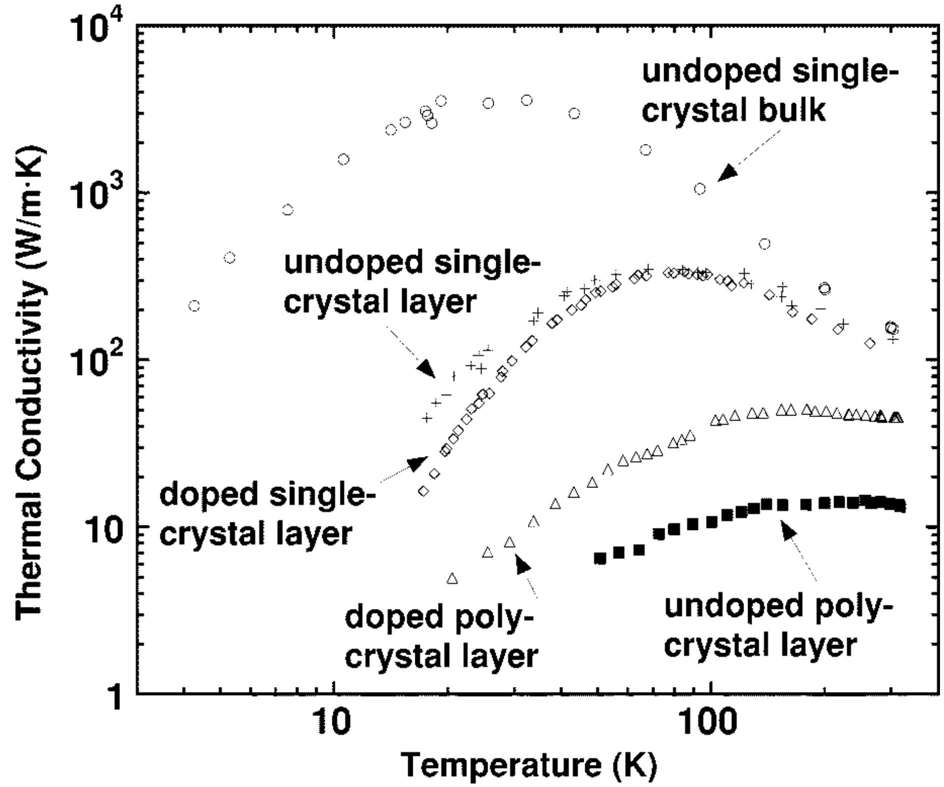
Steady state measurements - nano

Lithography

Thermal Conductivity of Doped Polysilicon Layers

Angela D. McConnell, Srinivasan Uma, *Member, IEEE*, and Kenneth E. Goodson, *Associate Member, IEEE*

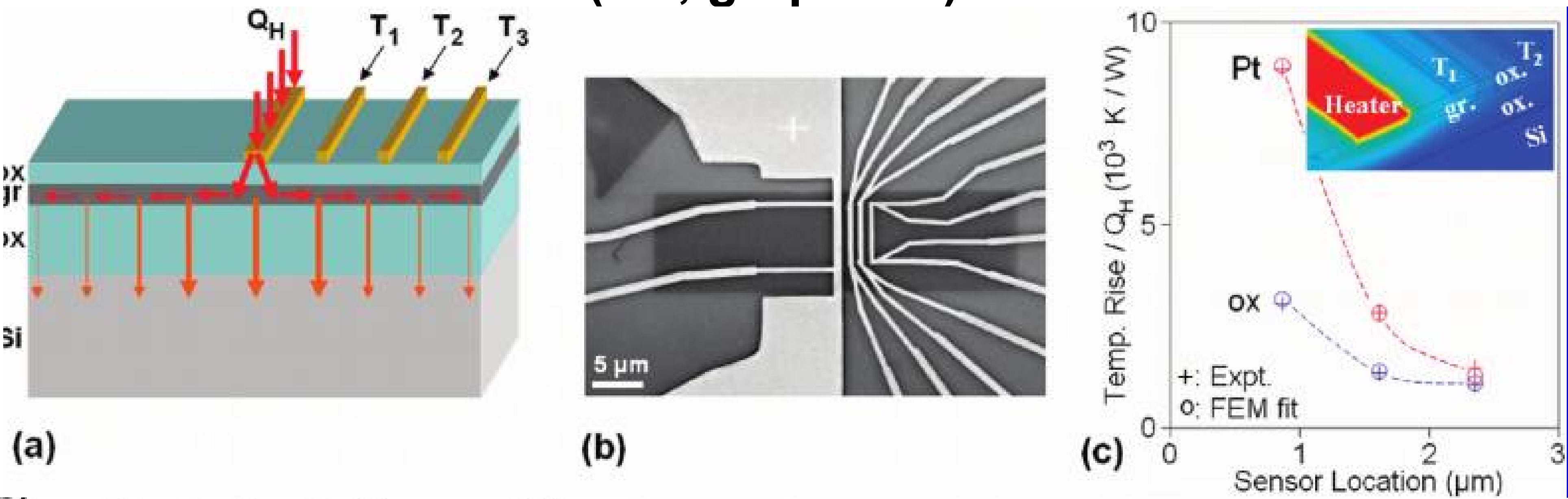


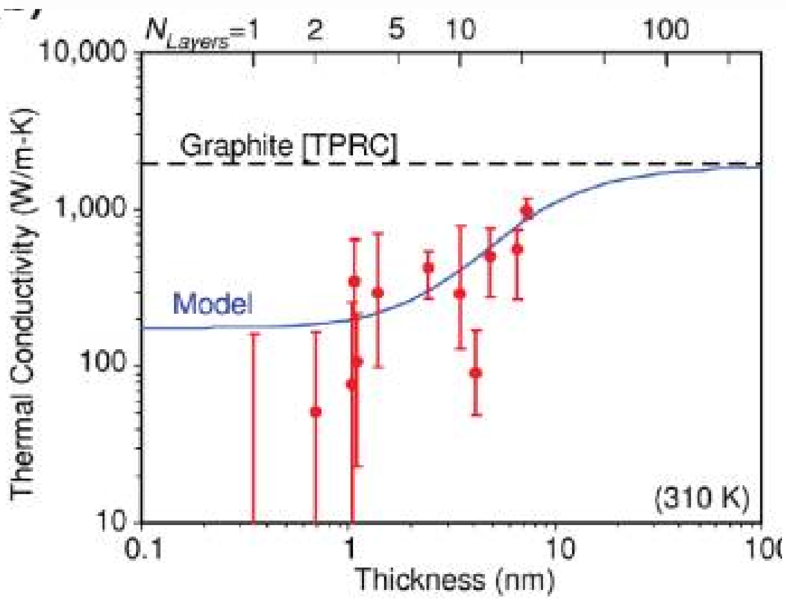


A. D. McConnell, S. Uma, and K. E. Goodson. Thermal conductivity of doped polysilicon layers. Journal of Microelectromechanical Systems, 10:360–369, 2001.

Steady state measurements of κ in nanosystems

In-plane thermal conductivity of "atomically thin" film (i.e., graphene)

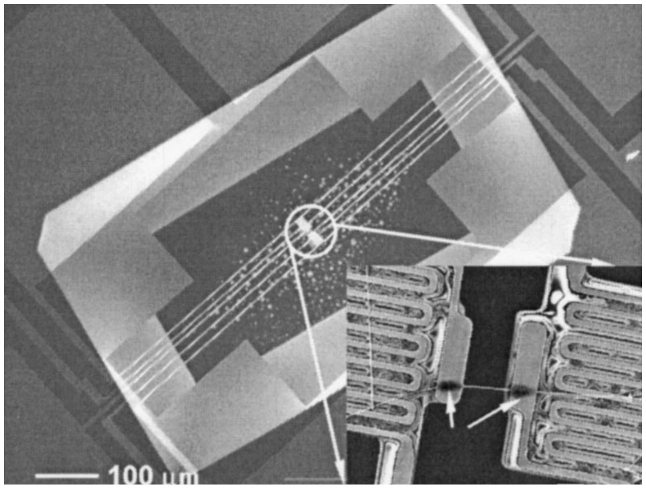


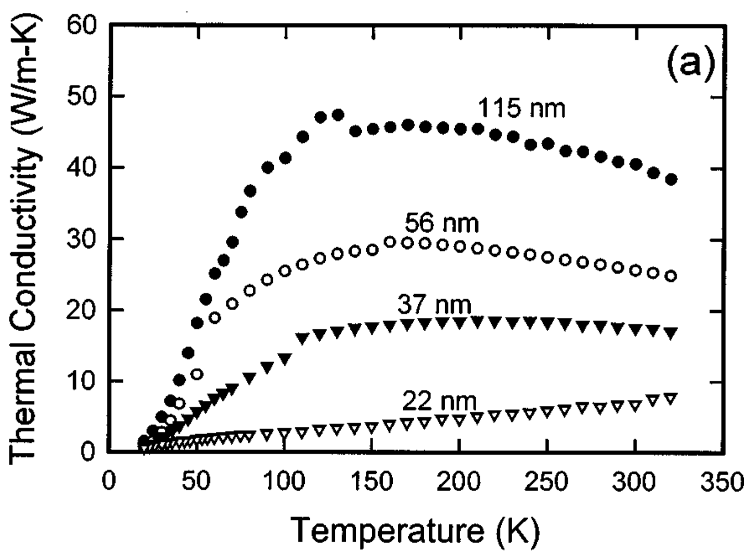


W. Jang, Z. Chen, W. Bao, C. N. Lau, and C. Dames. Thickness-dependent thermal conductivity of encased graphene and ultrathin graphene. **Nano Letters, 10:3909–3913, 2010.**

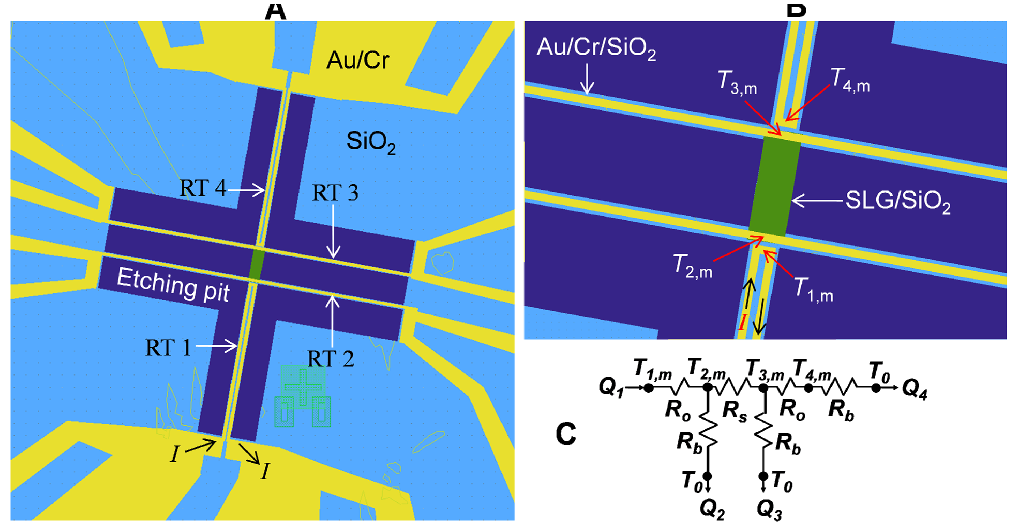
Steady state measurements of κ in nanosystems

Can be extended to suspended nanostructures (e.g., thin films, graphene, nanowires, etc)





D. Li, Y. Wu, P. Kim, L. Shi, P. Yang, and A. Majumdar. Thermal conductivity of individual silicon nanowires. **Applied Physics Letters, 83:2934–2936, 2003.**



J. H. Seol, I. Jo, A. L. Moore, L. Lindsay, Z. H. Aitken, M. T. Pettes, X. Li, Z. Yao, R. Huang, D. A. Broido, N. Mingo, R. S. Ruoff, and L. Shi. Two-dimensional phonon transport in supported graphene. **Science, 328:213–216, 2010.**

Overview: Methods for testing κ of thin films

- **Steady State resistivity approaches**
  - **No variation in time (“Fourier Law”)**

- **Transient reflectivity and optical methods**
  - **Time dependent (“The heat eq. w/ impulse response”)**

- **Modulated methods (“The heat eq. w/ frequency dep. source”)**
  - **3ω**
  - **Thermoreflectance-based techniques**
    - **FDTR**
    - **TDTR**

Steady state vs. transient

Steady state = The Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$

Transient = The Heat Equation

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial z^2} + q(t)$$

Heat capacity enters the picture

The source term can make a difference

Steady state, or long time, experiments, are subjected to MAJOR convection and conduction losses. High *T* issues (i.e., RT and above)

Source can be “single shot/impulse” or “periodic”

Transient measurements

"RC" techniques

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial z^2} + q(t)$$

Solution results in "thermal" time constant in exponential decay

$$\Delta T(t) = \Delta T_0 \exp\left[\frac{-t}{\tau}\right]$$

$$\tau = \frac{CV}{Ah}$$

← Thermal conductance

How does $\Delta T(t)$ change with *C, V, A,* and *h*?

Transient measurements

$$\tau = \frac{CV}{Ah}$$

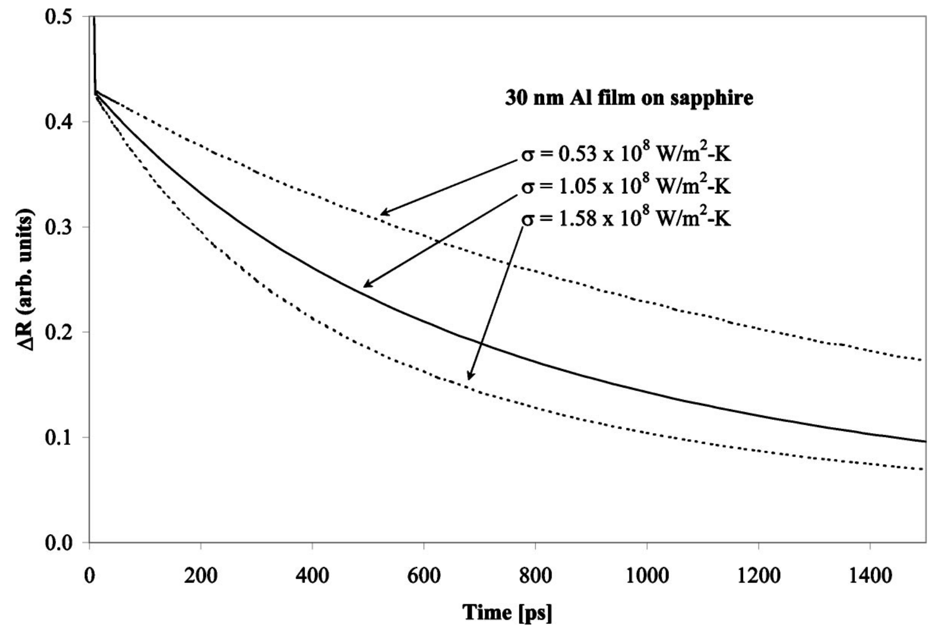


Fig. 2 Modeled thermal response of 30 nm Al film on a sapphire substrate with $\sigma = 1.05 \times 10^8$ W/m^2 K. The dotted lines are the thermal response for the same film with ±50% change in σ.

SO WHAT DO YOU MEASURE????

R. J. Stevens, A. N. Smith, and P. M. Norris. Measurement of thermal boundary conductance of a series of metal-dielectric interfaces by the transient thermoreflectance technique. Journal of Heat Transfer, 127(3):315–322, 2005.

Steady state vs. transient

Steady state = The Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$

Steady state techniques are the only measurements that are directly related to thermal conductivity

Transient = The Heat Equation

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial z^2} + q(t)$$

Transient techniques measure quantities that are related to the thermal diffusivity or thermal effusivity of the sample

$$D = \frac{\kappa}{C} \qquad E = \sqrt{\kappa C}$$

Thermal effusivity

Thermal effusivity
From Wikipedia, the free encyclopedia

A material's thermal effusivity is a measure of its ability to exchange thermal energy with its surroundings.

If two semi-infinite bodies initially at temperatures T1 and T2 are brought in perfect thermal contact, the temperature at the contact surface Tm will be given by their relative effusivities.

$$T_m = T_1 + (T_2 - T_1)\,\frac{E_2}{E_1+E_2}$$

This expression is valid for all times for semi-infinite bodies in perfect thermal contact. It is also a good first guess for the initial contact temperature for finite bodies.

Notes on heat capacity

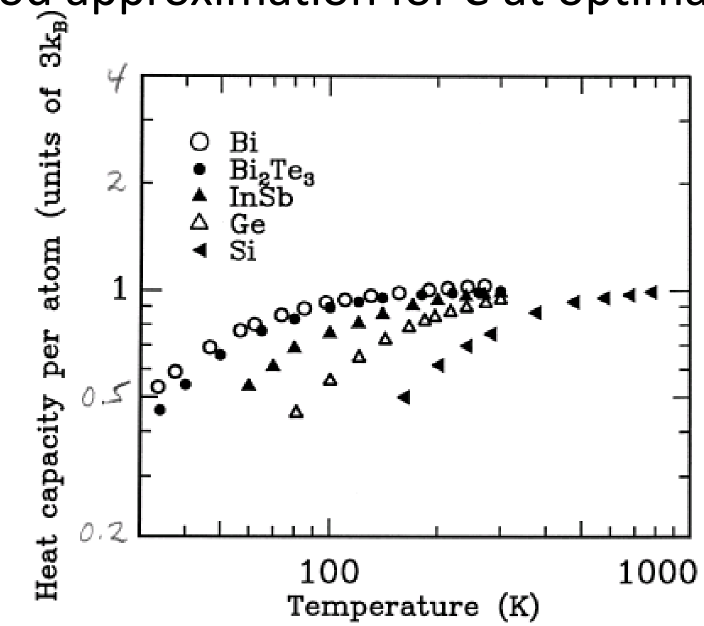
So if *C* is well known, then *k* can be "inferred" with transient measurements

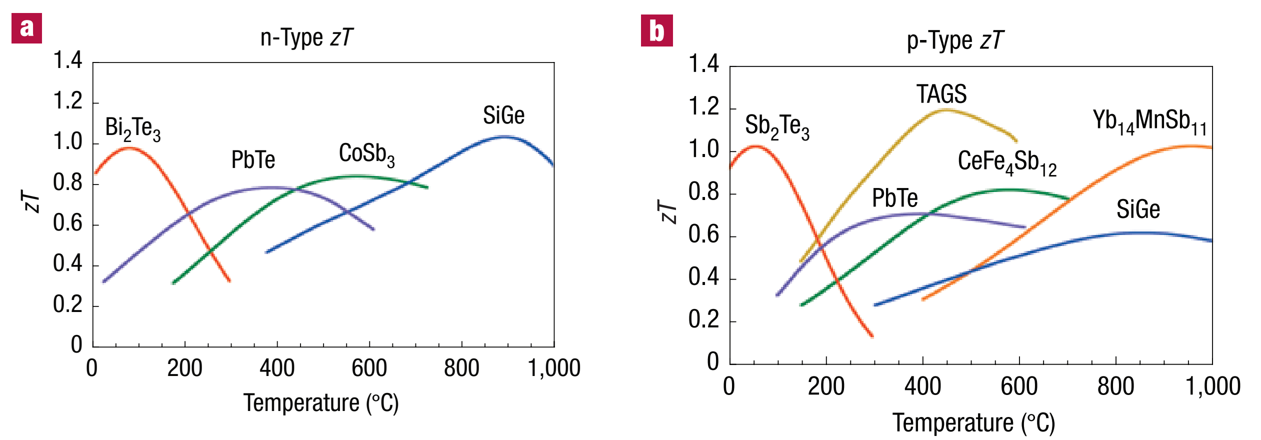
- In many cases, *C* is well known from careful measurements on bulk materials
- When scaled for porosity, *C* is ~ independent of microstructure (i.e., only atomic density is important)
- Materials with similar bonding and atomic weights have similar heat capacities
- Electronic heat capacity is too small to matter in most considerations

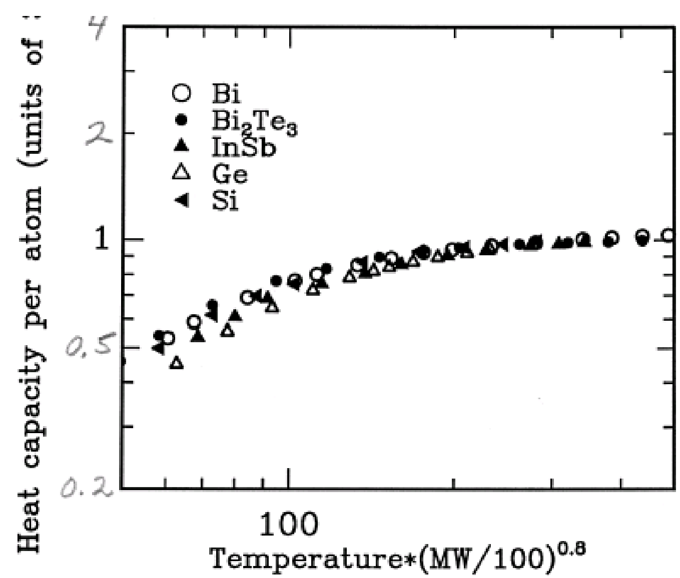
Notes on heat capacity

- For many semiconductors, classical equipartition is a good approximation for *C* at optimal thermoelectric operating temperatures

$$C = C_V = 3Nk_{\mathrm{B}}$$





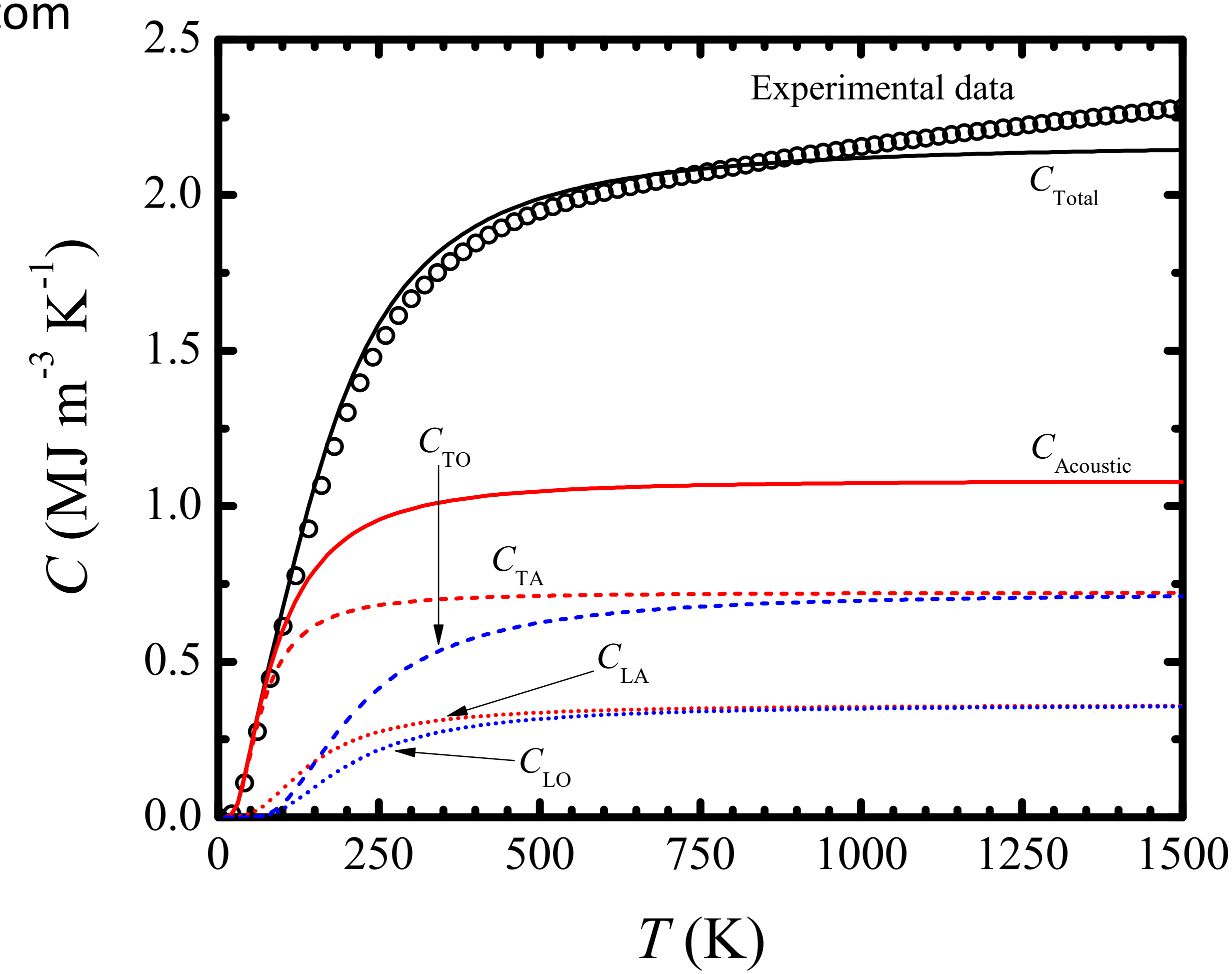


G. J. Snyder and E. S. Toberer. Complex thermoelectric materials. Nature Materials, 7:105–114, 2008.
David Cahill, “Measurement of Thermal conductivity,” “Thermal_School09.pdf”, on

http://users.mrl.illinois.edu/cahill/presentations.html

Notes on heat capacity

- At high temperatures, anharmonicity also increases the heat capacity
- Thermal expansion causes the vibrational modes to soften increasing the vibrational entropy per atom



Heat capacity of silicon

Grüneisen Parameters

TABLE I. The Grüneisen parameters $\gamma_{TA(X)}$ and $\gamma_{TA(L)}$ for Si, diamond, and Ge are calculated from Eq. (10). The estimated $\gamma_{TA(X)}$ and $\gamma_{TA(L)}$ are compared with experimental data (Ref. 1) and tight-binding calculation results.

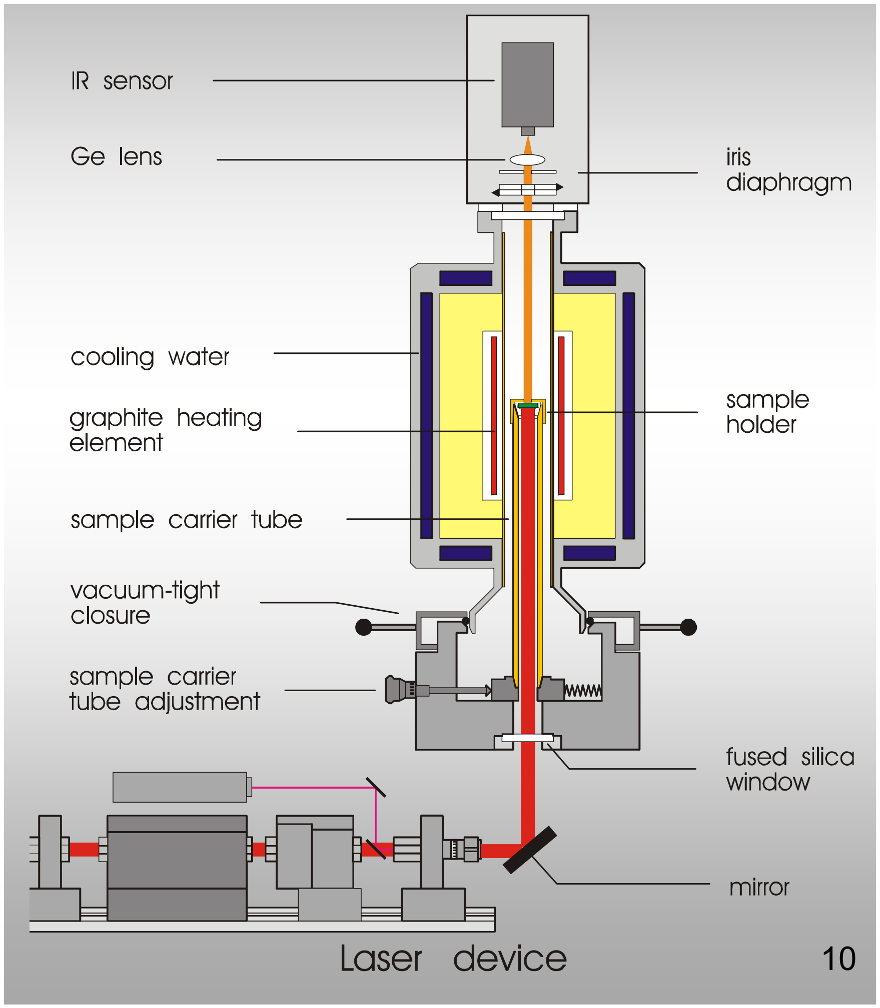
	Si	Diamond	Ge
$\gamma_{TA(X)}$ (*TB* calc.)	−1.08	0.042	
$\gamma_{TA(X)}$ (expt.)	−1.4		−1.53
$\gamma_{TA(X)}$ [Eq. (10)]	−1.67	0.017	−1.08
$\gamma_{TA(L)}$ (*TB* calc.)	−1.15	−0.047	
$\gamma_{TA(L)}$ (expt.)	−1.3		−0.4
$\gamma_{TA(L)}$ [Eq. (10)]	−1.54	−0.060	−0.77

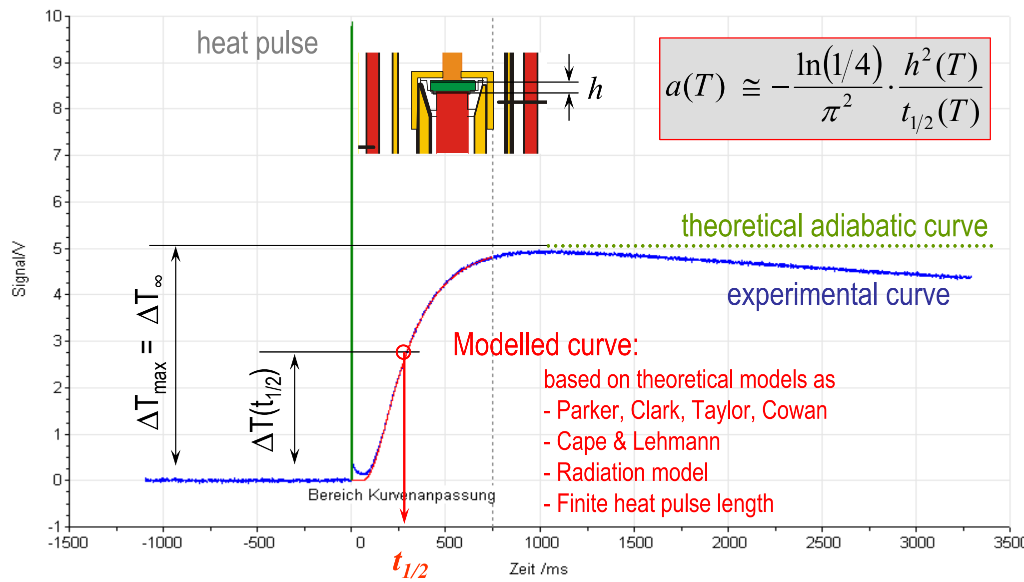
PRB 43, 5024 (1991)

What does this say about Debye temperatures?

Transient measurements

Laser Flash





- Very dependent on surface emissitivity
- Terrible sensitivity in nanosystems

Transient measurements

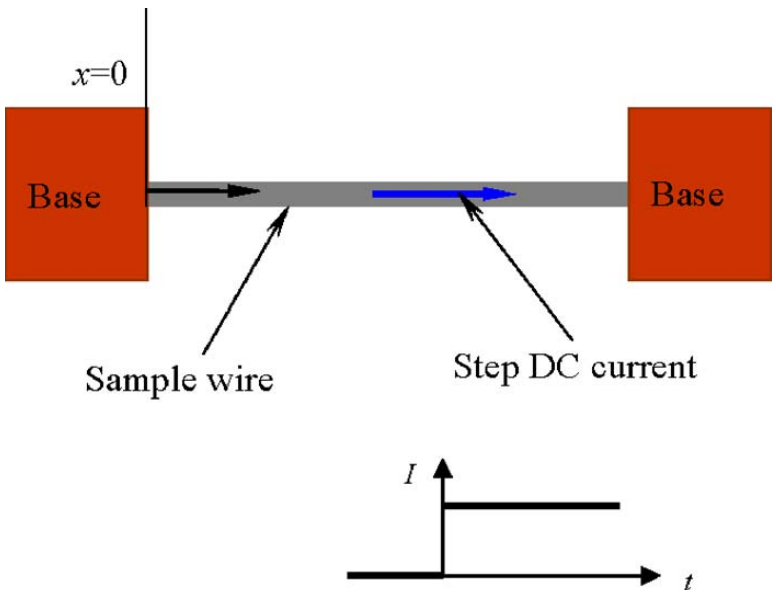
Transient electro-thermal technique (TET technique)

JOURNAL OF APPLIED PHYSICS **101**, 063537 (2007)

Thermal characterization of microscale conductive and nonconductive wires using transient electrothermal technique

Jiaqi Guo and Xinwei Wang[a)]
Department of Mechanical Engineering, N104 Walter Scott Engineering Center, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0656

Tao Wang
State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, People's Republic of China 310027



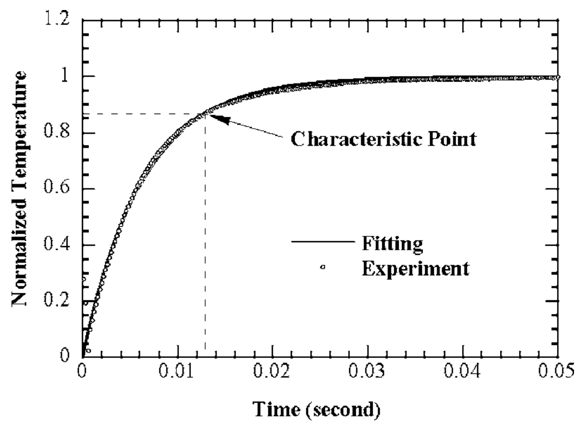


FIG. 5. The normalized temperature vs the theoretical fitting for the SWCNT bundle.

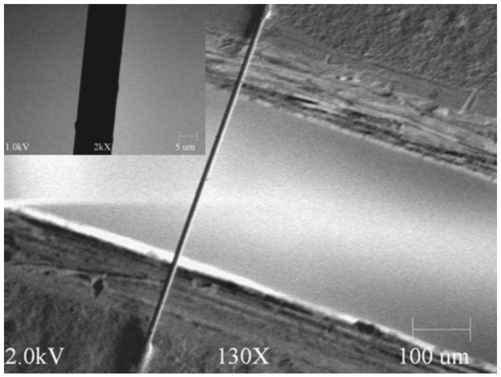
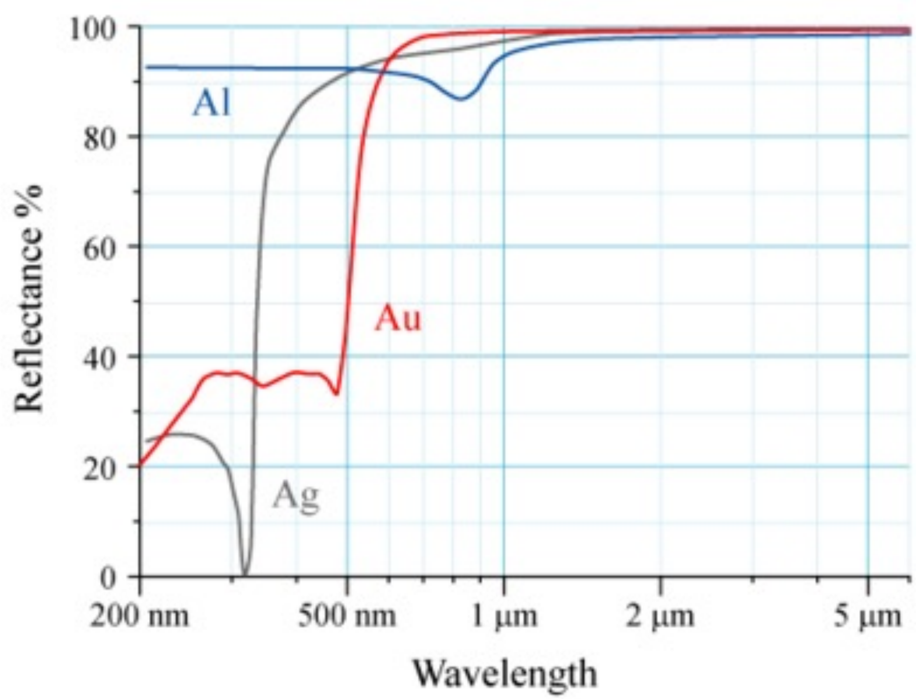
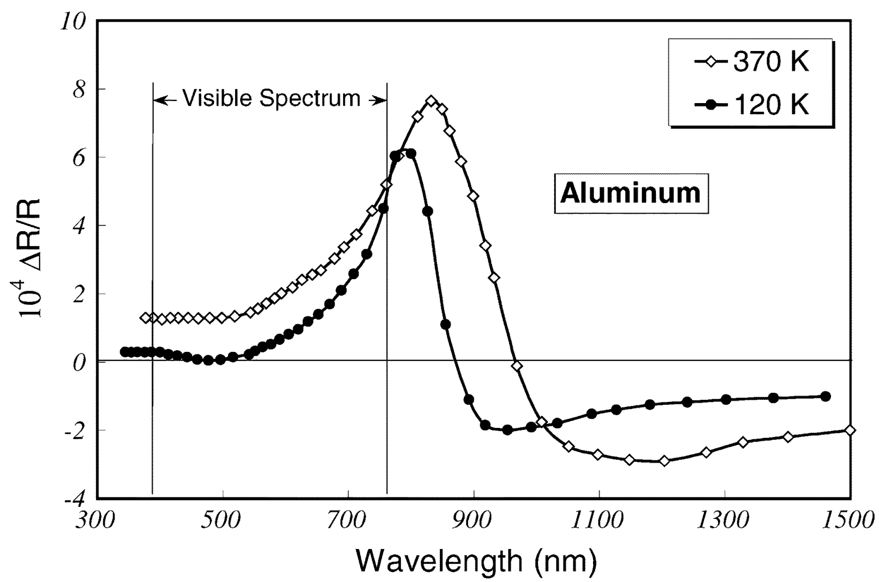


FIG. 6. SEM picture of coated polyester fiber (sample 2).

Transient measurements (optical)

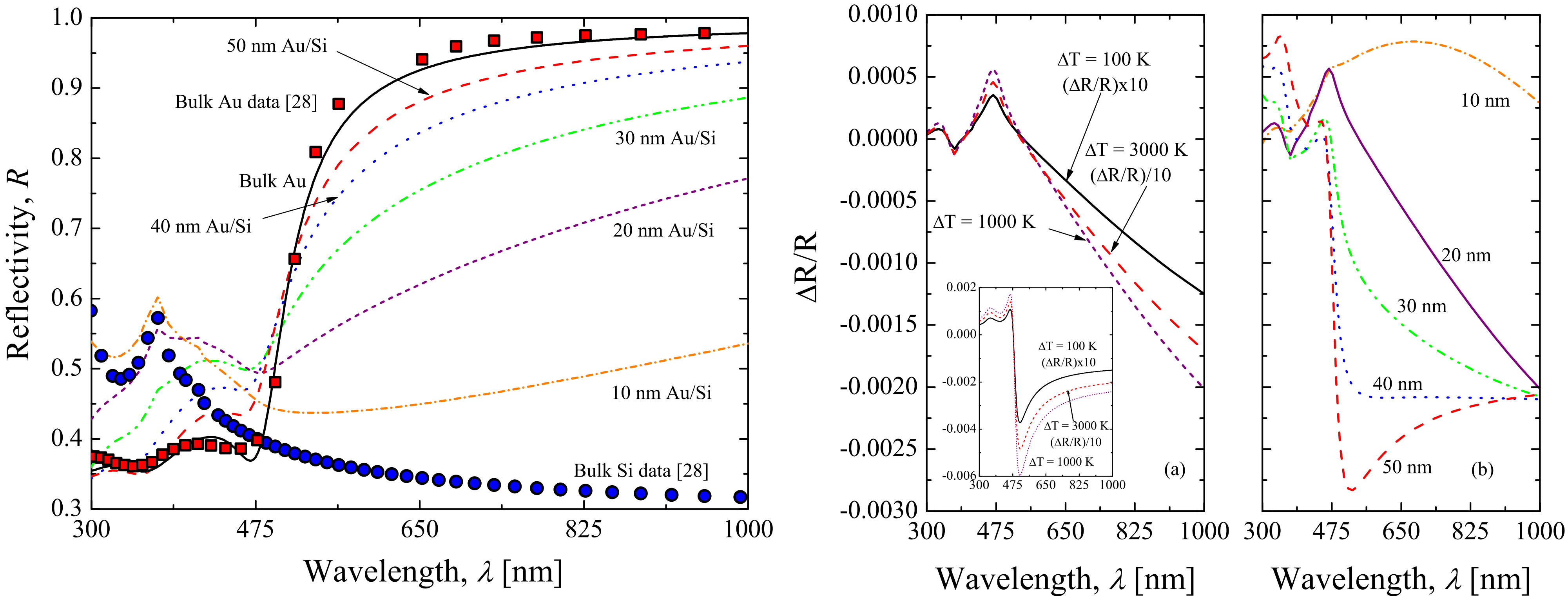
Reflectivity vs. Thermoreflectivity $\frac{\partial R}{\partial T}$





Transient measurements (optical)

Thermoreflectivity $\frac{\partial R}{\partial T}$



P. E. Hopkins. Influence of electron-boundary scattering on thermoreflectance calculations after intra- and interband transitions induced by short-pulsed laser absorption. Physical Review B, 81:035413, 2010.

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: nanosecond pump

Full relaxation of thermal energy before next pulse arrives

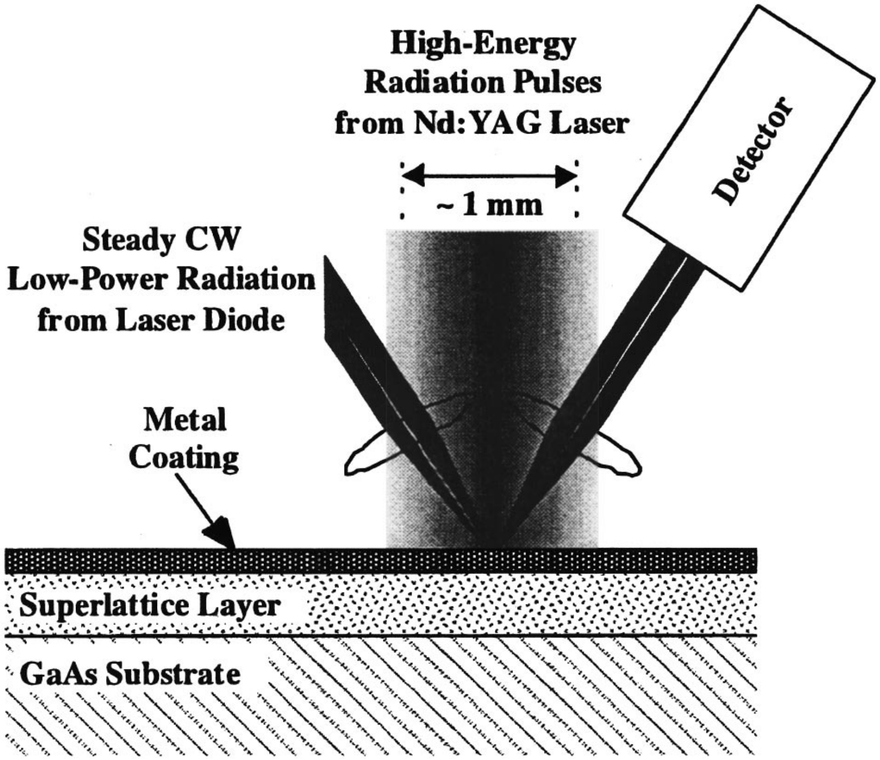


FIG. 1. The thermoreflectance method for measuring the vertical thermal resistance of Bi_2Te_3/Sb_2Te_3 superlattice layers.

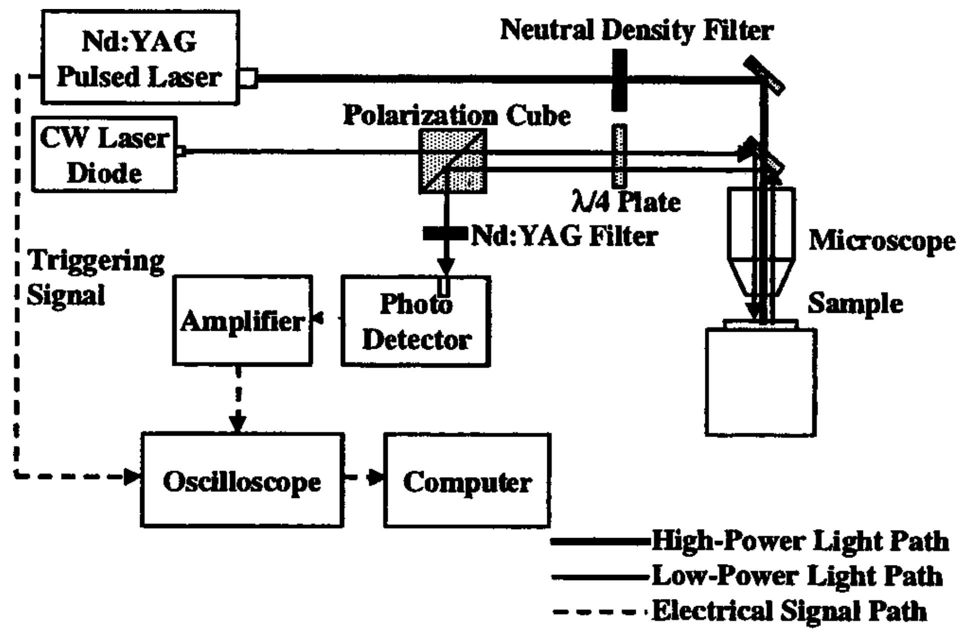
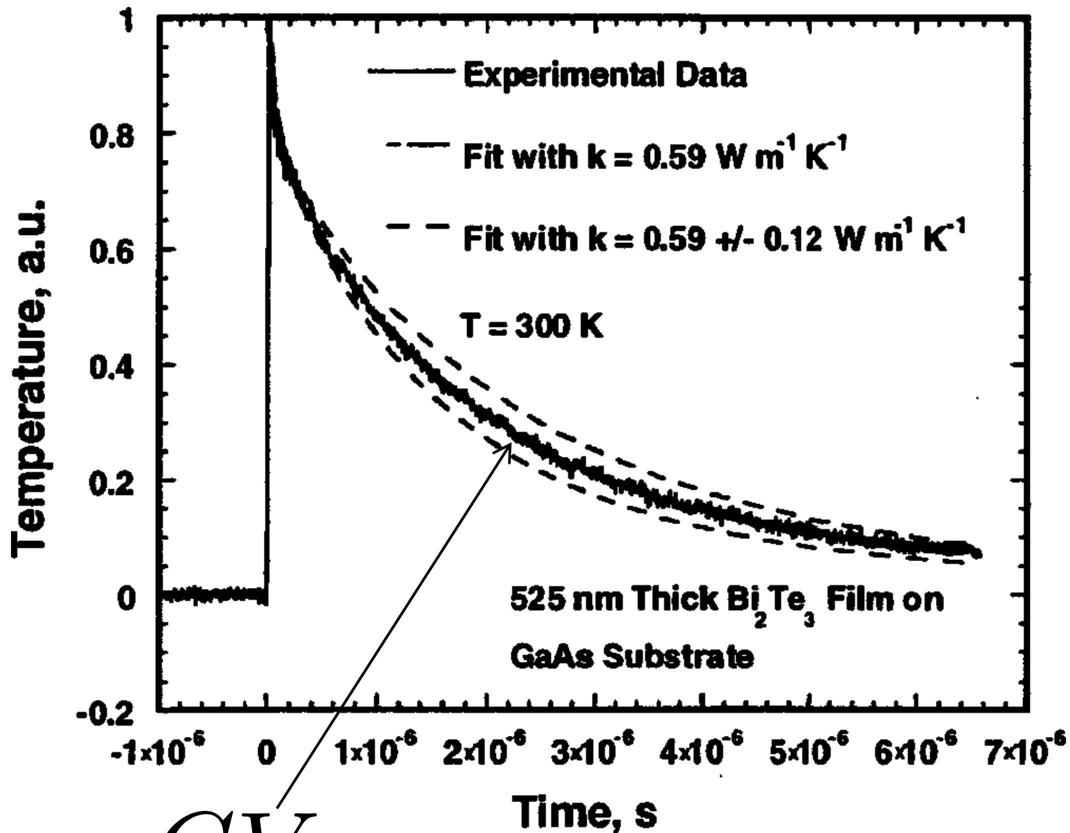


FIG. 2. Diagram describing the paths for radiation and electrical signals in the experimental setup.

M. N. Touzelbaev, P. Zhou, R. Venkatasubramanian, and K. E. Goodson. Thermal characterization of Bi_2Te_3/Sb_2Te_3 superlattices. Journal of Applied Physics, 90:763–767, 2001.

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: nanosecond pump



$$\tau = \frac{CV}{Ah}$$

Pump pulse gives heating event, and decay is monitored after heating event. Time of pulse dictates spatial resolution

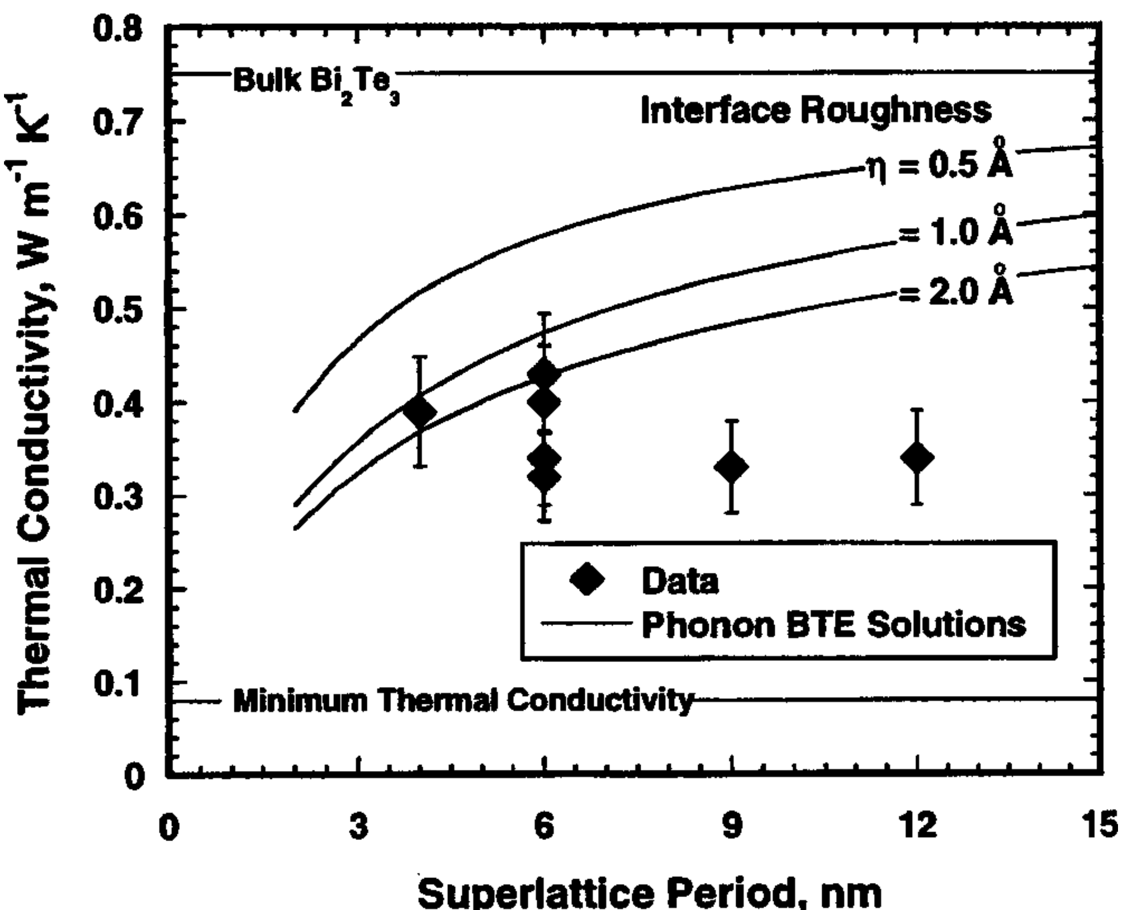
$$\tau = \frac{\delta^2 C}{\kappa}$$

$$\delta = \sqrt{\frac{\tau \kappa}{C}}$$

M. N. Touzelbaev, P. Zhou, R. Venkatasubramanian, and K. E. Goodson. Thermal characterization of Bi_2Te_3/Sb_2Te_3 superlattices. Journal of Applied Physics, 90:763–767, 2001.

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: nanosecond pump



M. N. Touzelbaev, P. Zhou, R. Venkatasubramanian, and K. E. Goodson. Thermal characterization of Bi_2Te_3/Sb_2Te_3 superlattices. Journal of Applied Physics, 90:763–767, 2001.

$$\delta = \sqrt{\frac{\tau \kappa}{C}}$$

For low diffusivity materials, assuming τ = 1 ns

$$\delta = 30\,\mathrm{nm}$$

For high diffusivity materials

$$\delta = 300\,\mathrm{nm}$$

BUT – limited by temporal processes > τ = 1 ns

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: femtosecond pump

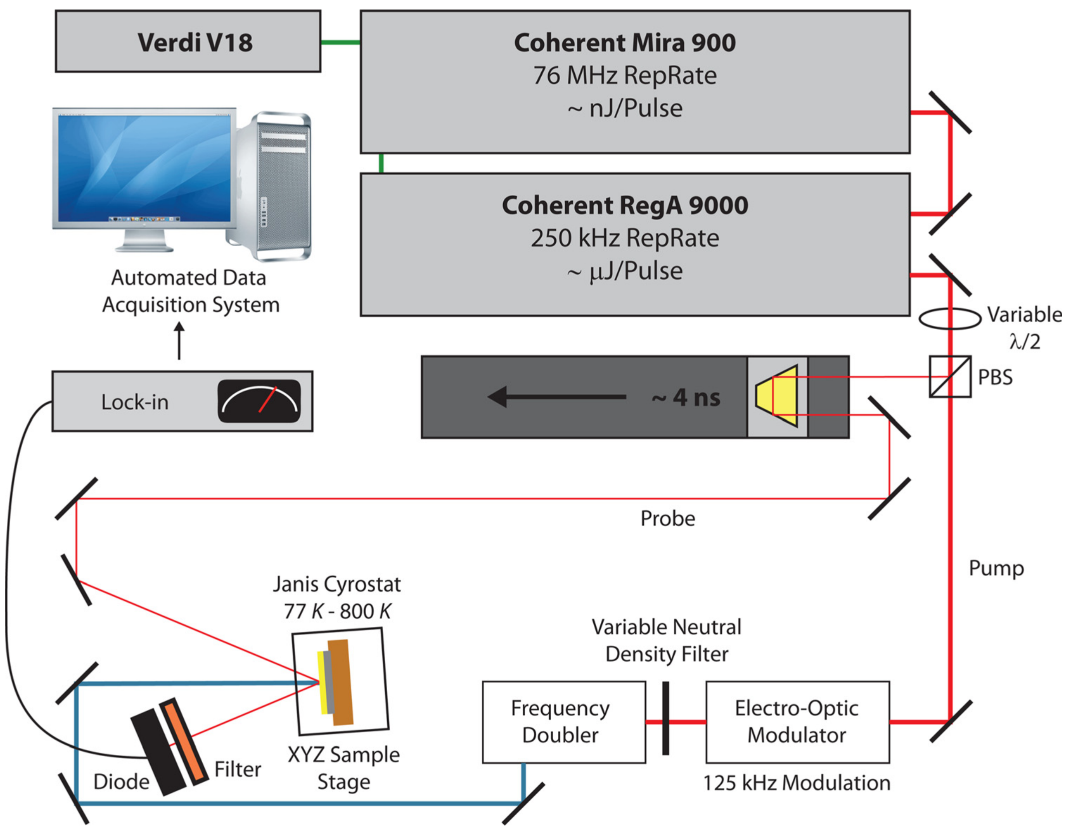


Fig. 2 Schematic of transient thermoreflectance setup at University of Virginia.

P. M. Norris, J. L. Smoyer, J. C. Duda, and P. E. Hopkins. Prediction and measurement of thermal transport across interfaces between isotropic solids and graphitic materials. Journal of Heat Transfer, 134:020910, 2012.

$$\delta = \sqrt{\frac{\tau \kappa}{C}}$$

For low diffusivity materials, assuming τ = 100 fs

$$\delta = 0.3\,\mathrm{nm}$$

For high diffusivity materials

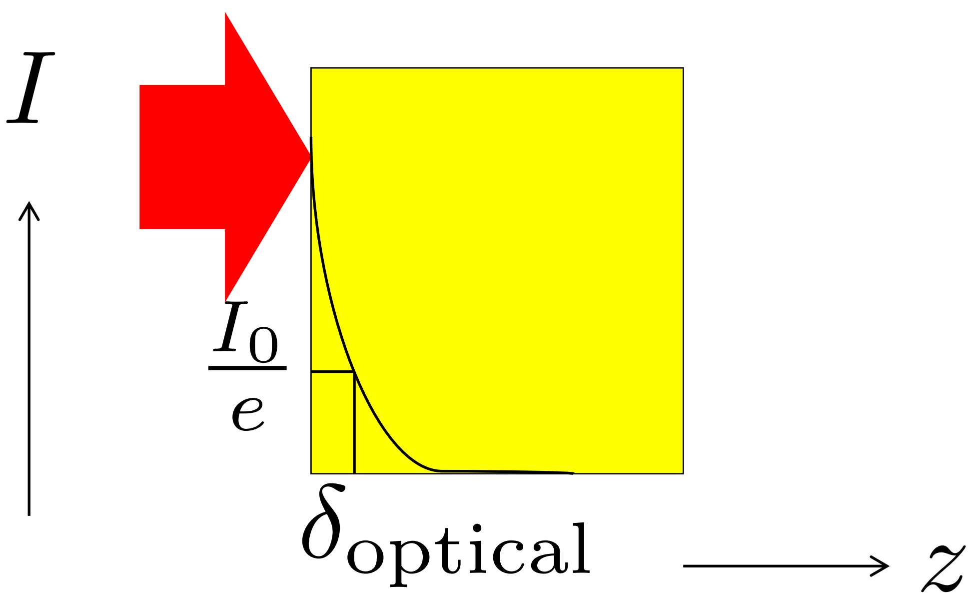
$$\delta = 3.0\,\mathrm{nm}$$

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: femtosecond pump

In this case, spatial resolution limited by optical penetration depth of metal

$$\delta_{\text{optical}} = \frac{\lambda}{4\pi k}$$



FANTASTIC temporal resolution (limited by pulse width)

Pulse absorption (~100 fs)

↓

Fermi relaxation and ballistic transport (few hundred fs)

↓

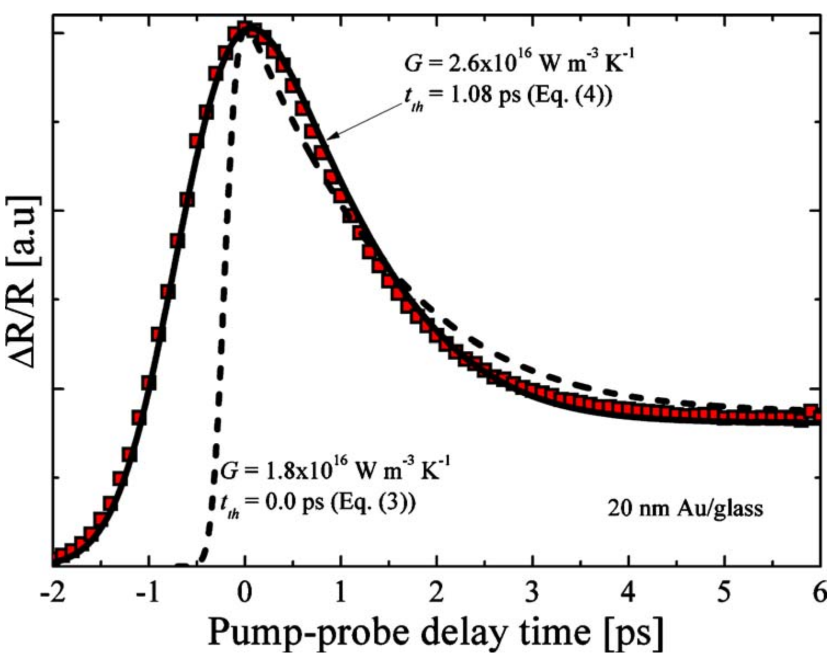
Electron-phonon coupling (a few ps)

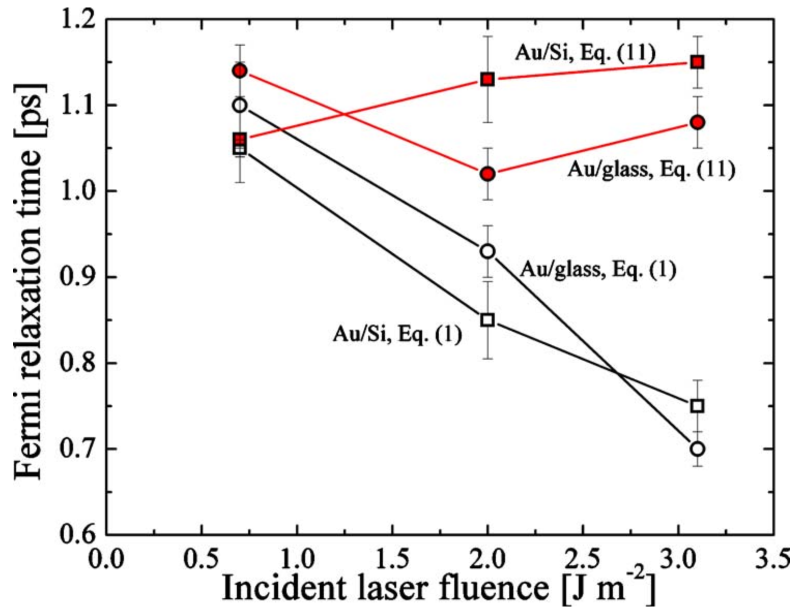
↓

Thermal diffusion (hundreds of ps to ns)

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: femtosecond pump

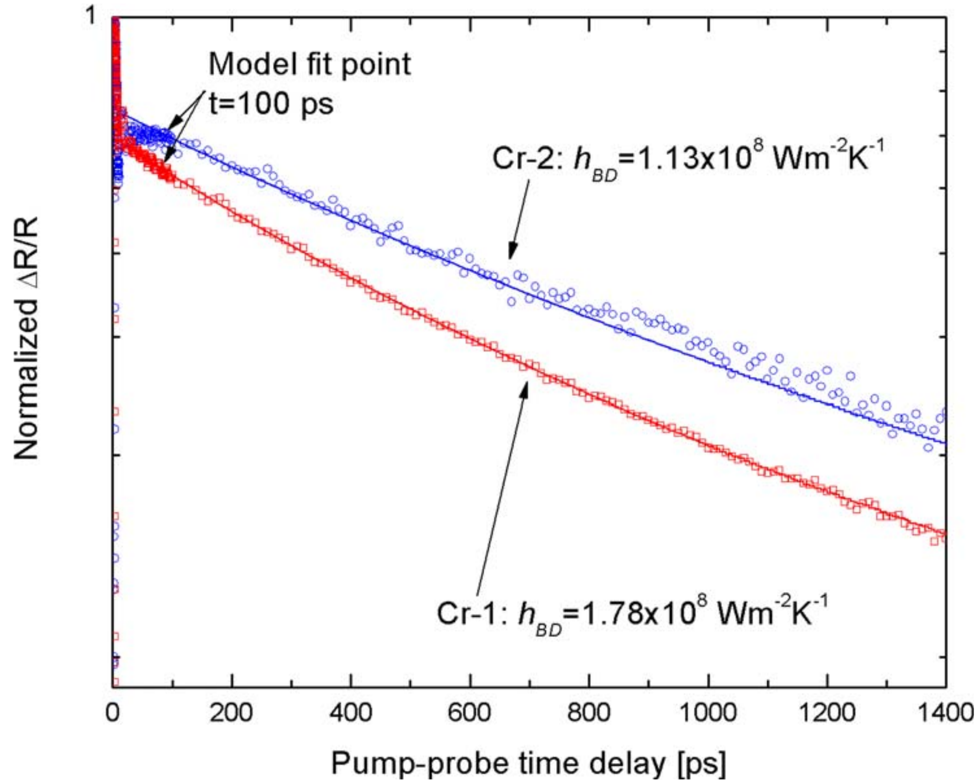


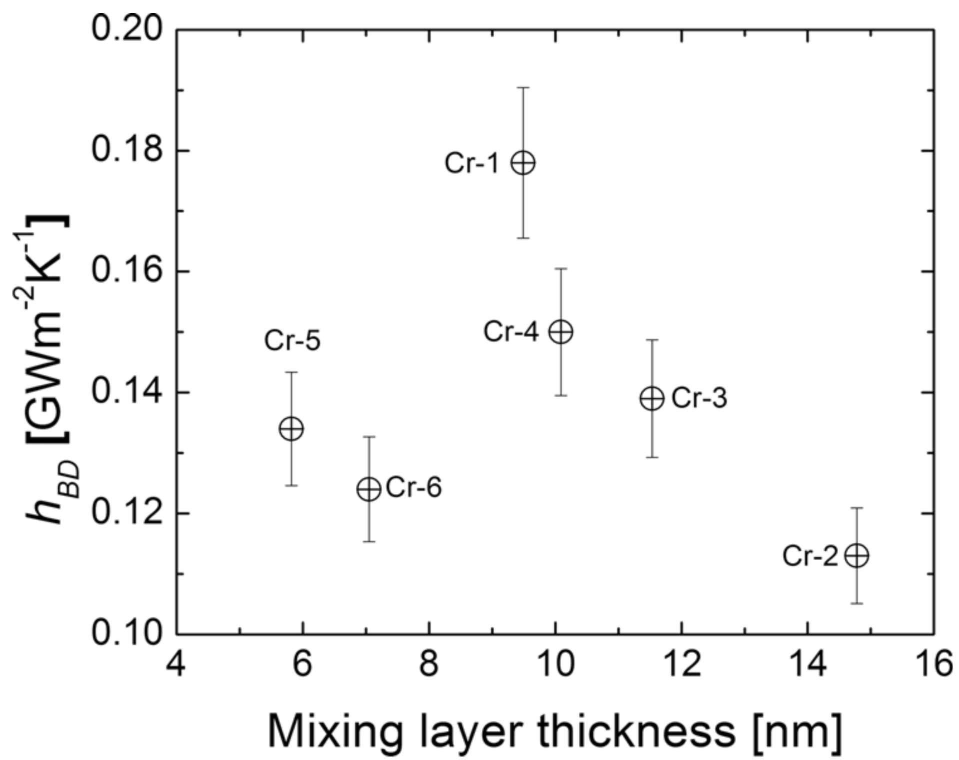


P. E. Hopkins, L. M. Phinney, and J. R. Serrano. Reexamining electron-fermi relaxation in gold films with a nonlinear thermoreflectance model. Journal of Heat Transfer, 133:044505, 2011

Transient ThermoReflectance (TTR) measurements (optical)

Pump-probe: femtosecond pump





P. E. Hopkins, P. M. Norris, R. J. Stevens, T. Beechem, and S. Graham. Influence of interfacial mix- ing on thermal boundary conductance across a chromium/silicon interface. Journal of Heat Transfer, 130:062402, 2008.

Overview: Methods for testing κ of thin films

- **Steady State resistivity approaches**
  - **No variation in time ("Fourier Law")**

- **Transient reflectivity and optical methods**
  - **Time dependent ("The heat eq. w/ impulse response")**

- **Modulated methods ("The heat eq. w/ frequency dep. source")**
  - **3ω**
  - **Thermoreflectance-based techniques**
    - **FDTR**
    - **TDTR**

Steady state vs. transient vs. periodic

Steady state = The Fourier Law

$$q = -\kappa \frac{\partial T}{\partial z}$$

Transient = The Heat Equation

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial z^2} + q(t)$$

Heat capacity enters the picture

The source term can make a difference $q\left(t\right)$ vs. $q\left(t, \omega\right)$

If source term is periodic (and not "single shot, or instantaneous), then you get a modulated temperature on your samples surface

1) This yields both steady state and transient components
2) Makes data analysis easier since you can work in frequency domain

Periodic measurements

What separates periodic measurements from everything else????

$$\delta_{\mathrm{thermal}} = \sqrt{\frac{\kappa}{\pi f C}} = \sqrt{\frac{2\kappa}{\omega C}}$$

Thermal penetration depth

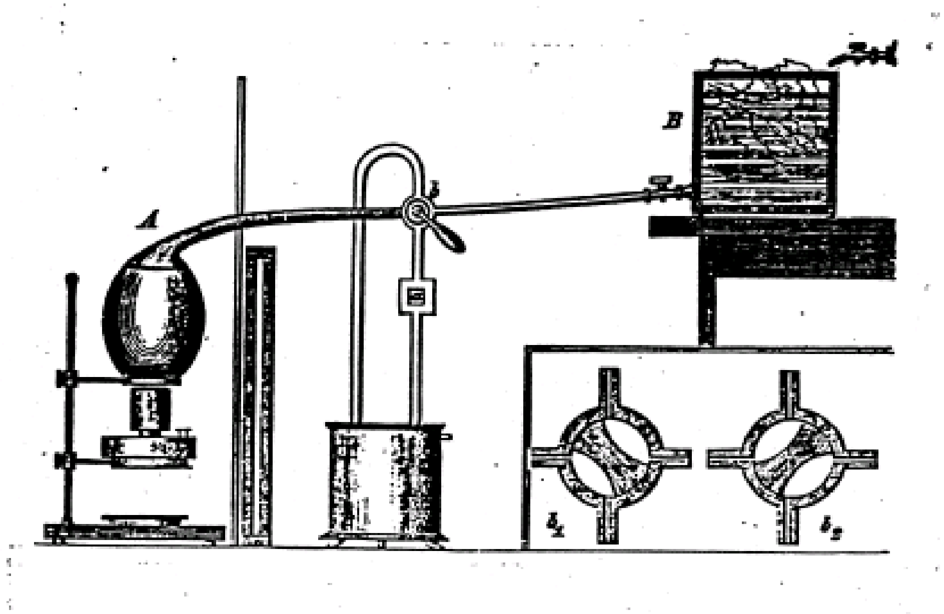
Angstrom method

Used fixed temperature boundary conditions

$$T\,(x = 0) = 0^{\circ}\mathrm{C} \qquad 0 < t < \Gamma/2$$

$$T\,(x = 0) = 100^{\circ}\mathrm{C} \qquad \Gamma/2 < t < \Gamma$$

where Γ is the period of temperature oscillations produced by alternating flow of ice water and steam



Frequency dependent temperature rise leads to temperature fluctuation at end of sample with some phase lag based on RC

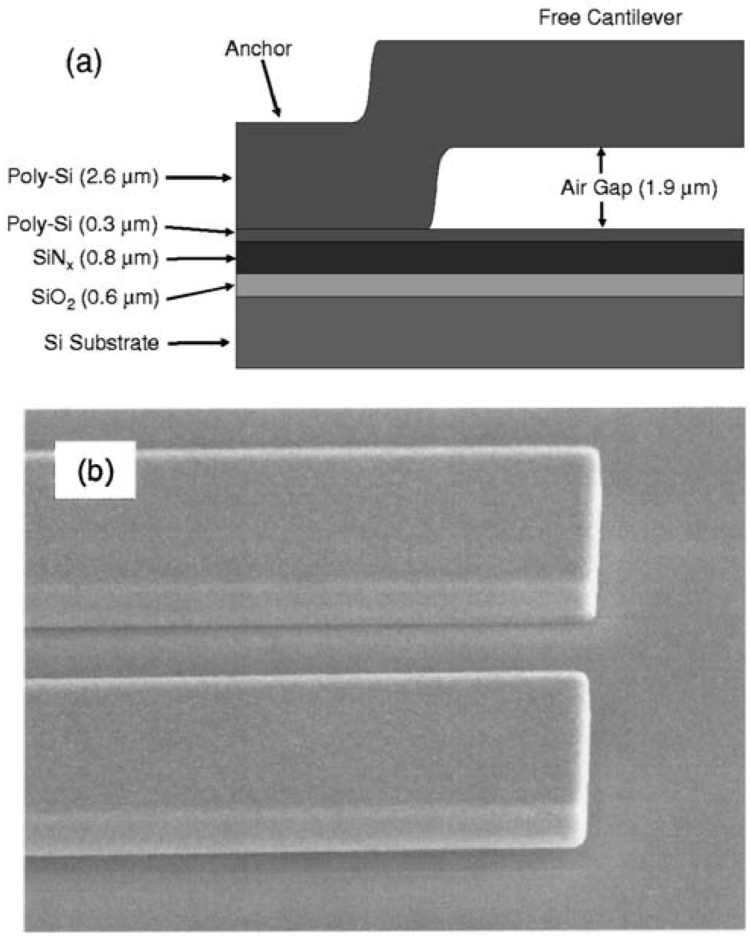
Modified Angstrom method

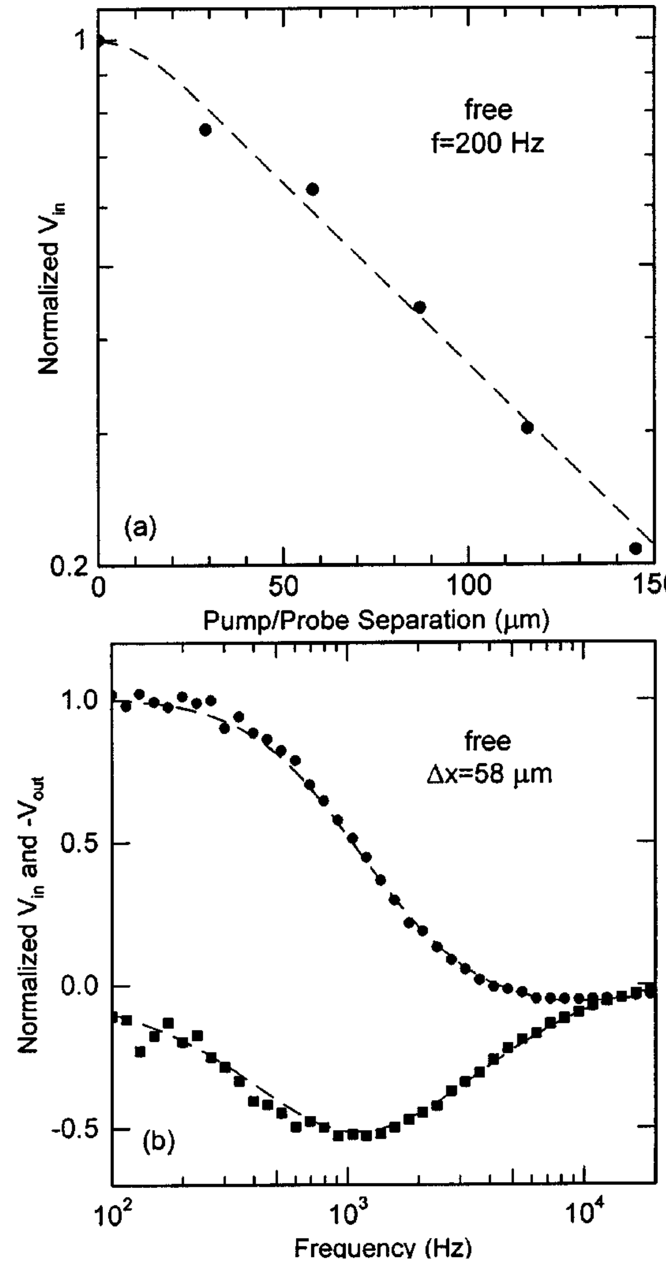
JOURNAL OF APPLIED PHYSICS VOLUME 95, NUMBER 4 15 FEBRUARY 2004

Thermal contact conductance of adhered microcantilevers

Scott T. Huxtable[a] and David G. Cahill
Department of Materials Science and Engineering and the Frederick Seitz Materials Research Laboratory, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801

Leslie M. Phinney[b]
Department of Mechanical and Industrial Engineering, University of Illinois, Urbana, Illinois 61801

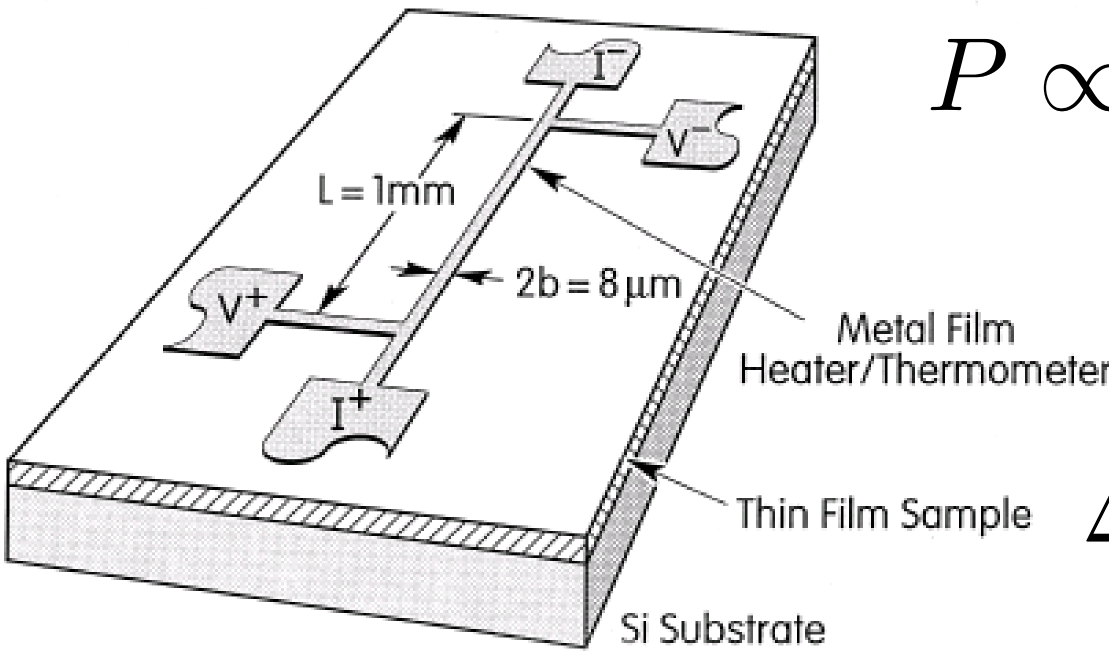




3ω technique

Uses single metal film for heater/thermometer (Birge, 1987); (Cahill, 1990).

$$I \propto \exp\left[i\omega t\right]$$

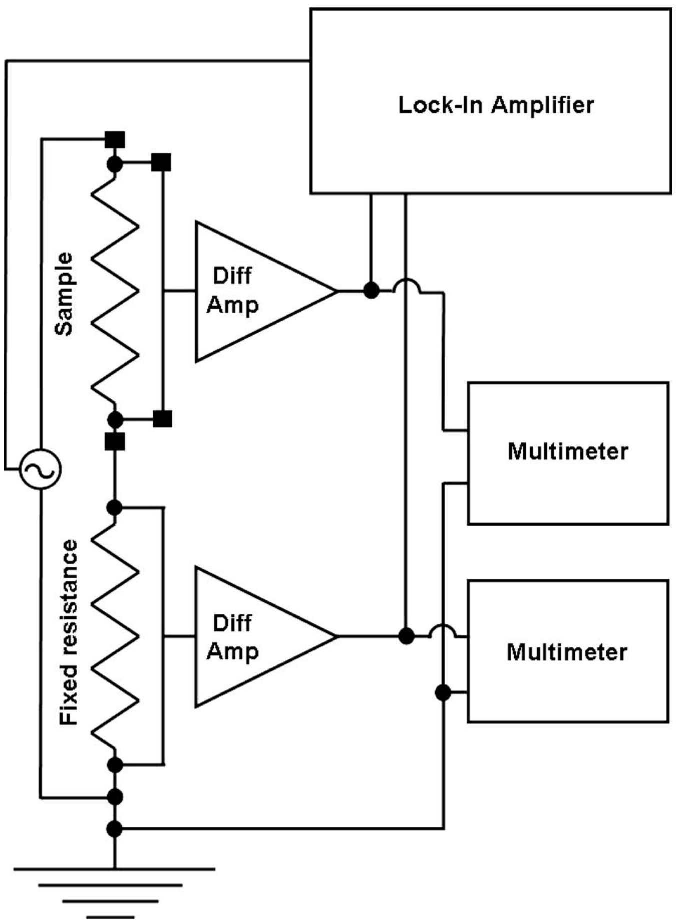


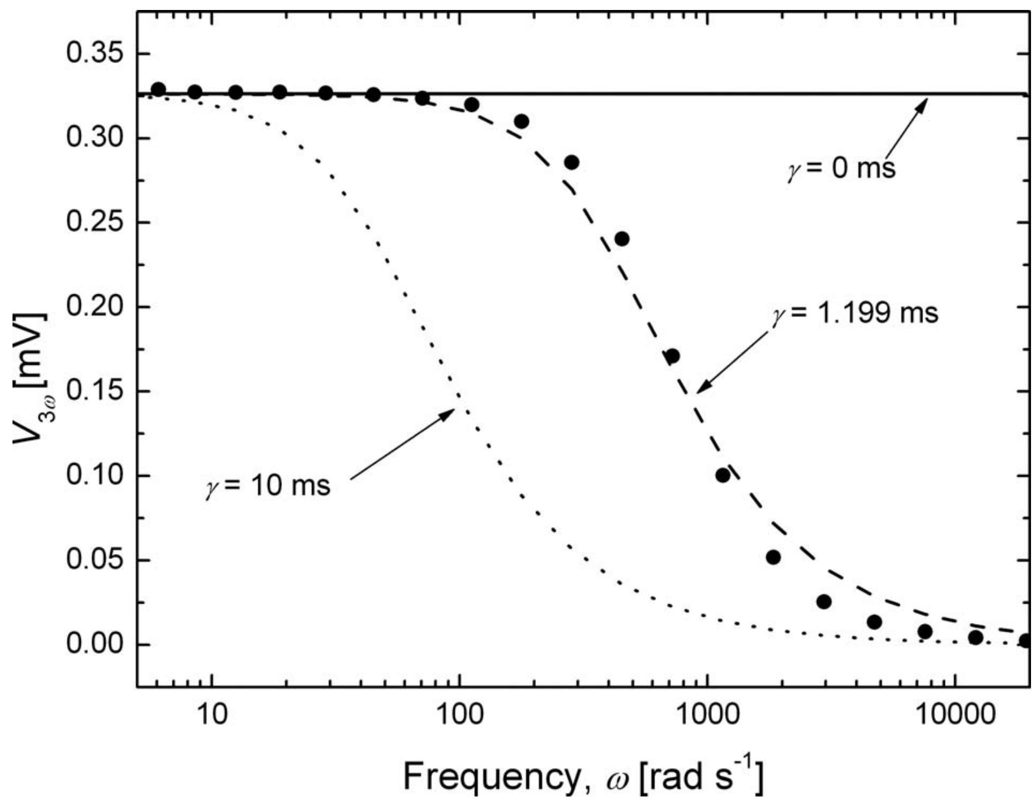
$$P \propto \Delta T \propto \exp\left[i2\omega t\right]$$

$$\Delta R \propto \exp\left[i2\omega t\right]$$

$$\Delta V = I\Delta R \propto \exp\left[i3\omega t\right]$$

3ω technique

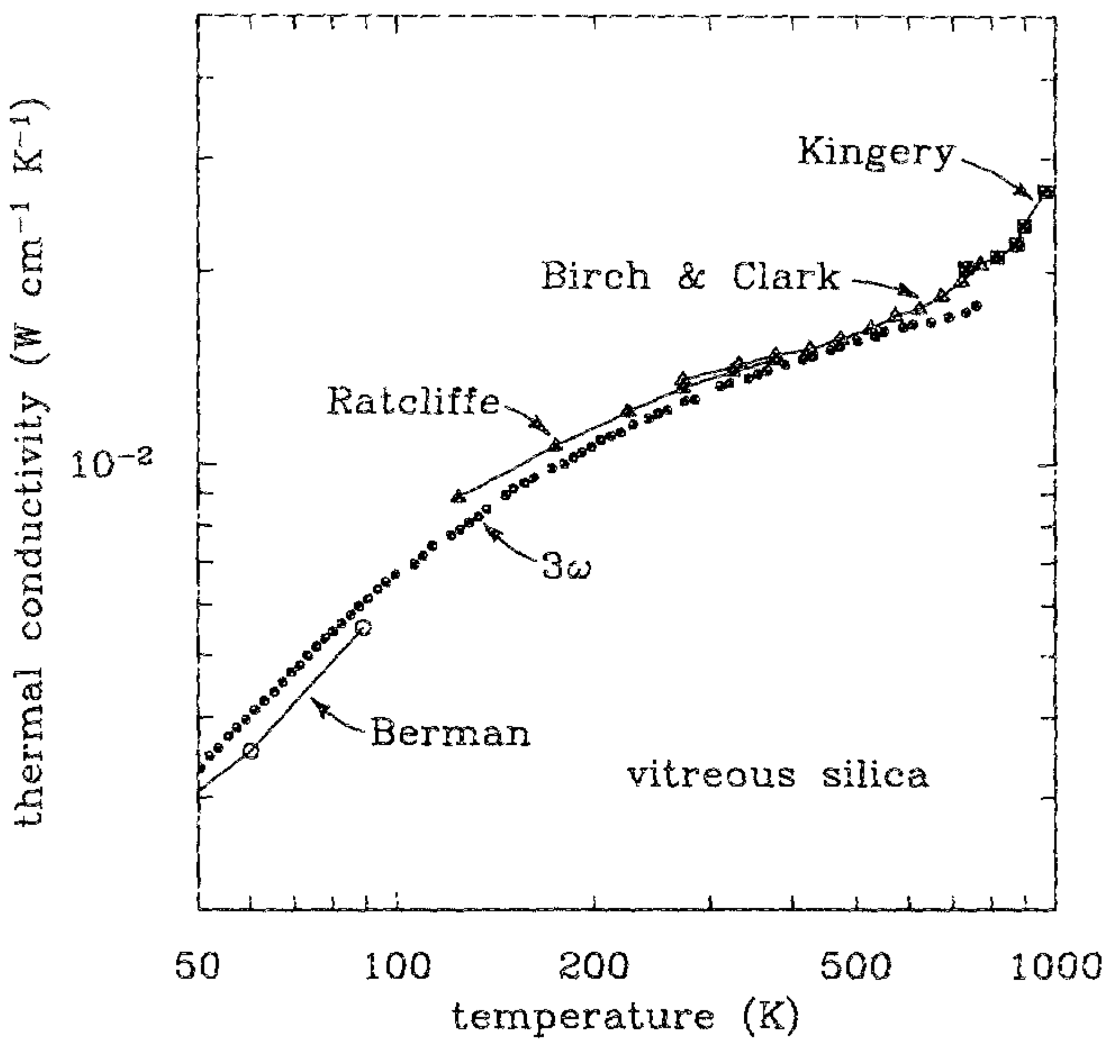




P. E. Hopkins and L. M. Phinney. Thermal conductivity measurements on polycrystalline silicon micro- bridges using the 3w technique.

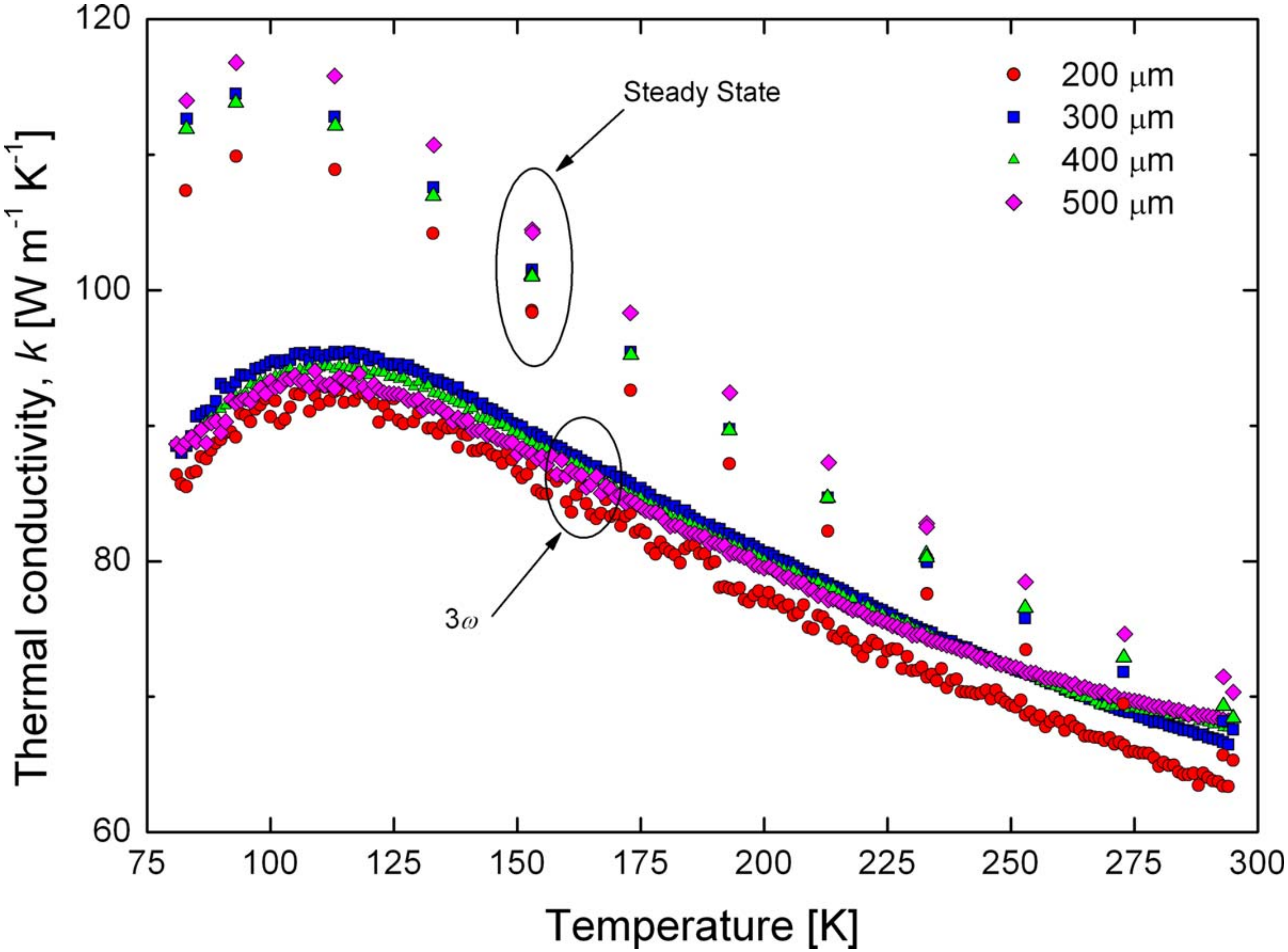
Journal of Heat Transfer, 131:043201, 2009.

3ω technique



D. G. Cahill. Thermal conductivity measurement from 30 to 750 K: The 3ω method. Review of Scientific Instruments, 61:802–808, 1990.

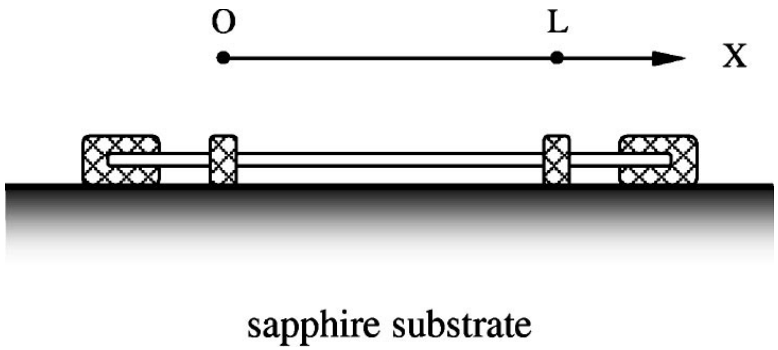
3ω vs. steady state



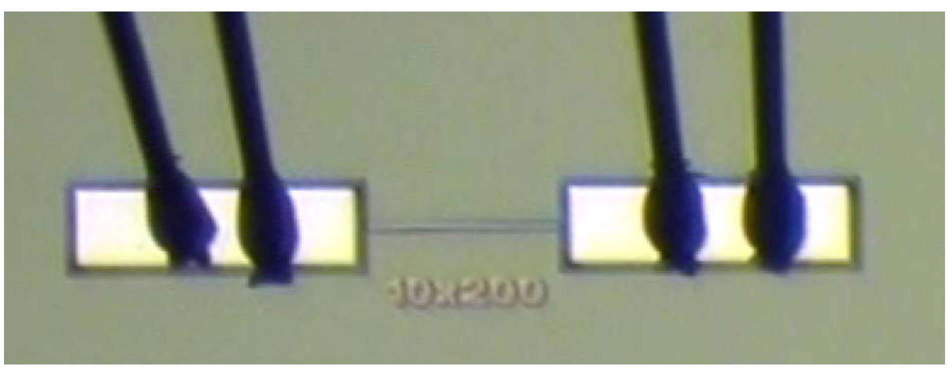
What could differences be caused by?

Frequency-domain measurements of κ in thin films

3ω technique – been used extensively for thin films, including suspended films and nanostructures



L. Lu, W. Yi, and D. L. Zhang. 3 omega method for specific heat and thermal conductivity measurements. **Review of Scientific Instruments, 72(7):2996–3003, 2001.**



P. E. Hopkins and L. M. Phinney. Thermal conductivity measurements on polycrystalline silicon micro- bridges using the 3w technique. **Journal of Heat Transfer, 131:043201, 2009.**

<u>Review of electrical resistivity-based thermal conductivity measurement techniques for nanosystems</u>

CHAPTER 2

MEASURING THE THERMAL CONDUCTIVITY OF THIN FILMS: 3 OMEGA AND RELATED ELECTROTHERMAL METHODS

Chris Dames

Department of Mechanical Engineering, University of California at Berkeley, 6107 Etcheverry Hall, Berkeley CA 94720-1740, USA; E-mail: cdames@berkeley.edu

C. Dames. Measuring the thermal conductivity of thin films: 3 omega and related electrotehrmal methods. **Annual Review of Heat Transfer, 16:7–49, 2013.**

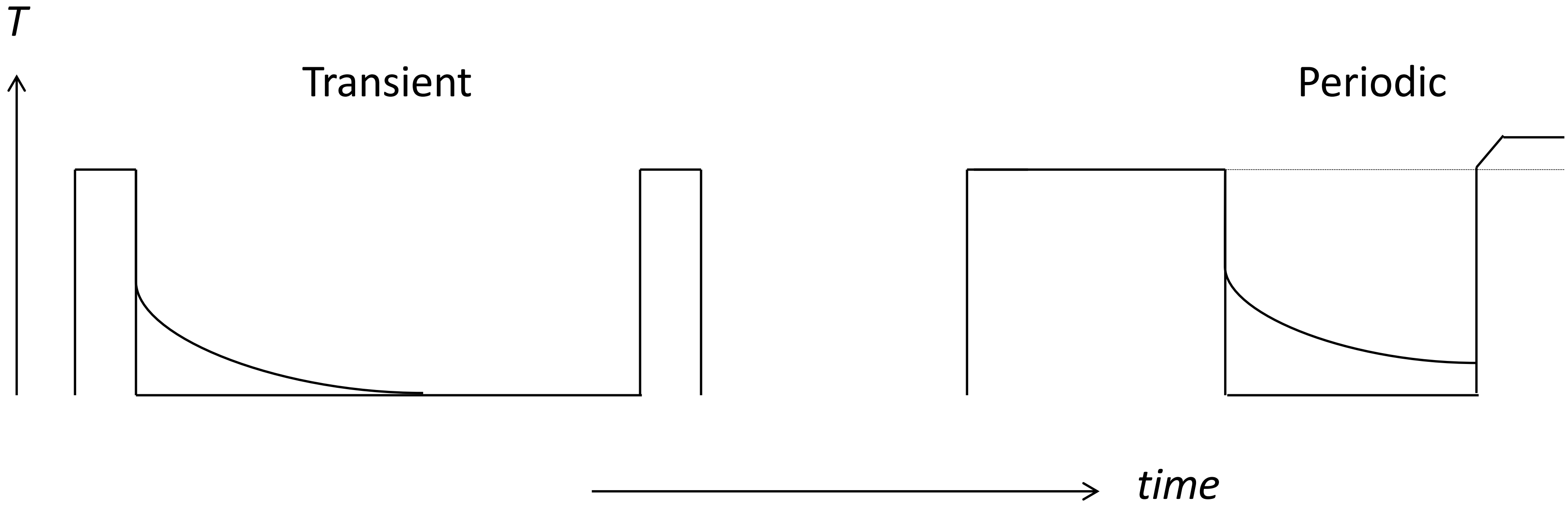
What about optical periodic heating techniques?

$$q\left(t\right) \;\text{ vs. }\; q\left(t,\omega\right)$$

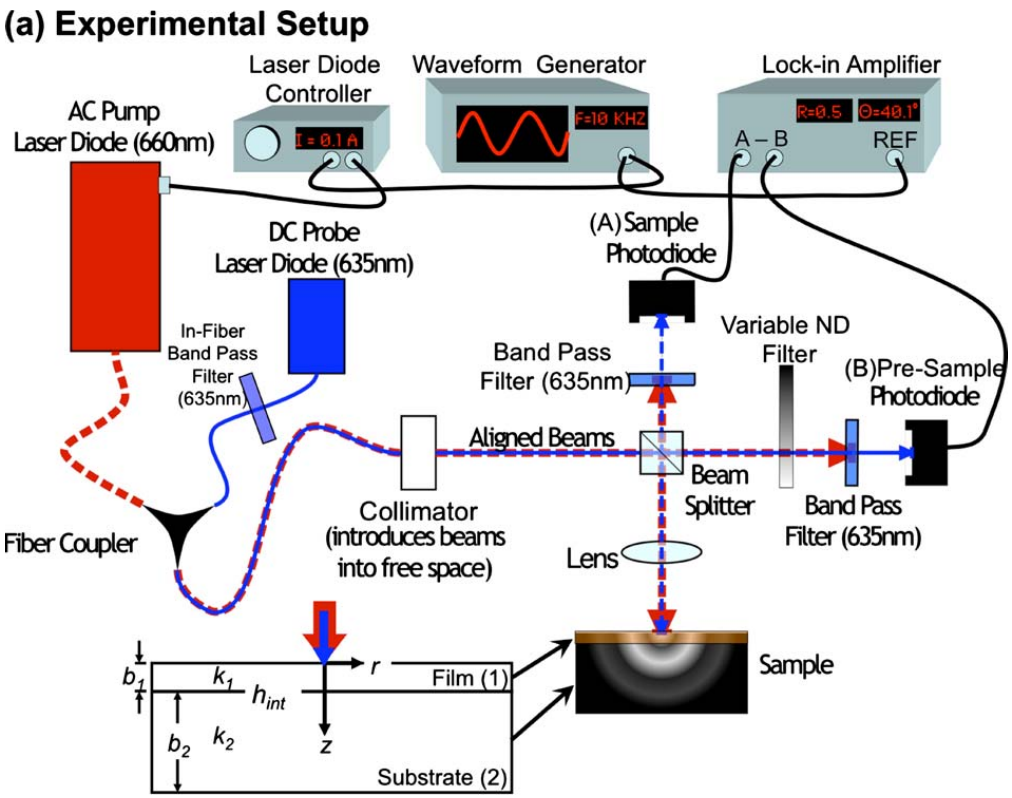
If source term is periodic (and not “single shot, or instantaneous), then you get a modulated temperature on your samples surface

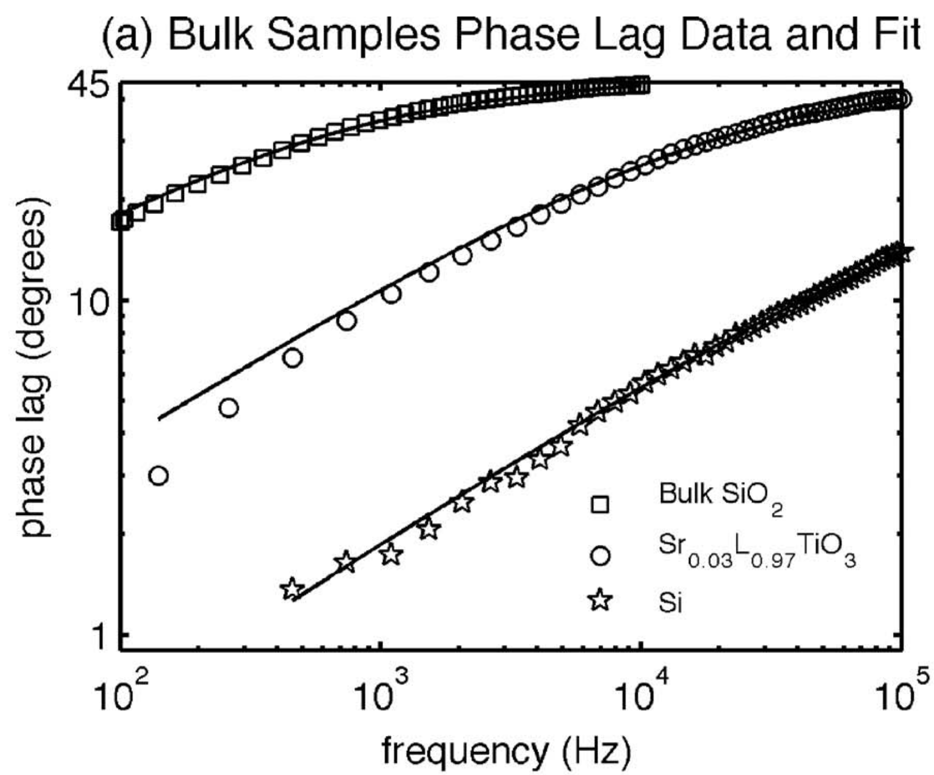
THE KEY IS THE DUTY CYCLE!!!!

NEED HEATING EVENT TO BE “FELT” BY NEXT HEATING EVENT



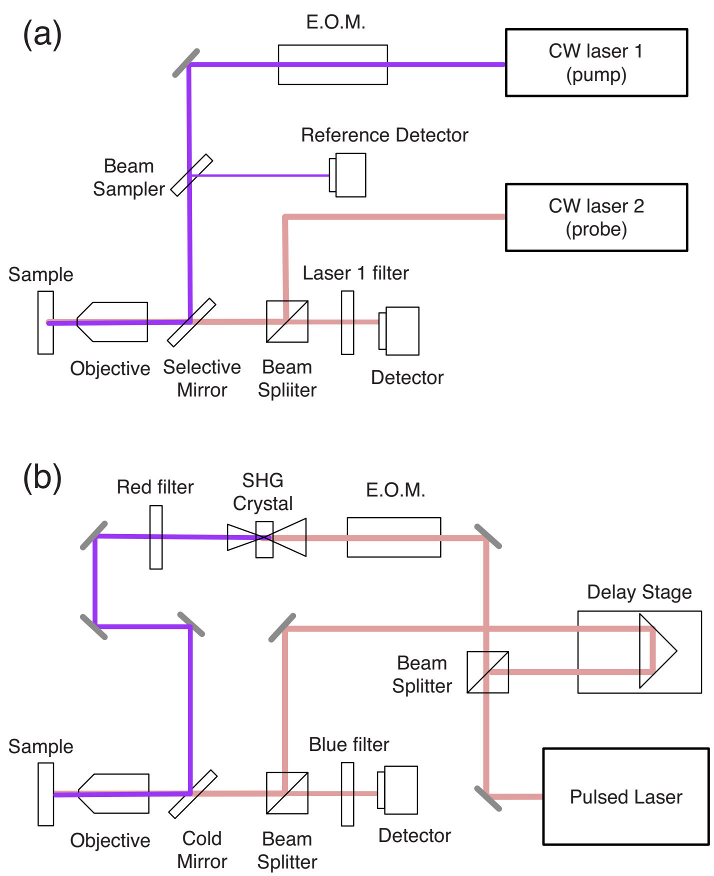
CW-Frequency domain thermoreflectance

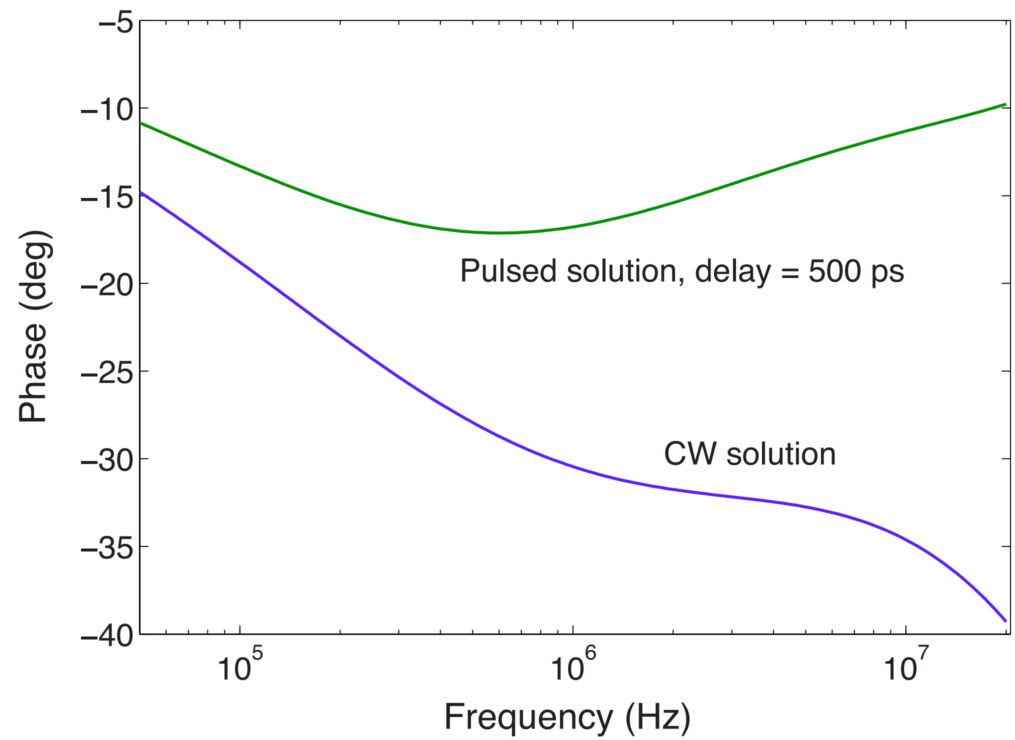




J. A. Malen, K. Baheti, T. Tong, Y. Zhao, J. A. Hudgings, and A. Majumdar. Optical measurement of thermal conductivity using fiber aligned frequency domain thermoreflectance. Journal of Heat Transfer, 133(8):081601, 2011.

FDTR – cw vs. pulsed



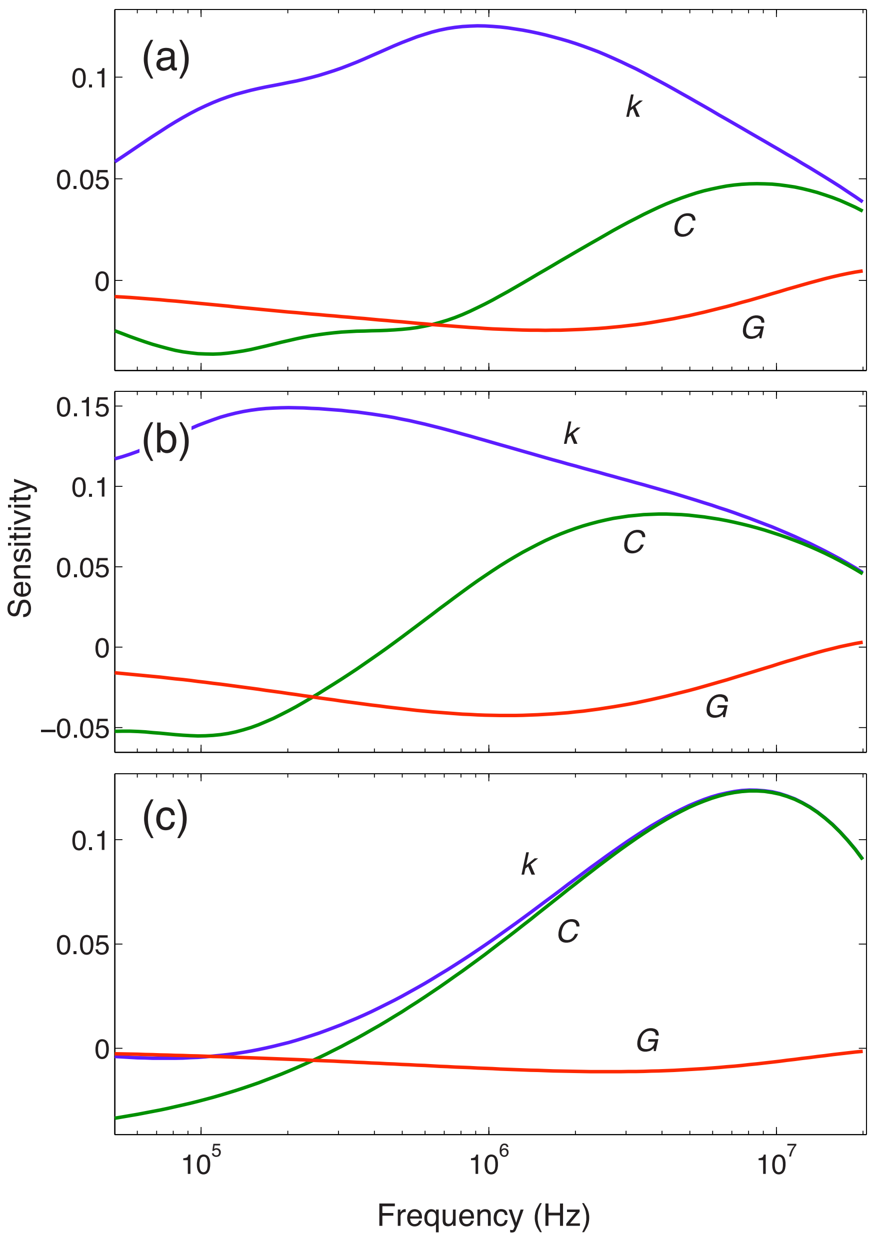


$$\delta_{\mathrm{cw}} = \sqrt{\frac{\kappa}{\pi f C}}$$

$$\delta_{\mathrm{pulsed}} \propto \sqrt{\tau \frac{\kappa}{C}}$$

A. J. Schmidt, R. Cheaito, and M. Chiesa. A frequency-domain thermoreflectance method for the characterization of thermal properties. Review of Scientific Instruments, 80:094901, 2009.

FDTR – pulsed



$$D = \frac{\kappa}{C} \qquad E = \sqrt{\kappa C}$$

Silicon

Where are the diffusivity and effusivity regimes?

Sapphire

In what materials can you accruately determine both C and κ?

Pyrex (SiO_2)

A. J. Schmidt, R. Cheaito, and M. Chiesa. A frequency-domain thermoreflectance method for the characterization of thermal properties. Review of Scientific Instruments, 80:094901, 2009.

FDTR – pulsed

JOURNAL OF APPLIED PHYSICS **107**, 024908 (2010)

Characterization of thin metal films via frequency-domain thermoreflectance

Aaron J. Schmidt,[1,2,a)] Ramez Cheaito,[2] and Matteo Chiesa[2]
[1]*Department of Mechanical Engineering, Massachusetts Institute of Technology Cambridge, Massachusetts, USA*
[2]*Department of Mechanical Engineering, Masdar Institute of Science and Technology, Abu Dhabi, United Arab Emirates*

(Received 30 October 2009; accepted 12 December 2009; published online 27 January 2010)

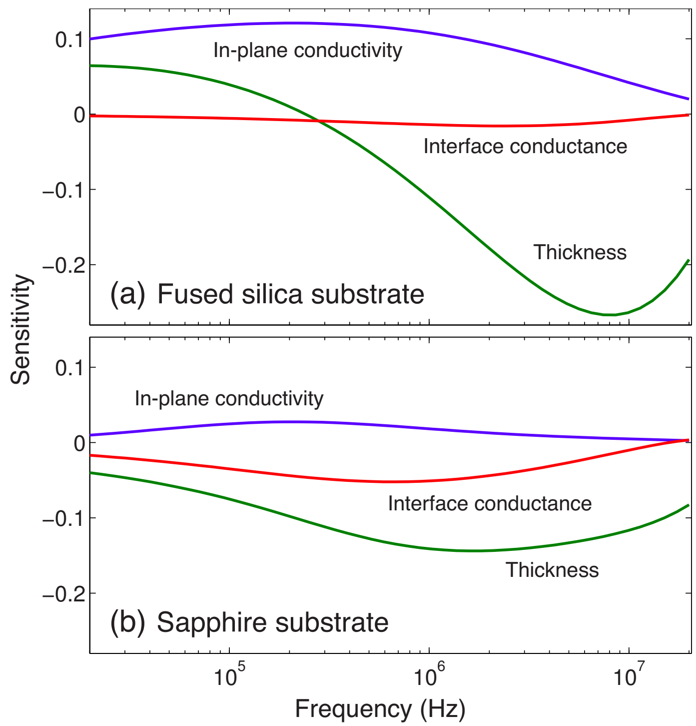


FIG. 5. (Color online) (a) The sensitivity parameter, Eq. (5), for in-plane thermal conductivity, metal-substrate boundary conductance, and metal thickness, for a sample consisting of an 80 nm film of Au deposited on a fused silica substrate. (b) The same sensitivities, calculated for a sapphire substrate. The phase angle in Eq. (5) is taken in radians.

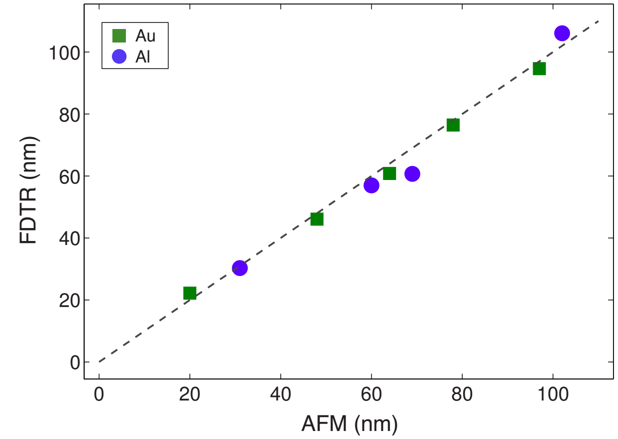


FIG. 3. (Color online) Film thickness data obtained for Au and Al films on fused silica substrates. The ordinate is the thickness determined from AFM cross sections while the abscissa is the FDTR value. Error bars based on two standard deviations are approximately the size of the symbols used.

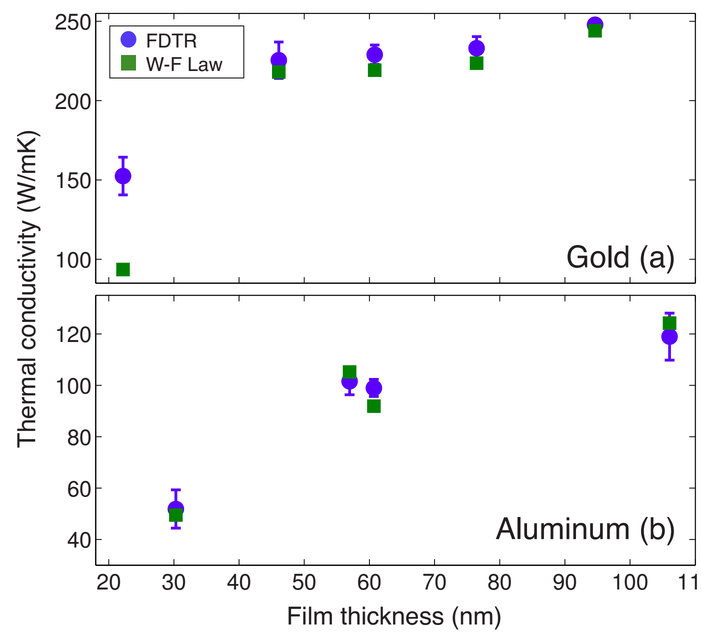
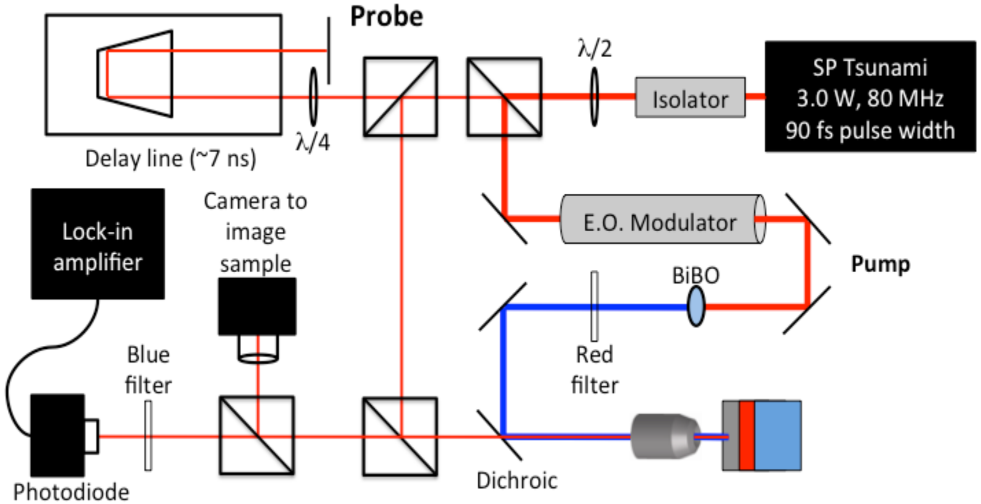


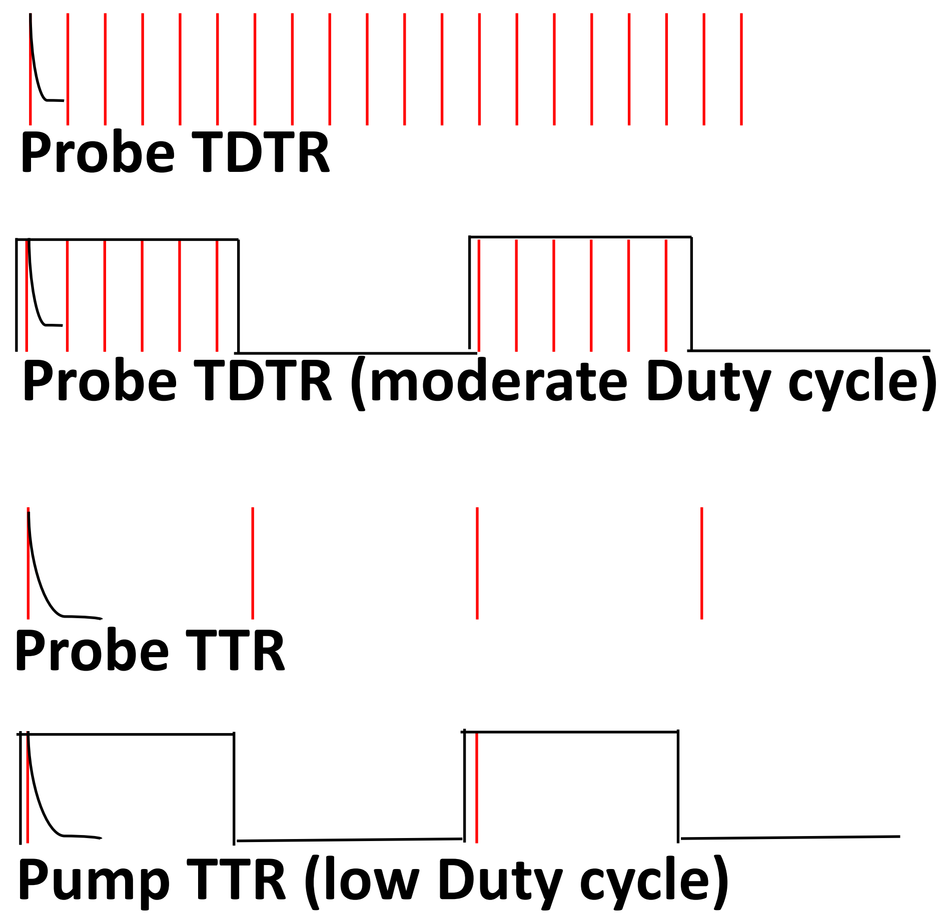
FIG. 4. (Color online) Thermal conductivity data obtained for (a) Au and (b) Al films on fused silica substrates. Circles are values obtained with the FDTR method, while the squares are values computed from electrical conductivity measurements using the WF law.

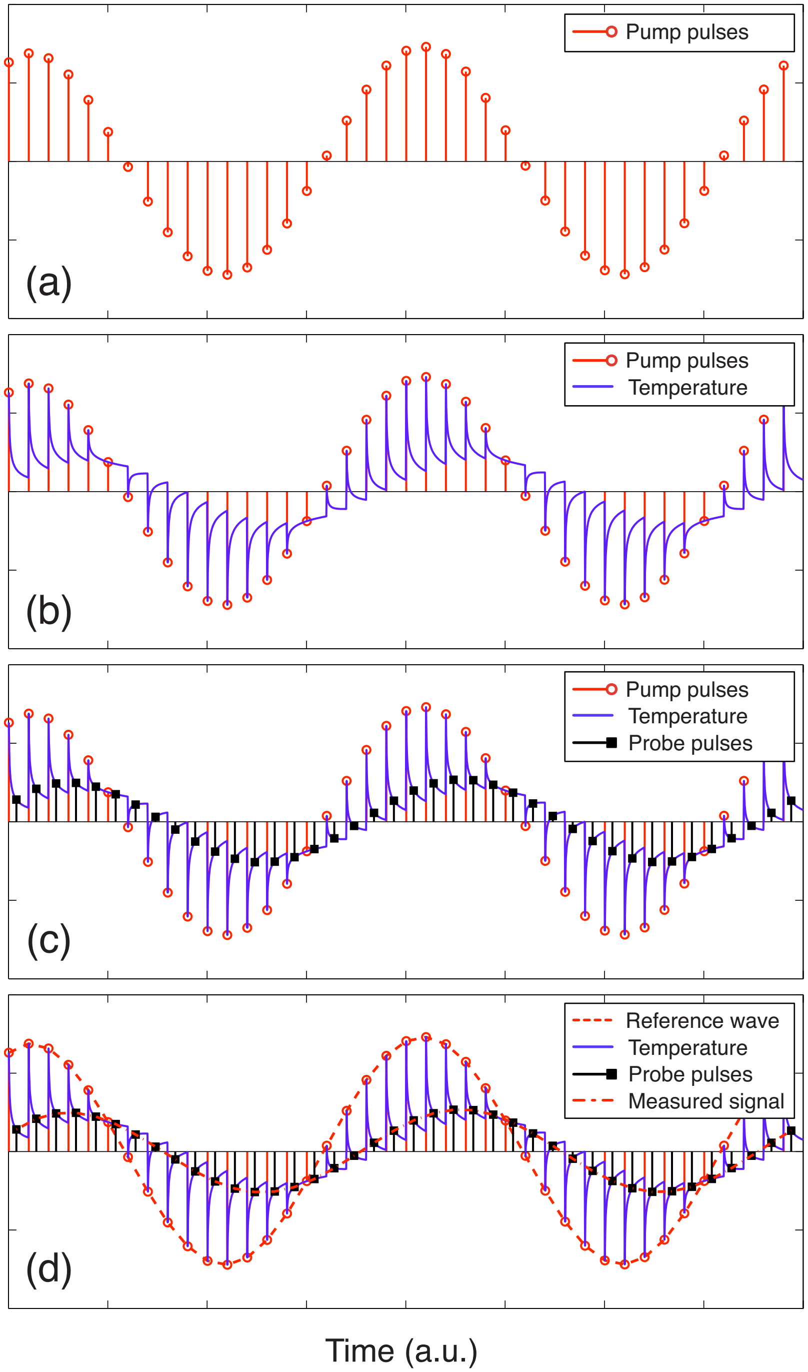
Can we achieve transient AND periodic thermometry?

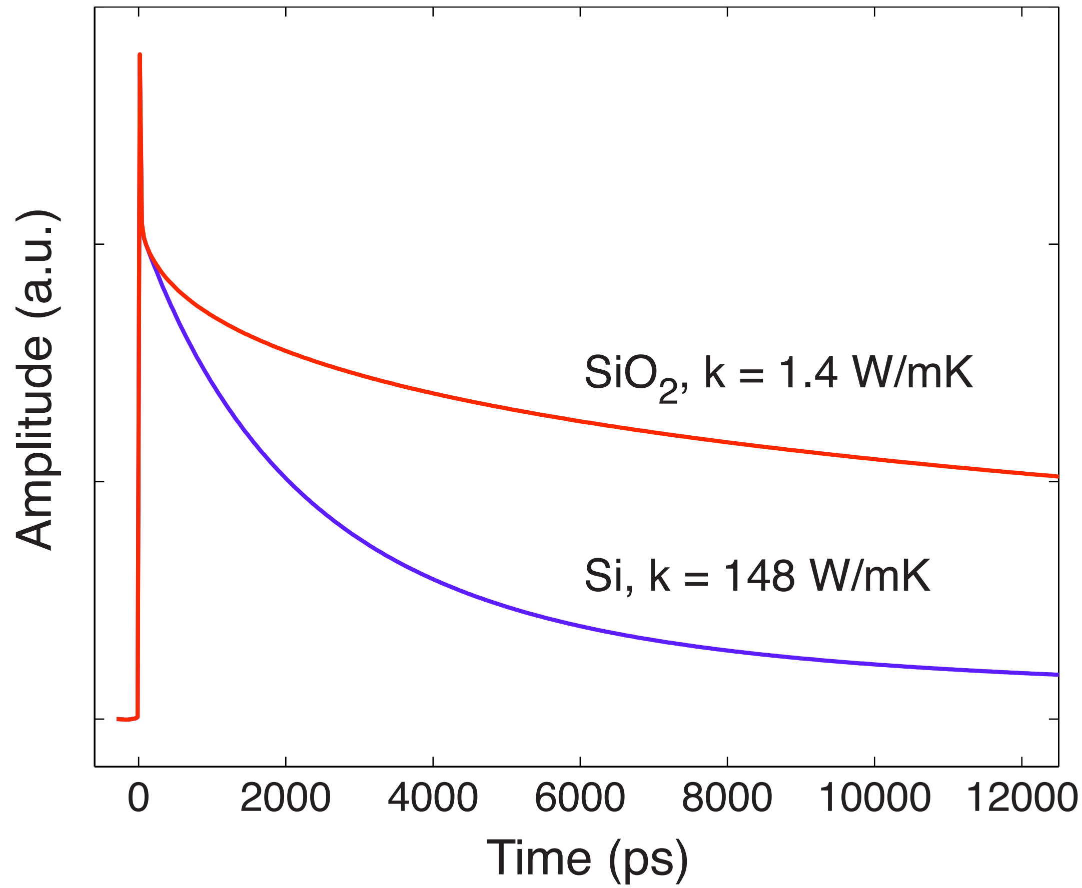
Time domain thermoreflectance



Use both the transient AND periodic response from the short pulsed heating event. Use high rep. rate laser and modulate at some frequency with moderate to high Duty cycle.







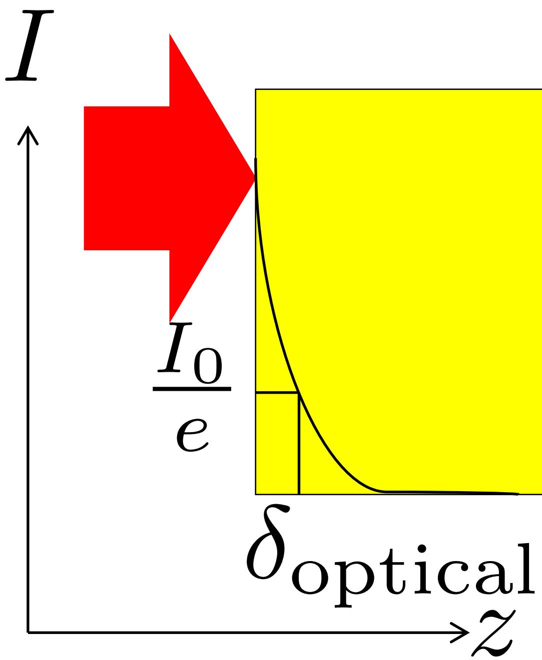
A. J. Schmidt, X. Chen, and G. Chen. Pulse accumulation, radial heat conduction, and anisotropic ther- mal conductivity in pump-probe transient thermoreflectance. Review of Scientific Instruments,

79:114902, 2008.

Time domain thermoreflectance (TDTR)

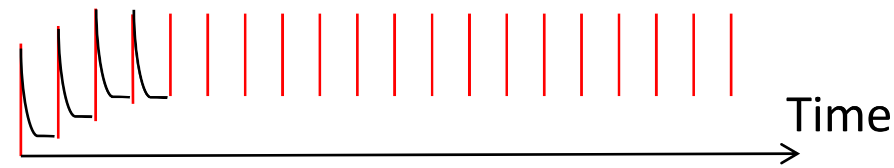
Spatial regimes in TDTR

Coat surfaces with metals to achieve near-surface absorption high *opto-spatial* resolution (i.e., surface localized heat source)

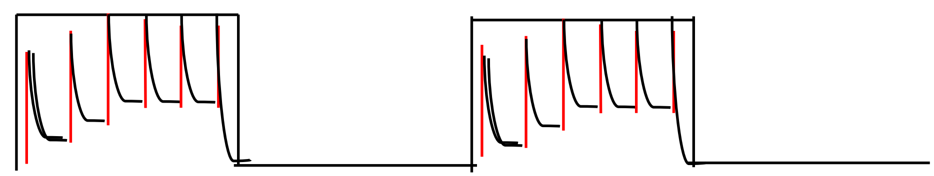


$$\delta_{\text{optical}} = \frac{\lambda}{4\pi k}$$

Modulated heat transfer regime achieves variable *thermo-spatial* resolution (i.e., variable temperature gradient via distance)



Probe TDTR
(accumulates due to 80 MHz rep. rate)



Accumulation leads to *MODULATED* heating event

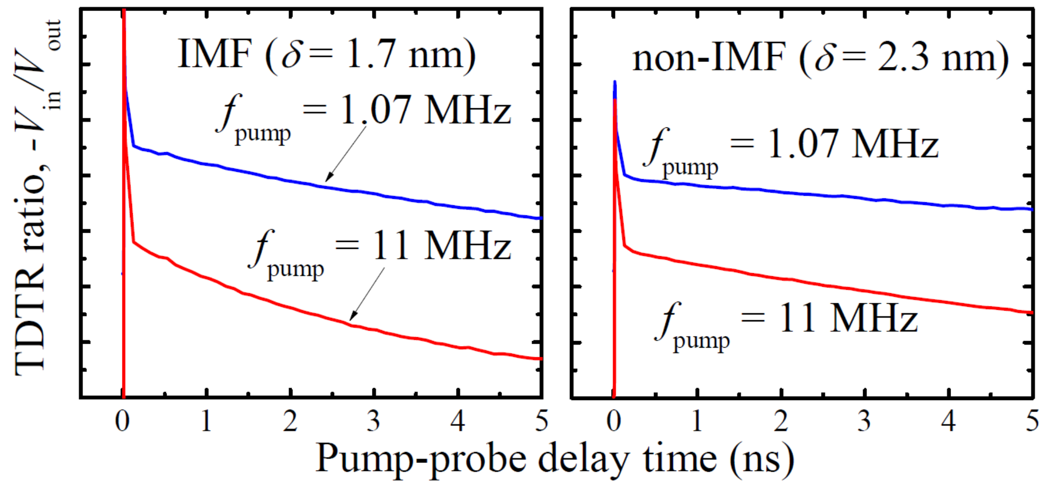
$$\delta_{\text{thermal}} = \sqrt{\frac{\kappa}{\pi f C}} = \sqrt{\frac{2\kappa}{\omega C}}$$

TDTR – depth profiling by enhancing sensitivities to different parameters

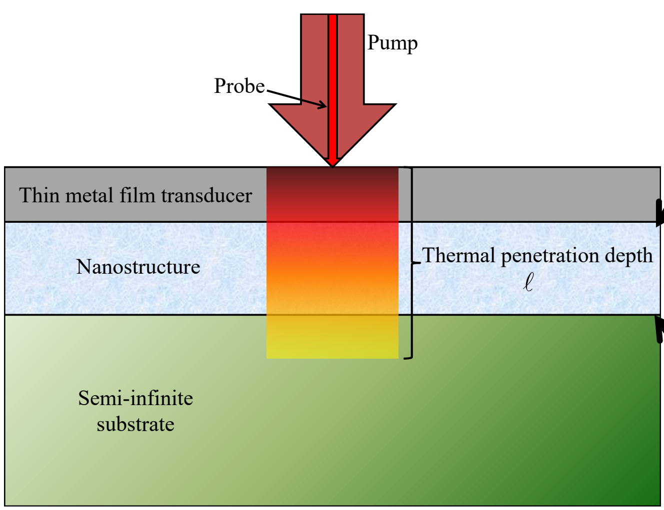
Effect of dislocation density on thermal boundary conductance across GaSb/GaAs interfaces

Patrick E. Hopkins,[1,2,a)] John C. Duda,[1,2] Stephen P. Clark,[3] Christopher P. Hains,[3] Thomas J. Rotter,[3] Leslie M. Phinney,[1] and Ganesh Balakrishnan[3]
[1]*Sandia National Laboratories, Albuquerque, New Mexico 87185, USA*
[2]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[3]*Department of Electrical and Computer Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA*

(Received 15 February 2011; accepted 30 March 2011; published online 22 April 2011)



Increase how "deep" you probe in TDTR by decreasing the frequency



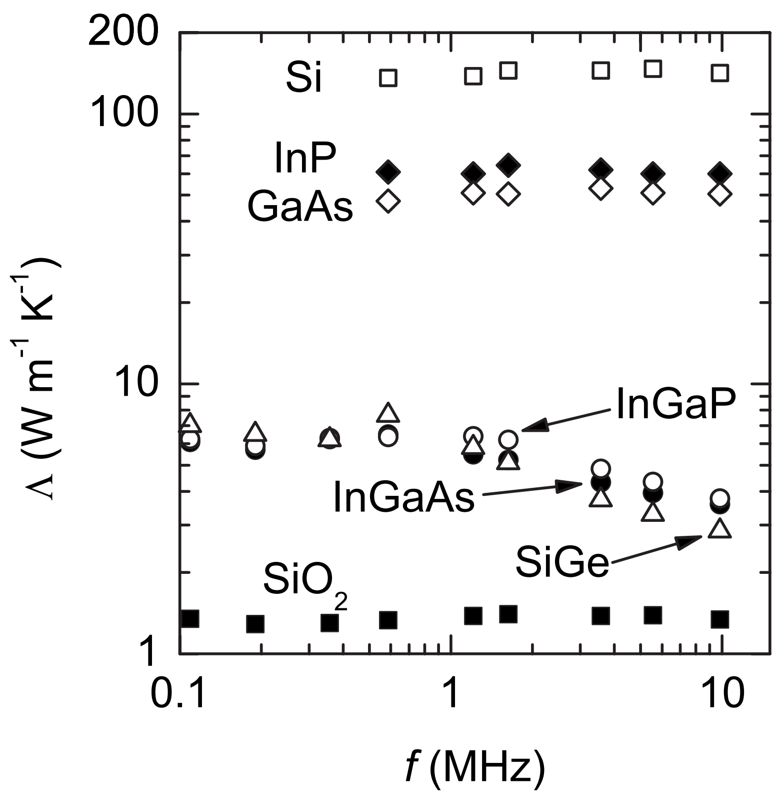
h_K – Al/GaSb (f = 11 MHz)

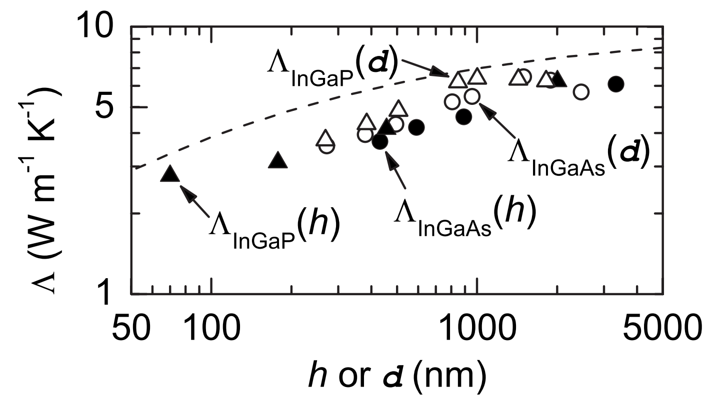
$$\ell \approx \sqrt{\frac{\kappa}{\pi C f_{\mathrm{pump}}}}$$

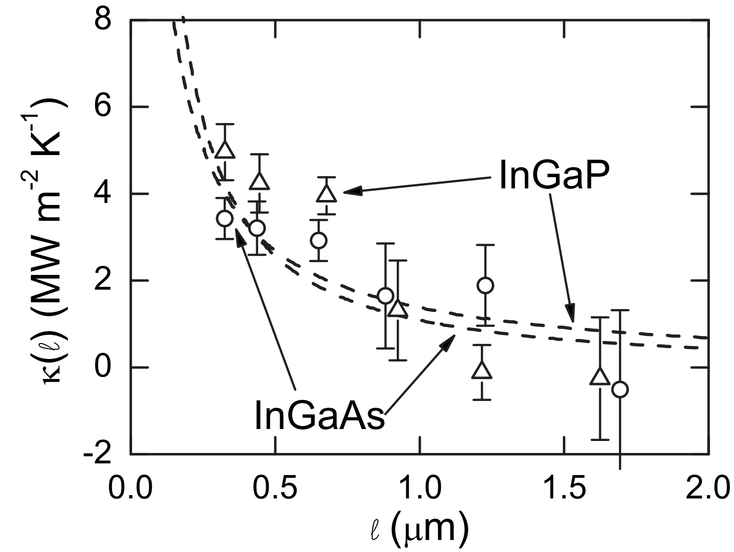
h_K – GaSb/GaAs (f = 1.07 MHz)

TDTR – modulation frequency dependence

$$\delta_{\text{thermal}} = \sqrt{\frac{\kappa}{\pi f C}} = \sqrt{\frac{2\kappa}{\omega C}}$$



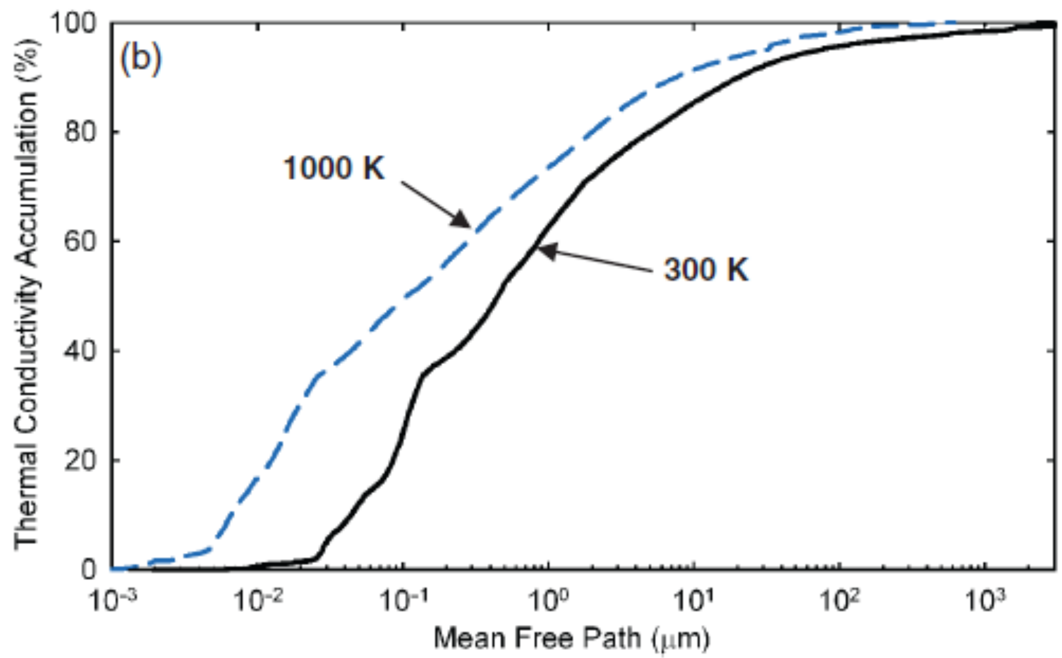




Y. K. Koh and D. G. Cahill. Frequency dependence of the thermal conductivity of semiconductor alloys. Physical Review B, 76:075207, 2007.

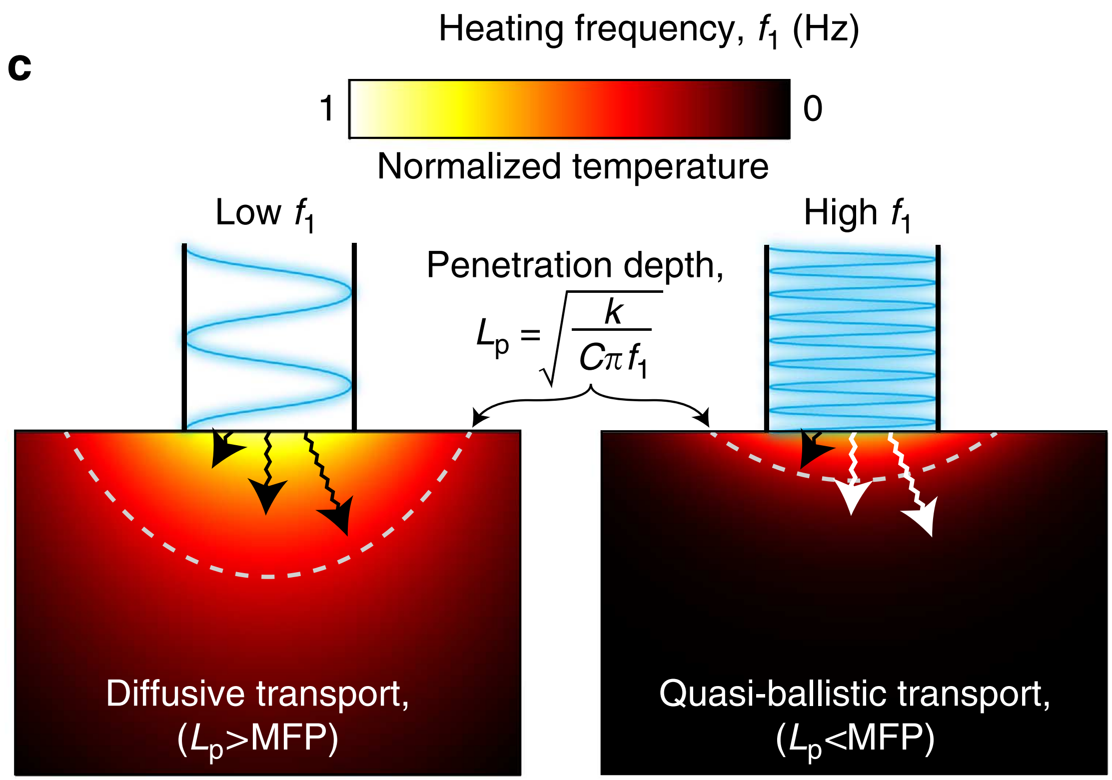
Phonon "escape" from thermal penetration depth

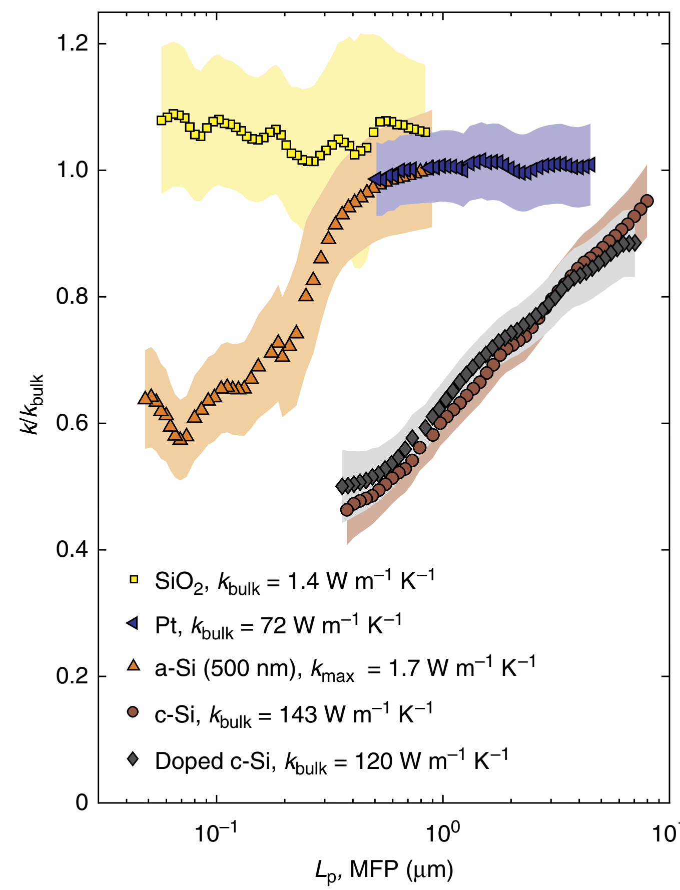
The “thermal conductivity accumulation function”



Henry and Chen, *J. Computational and Theoretical Nanoscience* **5**, 1 (2008).

So why can't we do this with FDTR? We can!

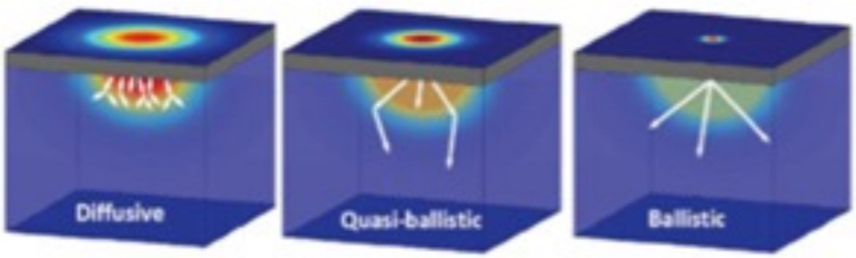


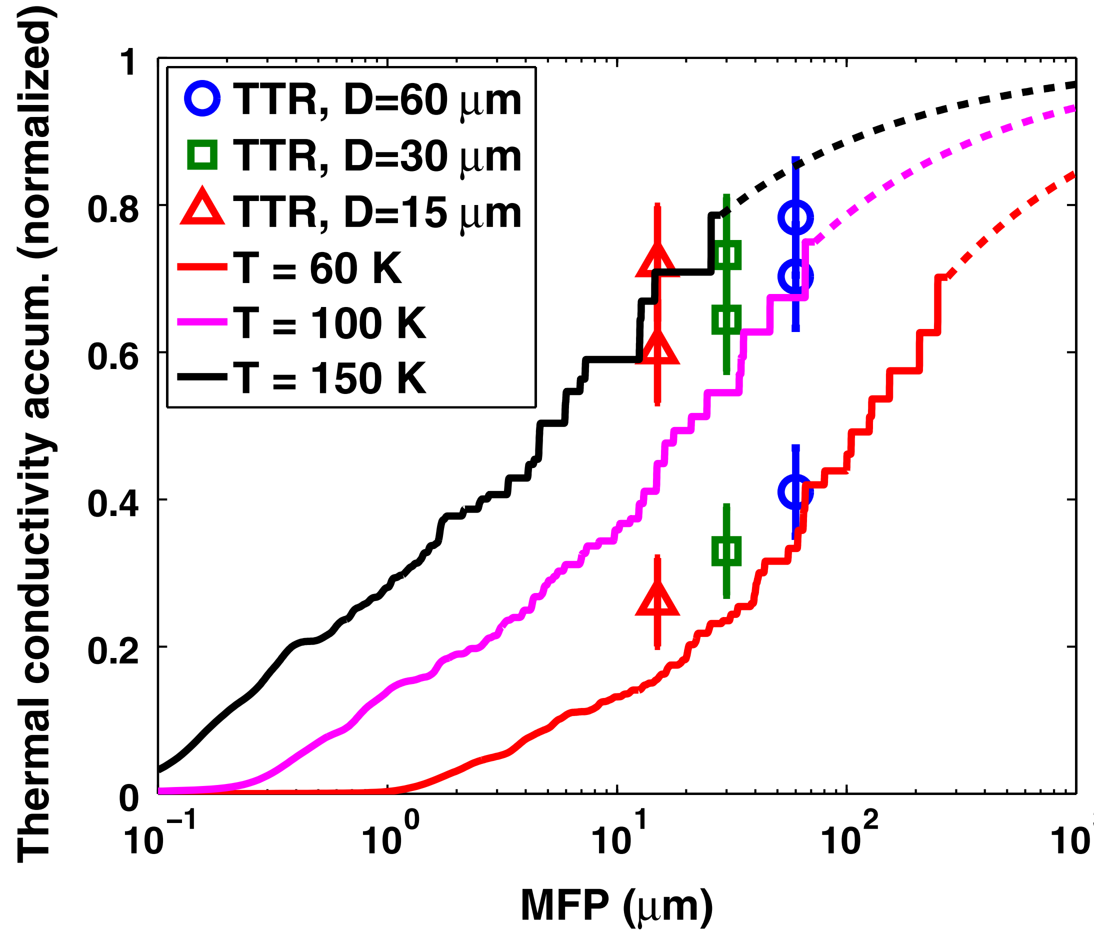


Thermal penetration depth (modulation frequency) varies number of "phonons" sampled in volume (same as Koh and Cahill 2 slides prev)

Regner *et al*, *Nat. Comm.* **4**, 1640 (2013).

So why can't we do this with spot size? We can!



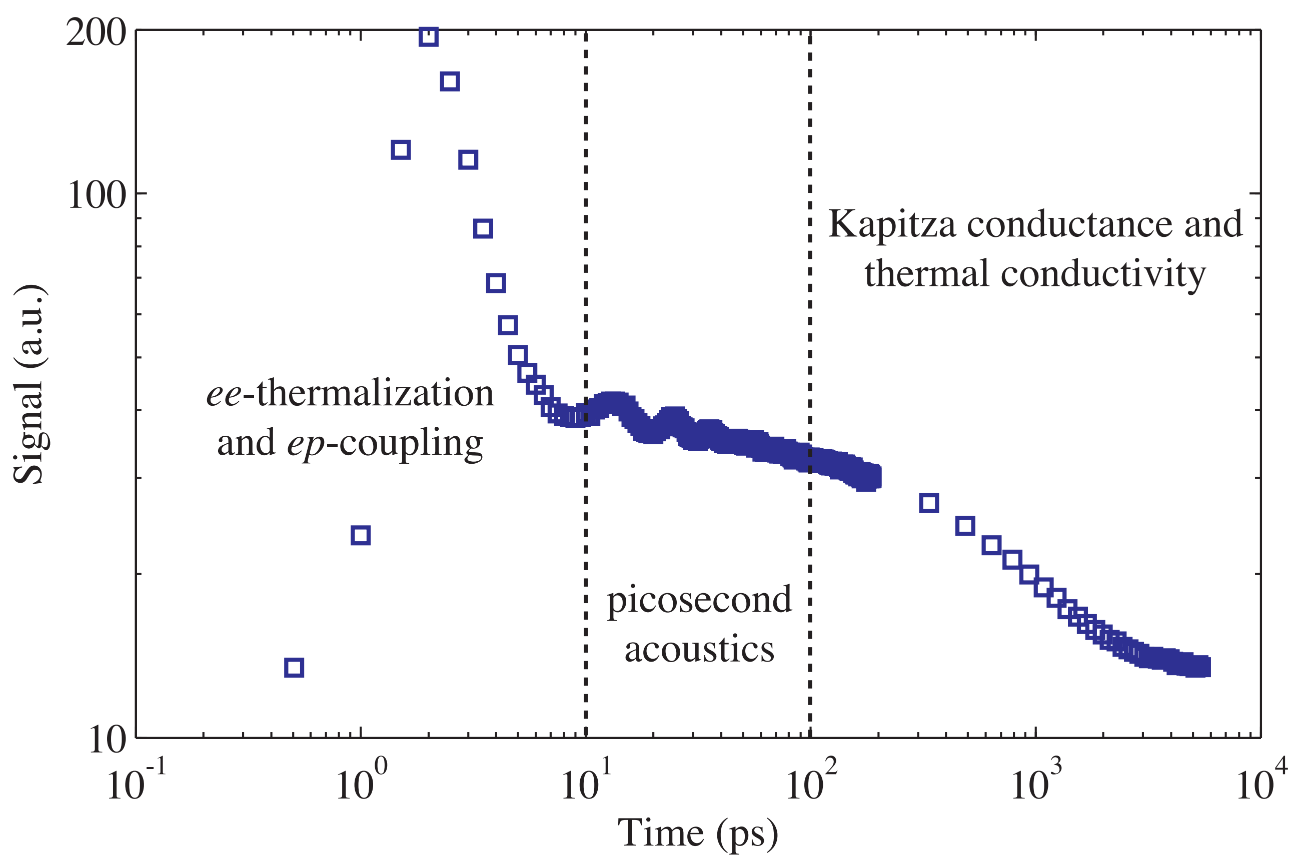


Spot size varies number of "phonons" sampled in volume

Minnich *et al*, *PRL* **107**, 095901 (2011).

Temporal regimes in TDTR

FANTASTIC temporal resolution (limited by pulse width)

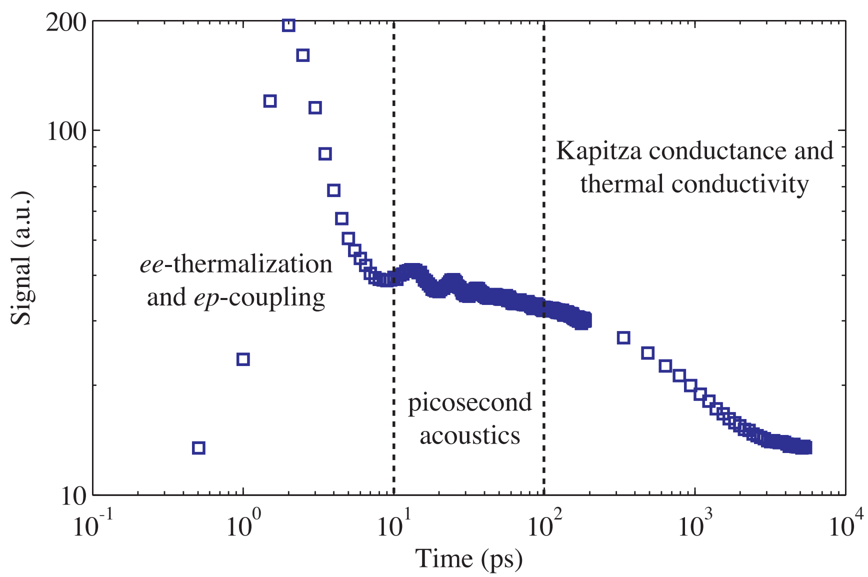


Pulse absorption (~100 fs)
↓
Fermi relaxation and ballistic transport (few hundred fs)
↓
Electron-phonon coupling (a few ps)
↓
Strain propagation in film (10's of ps)
↓
Thermal diffusion (hundreds of ps to ns)

Now let's look at a few specific examples….

Some TDTR References

- Cahill, "Analysis of heat flow in layered structures for time-domain thermoreflectance," *Review of Scientific Instruments* **75**, 5119 (2004)
- Schmidt *et al*., "Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance," *Review of Scientific Instruments* **79**, 114902 (2008)
- Hopkins *et al*., "Criteria for cross-plane dominated thermal transport in multilayer thin film systems during modulated laser heating," *Journal of Heat Transfer* **132**, 081302 (2010)
- Hopkins *et al*., "Measuring the thermal conductivity of porous, transparent SiO2 film with time domain thermoreflectance," *Journal of Heat Transfer* **133**, 061601 (2011)
- Schmidt, "Pump-probe thermoreflectance," *Annual Review of Heat Transfer* **16**, 159 (2013)



So what can we can measure with TDTR?

Time domain thermoreflectance (TDTR)

Electron thermalization and scattering (<10 ps)

Journal of Heat Transfer

APRIL 2011, Vol. 133 / **044505-1**

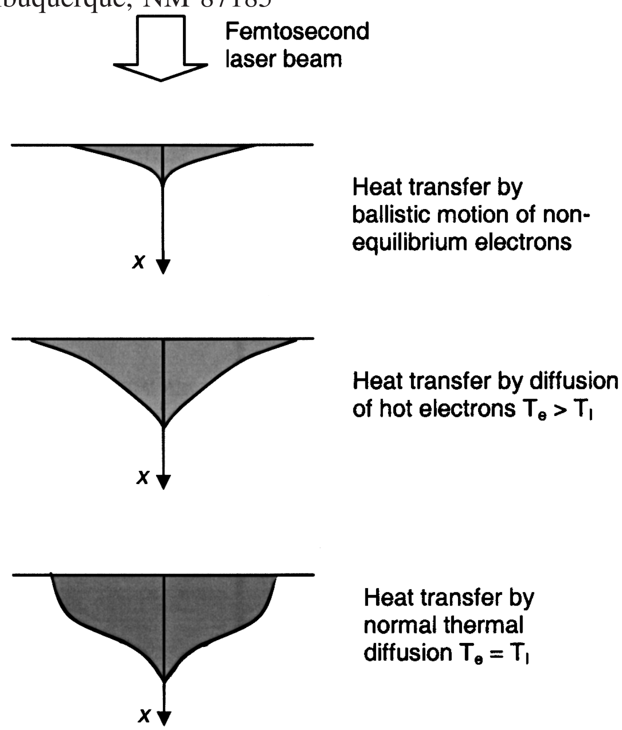
Re-examining Electron-Fermi Relaxation in Gold Films With a Nonlinear Thermoreflectance Model

Patrick E. Hopkins
e-mail: pehopki@sandia.gov

Leslie M. Phinney

Justin R. Serrano

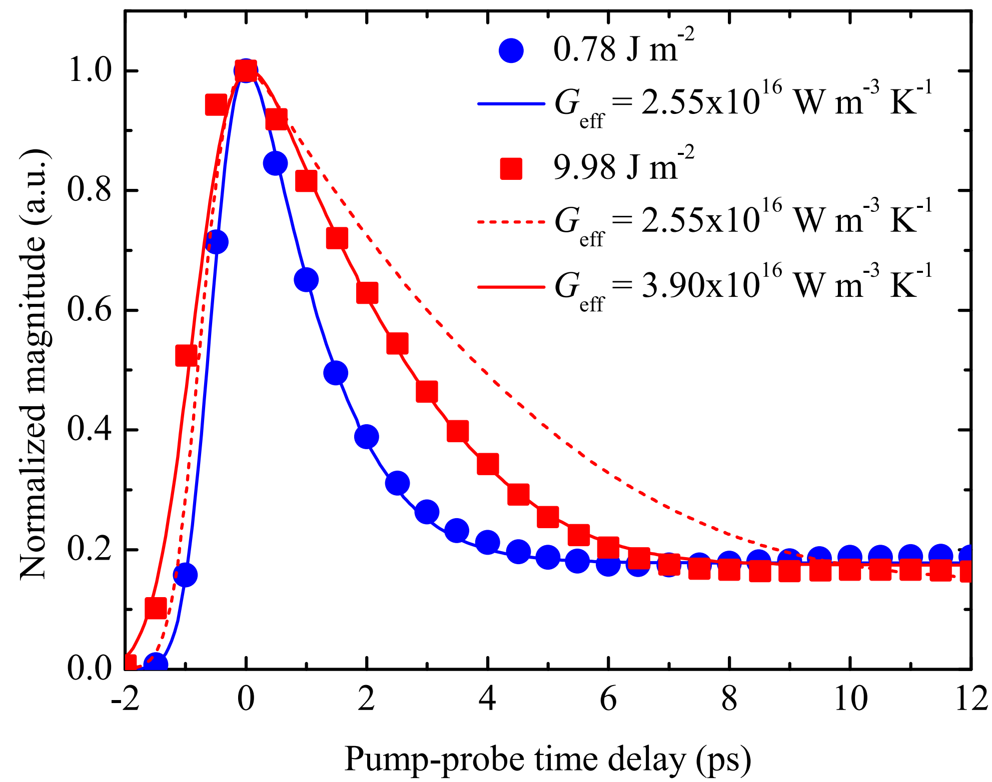
Sandia National Laboratories,
Albuquerque, NM 87185



APPLIED PHYSICS LETTERS **103**, 211910 (2013)

Ultrafast and steady-state laser heating effects on electron relaxation and phonon coupling mechanisms in thin gold films

Patrick E. Hopkins,[1,a)] John C. Duda,[1] Bryan Kaehr,[2,3] Xiao Wang Zhou,[4] C.-Y. Peter Yang,[4] and Reese E. Jones[4]
[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico 87106, USA*
[3]*Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA*
[4]*Sandia National Laboratories, Livermore, California 94550, USA*

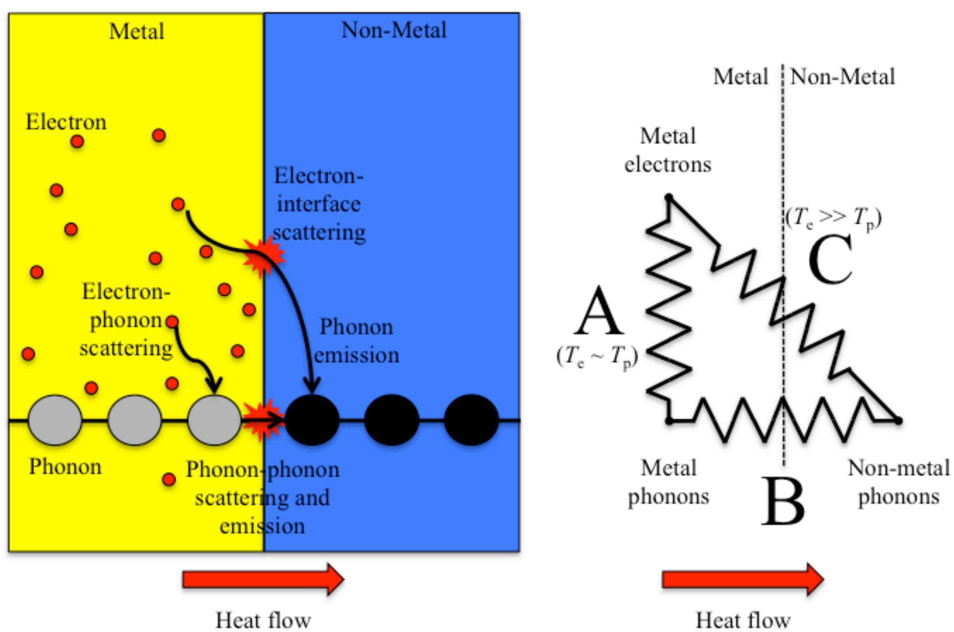


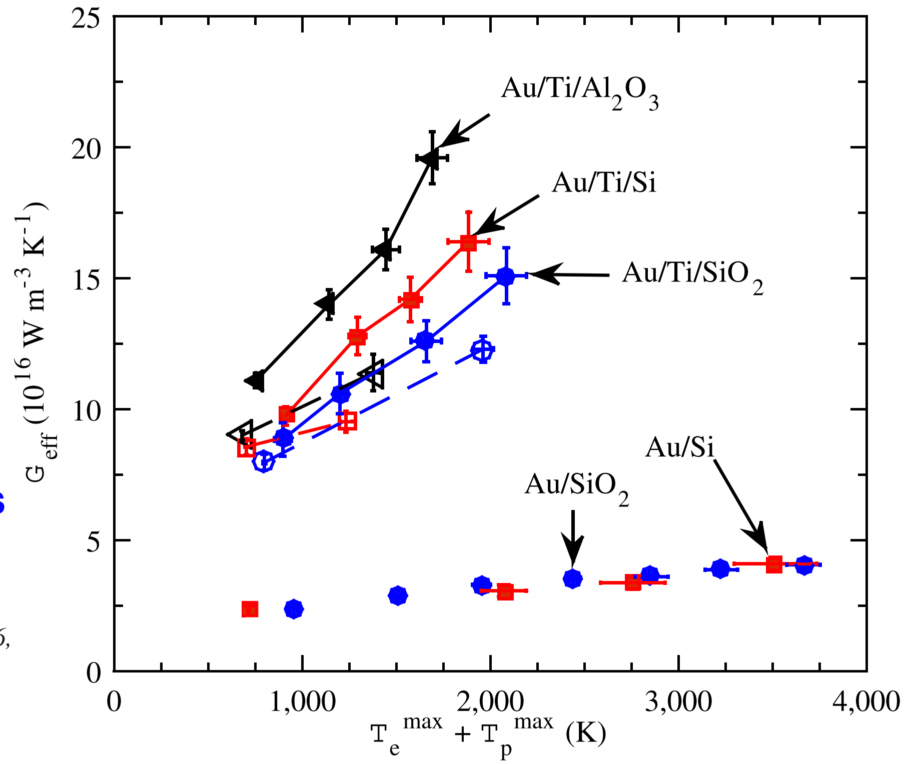
Electron-phonon scattering at interfaces

JOURNAL OF APPLIED PHYSICS **117**, 105105 (2015)

Mechanisms of nonequilibrium electron-phonon coupling and thermal conductance at interfaces

Ashutosh Giri,[1] John T. Gaskins,[1] Brian F. Donovan,[1] Chester Szwejkowski,[1] Ronald J. Warzoha,[2] Mark A. Rodriguez,[3] Jon Ihlefeld,[3] and Patrick E. Hopkins[1,a)]
[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Department of Mechanical Engineering, United States Naval Academy, Annapolis, Maryland 21401, USA*
[3]*Sandia National Laboratories, Albuquerque, New Mexico 87123, USA*



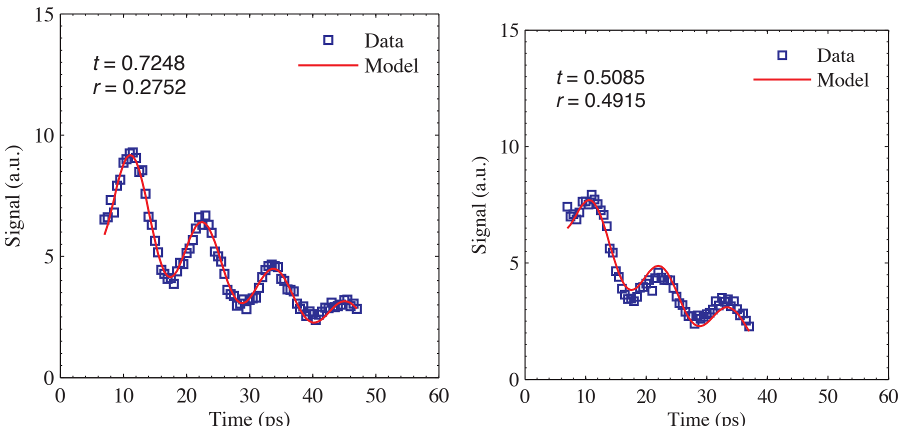


JOURNAL OF APPLIED PHYSICS **105**, 023710 (2009)

Effects of electron scattering at metal-nonmetal interfaces on electron-phonon equilibration in gold films

Patrick E. Hopkins,[a)] Jared L. Kassebaum, and Pamela M. Norris
Department of Mechanical and Aerospace Engineering, University of Virginia, P.O. Box 400746, Charlottesville, Virginia 22904-4746, USA

Acoustic transmission across interfaces (picosecond acoustics/ultrasonics: 10's of ps)



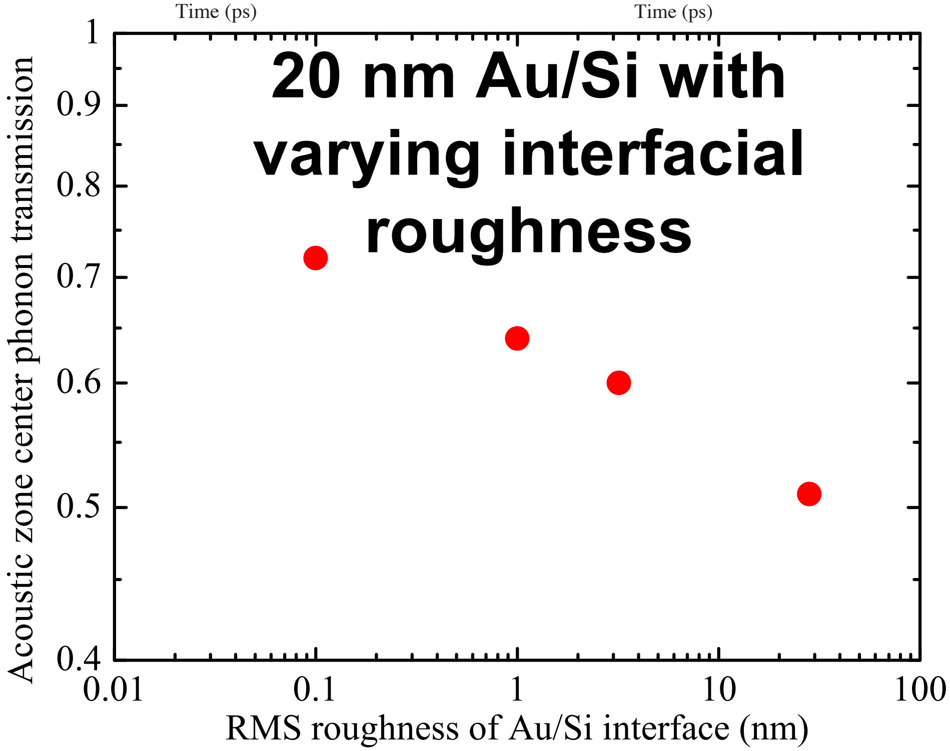
APPLIED PHYSICS LETTERS 102, 081902 (2013)

Influence of interfacial properties on thermal transport at gold:silicon contacts

J. C. Duda,[1,a)] C.-Y. P. Yang,[2] B. M. Foley,[1] R. Cheaito,[1] D. L. Medlin,[2] R. E. Jones,[2] and P. E. Hopkins[1,b)]

[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*

[2]*Sandia National Laboratories, Livermore, California 94550, USA*



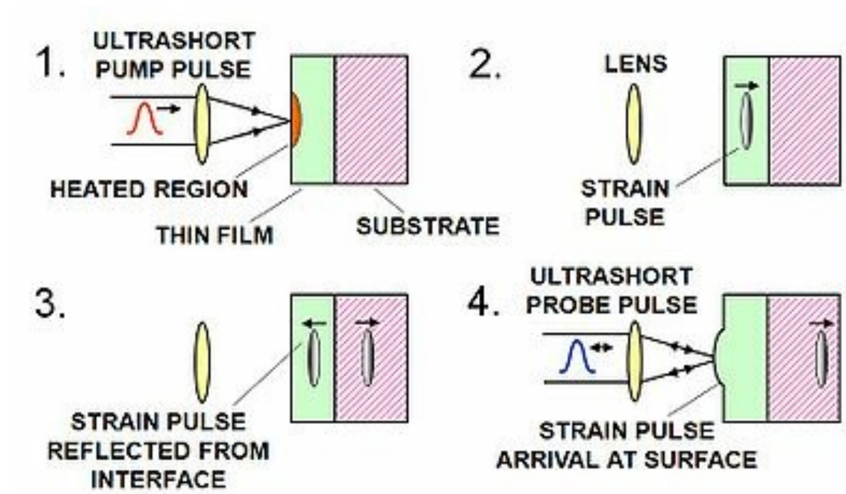


Image from Wikipedia:

http://en.wikipedia.org/wiki/Picosecond_ultrasonics

Time domain thermoreflectance (TDTR)

Thermal conductivity of thin films (100's ps – ns)

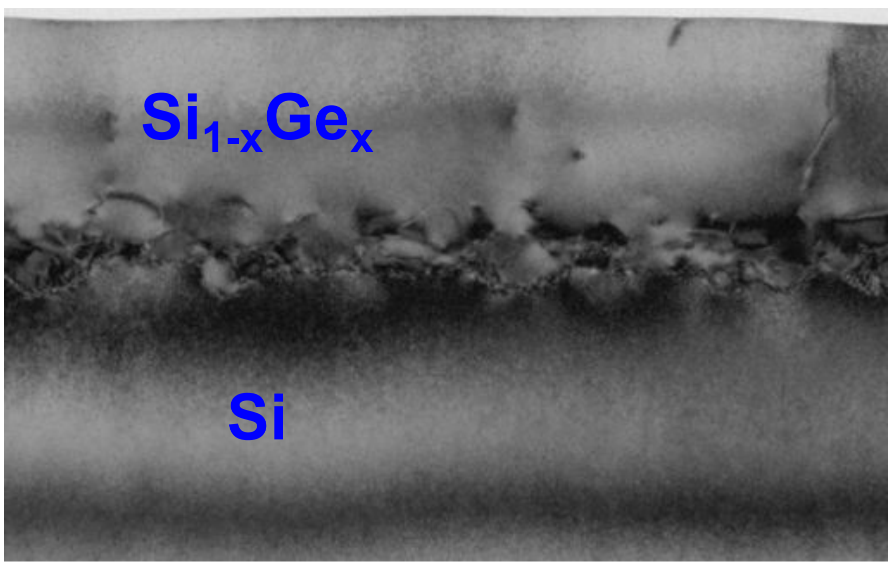
PRL **109,** 195901 (2012) PHYSICAL REVIEW LETTERS week ending 9 NOVEMBER 2012

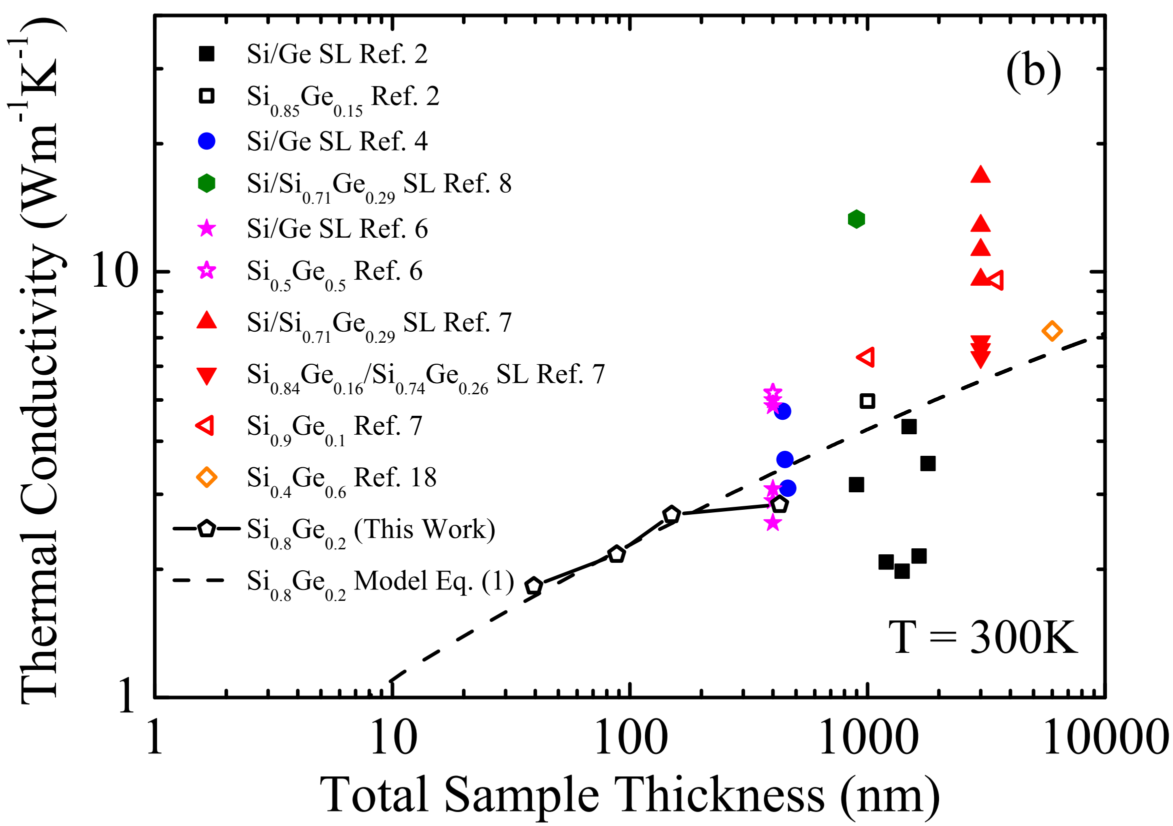
Experimental Investigation of Size Effects on the Thermal Conductivity of Silicon-Germanium Alloy Thin Films

Ramez Cheaito,[1] John C. Duda,[1,2] Thomas E. Beechem,[2] Khalid Hattar,[2] Jon F. Ihlefeld,[2] Douglas L. Medlin,[3] Mark A. Rodriguez,[2] Michael J. Campion,[2,4] Edward S. Piekos,[2] and Patrick E. Hopkins[1,*]

[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Sandia National Laboratories, Albuquerque, New Mexico 87123, USA*
[3]*Sandia National Laboratories, Livermore, California 94550, USA*
[4]*Massachusetts Institute of Technology, Department of Material Science and Engineering, Cambridge, Massachusetts 02139, USA*

(Received 1 June 2012; published 8 November 2012)





Time domain thermoreflectance (TDTR)

Thermal conductivity of superlattices (100's ps – ns)

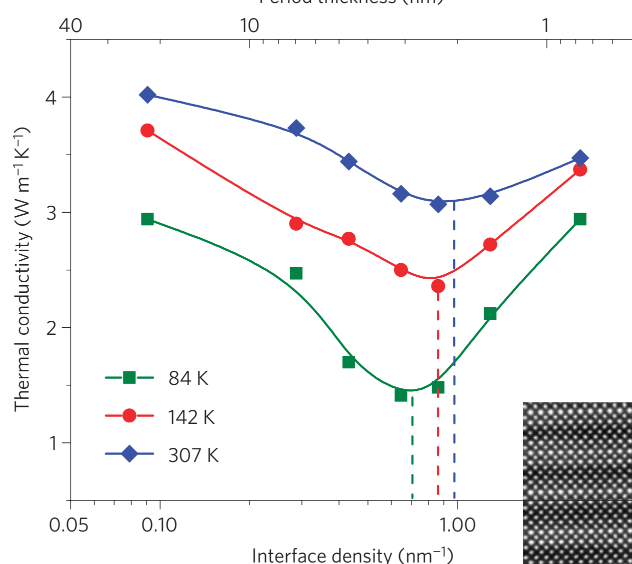
Coherent Phonon Heat Conduction in Superlattices

Maria N. Luckyanova,[1]* Jivtesh Garg,[1]* Keivan Esfarjani,[1] Adam Jandl,[2] Mayank T. Bulsara,[2] Aaron J. Schmidt,[3] Austin J. Minnich,[4] Shuo Chen,[5] Mildred S. Dresselhaus,[6,7] Zhifeng Ren,[5] Eugene A. Fitzgerald,[2] Gang Chen[1]†

LETTERS
PUBLISHED ONLINE: 8 DECEMBER 2013 | DOI: 10.1038/NMAT3826
nature materials

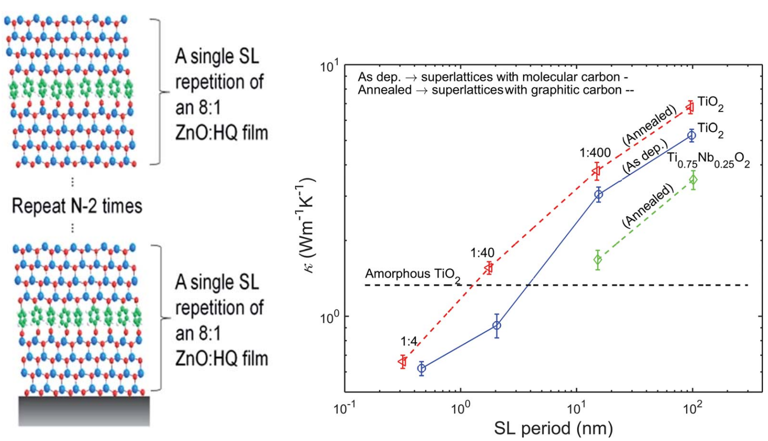
Crossover from incoherent to coherent phonon scattering in epitaxial oxide superlattices

Jayakanth Ravichandran[1,2†‡], Ajay K. Yadav[2,3‡], Ramez Cheaito[4‡], Pim B. Rossen[3], Arsen Soukiassian[5], S. J. Suresha[2], John C. Duda[4], Brian M. Foley[4], Che-Hui Lee[5], Ye Zhu[6], Arthur W. Lichtenberger[7], Joel E. Moore[2,8], David A. Muller[6,9], Darrell G. Schlom[5,9], Patrick E. Hopkins[4], Arun Majumdar[10], Ramamoorthy Ramesh[1,2,3,8,11]★ and Mark A. Zurbuchen[12,13,14]★

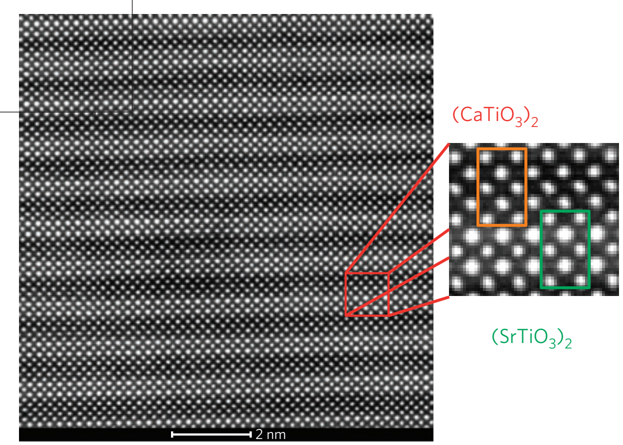


Ultra-low thermal conductivity in TiO_2:C superlattices

Janne-Petteri Niemelä,[a] Ashutosh Giri,[b] Patrick E. Hopkins[b] and Maarit Karppinen*[a]



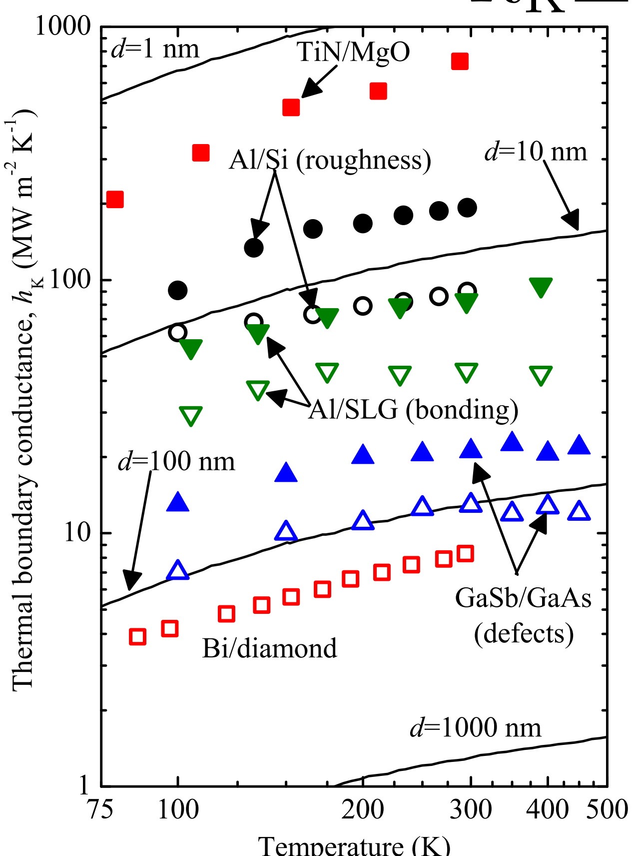
Cite this: *J. Mater. Chem. A*, 2015, **3**, 11527



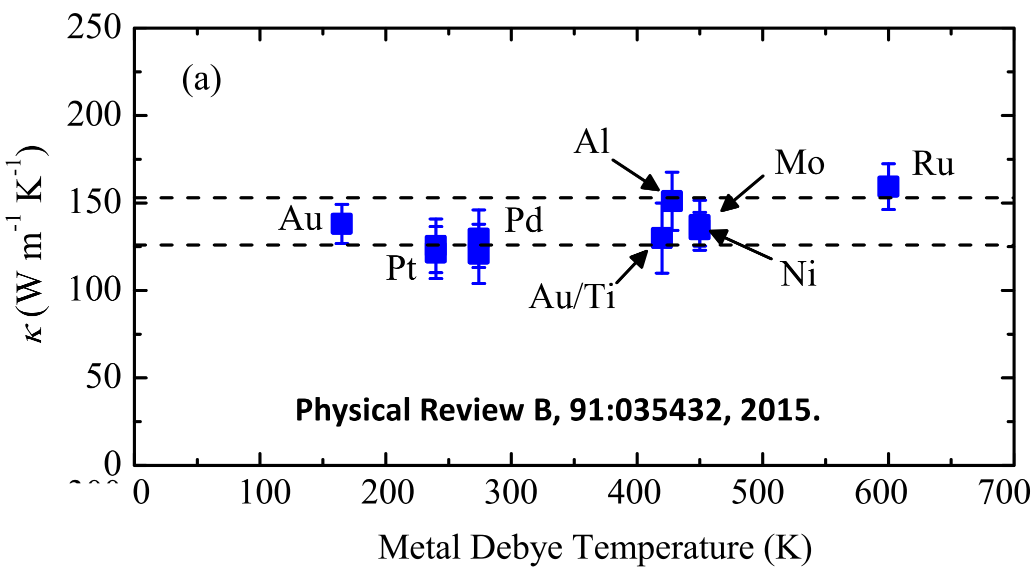
Time domain thermoreflectance (TDTR)

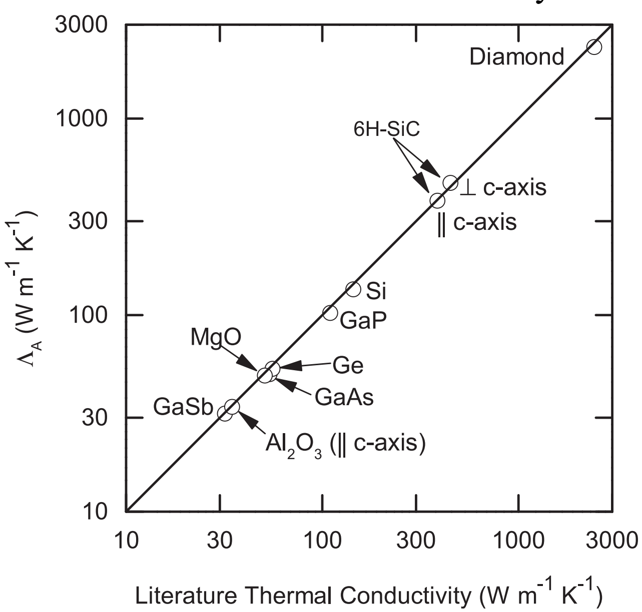
Thermal boundary conductance and bulk thermal conductivity (100's ps – ns)

$$q_{\mathrm{int}} = h_{\mathrm{K}}\Delta T = \frac{1}{R_{\mathrm{K}}\Delta T}$$



Hopkins, "Thermal transport across solid interfaces with nanoscale imperfections: Effects of roughness, disorder, dislocations and bonding on thermal boundary conductance," ***ISRN Mechanical Engineering*** **2013, 682586 (2013)**

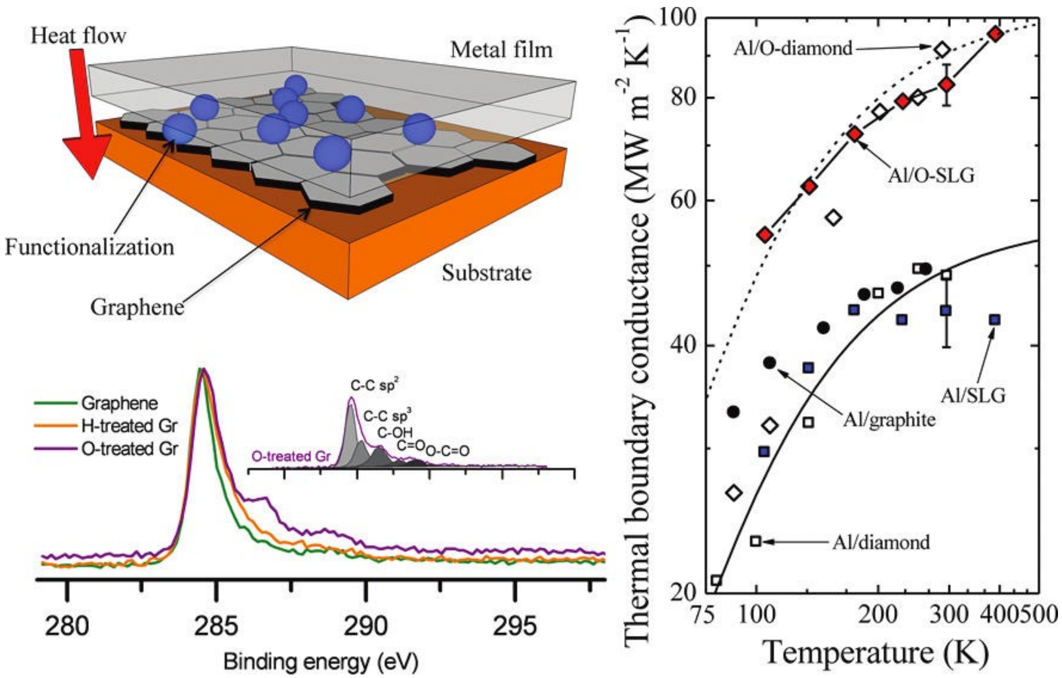


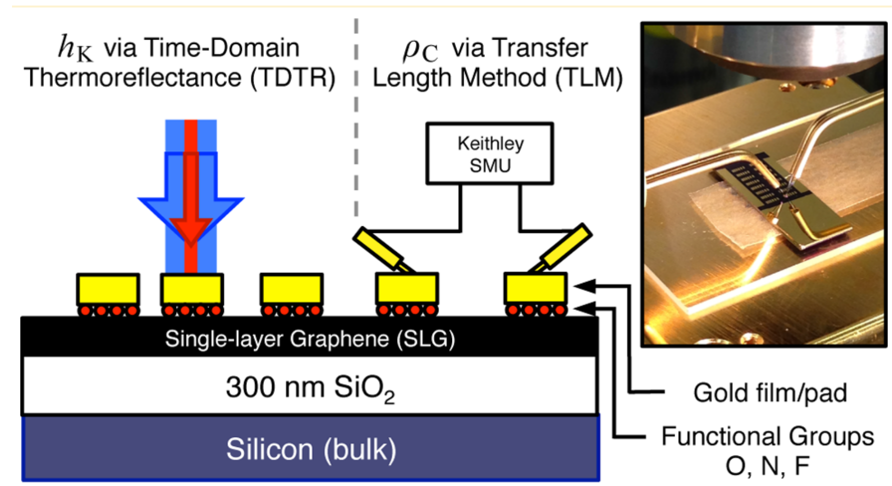


Applied Physics Letters, 107(20):203112, 2015

Time domain thermoreflectance (TDTR)

2D materials (e.g., graphene and their contacts)

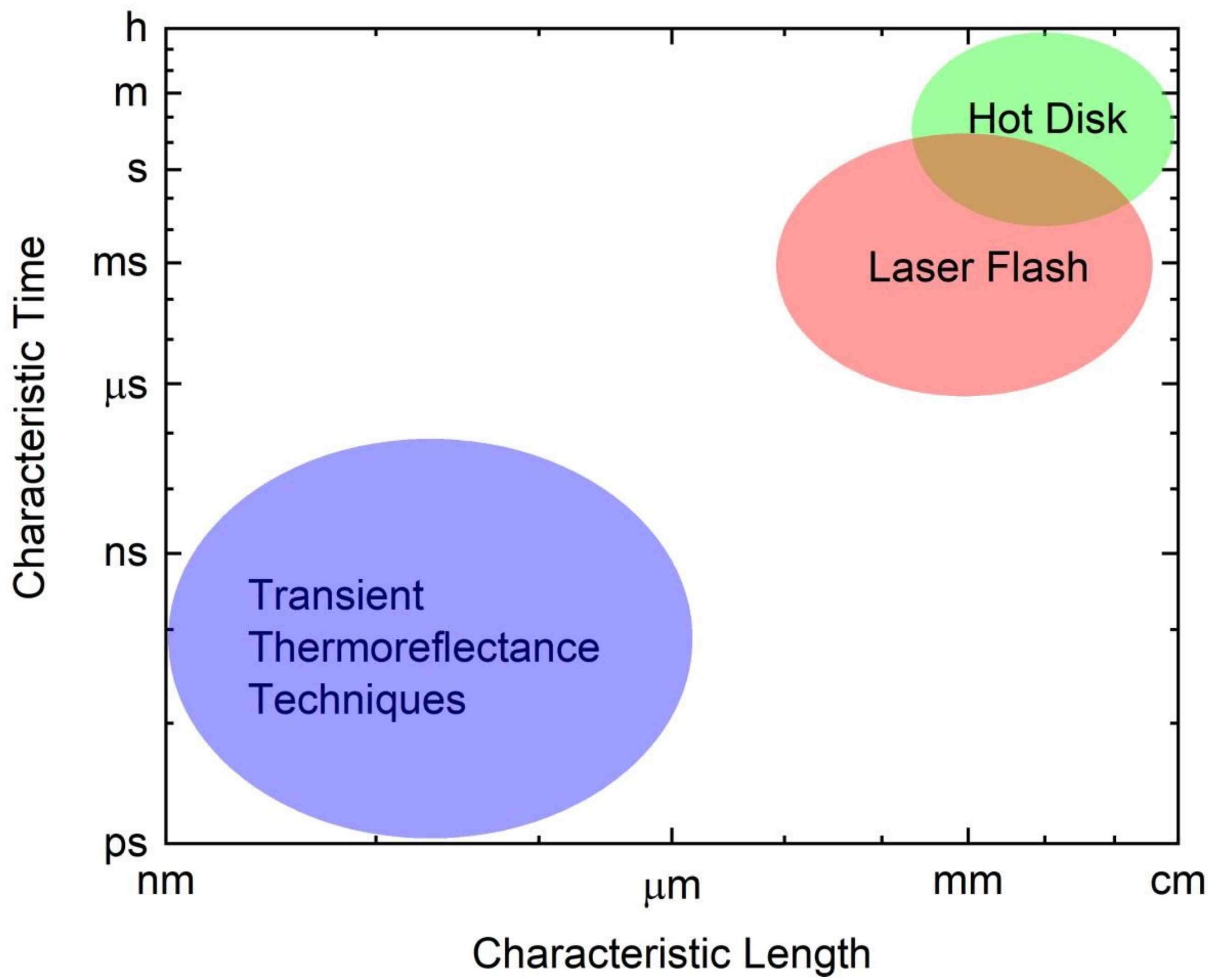




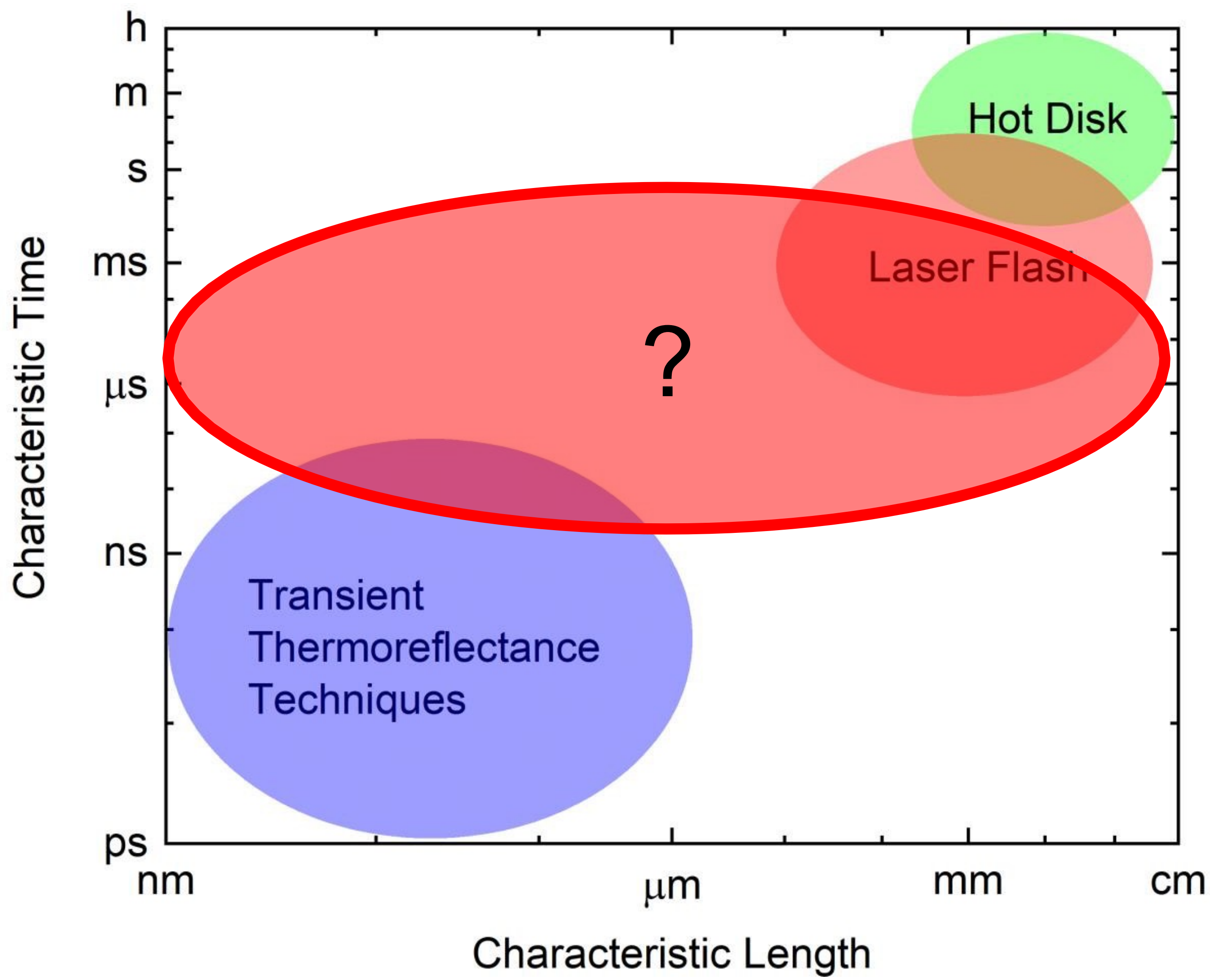
P. E. Hopkins, M. Baraket, E. V. Barnat, T. E. Beechem, S. P. Kearney, J. C. Duda, J. T. Robin- son, and S. G. Walton. Manipulating thermal conductance at metal-graphene contacts via chemical functionalization. **Nano Letters, 12:590–595, 2012.**

B. M. Foley, S. C. Hern ́andez, J. C. Duda, J. T. Robinson, S. G. Walton, and P. E. Hopkins. Modifying surface energy of graphene via plasma-based chemical functionalization to tune thermal and electrical transport at metal interfaces. **Nano Letters, 15(8):4876–4882, 2015.**

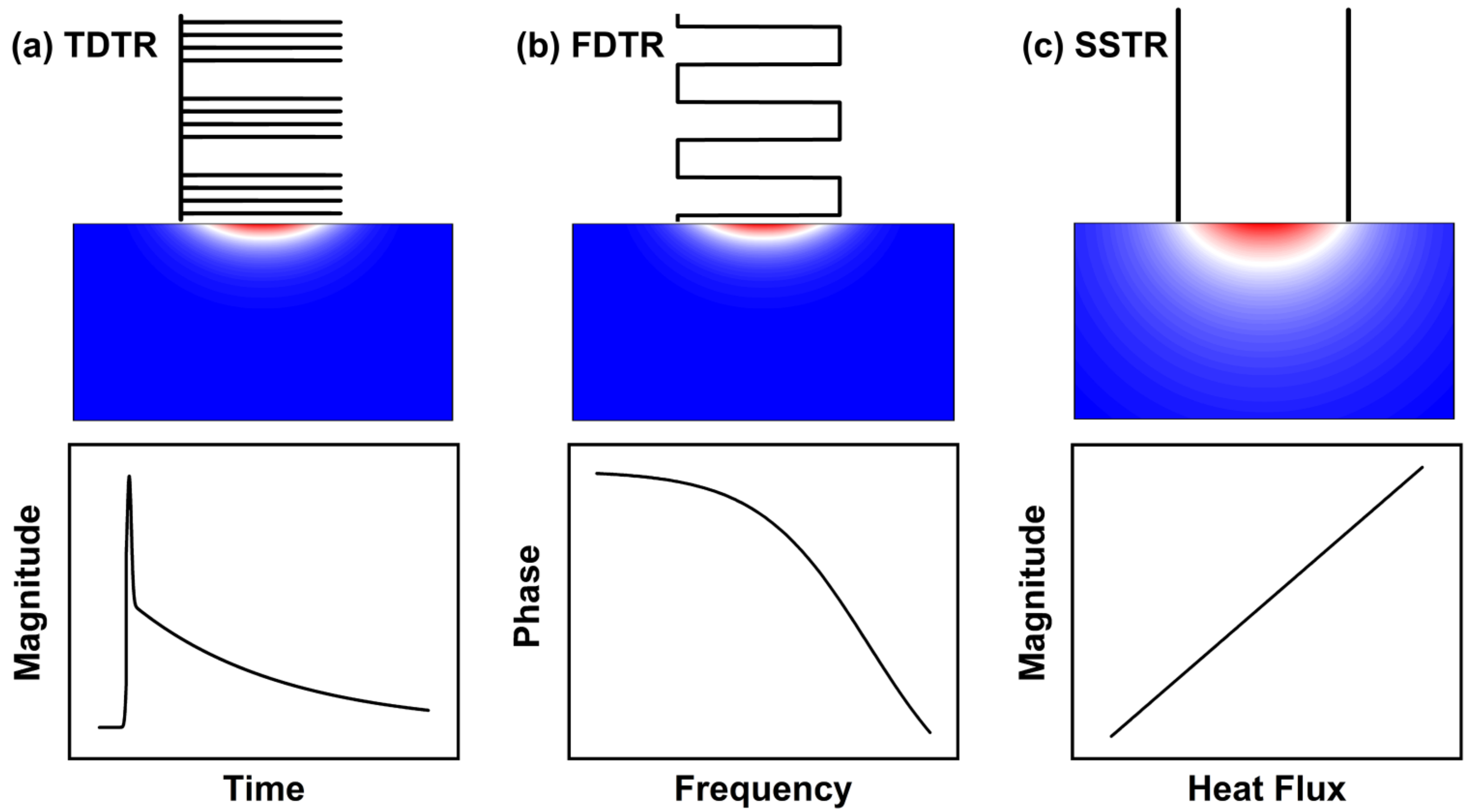
Regimes of thermal conductivity "tools"



Regimes of thermal conductivity “tools”



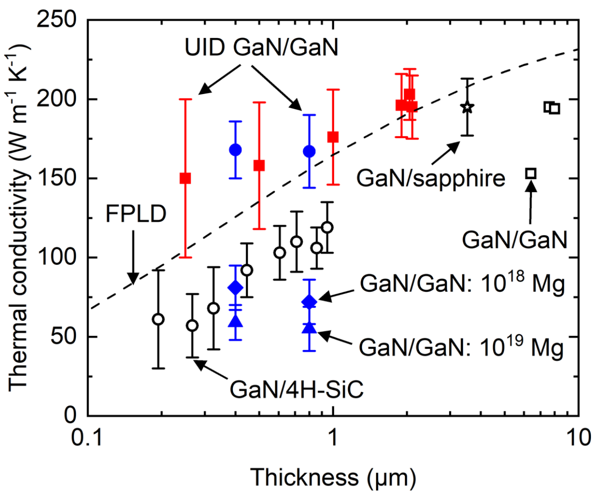
The “flavors” of thermoreflectance



J. Appl. Phys. **126**, 150092

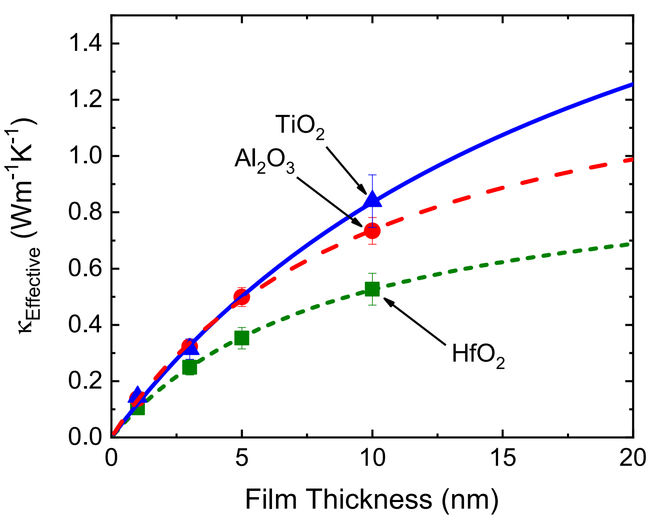
Reviewing the "power" of pump-probe thermoreflectance

Bulk materials and coatings



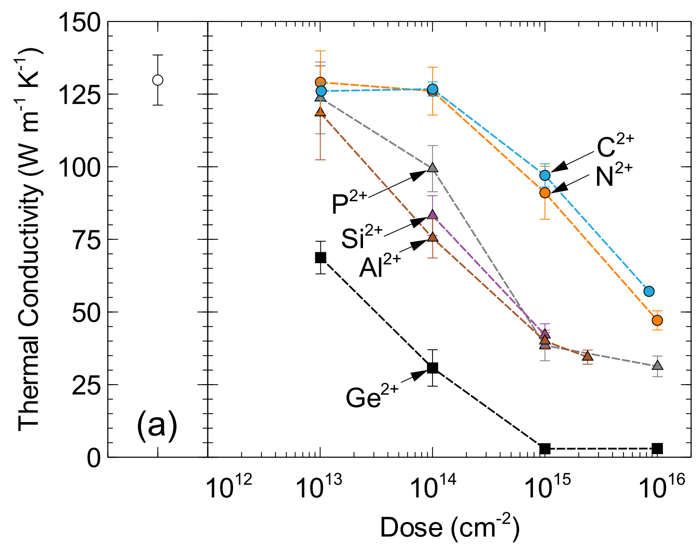
Phys. Rev. Mat. **5**, 104604

Thin films
(even *really really* thin films)



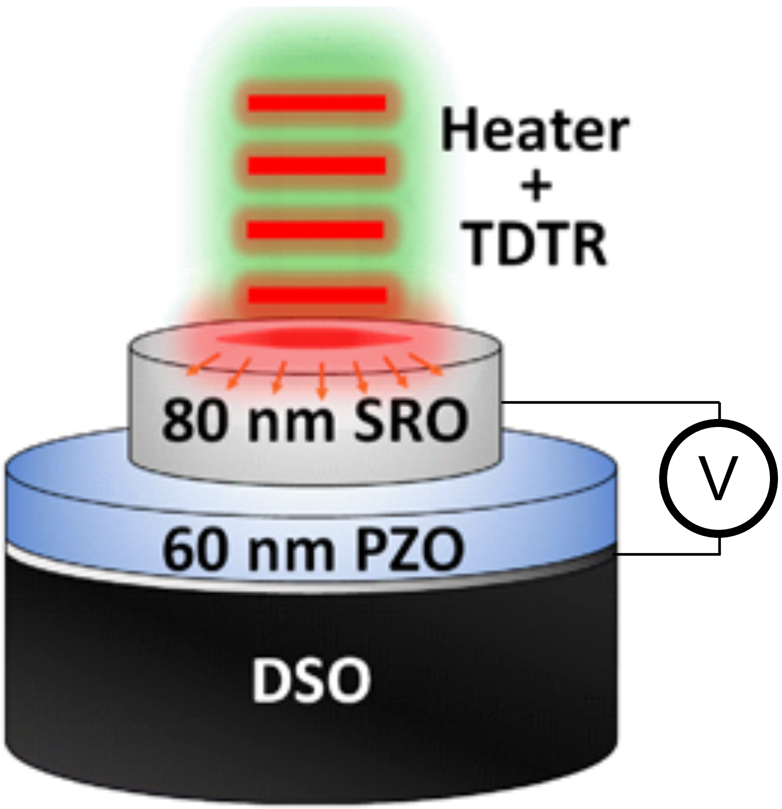
APL Materials **6**, 058302

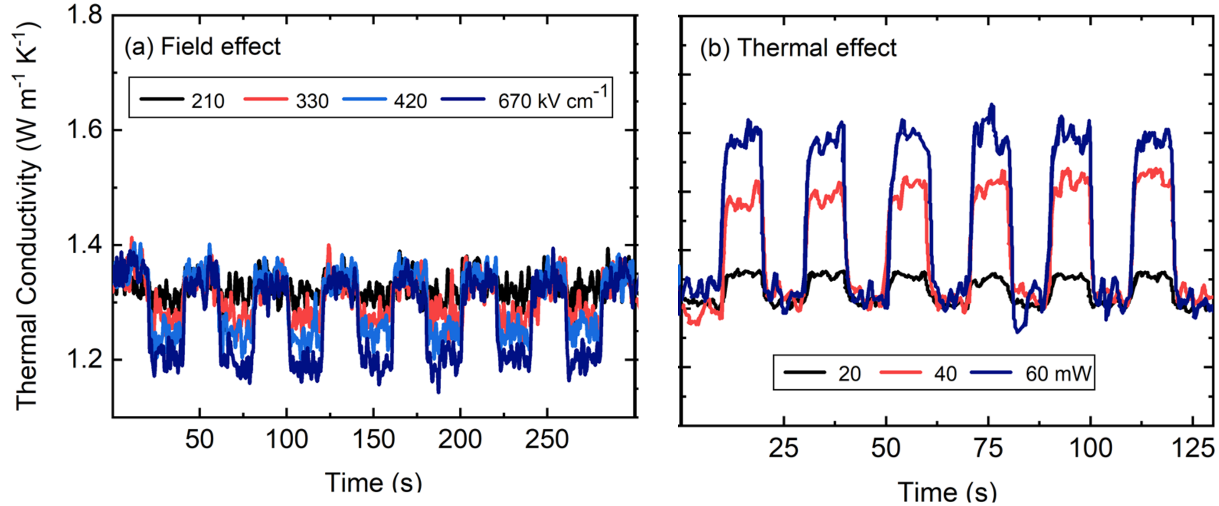
Evaluation of irradiated materials and defects



Phys. Rev. B **104**, 134306

In operando: thermal conductivity under bias



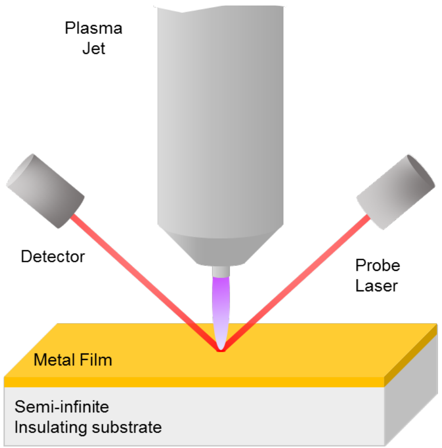


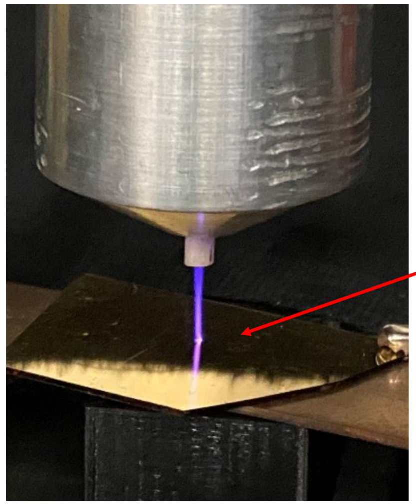
nature COMMUNICATIONS
ARTICLE
https://doi.org/10.1038/s41467-022-29023-y OPEN

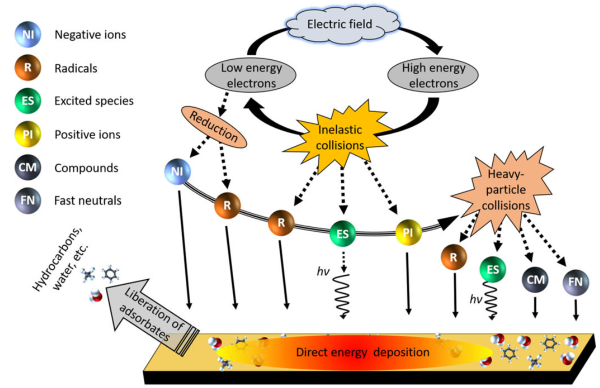
Observation of solid-state bidirectional thermal conductivity switching in antiferroelectric lead zirconate ($PbZrO_3$)

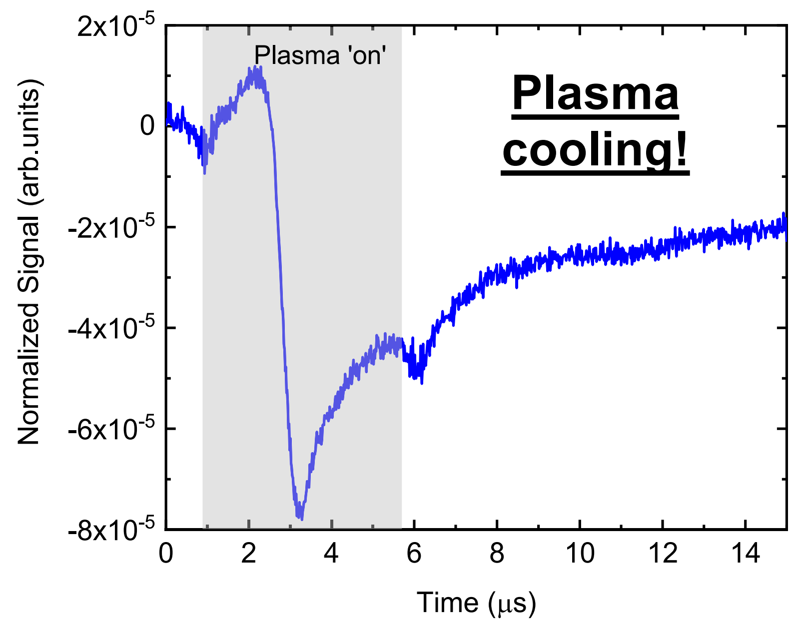
Kiumars Aryana[1], John A. Tomko[1], Ran Gao[2], Eric R. Hoglund[3], Takanori Mimura[3], Sara Makarem[3], Alejandro Salanova[3], Md Shafkat Bin Hoque[1], Thomas W. Pfeifer[1], David H. Olson[1], Jeffrey L. Braun[1], Joyeeta Nag[4], John C. Read[4], James M. Howe[3], Elizabeth J. Opila[1,3], Lane W. Martin[2,5], Jon F. Ihlefeld[3,6]✉ & Patrick E. Hopkins[1,3,7]✉

In situ: thermal conductivity during external stimuli









nature COMMUNICATIONS

ARTICLE

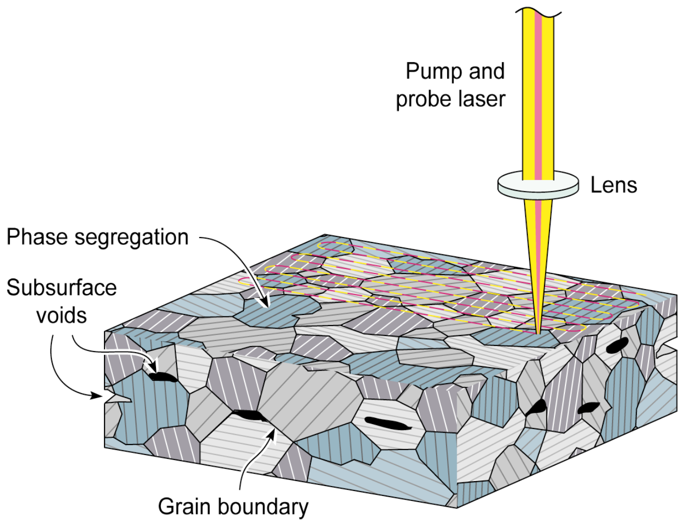
Check for updates

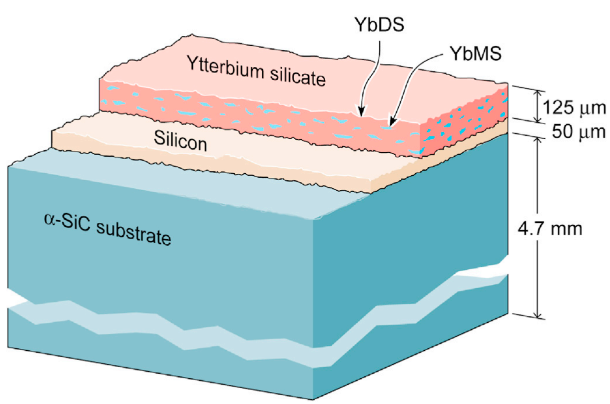
https://doi.org/10.1038/s41467-022-30170-5 OPEN

Plasma-induced surface cooling

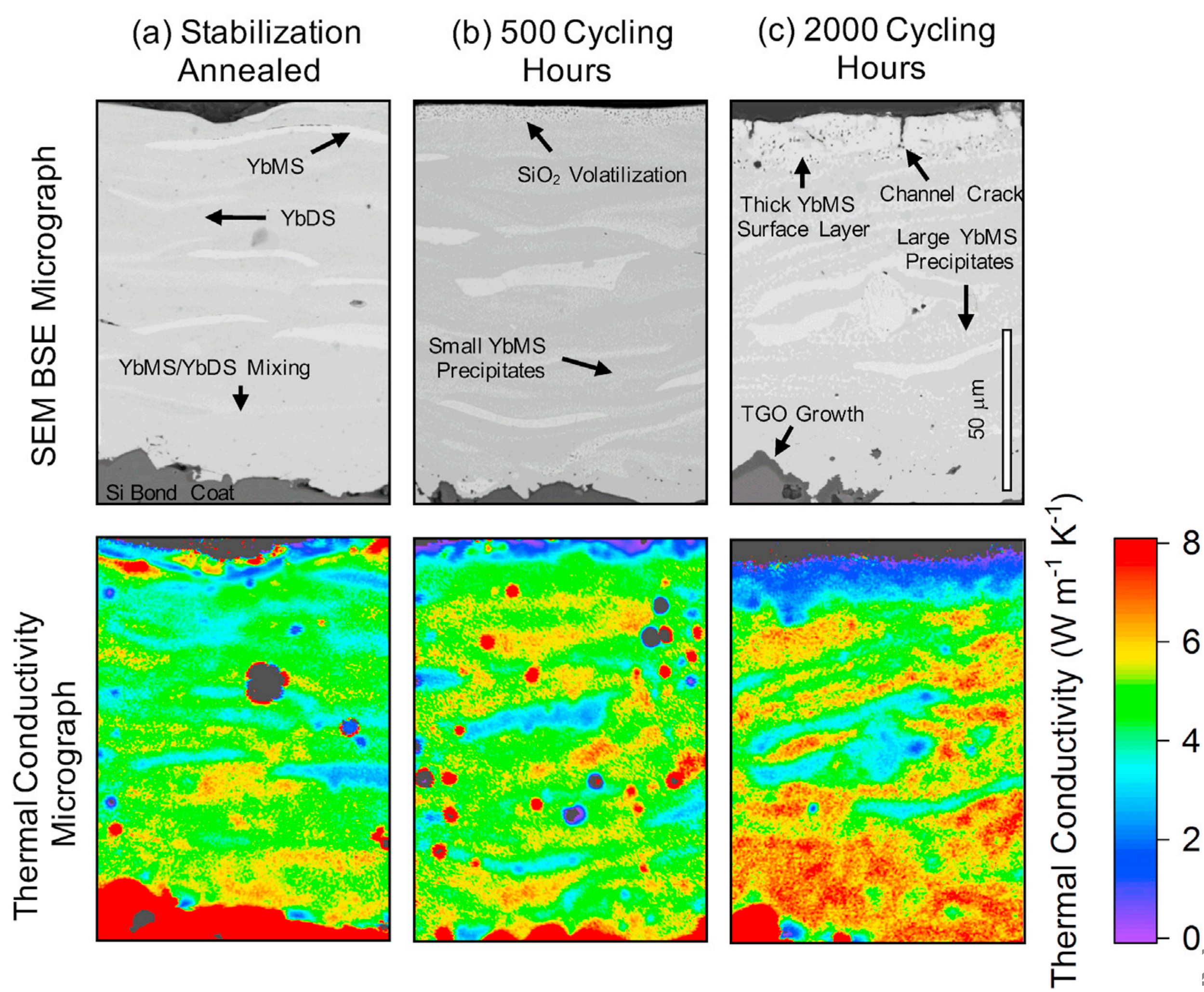
John A. Tomko [1✉], Michael J. Johnson[2], David R. Boris[3], Tzvetelina B. Petrova[3], Scott G. Walton [3✉] & Patrick E. Hopkins [1,4,5✉]

Thermal conductivity mapping

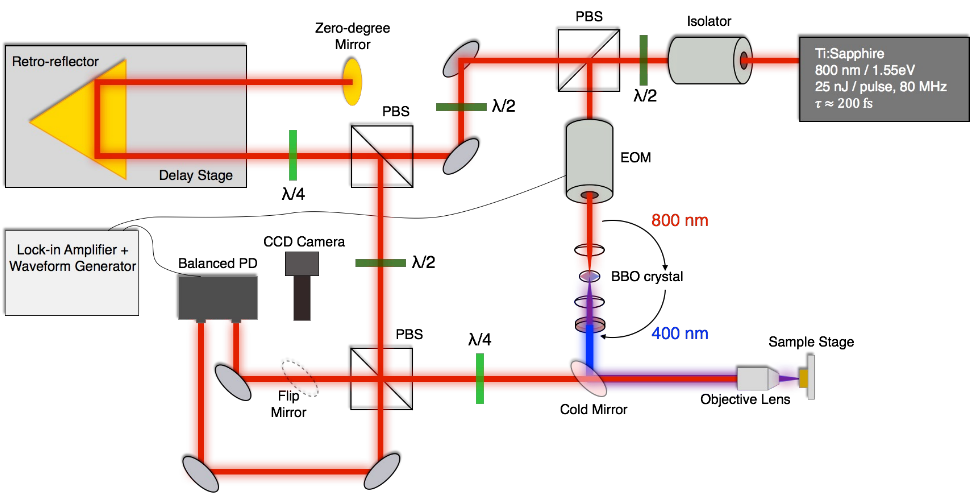


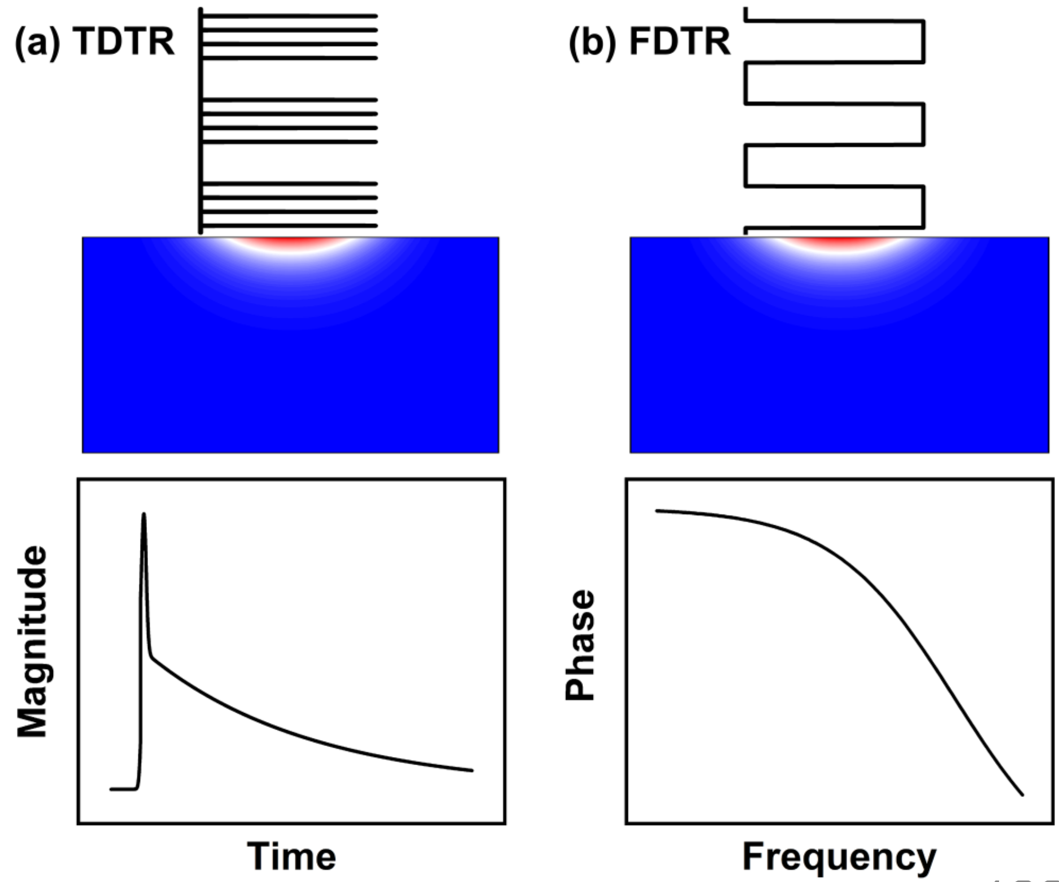


Mat. Today Phys. **17**, 100304



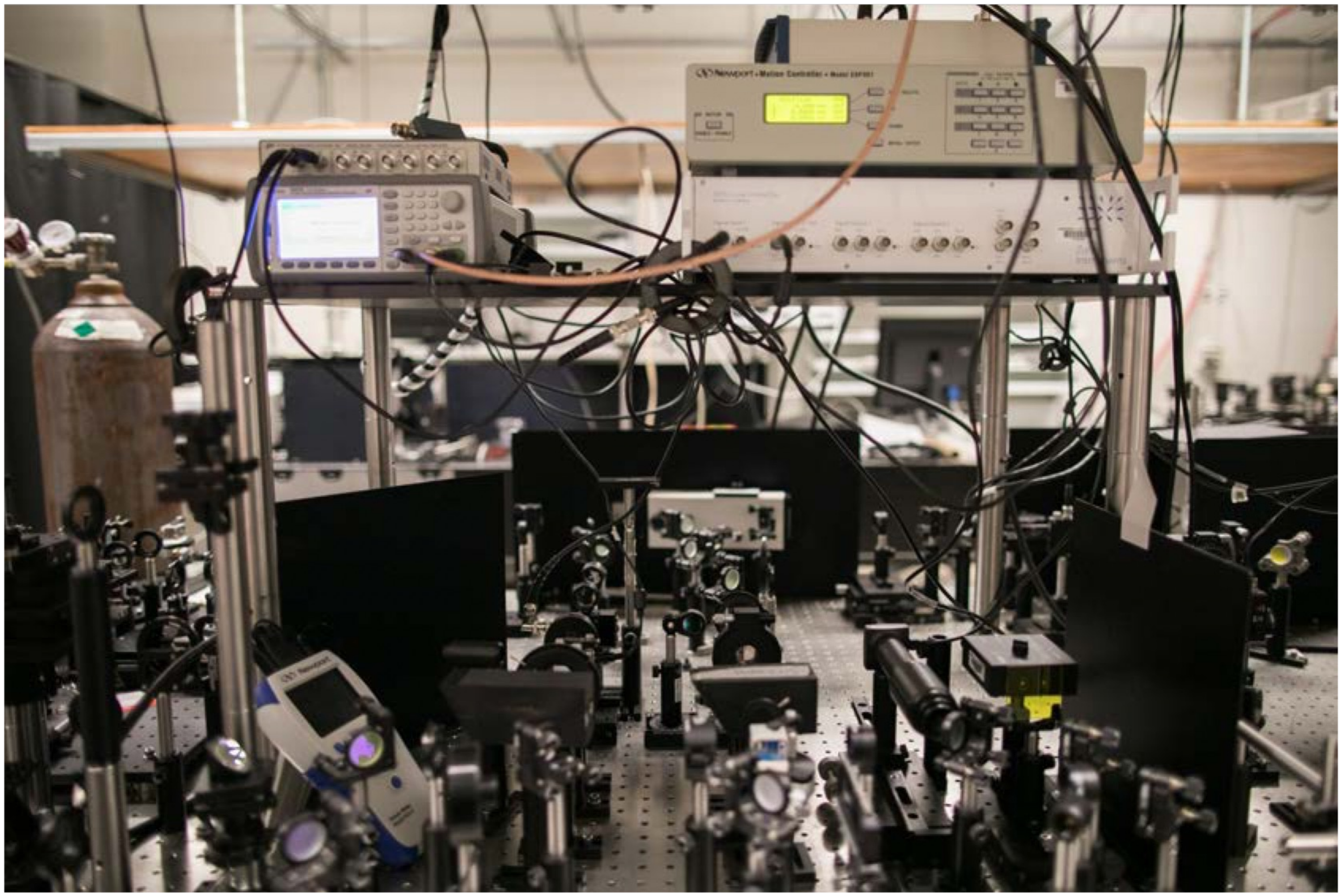
TDTR and FDTR (traditional)





- Thermoreflectance: change in reflectivity with change in temperature
- Pump-probe techniques: pump induces small temperature rise, probe measures material response
- TDTR: Time domain thermoreflectance
- FDTR: Frequency domain thermoreflectance
- Measurement of thermal effusivity/diffusivity **(*not direct thermal conductivity measurement*)**

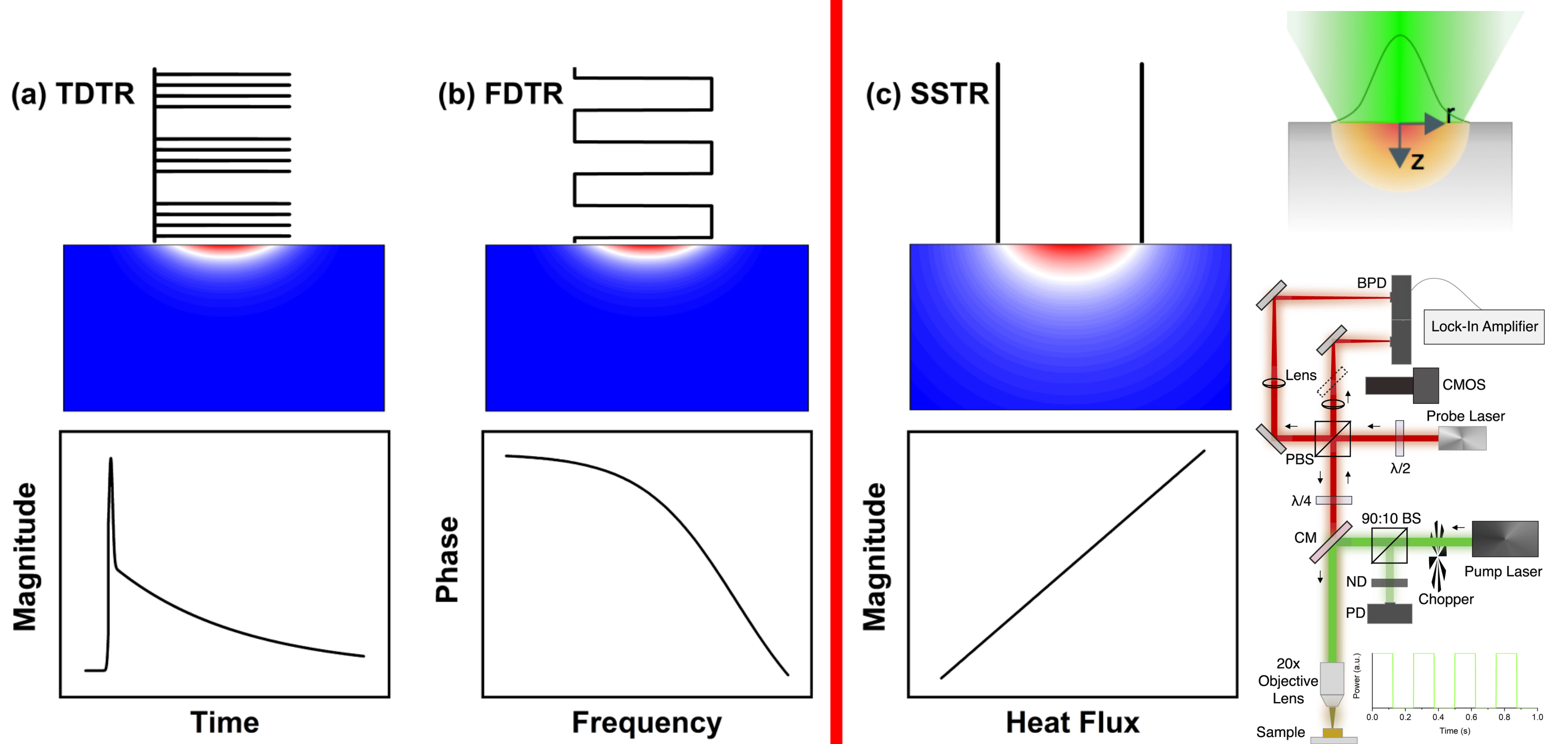
TDTR and FDTR: Practical limitations



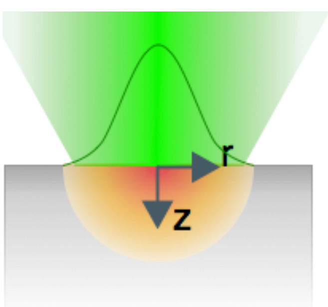
- But generally quite complicated, both experimental build and accurate analysis of data
- Daily upkeep needed
- Typically takes a PhD student their entire degree program to understand/independently operate/maintain a system like TDTR shown here

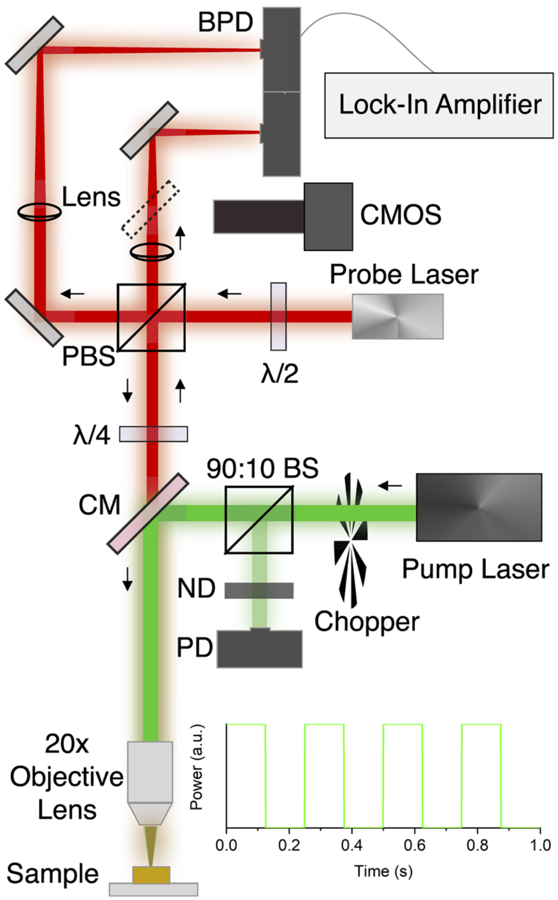
Steady state thermoreflectance (SSTR)

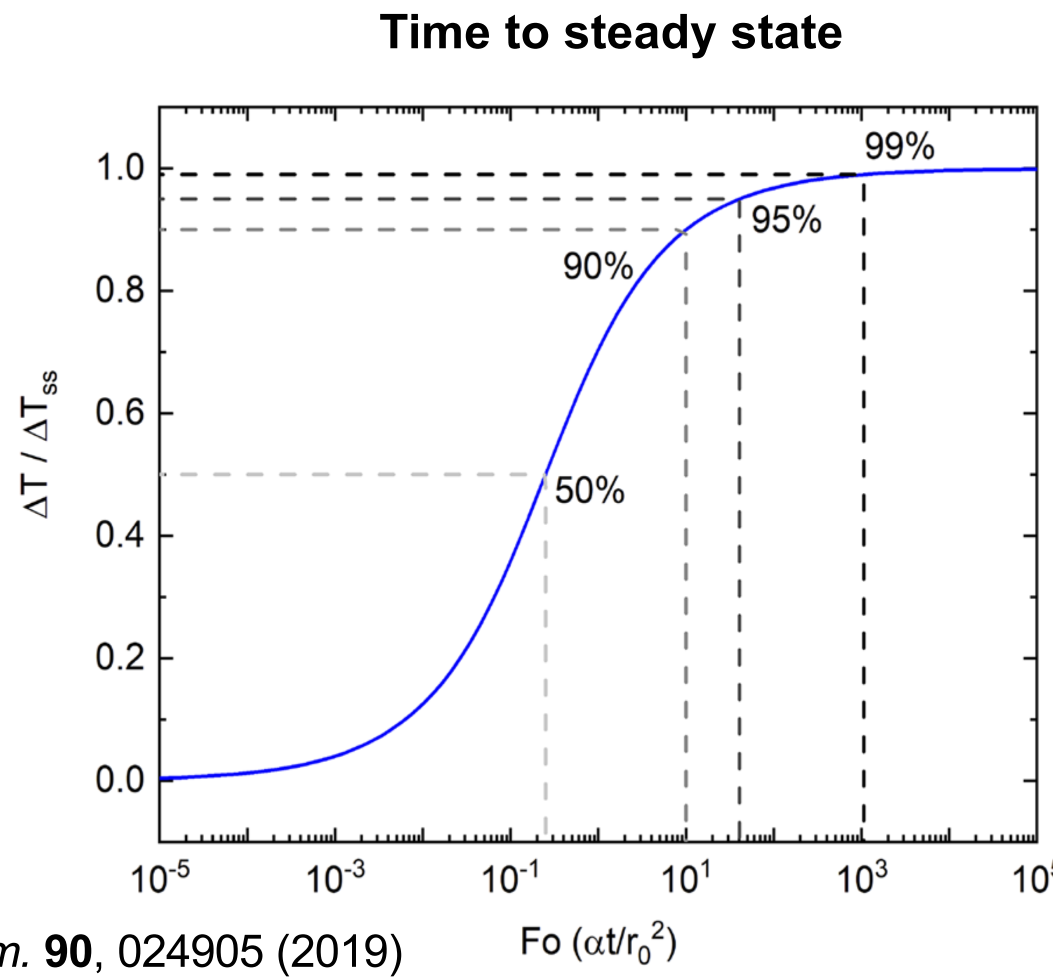
Steady-state thermoreflectance
Rev. Sci. Intr. **90**, 024905 (2019)



Steady state thermoreflectance (SSTR)



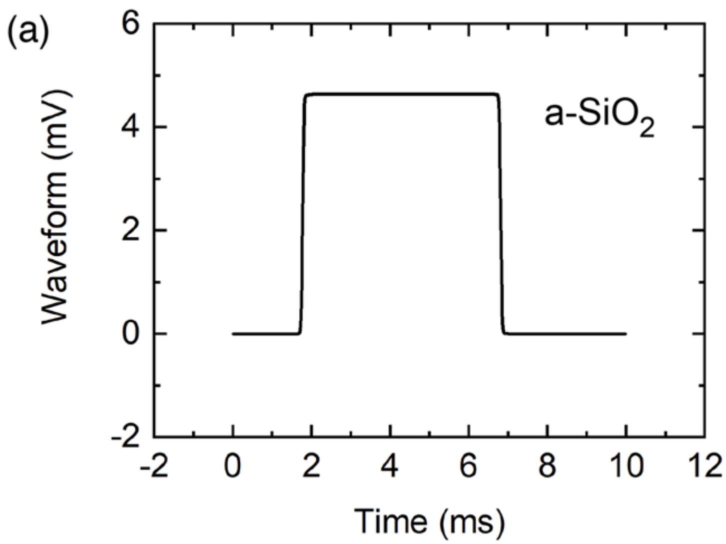




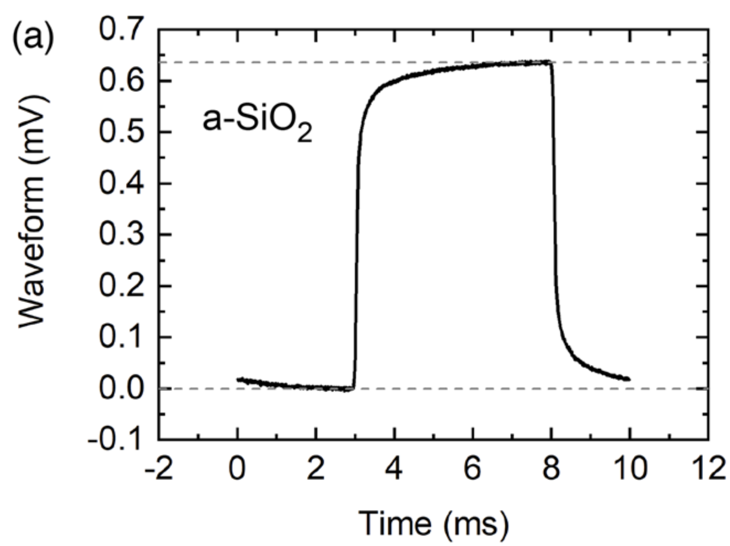
Rev. Sci. Instrum. **90**, 024905 (2019)

Steady state thermoreflectance (SSTR)

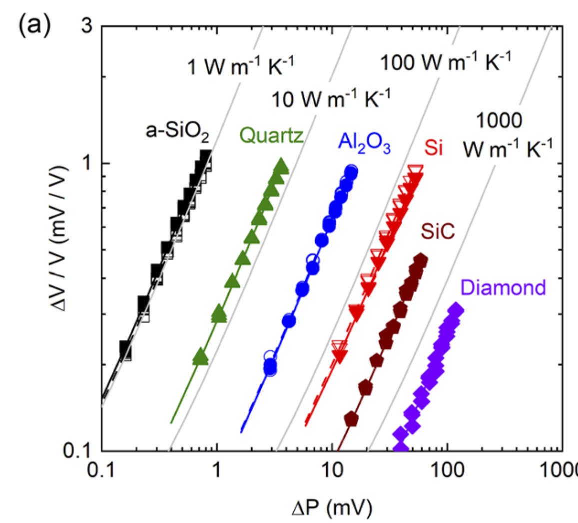
Pump waveform



Probe response

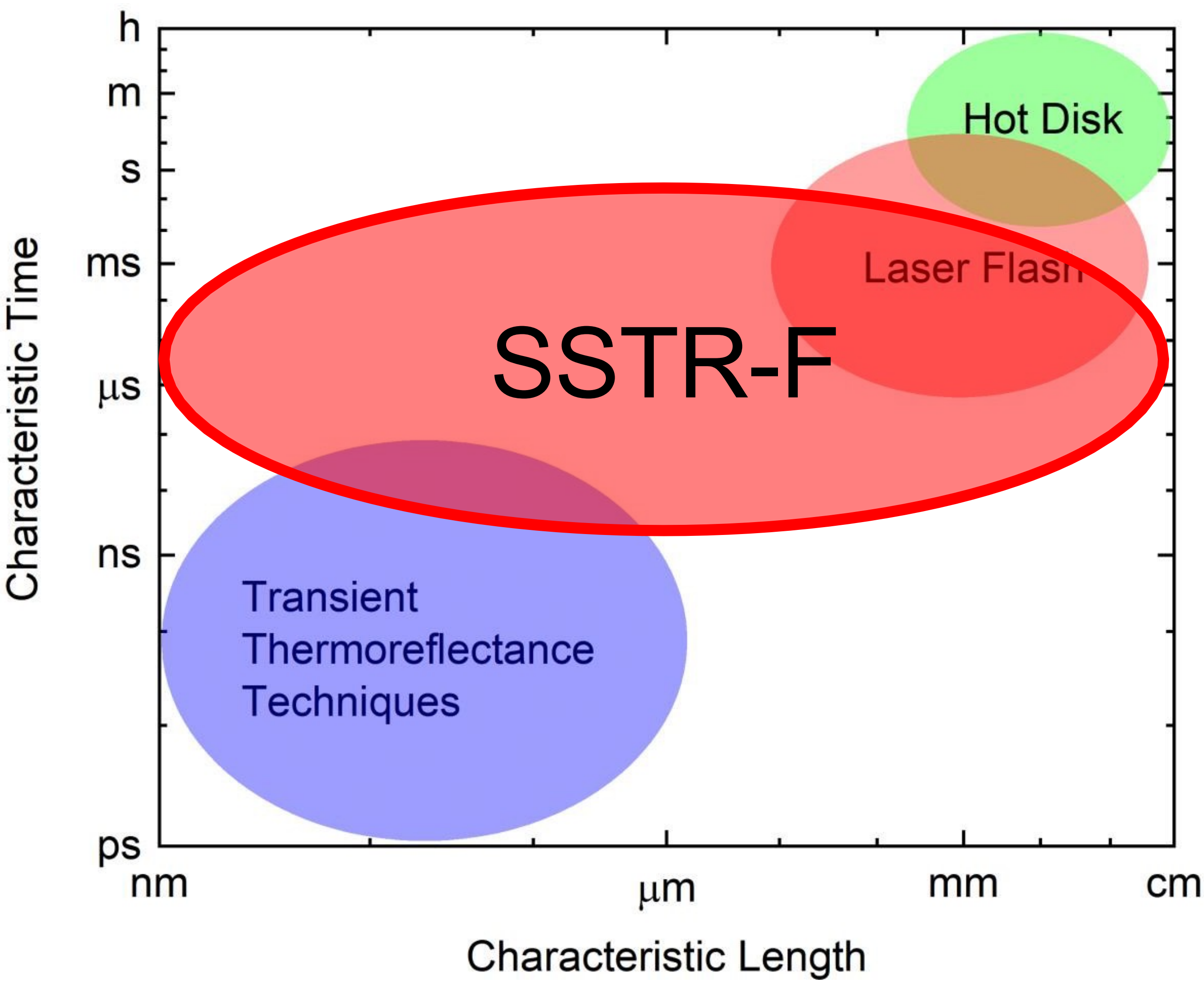


Fourier's Law



Rev. Sci. Instrum. **90**, 024905 (2019)

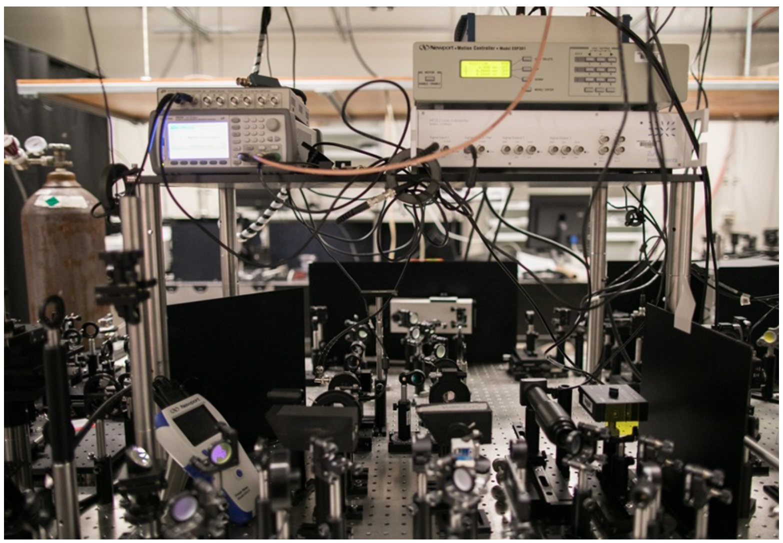
Steady state thermoreflectance (SSTR)



- Geometric limitations (thin, small, etc.)
  - Bulk measurements at nano/micro scales
- Testing times (thick/insulating)
  - Sub-minute regardless of material
- High and low thermal conductivity materials
  - As high as diamond (~2,000 W m^{-1} K^{-1})
  - As low as PCBM (~0.05 W m^{-1} K^{-1})
- Expert User
  - Full-automation
- High Cost (upkeep, labor)
  - No optics experience needed, tech level testing with automation

Lsaer Thermal's SSTR-F

Academic State of the Art vs. Laser Thermal's SSTR-F



Current lab-based systems are complex with advanced optics and instrumentation knowledge needed for use.

Shown Above: Time-domain Thermoreflectance (TDTR)
Expertise required: PhD-level operator.

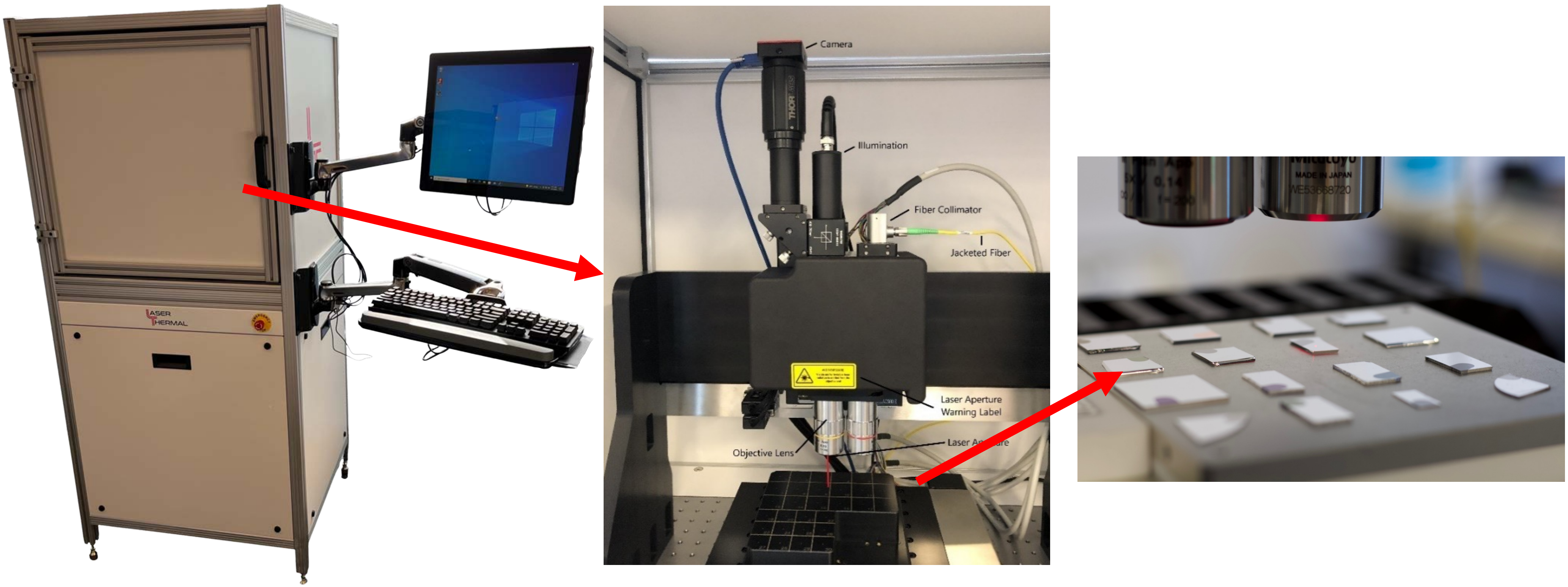
Laser Thermal's SSTR-F is simple, enabling fully automated push-button testing.

Expertise required: Associates degree technician level operator.

LASER THERMAL

Tutorials and demos available online
https://laserthermal.com

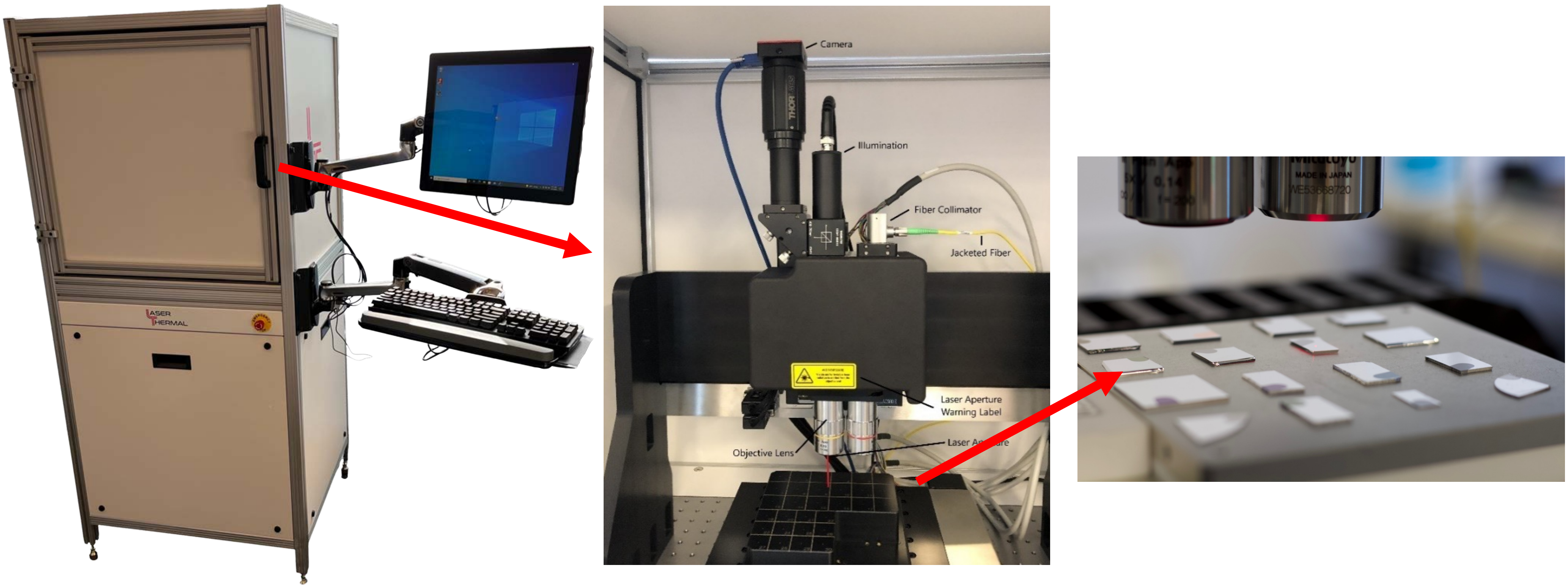
SSTR-F: Fully automated thermal conductivity testing and analysis



LASER THERMAL

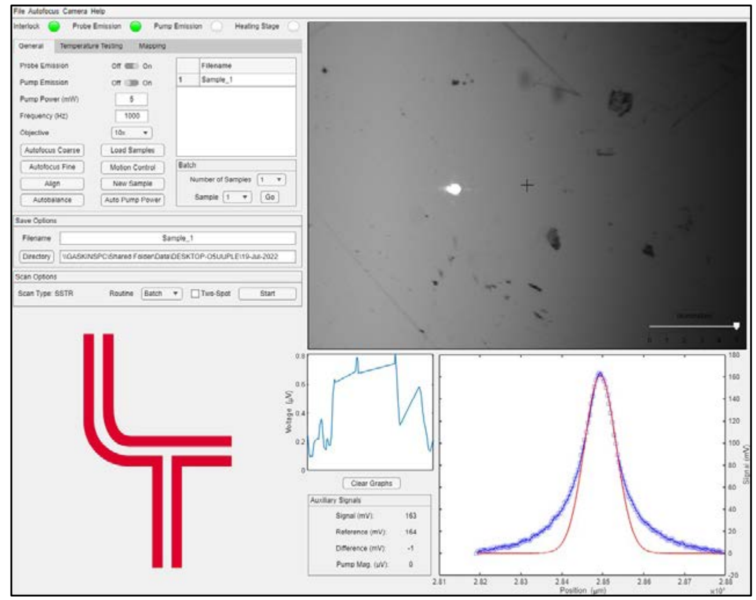
Tutorials and demos available online
https://laserthermal.com

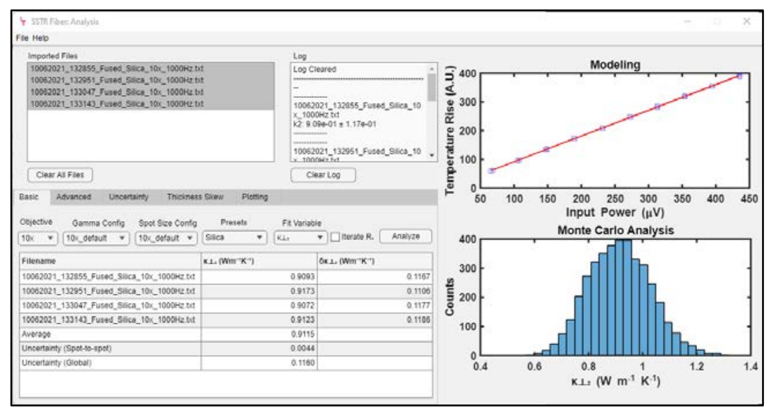
SSTR-F: Fully automated thermal conductivity testing and analysis



Tutorials and demos available online
https://laserthermal.com

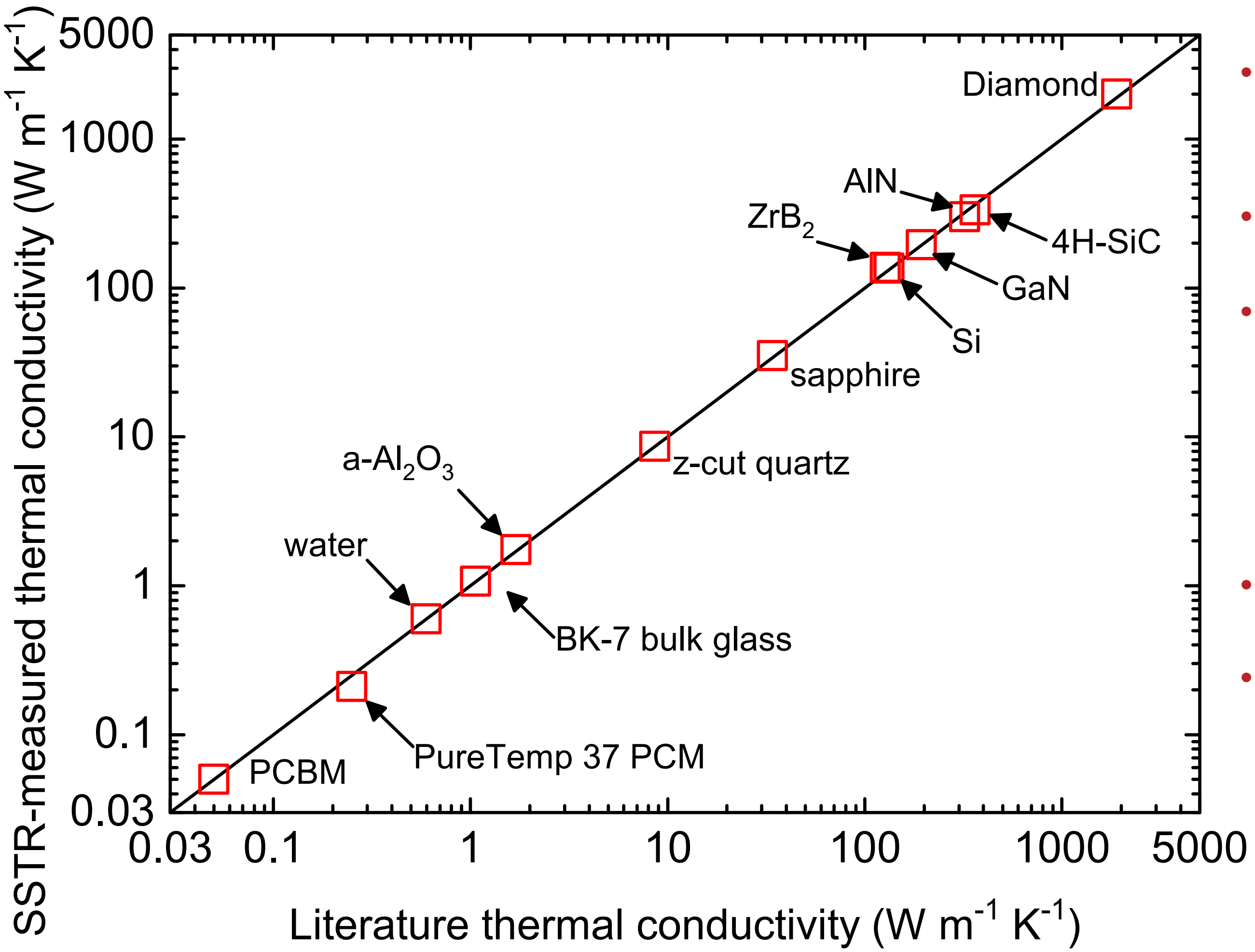
SSTR-F: Fully automated thermal conductivity testing and analysis





Tutorials and demos available online
https://laserthermal.com

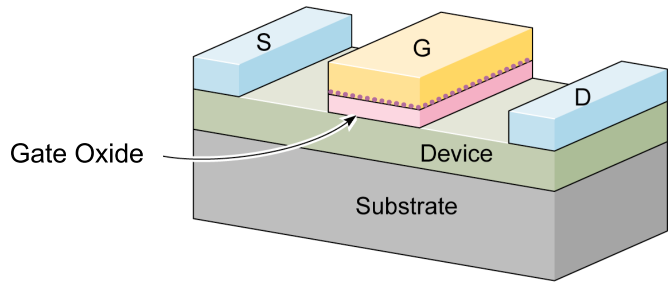
SSTR-F capabilities

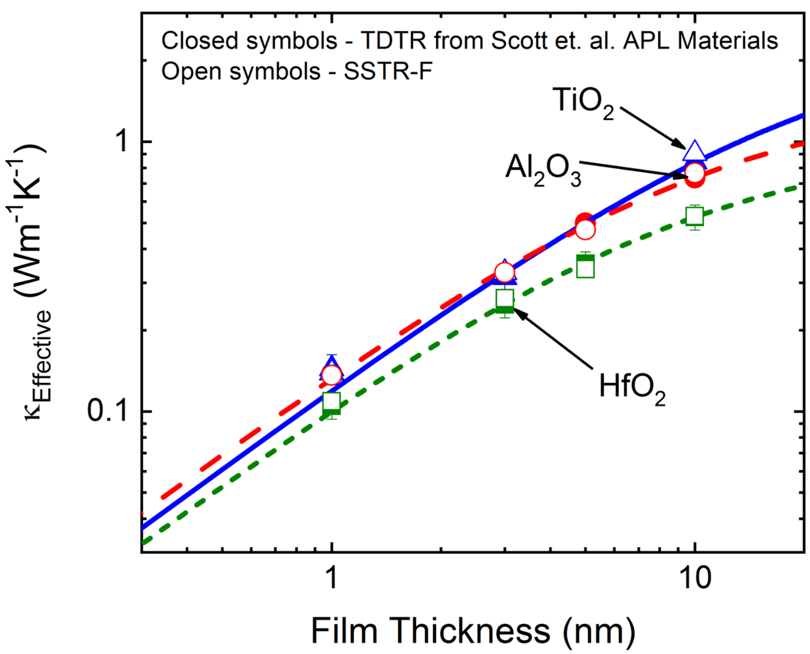


- Geometric limitations (thin, small, etc.)
  - Bulk measurements at nano/micro scales
- Testing times (thick/insulating)
  - Sub-minute regardless of material
- High and low thermal conductivity materials
  - As high as diamond ($\sim$2,000 W m^{-1} K^{-1})
  - As low as PCBM ($\sim$0.05 W m^{-1} K^{-1})
- Expert User
  - Full-automation
- High Cost (upkeep, labor)
  - No optics experience needed, tech level testing with automation

SSTR-F capabilities

Thermal conductivity of dielectric films as thin as 1 nm



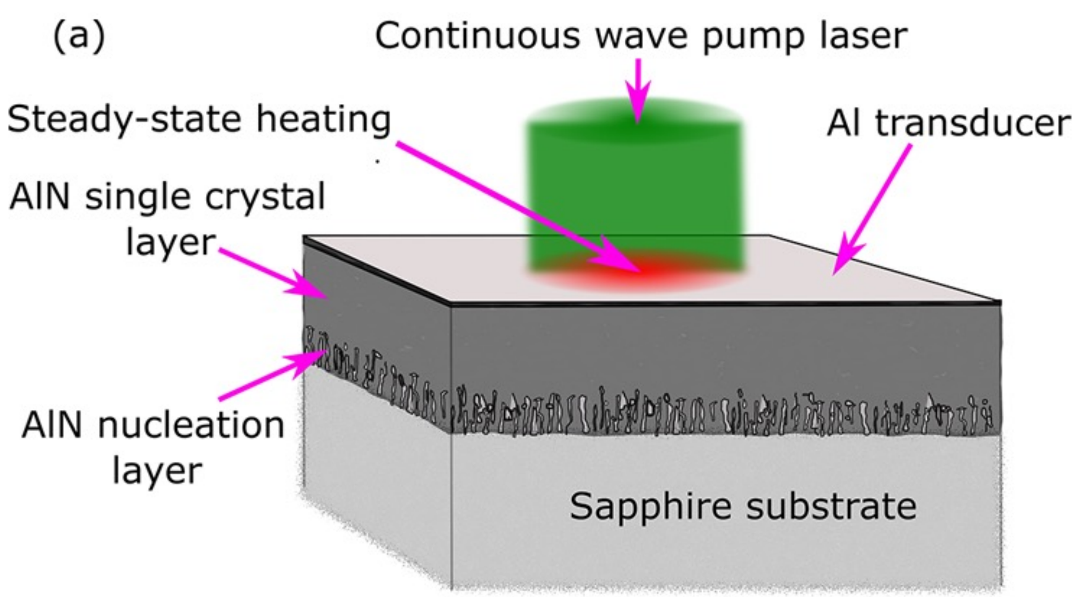


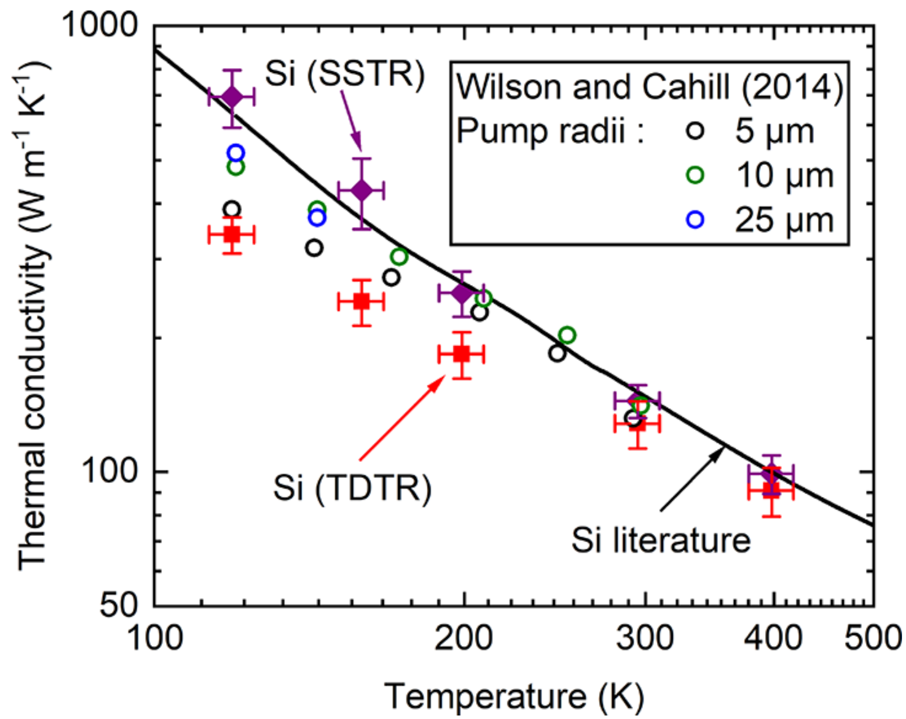
- Verified over three materials systems using SSTR-F
- Matches existing TDTR measurements
- Measuring resistance from interfaces and from material resistance in this case

APL Materials **6**, 058302 (2018)

SSTR-F capabilities

Temperature dependent thermal conductivity

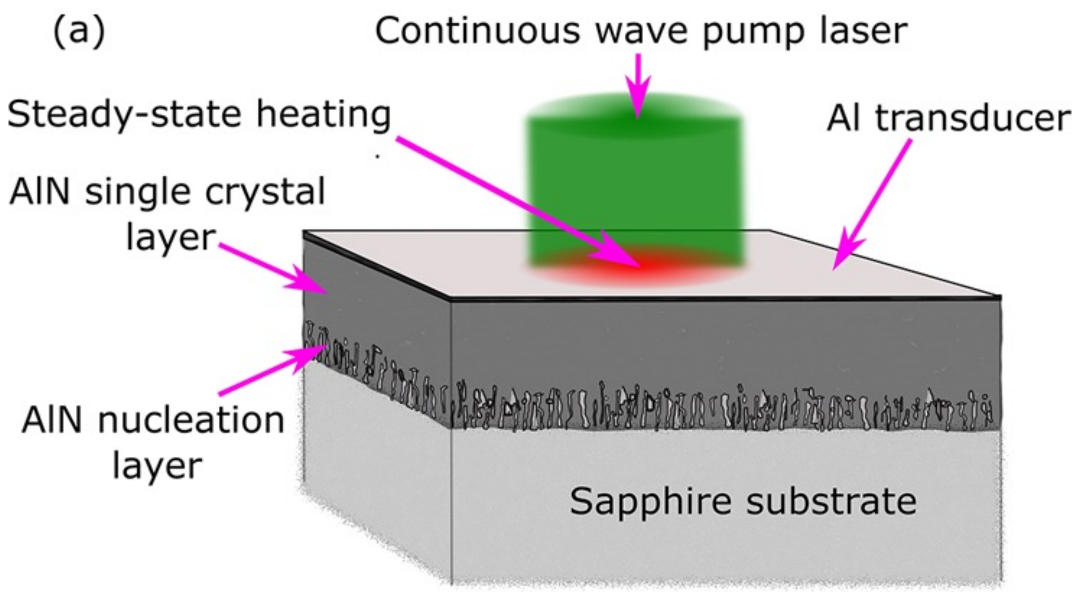


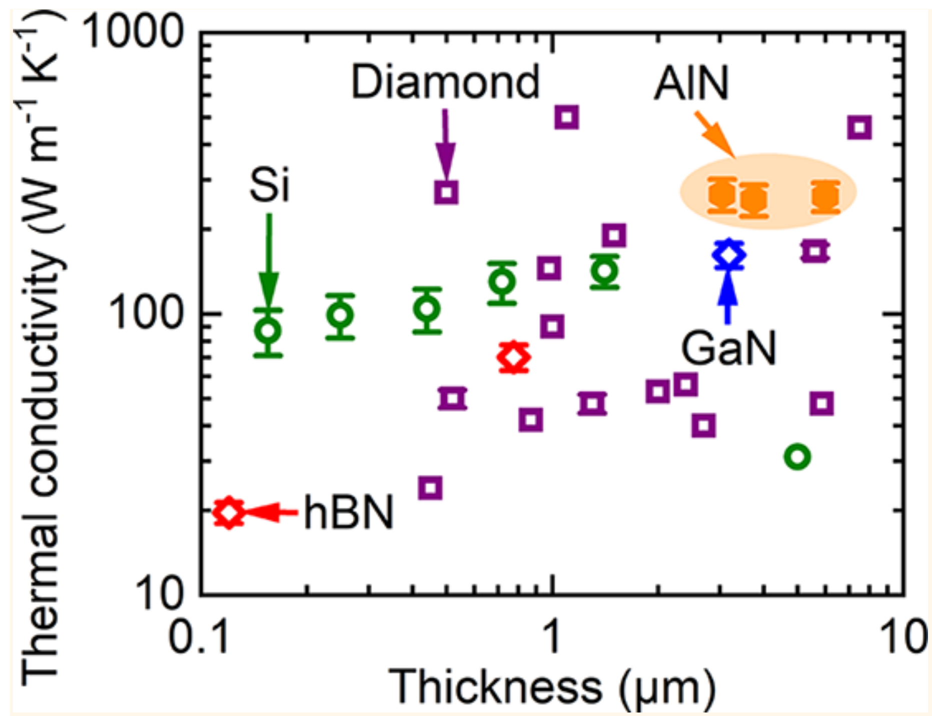


ACS Nano **15**, 9588 (2021)

SSTR-F capabilities

In-plane thermal conductivity of thin films
e.g., anisotropy effects in AlN thin films





ACS Nano **15**, 9588 (2021)

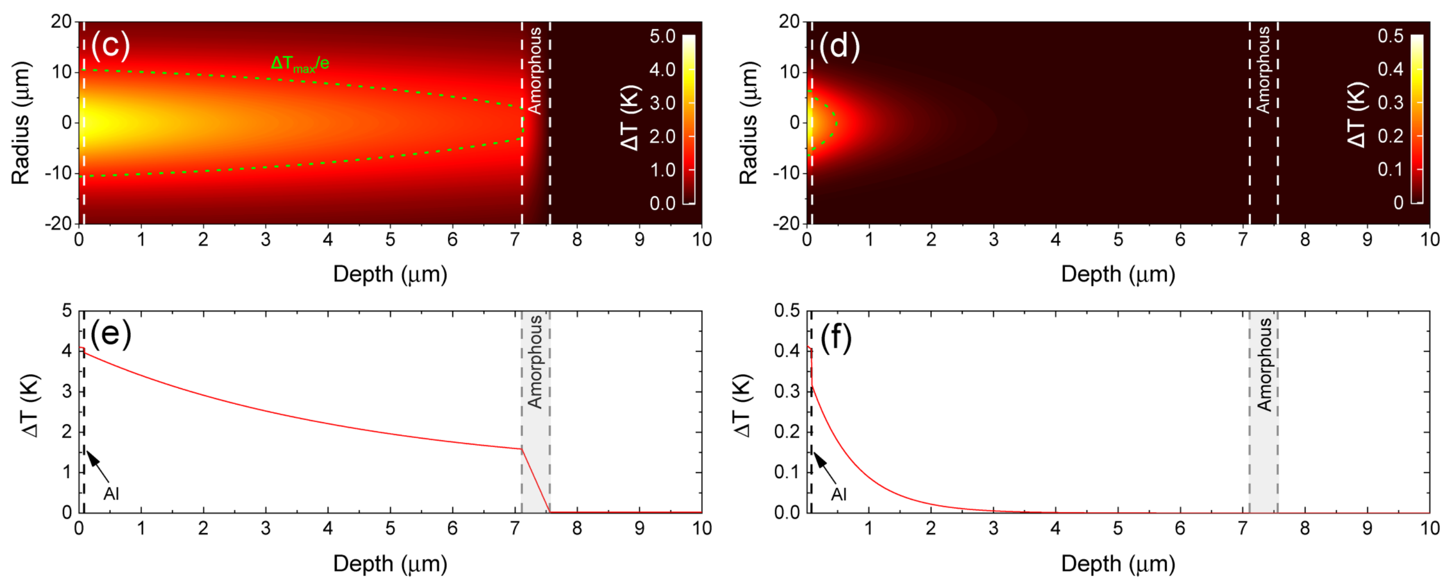
SSTR-F capabilities

Sub-surface defect detection
e.g., measure thermal conductivity of thin region with point defects 7 μm under diamond surface

J. Appl. Phys. **15**, 9588

SSTR can measure at variable depths under the surface controlled by the laser spot size

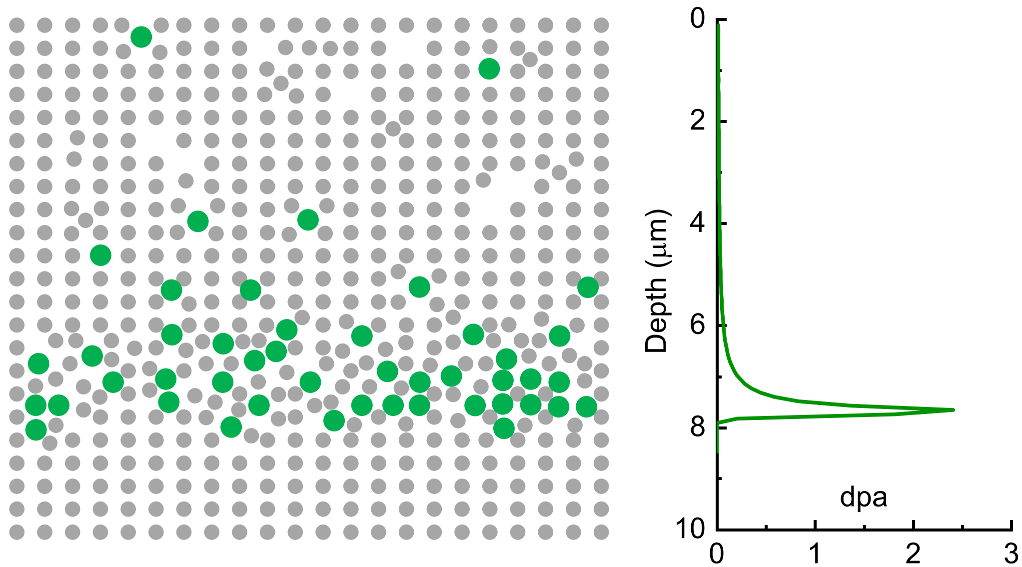
TDTR/FDTR restricted to ~1 μm beneath surface in diamond

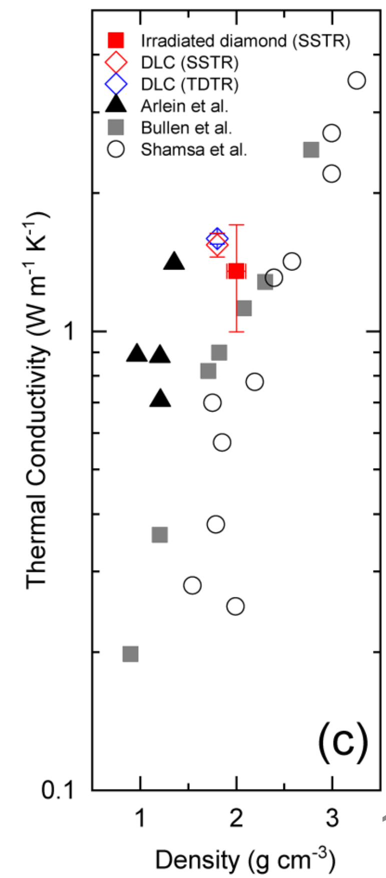


SSTR-F capabilities

Sub-surface defect detection
e.g., measure thermal conductivity of thin region with point defects 7 μm under diamond surface

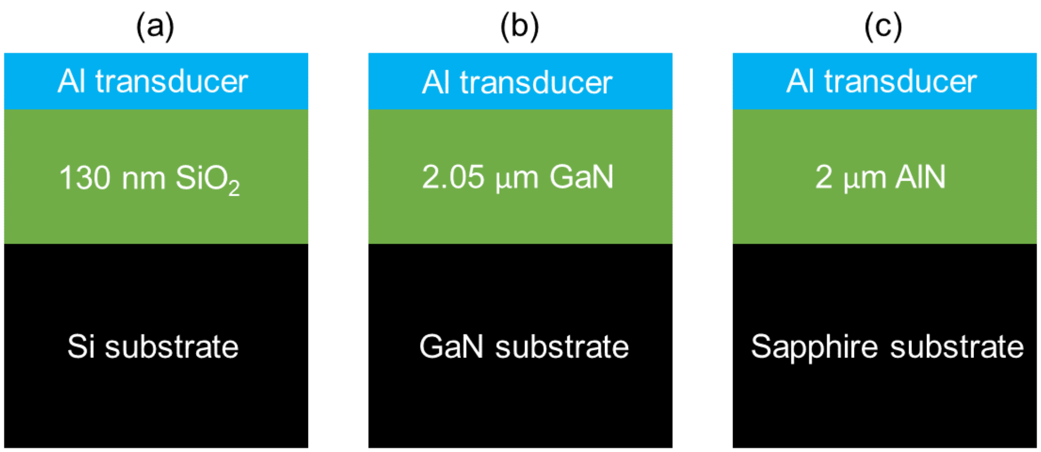
J. Appl. Phys. **15**, 9588





SSTR-F capabilities

Sub-surface interfaces and heat sinks
e.g., measure thermal conductivity of buried interfaces, sub-mounts & substrates under GaN and AlN thin films



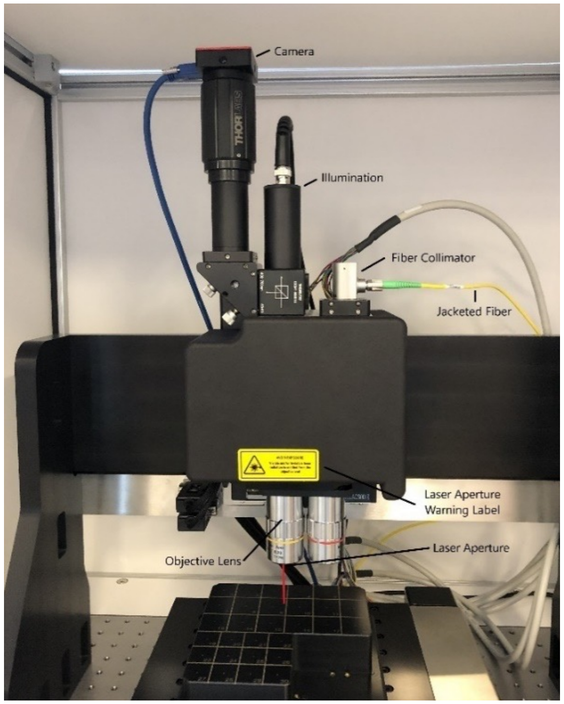
- Automated variable spot size in SSTR-F allows for control over testing depth
- Measurement of layer-by-layer thermal conductivity in material stack/composite

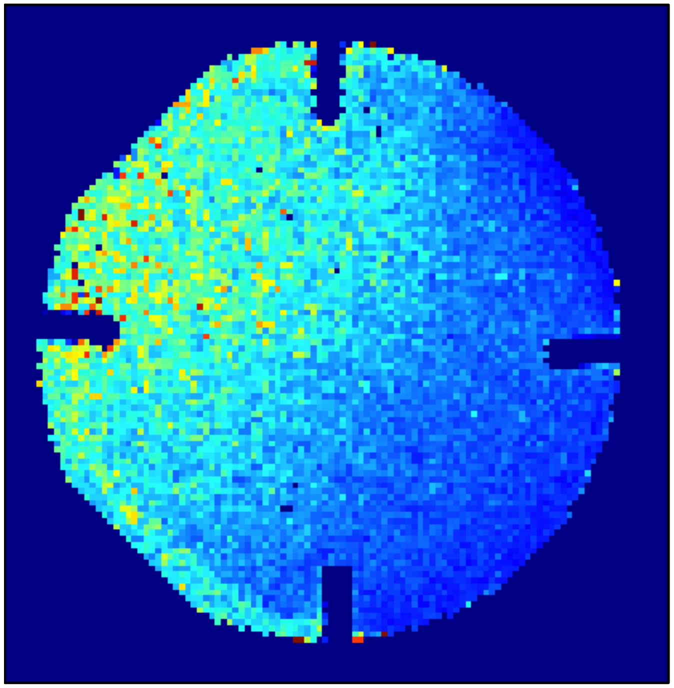
Substrates	Thermal conductivity ($W\ m^{-1}\ K^{-1}$)		
	spot size 10 μm	spot size 20 μm	literature
Si	141 ± 27	140 ± 18	140[30]
GaN	194 ± 27	185 ± 16	195[41]
Sapphire	35.1 ± 5.9	34.5 ± 4.2	35[42]

Rev. Sci. Instrum. **92**, 064906

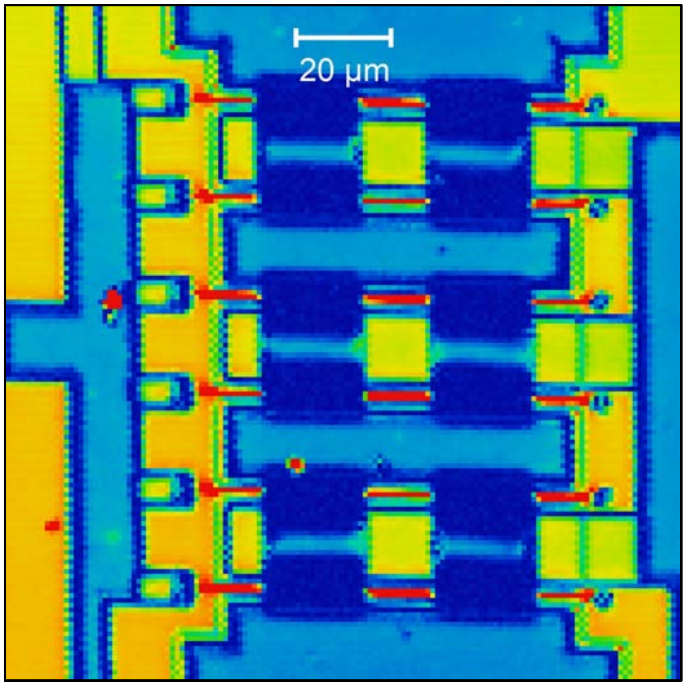
SSTR-F capabilities

Thermal Mapping of Wafers, Devices, etc.
w/ Lateral Resolution Down to ~1 micron





ALD film on 4” diameter silicon waver



6-finger GaAs pHEMT on a MMIC power amplifier

Laser Thermal's SSTR-F

- Thermal conductivity and resistance testing services from thin films to heat sinks

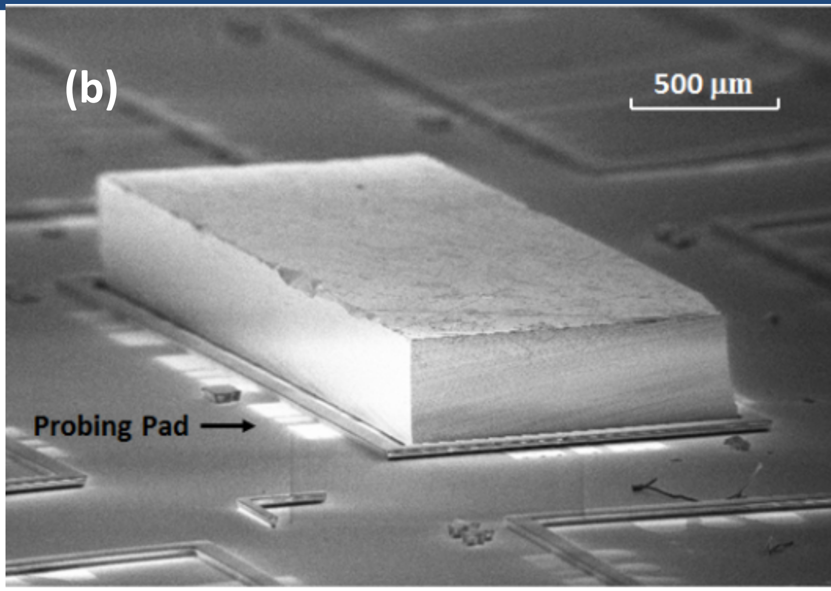
- Thermal conductivity measurement systems for thin films and bulk materials (SSTR-F)

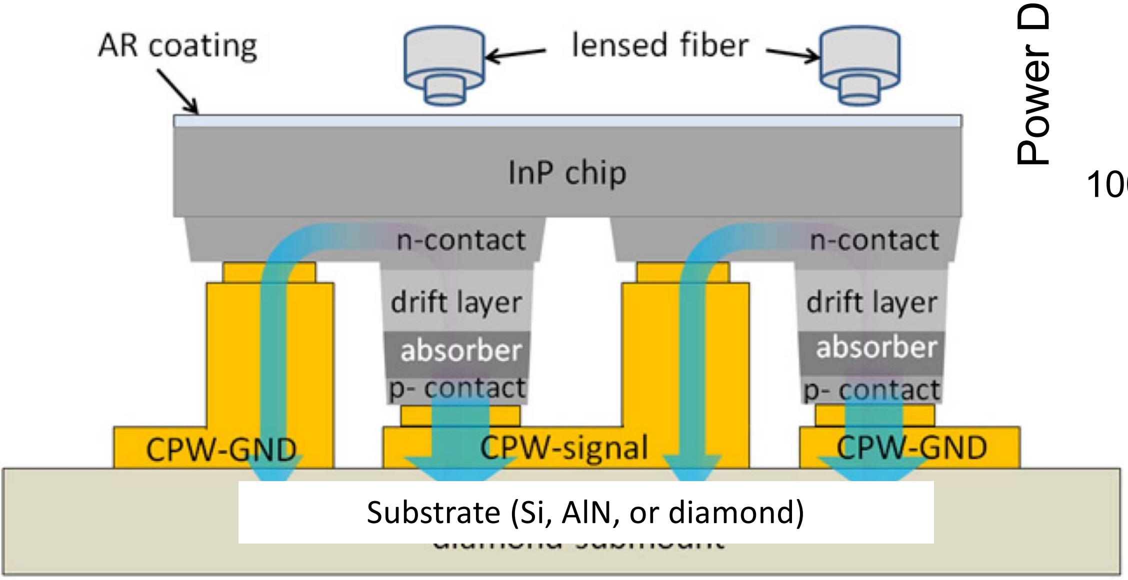
- Tutorials and demos available online https://laserthermal.com

Outline

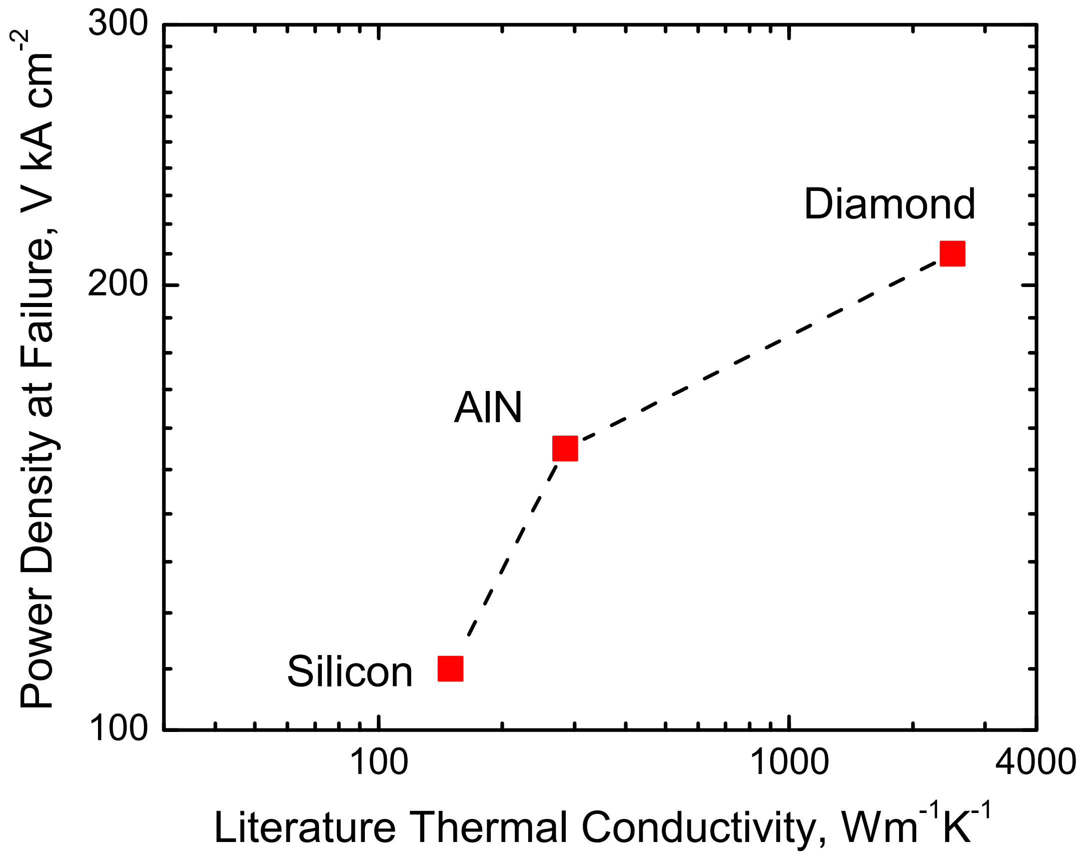
1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

High power device thermal management - traditional





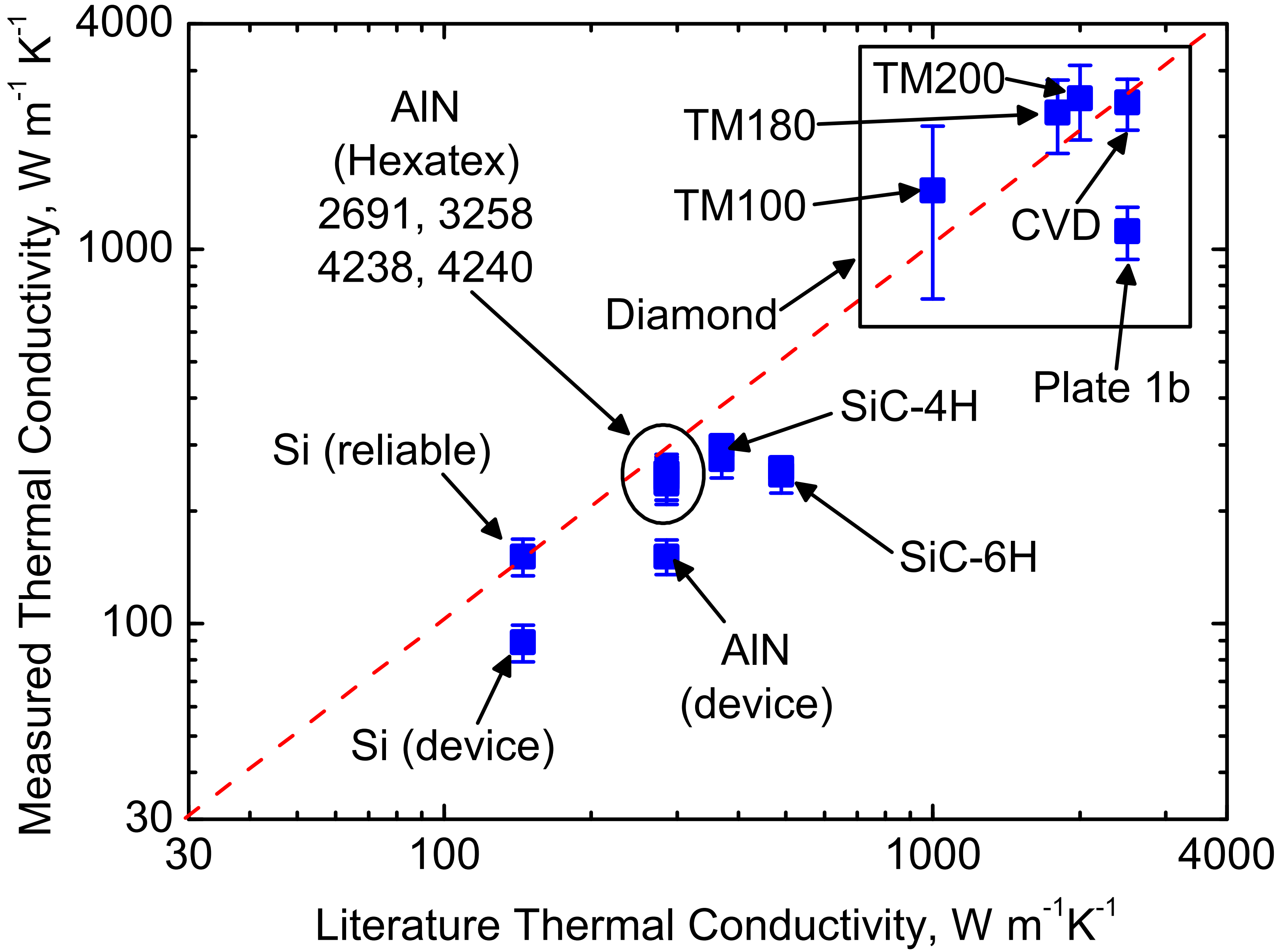
(c)



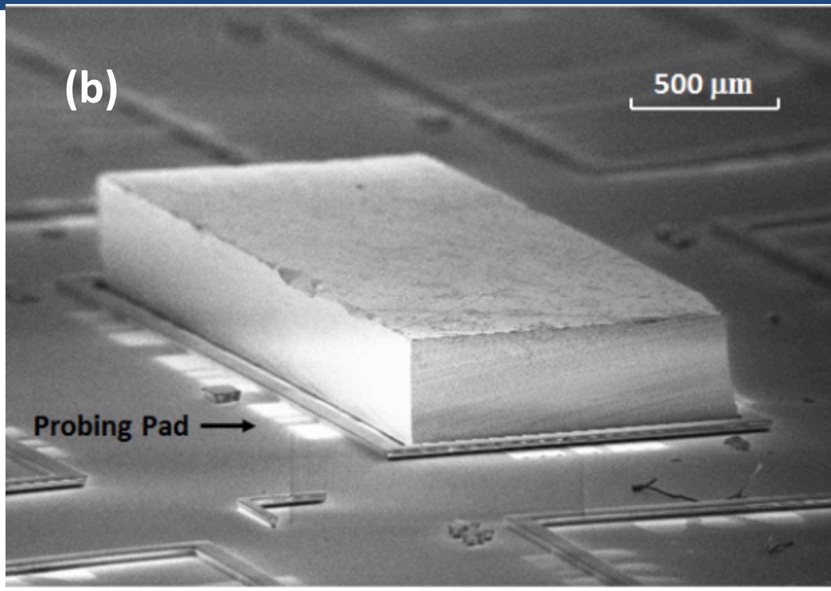
Collaboration with Joe Campbell (UVA)

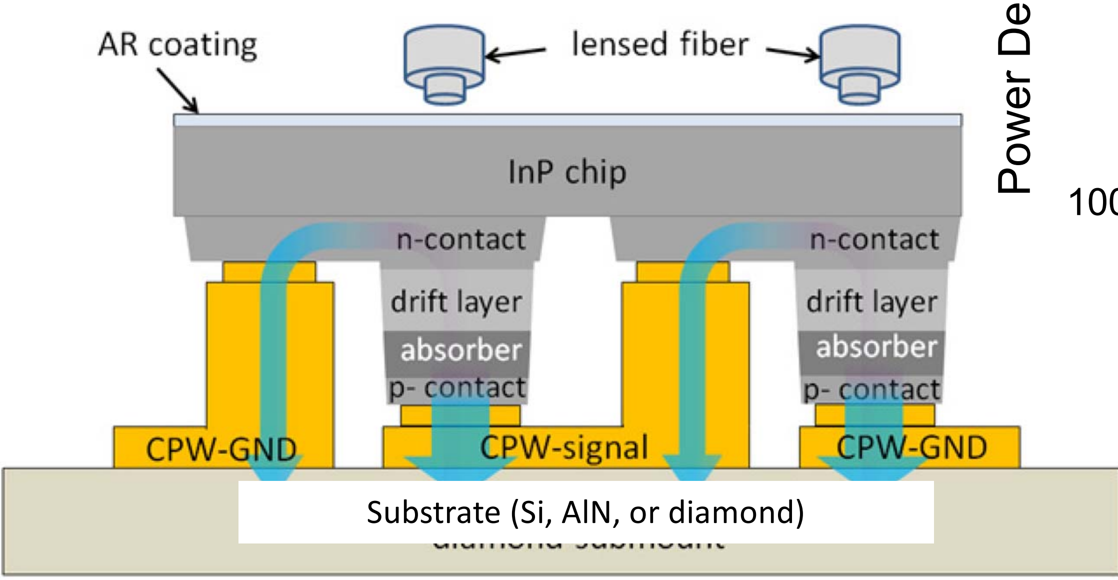
J. Lightwave Technology **35**, 4242 (2017)

Example: thermal conductivity of common high κ substrates

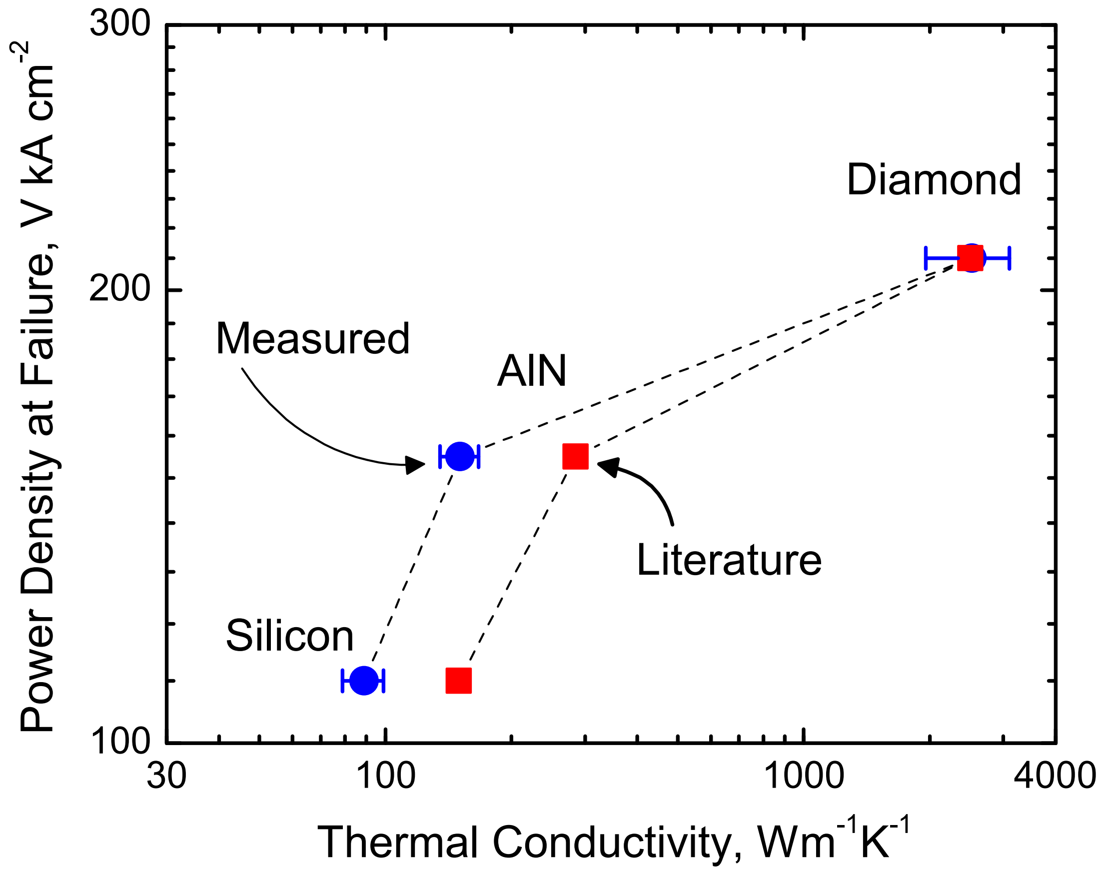


High power device thermal management – substrate effects





(c)



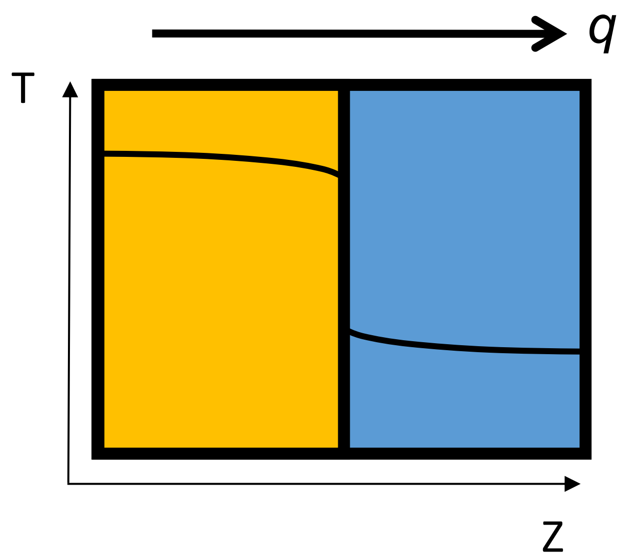
Still does not scale with heat sink thermal conductivity

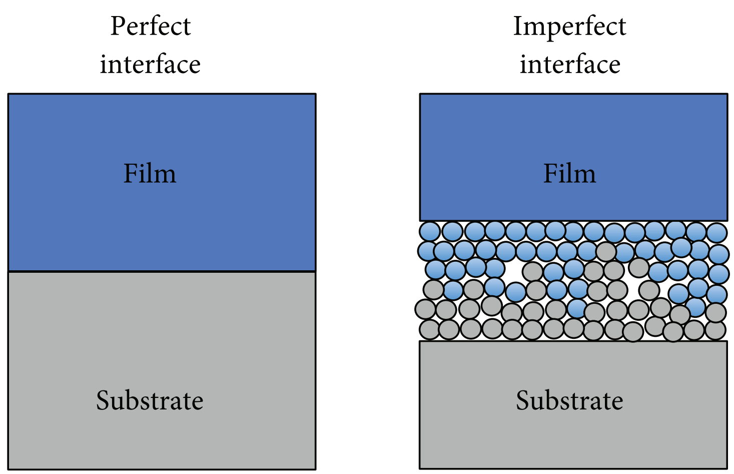
Collaboration with Joe Campbell (UVA)

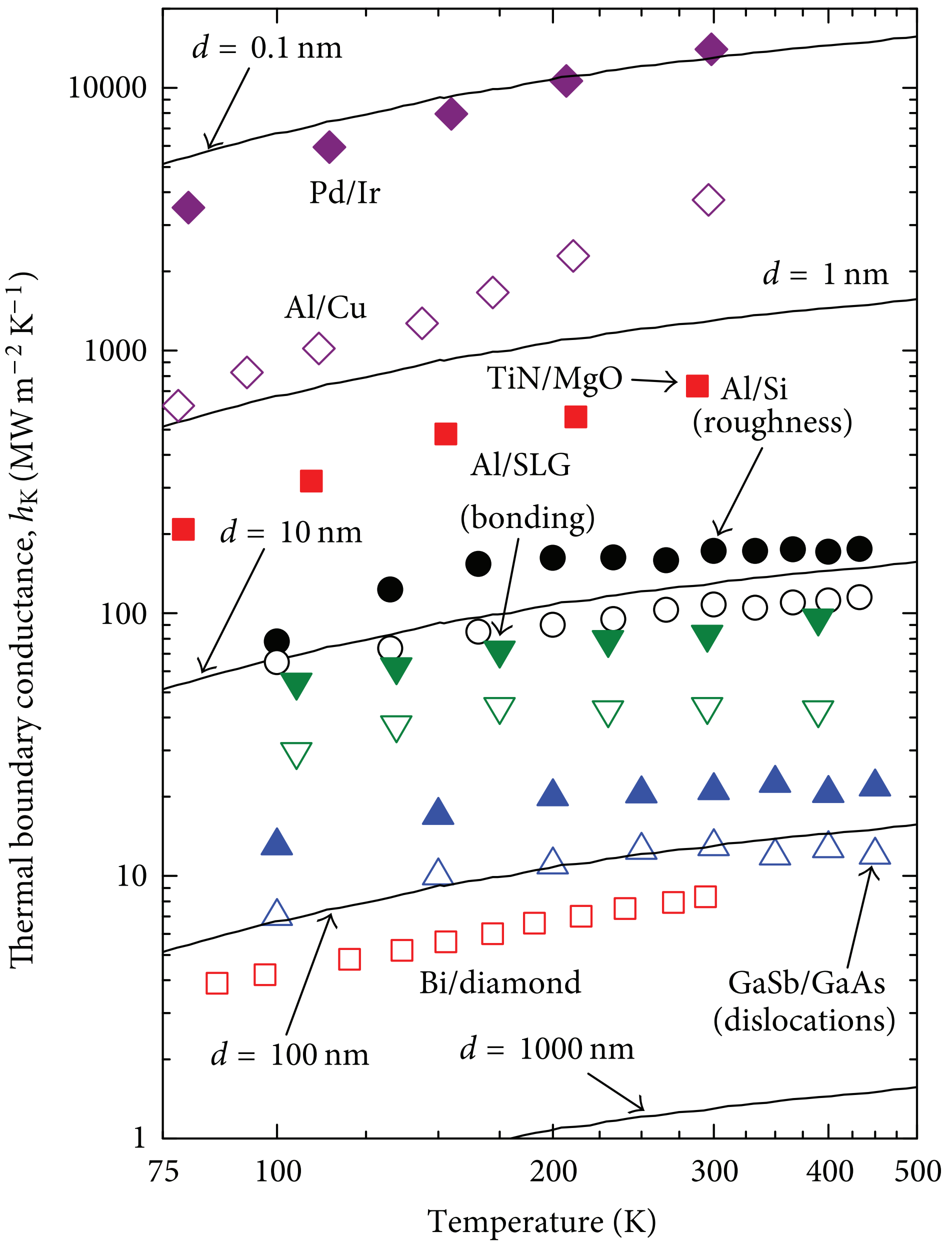
J. Lightwave Technology **35**, 4242 (2017)

Thermal boundary conductance – nanoscale resistances

$$q = h_K \Delta T = \frac{1}{R_K}\Delta T$$

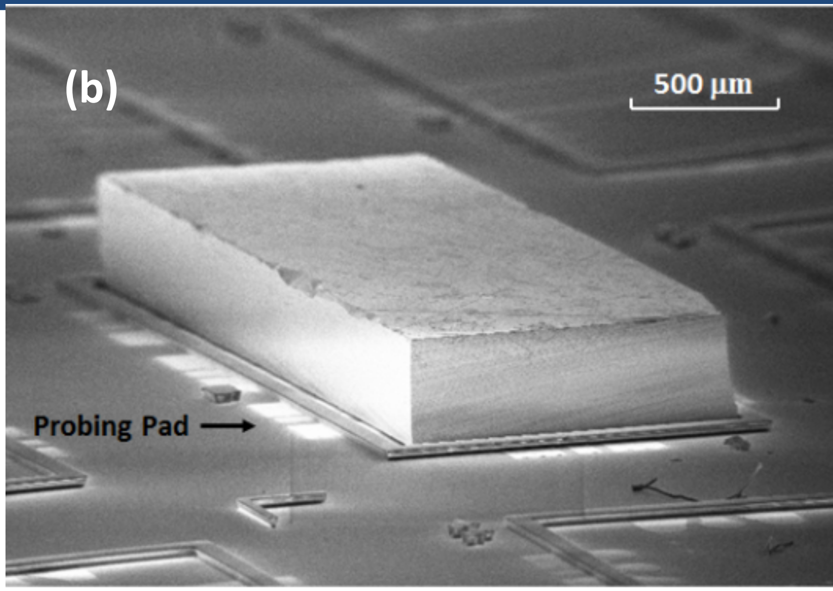


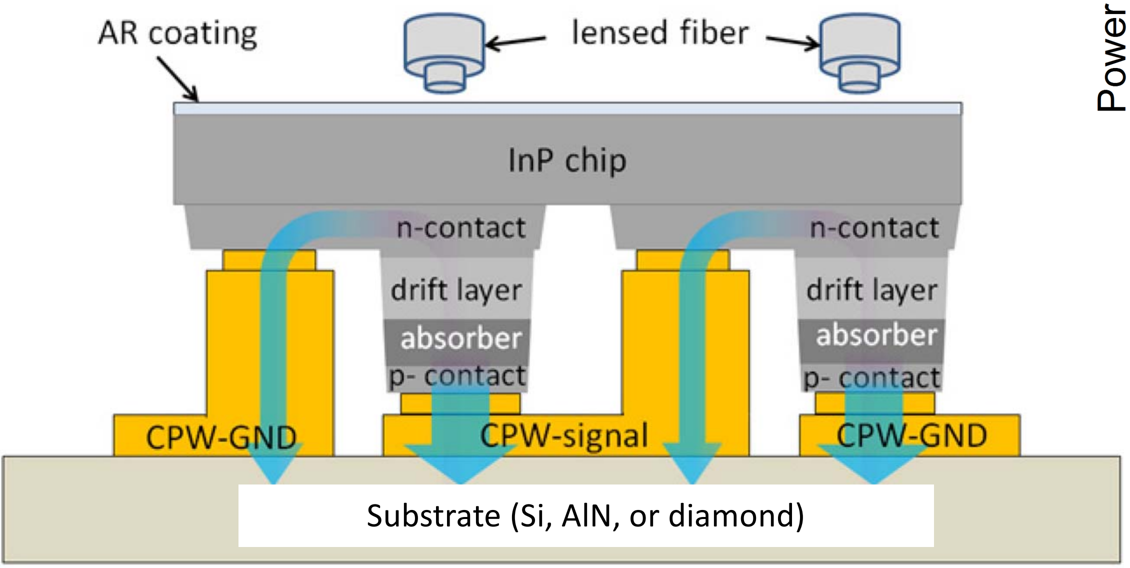




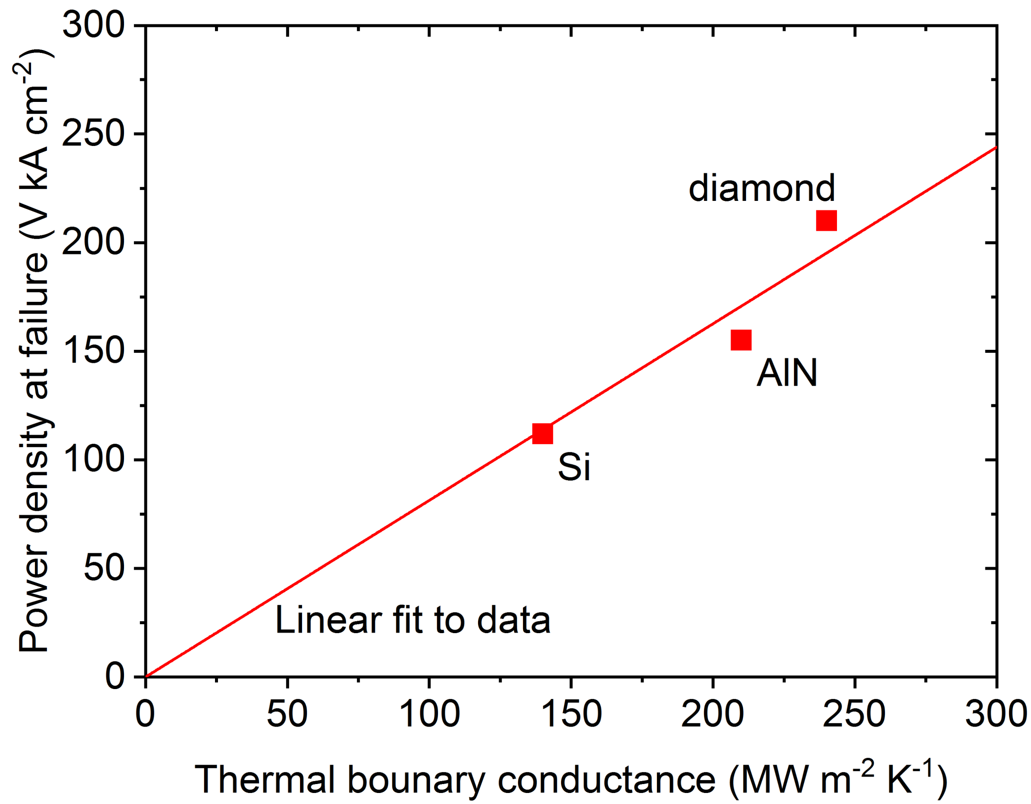
ISRN Mech. Eng. **2013**, 682586

High power device thermal management - nanoscale





(c)



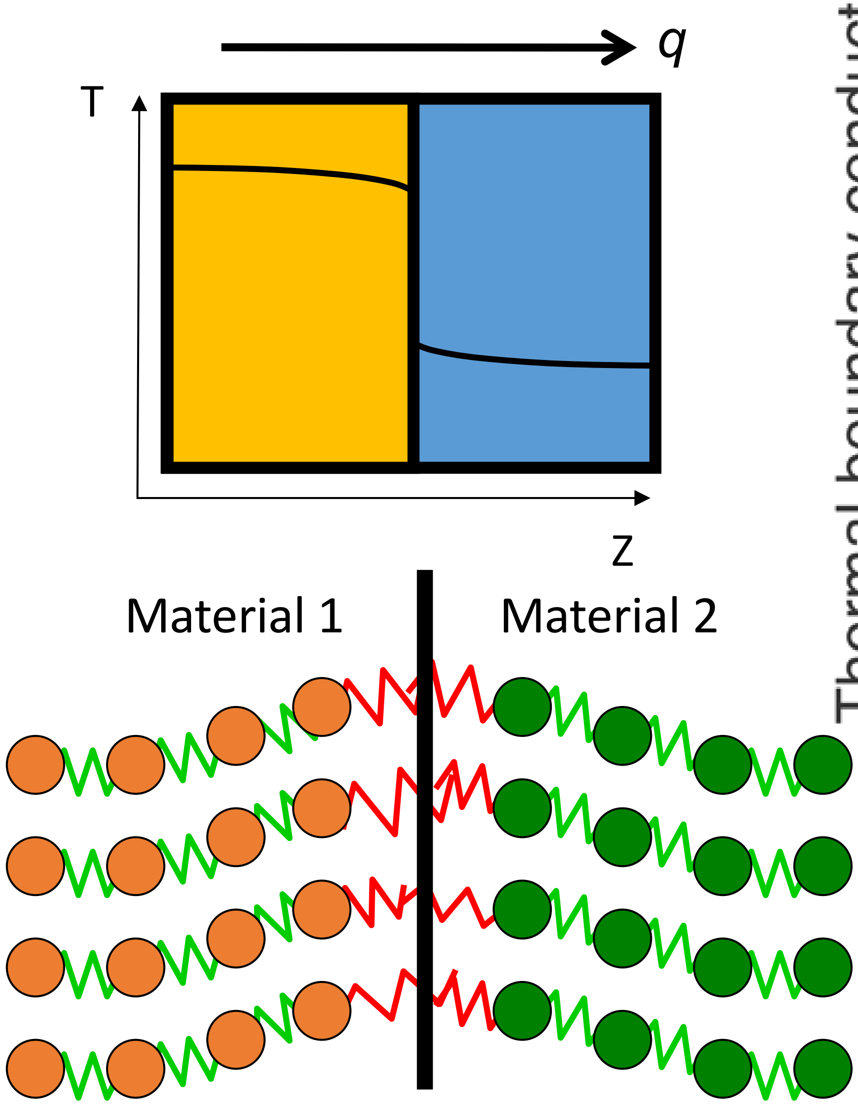
TBC plays direct role in power density at failure

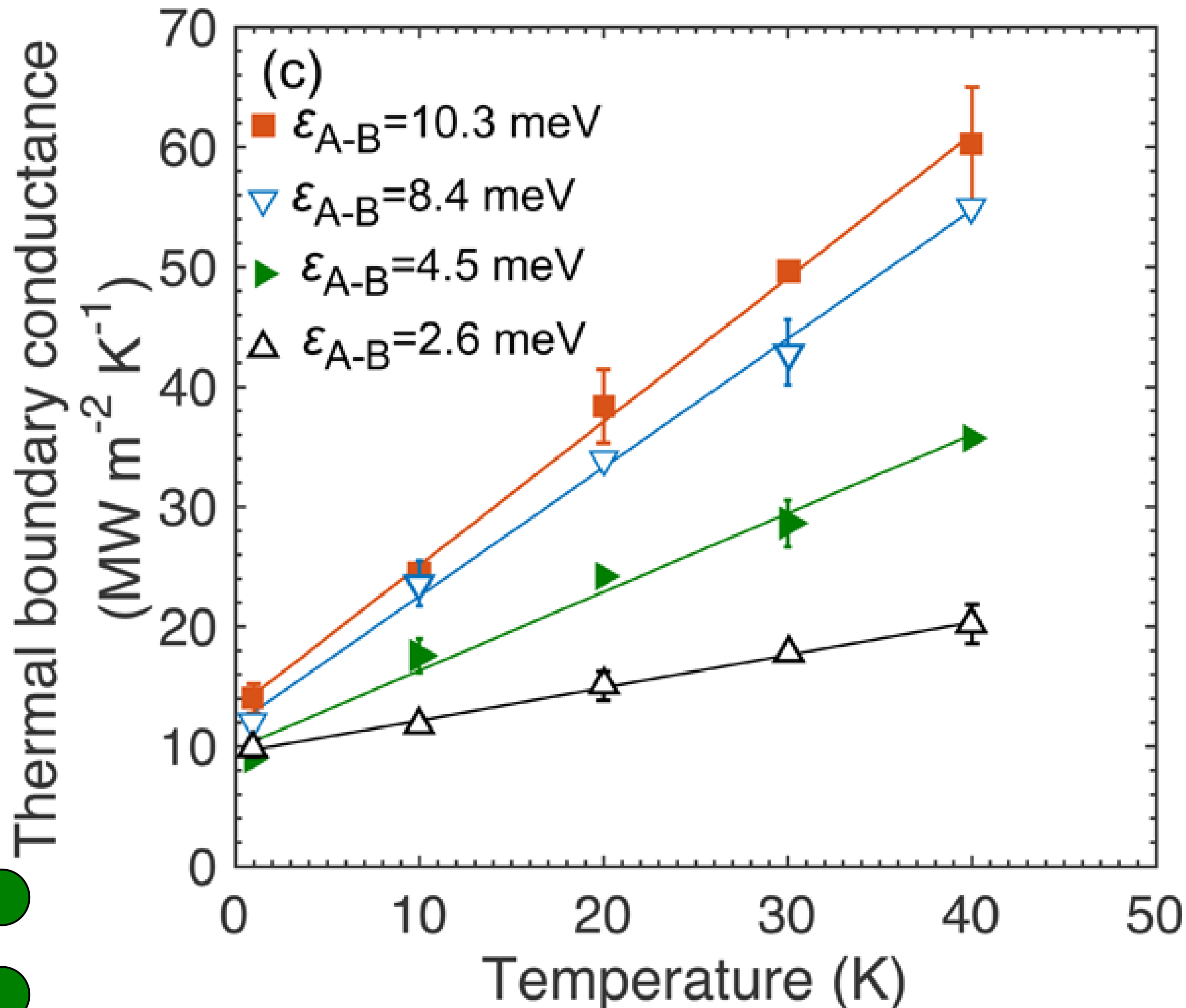
Collaboration with Joe Campbell (UVA)

J. Lightwave Technology **35**, 4242 (2017)

Increase in bonding increases solid/solid TBC

$$q = h_K \Delta T = \frac{1}{R_K} \Delta T$$

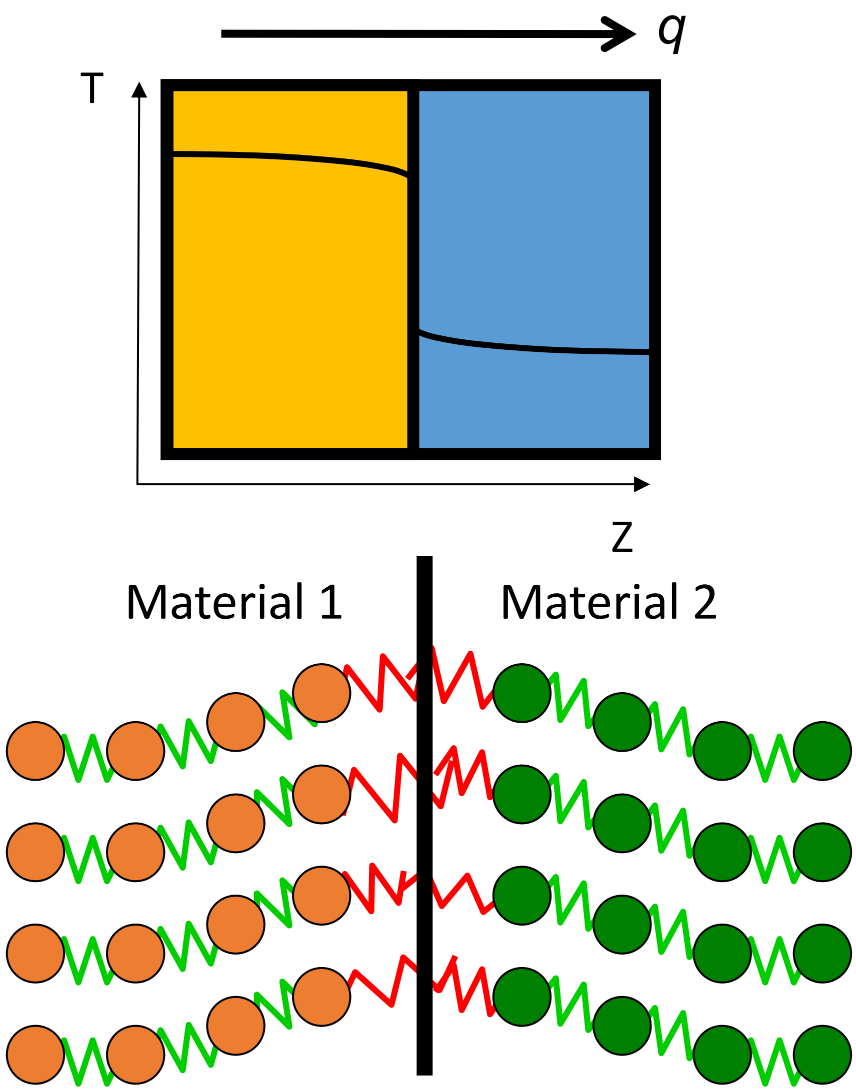




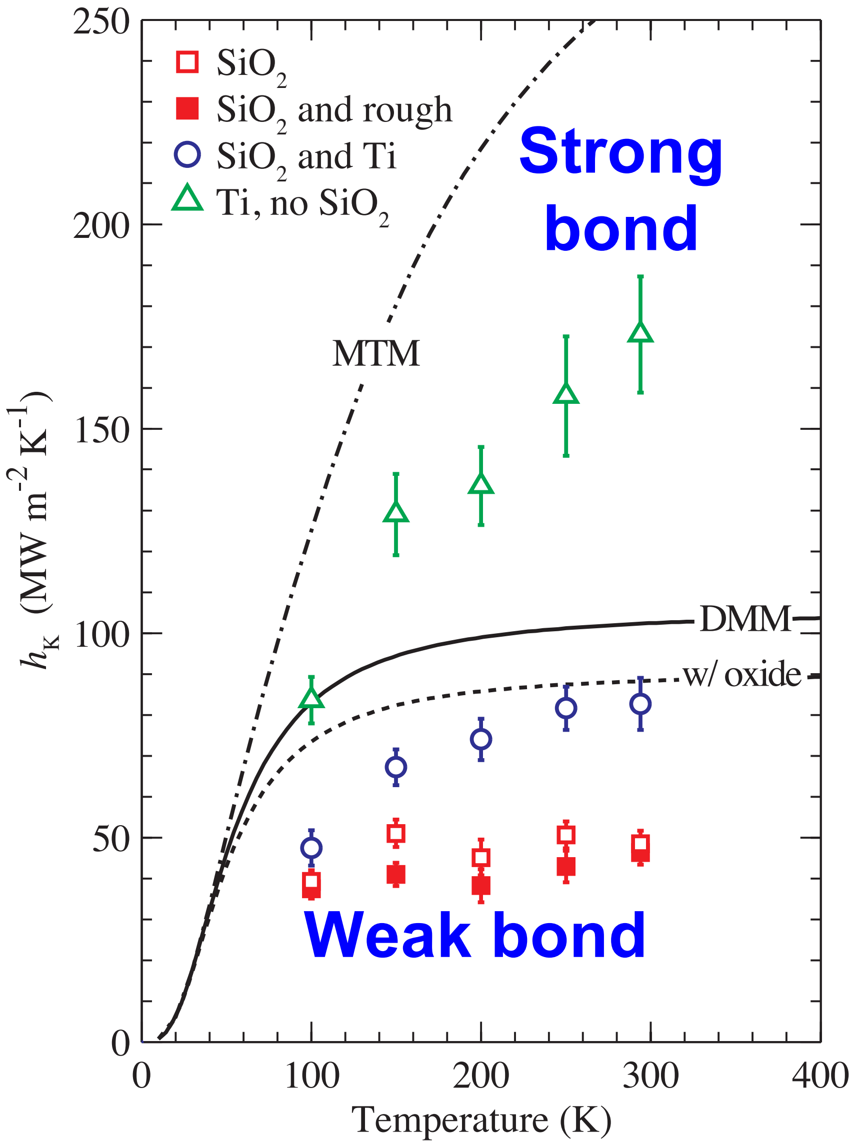
J. Phys. Chem. C **120**, 24847

Increase in bonding increases solid/solid TBC

$$q = h_K \Delta T = \frac{1}{R_K} \Delta T$$

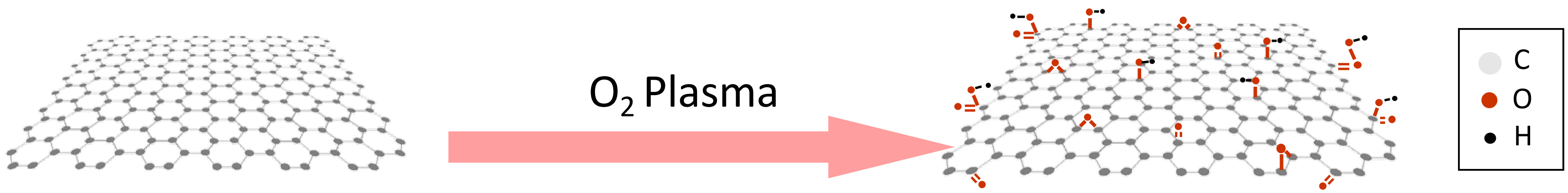


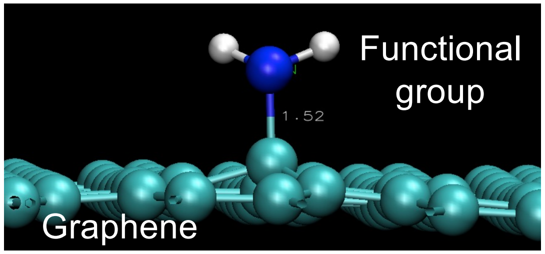
Ti adhesion layer effects on Au/Si TBC



Appl. Phys. Lett. **102**, 081902

Atmospheric plasma functionalization of graphene surfaces



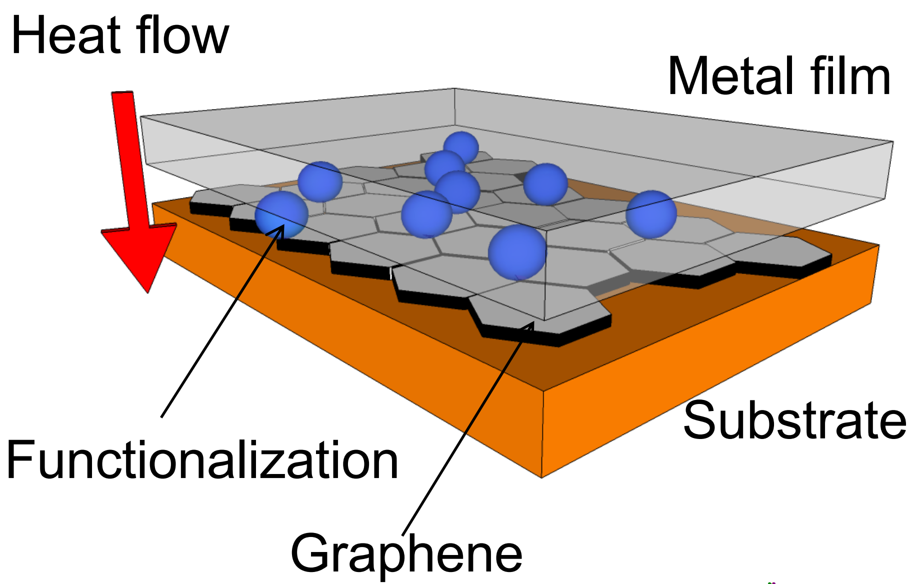


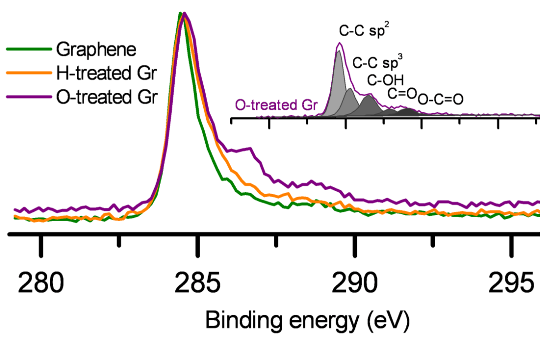
Functional groups covalently bound to graphene
Reversible after anneal

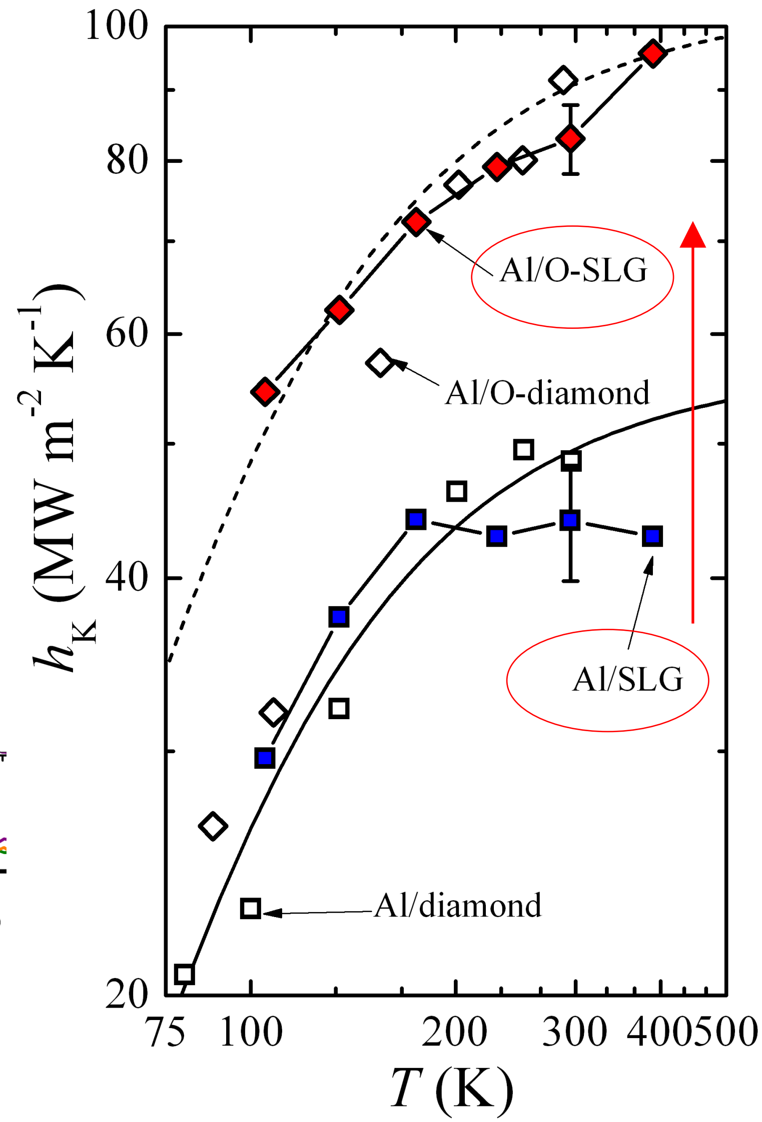
Appl. Phys. Lett. **96**, 231501

Collaboration: Scott Walton (NRL)

Atmospheric plasma functionalization of graphene surfaces





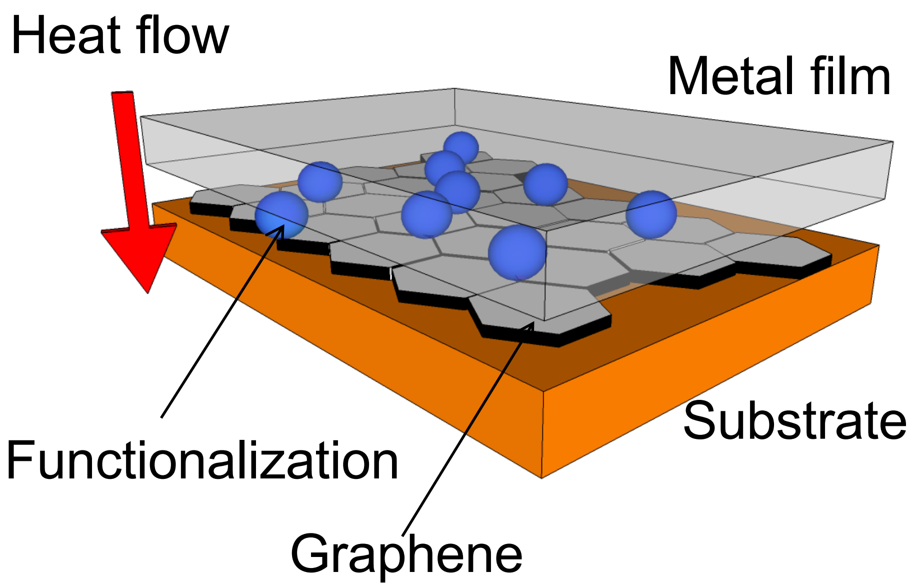


Nano Lett. **12**, 590 (2012)

- **Al/graphene interaction increased with oxygen bond (Al-O bond)**
- **But what implications does this have for SLG devices?**

Collaboration: Scott Walton (NRL)

Au/graphene electronic contacts

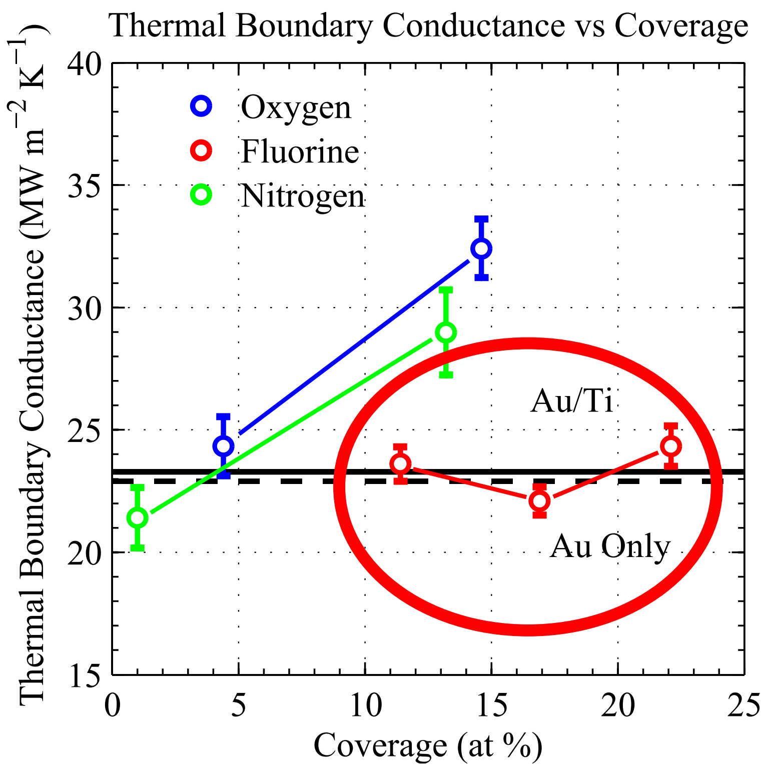


<u>Thermal boundary conductance results</u>

Similar trends for oxygen as Al

Ti adhesion layer does nothing

Fluorine does nothing

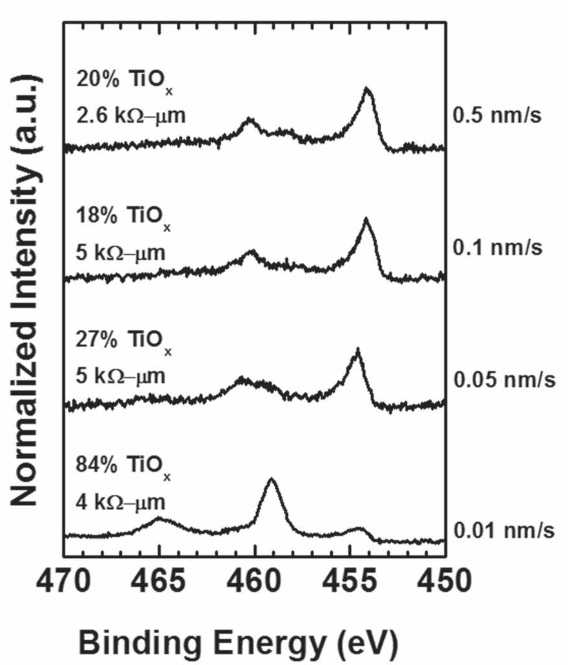


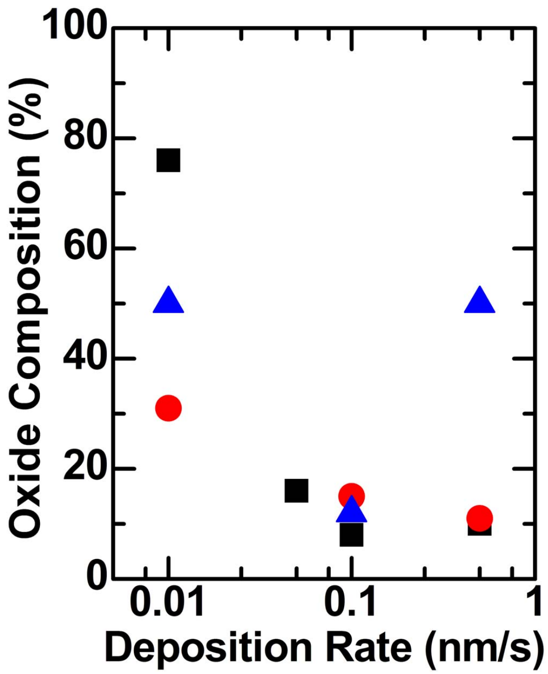
C-F bond inert does not want to interact!

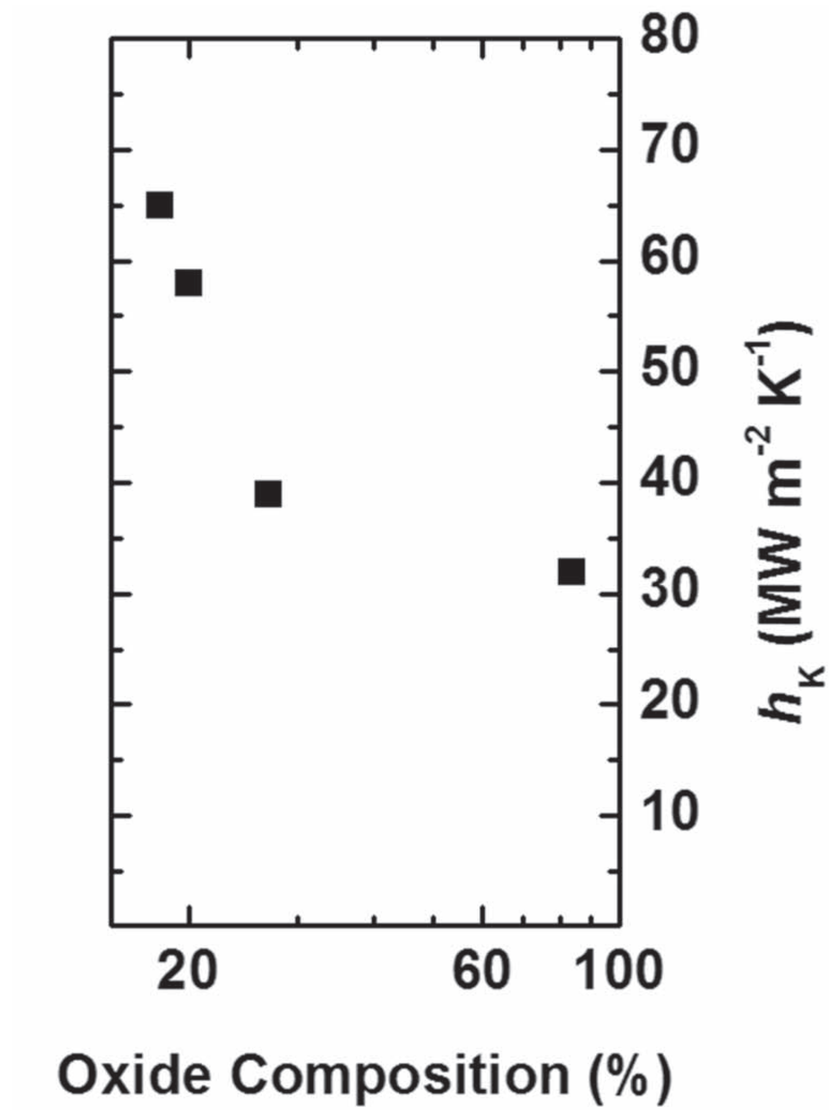
Nano Lett. **15**, 4876 (2015)

Collaboration: Scott Walton (NRL)

Chemistry effects on the TBC across Au/Ti/graphene





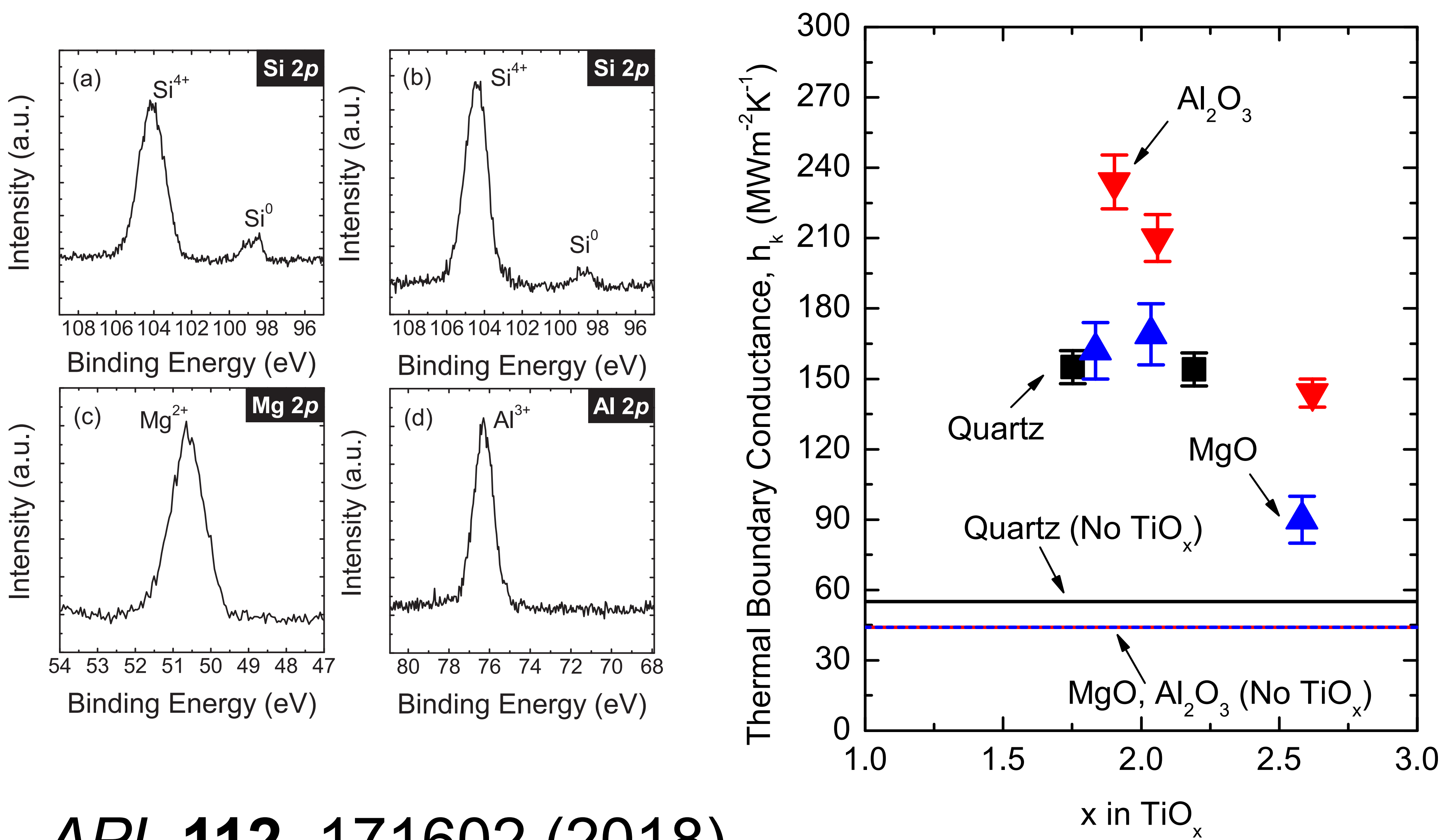


Nanotechnology (in press)

Collaboration: Stephen McDonnell (UVA)

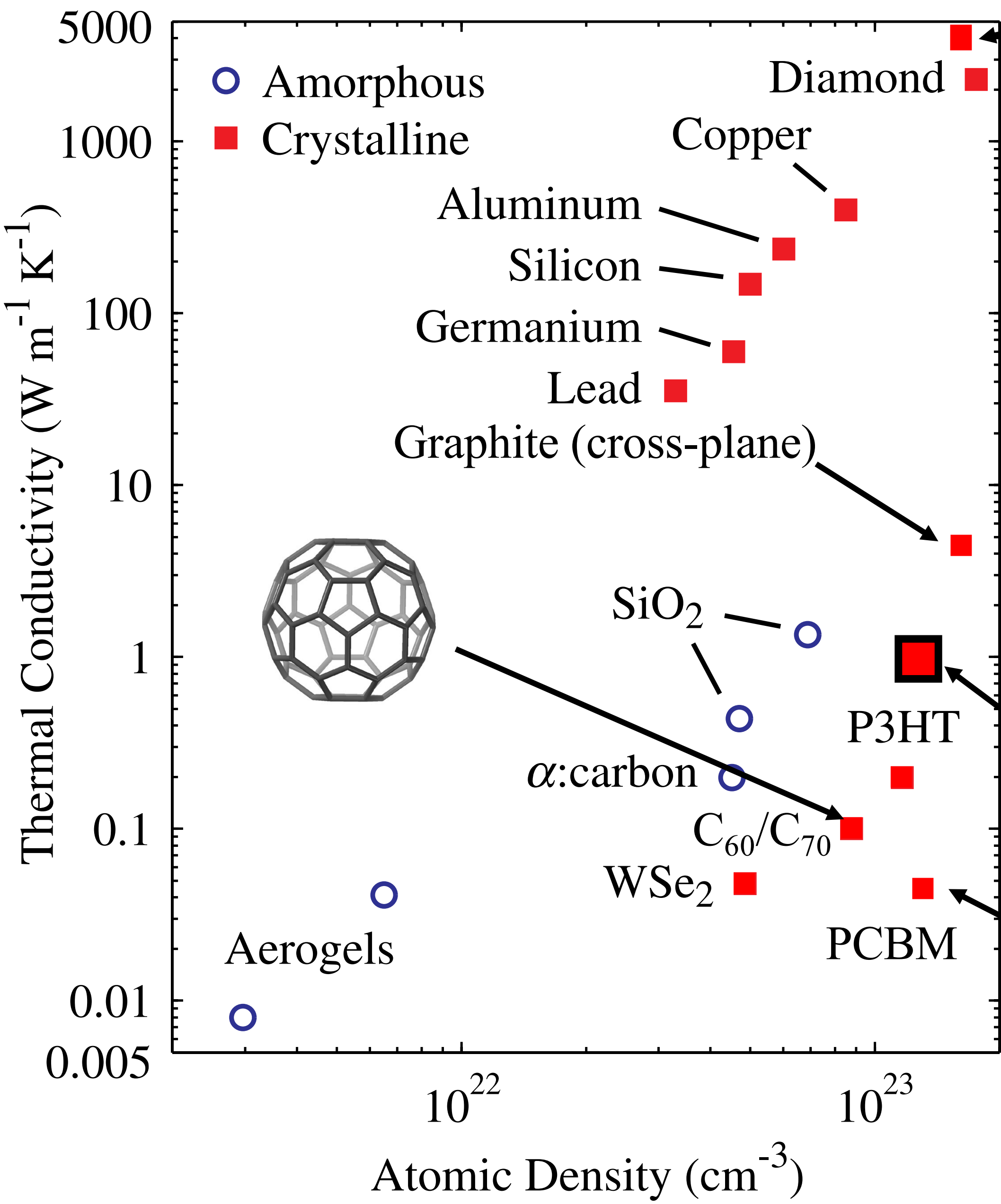
Chemical interactions influence bonding and TBC

Turns out defects can be a driving force that can drive chemical bonding and in turn influence TBC

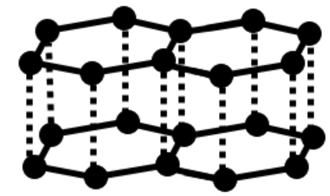


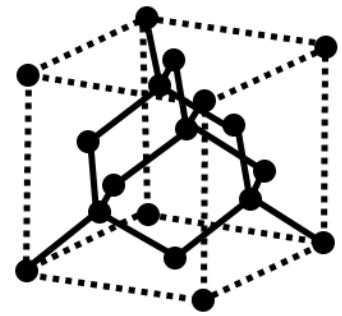
APL **112**, 171602 (2018)

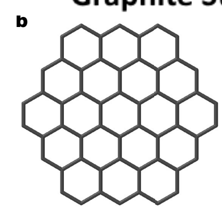
Let's talk about PCBM again in terms of vibrational mismatch

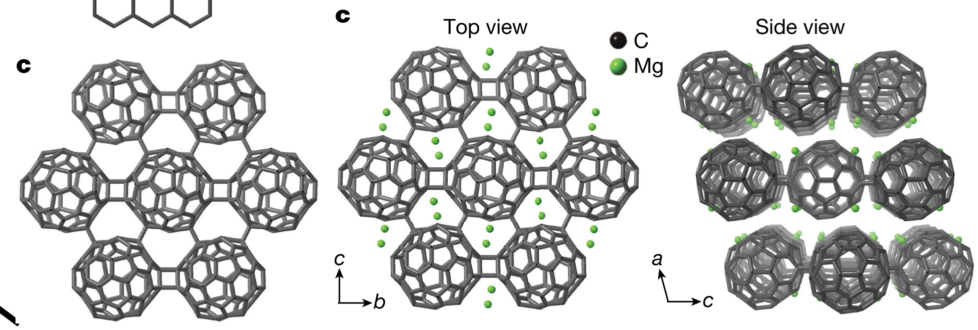


PRL **110**, 015902 (2013)

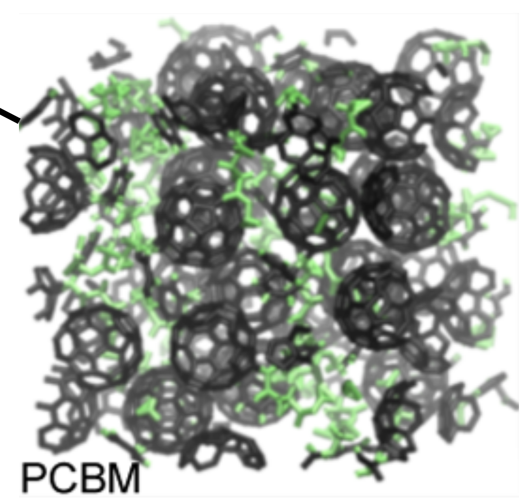




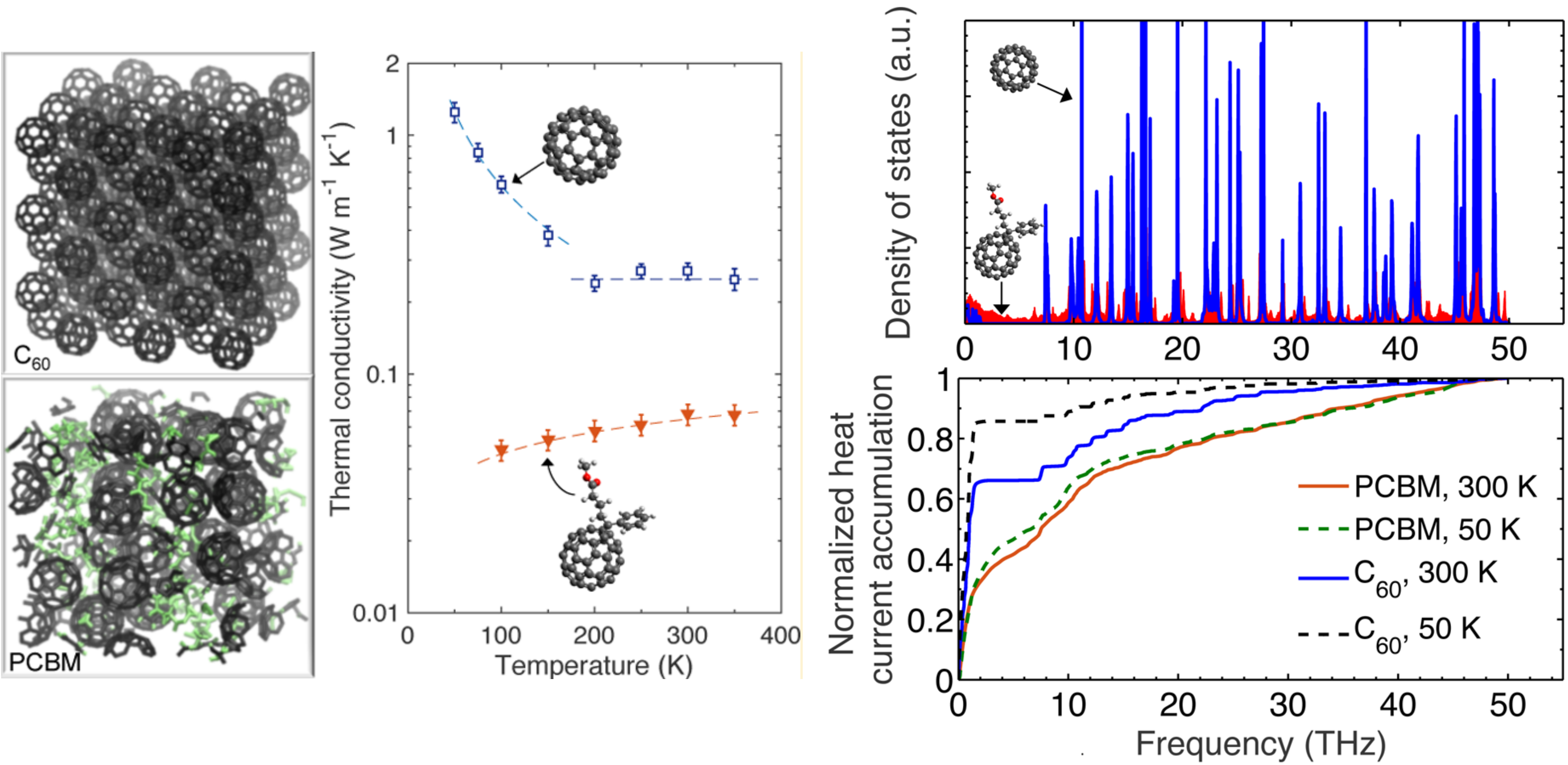




Graphullerne *Nature* 613, 71 (2023)



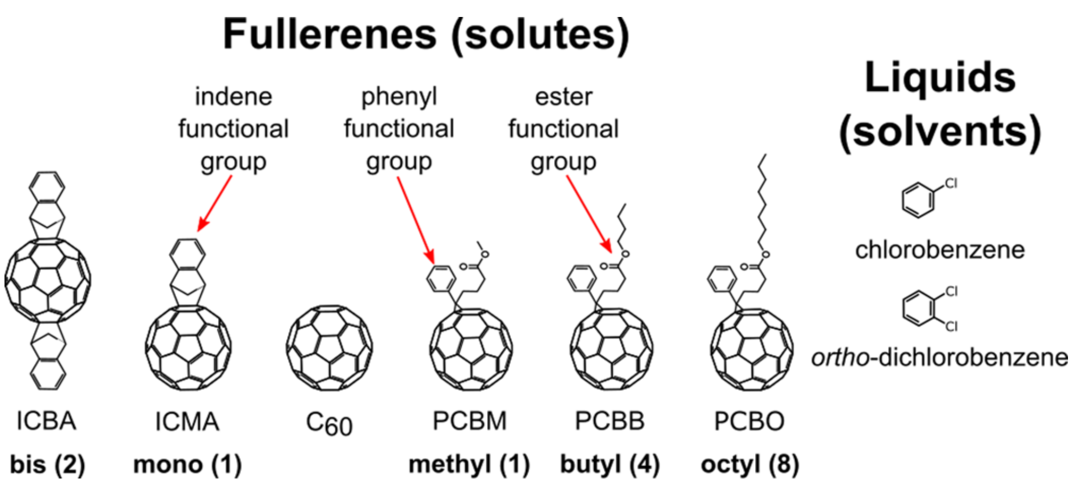
Turn to molecular dynamics simulations

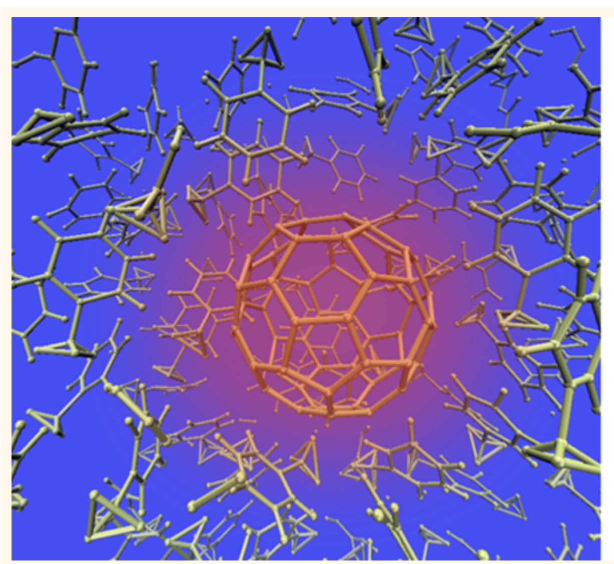


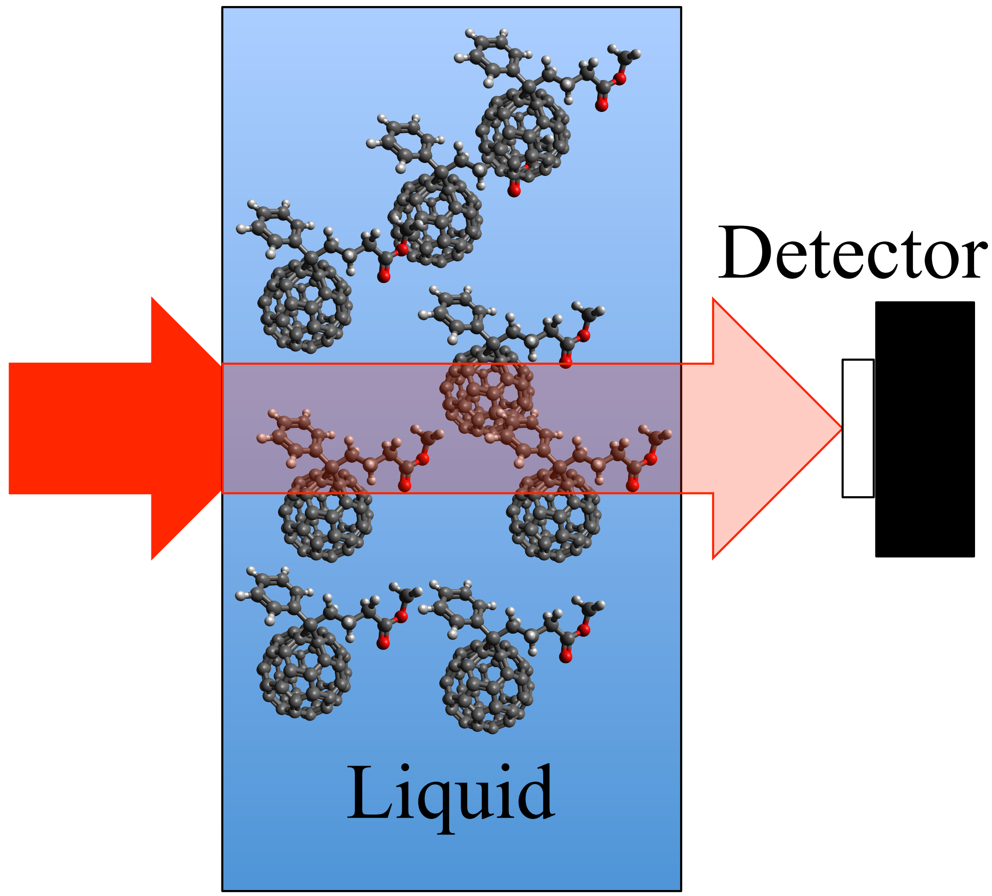
J. Phys. Chem. Lett. **8**, 2153 (2017)
Phys. Rev. B **96**, 220303 (2017)

Molecule/liquid thermal energy transfer in fullerenes

Can we study this spectral effects in fullerene derivatives more explicitly?



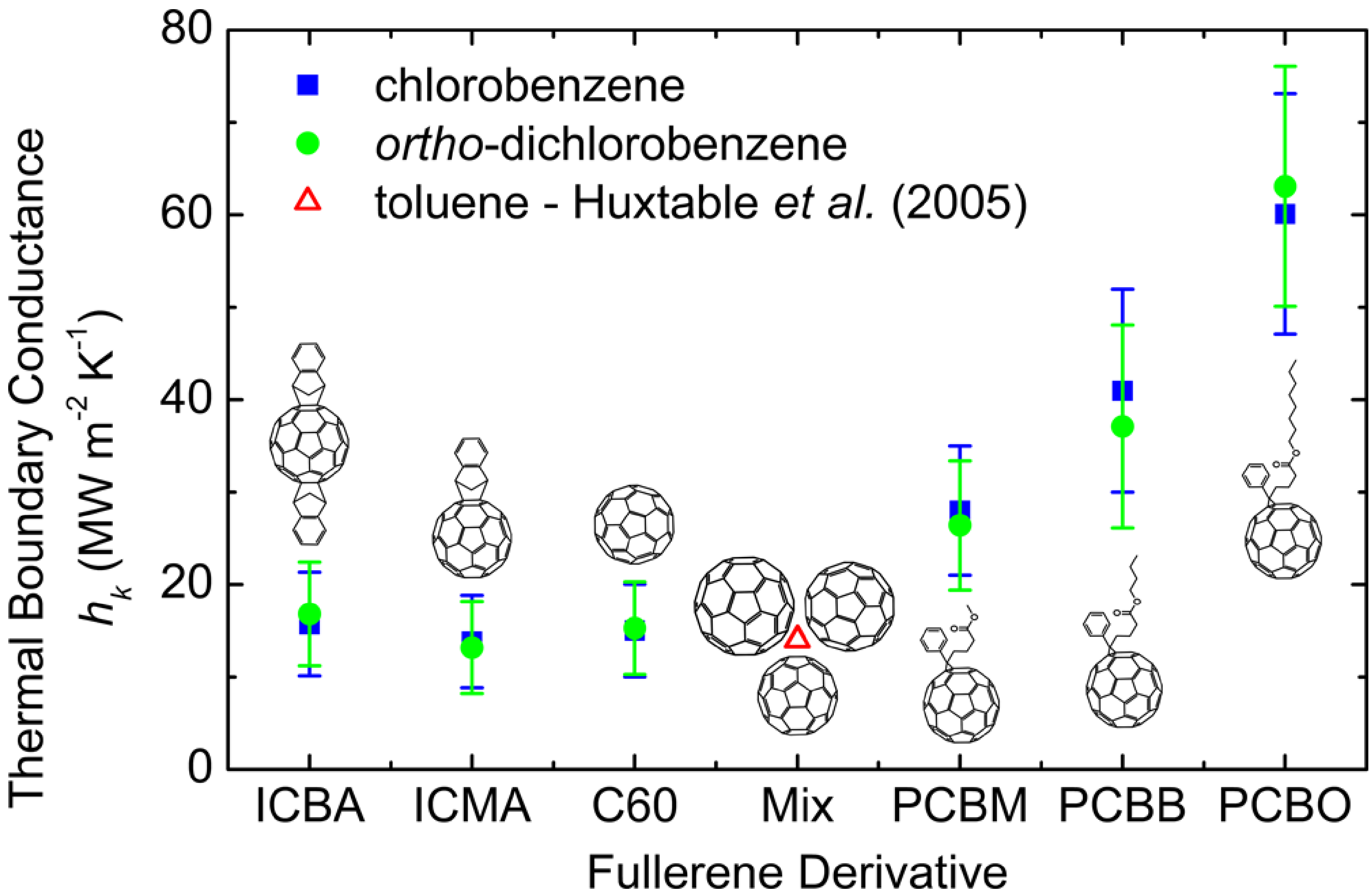




ACS Nano **11**, 1389

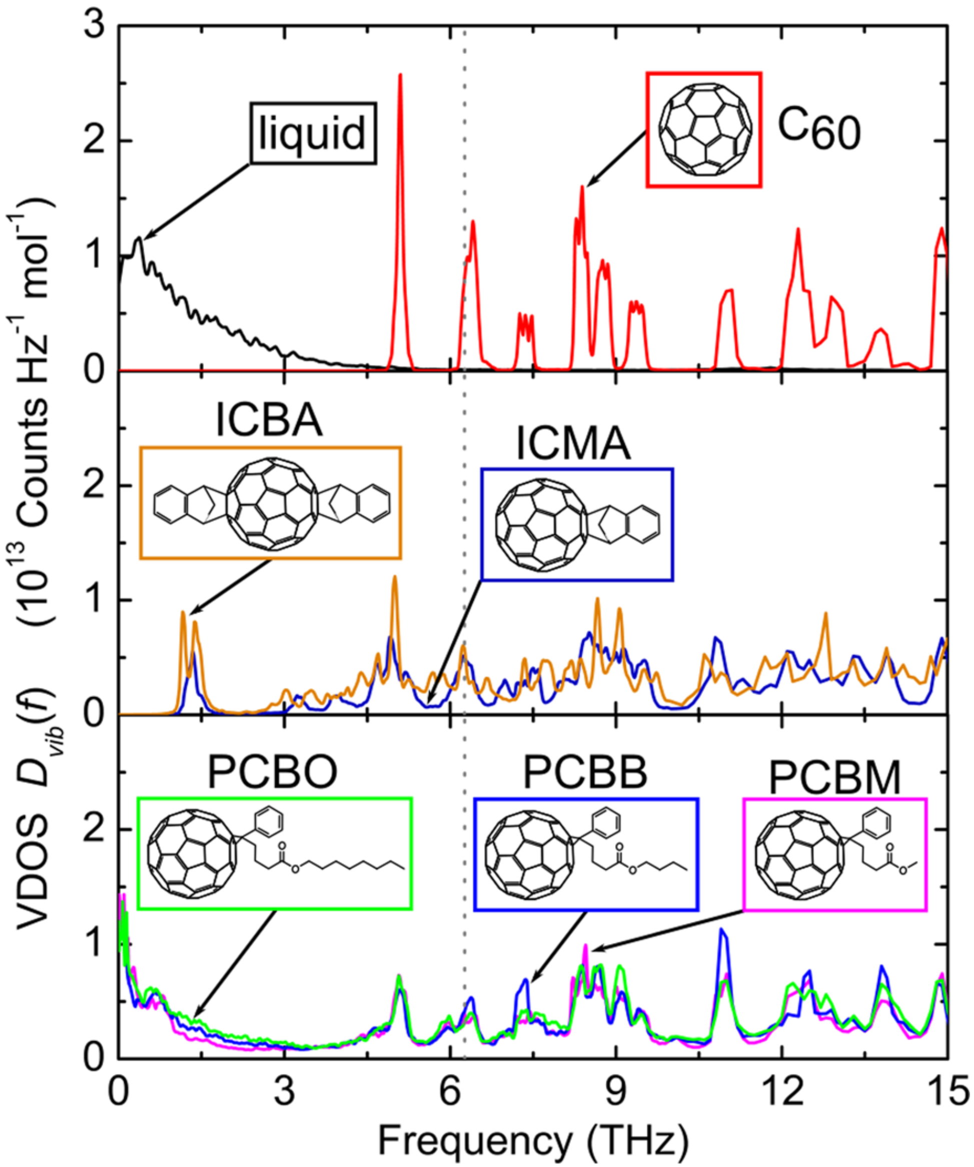
Molecule/liquid thermal energy transfer in fullerenes

Chemical moiety controls fullerene derivative vibrational energy coupling to a liquid



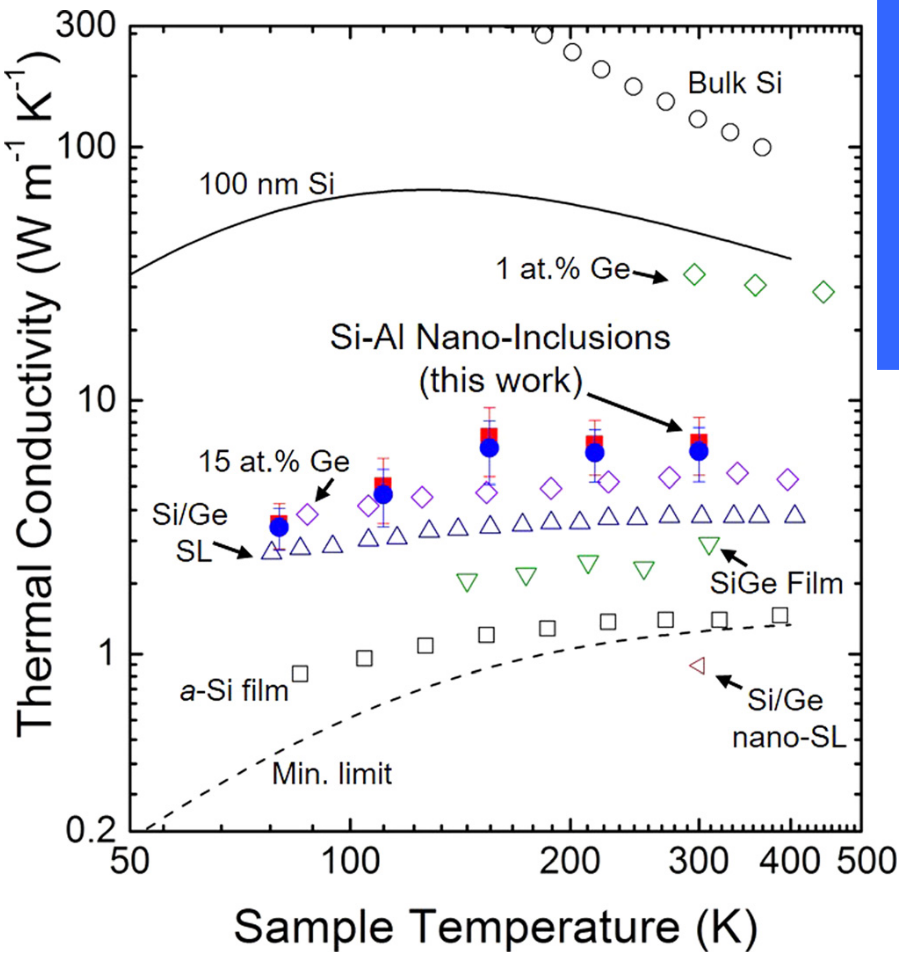
ACS Nano **11**, 1389

Molecule/liquid thermal energy transfer in fullerenes

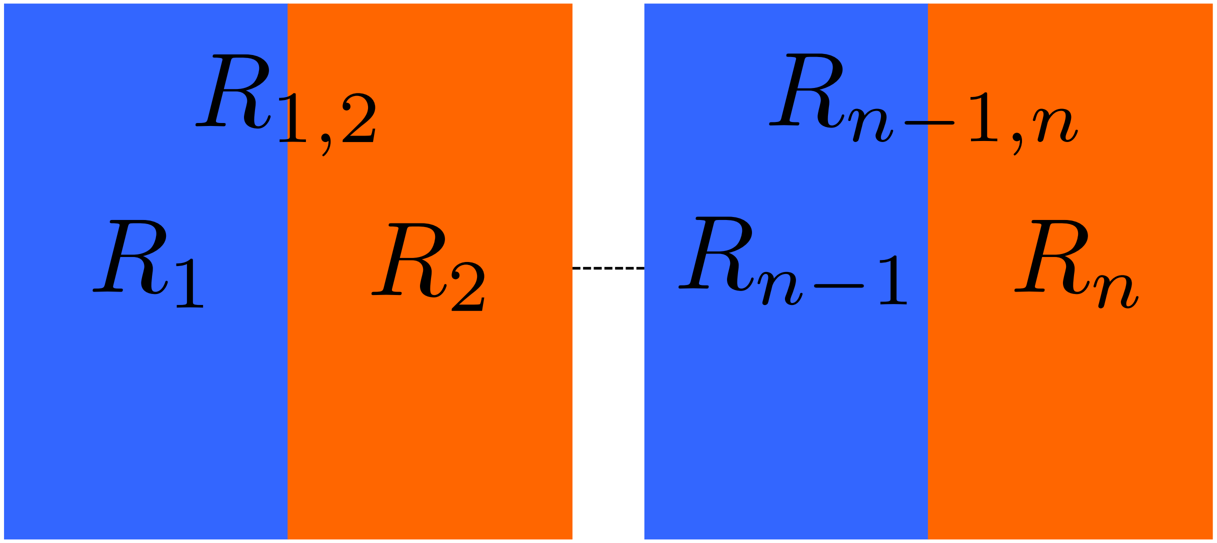


ACS Nano **11**, 1389

Phonon thermal conductivity in superlattices: incoherent



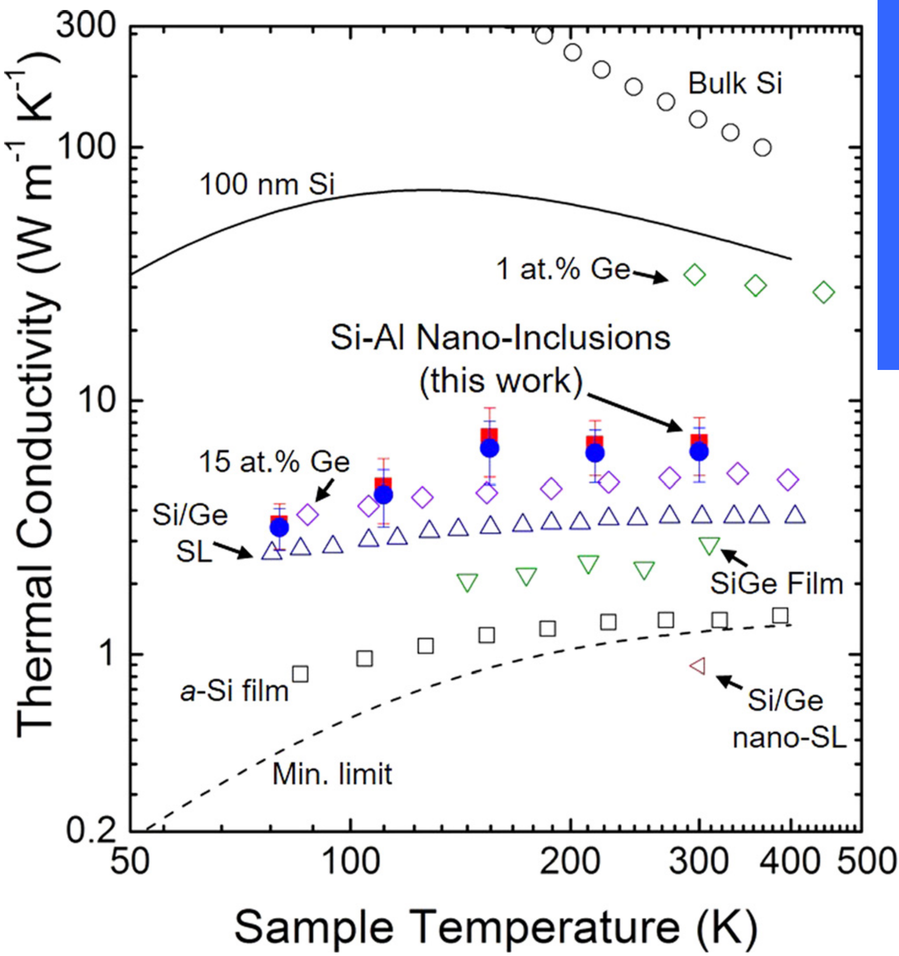
APL **112**, 213103



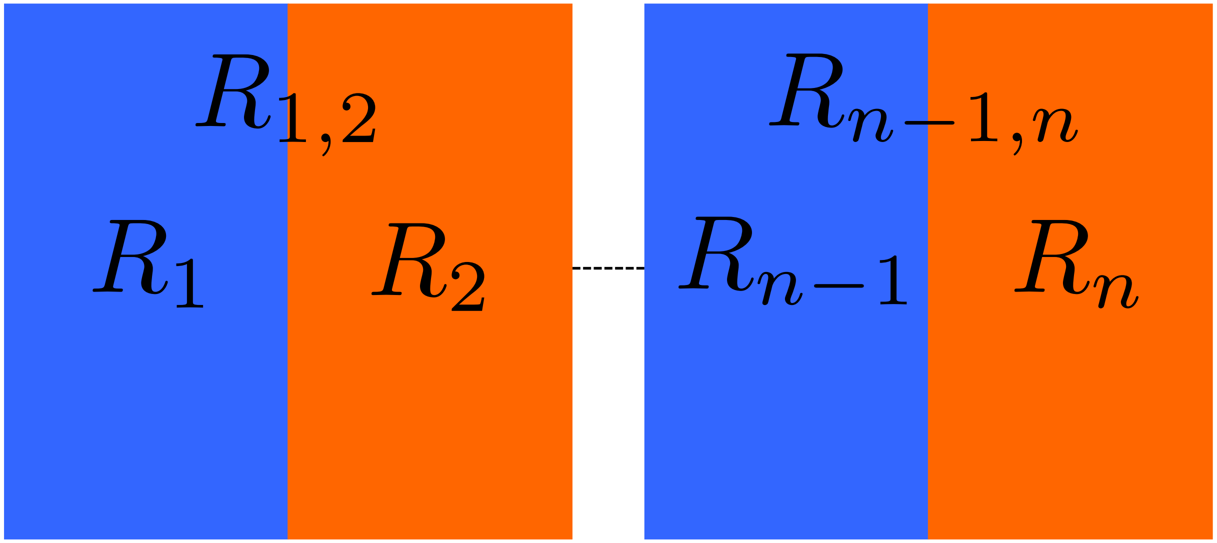
$$1/\kappa \propto 1/h = \sum_n R_j$$

Phonons scatter at every interface, and thus each interface offers a resistance to the overall thermal conductivity

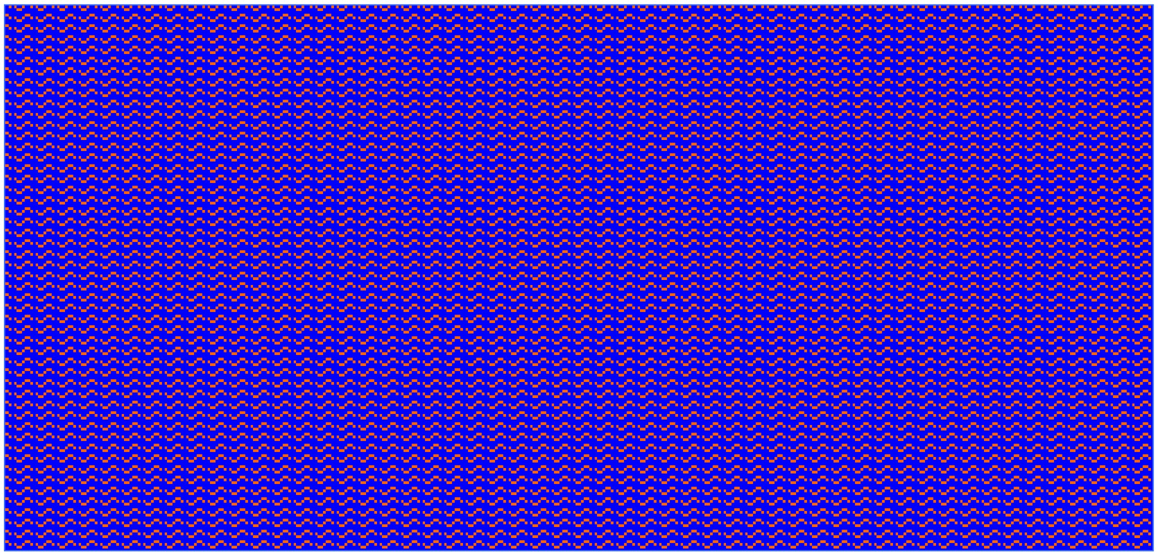
Phonon thermal conductivity in superlattices: coherent



APL **112**, 213103



What if layers are "linked"?
– coherent transport



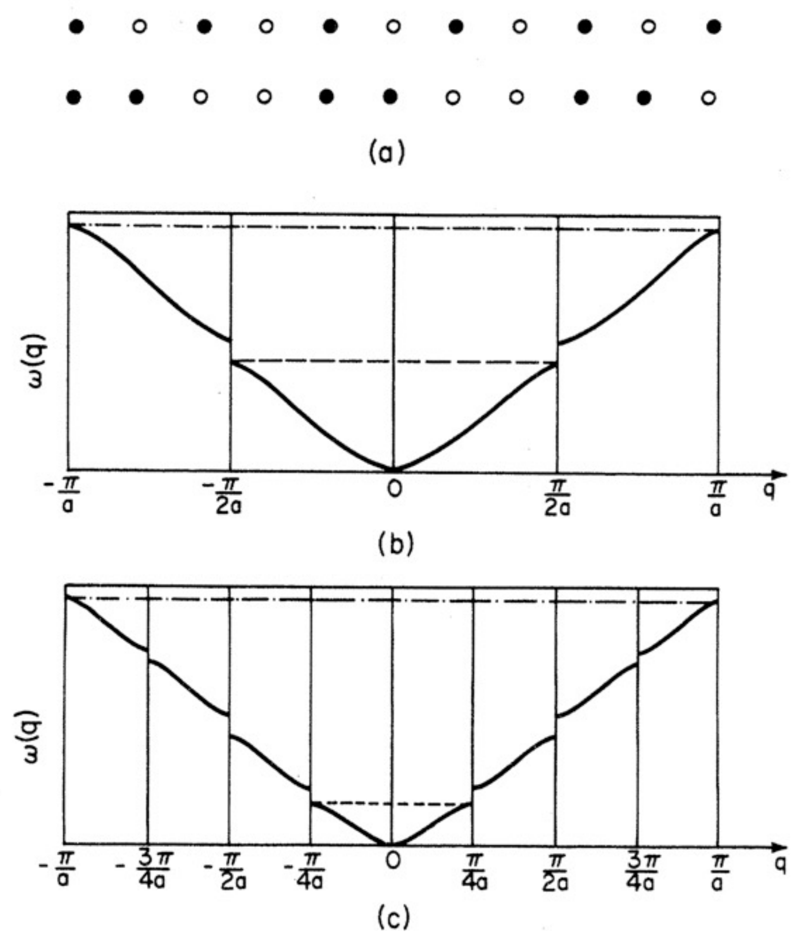
The minimum thermal conductivity of superlattices

PHYSICAL REVIEW B VOLUME 25, NUMBER 6 15 MARCH 1982

Thermal conductivity of superlattices

Shang Yuan Ren* and John D. Dow
Department of Physics and Coordinated Science Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

(Received 21 September 1981)



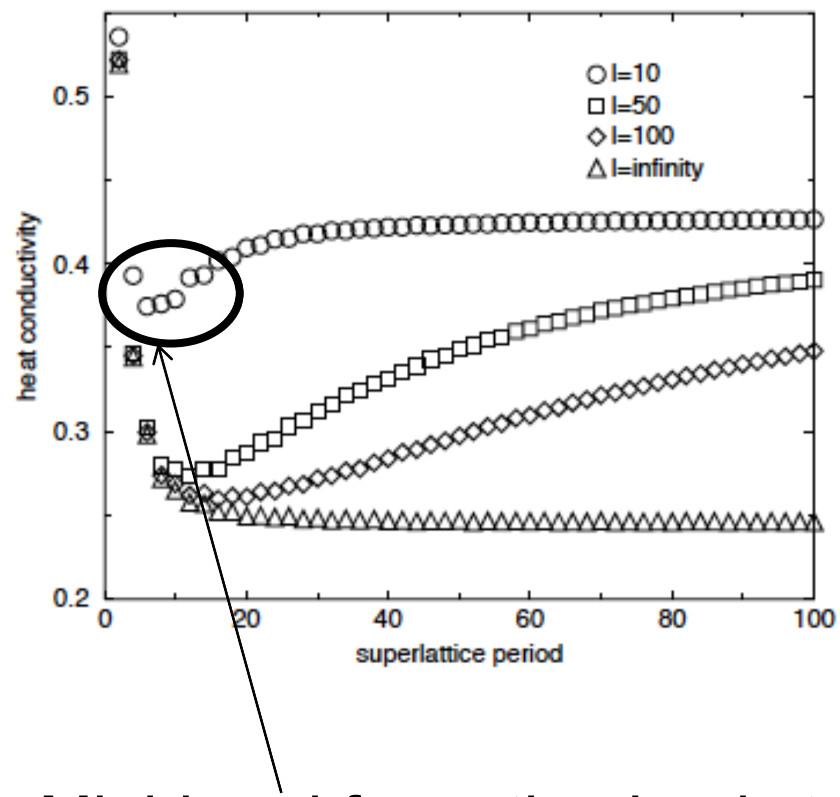
Interfacial periodicity can lead to “mini-band” formation

VOLUME 84, NUMBER 5 PHYSICAL REVIEW LETTERS 31 JANUARY 2000

Minimum Thermal Conductivity of Superlattices

M. V. Simkin and G. D. Mahan
Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996-1200
and Solid State Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831

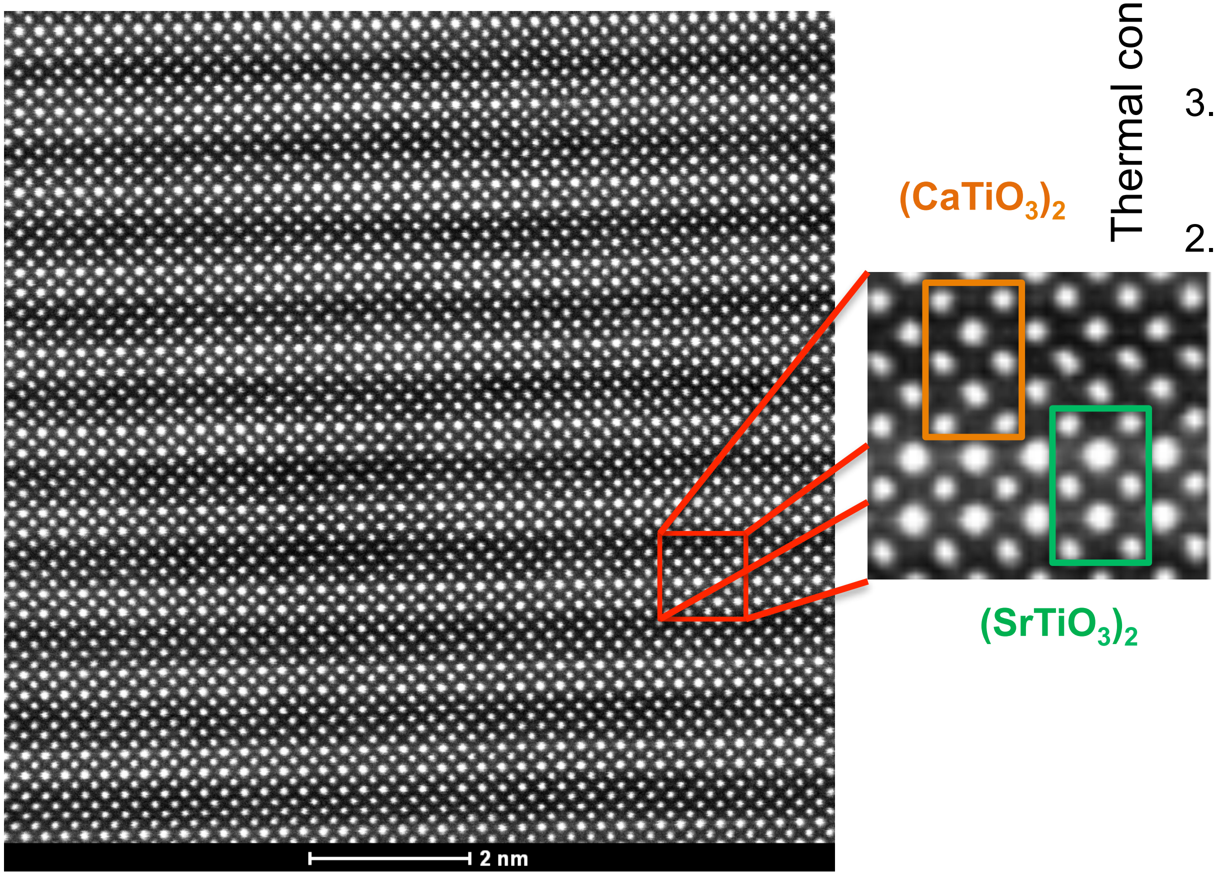
(Received 23 July 1999)

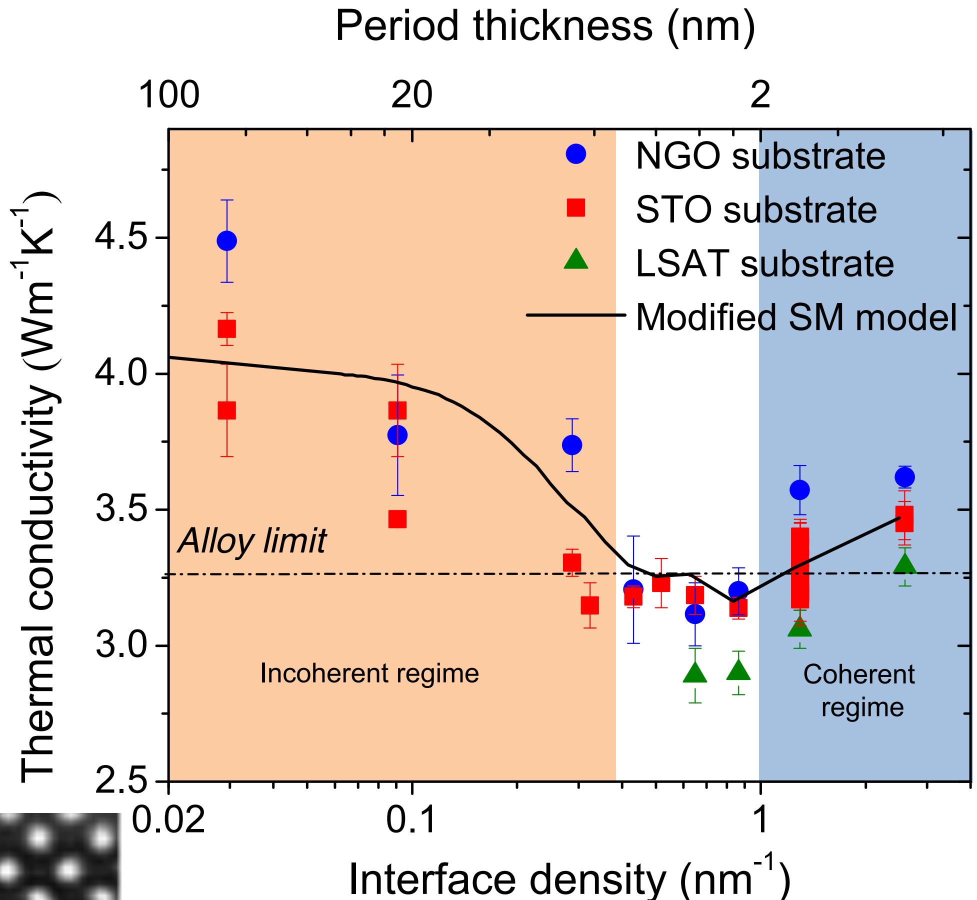


Mini-band formation leads to a minimum in the superlattice thermal conductivity

Experimental evidence of minimum thermal conductivity

Nature Materials **13**, 168 (2013)

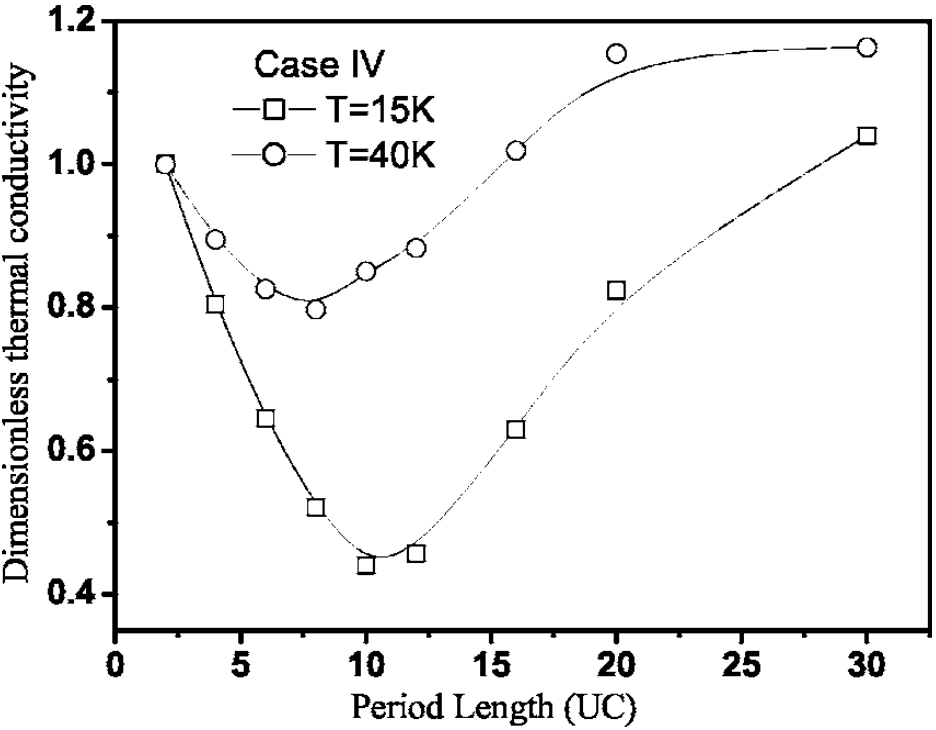


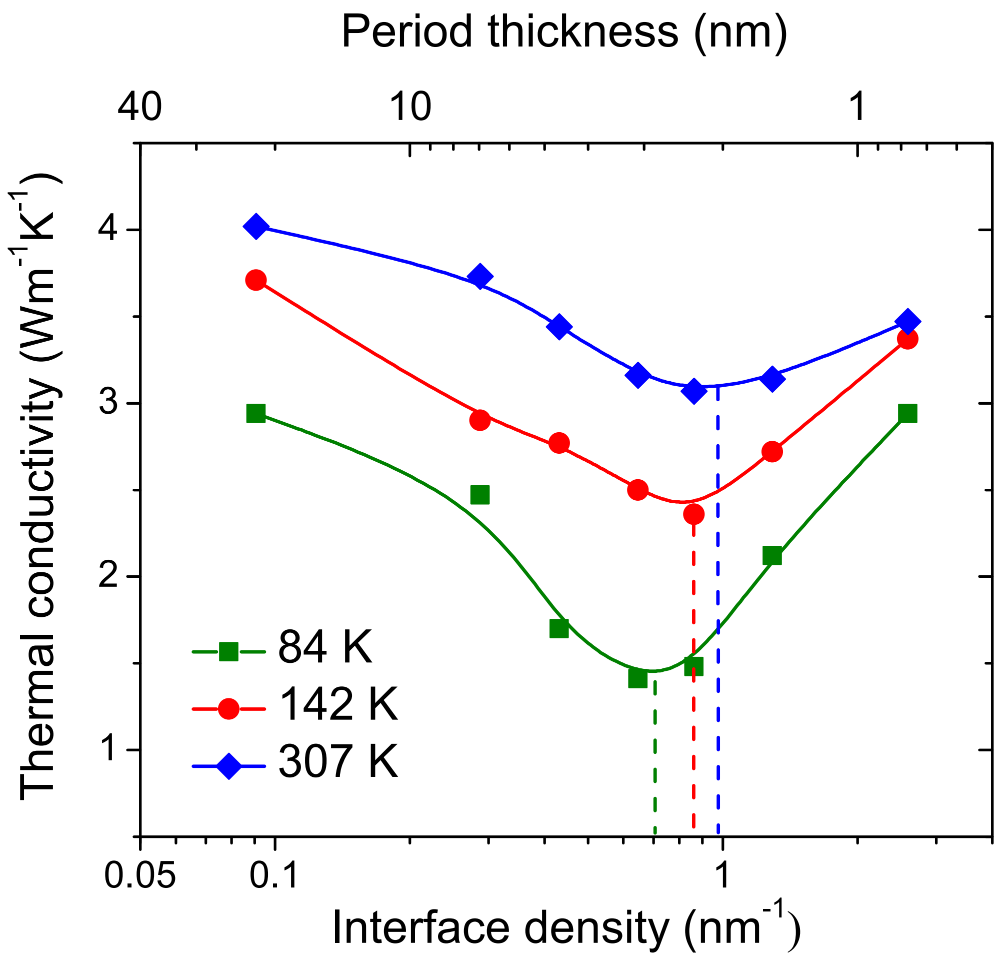


Growth PLD:
J. Ravichandran
A. Yadav
R. Ramesh
A. Majumdar

Experimental evidence of minimum thermal conductivity

- More pronounced minimum at low T, thermal conductivity measurements show trends of mini-band formation
- MD simulation (left), mini-band = phonon bandgap (PRB **72**, 174302)

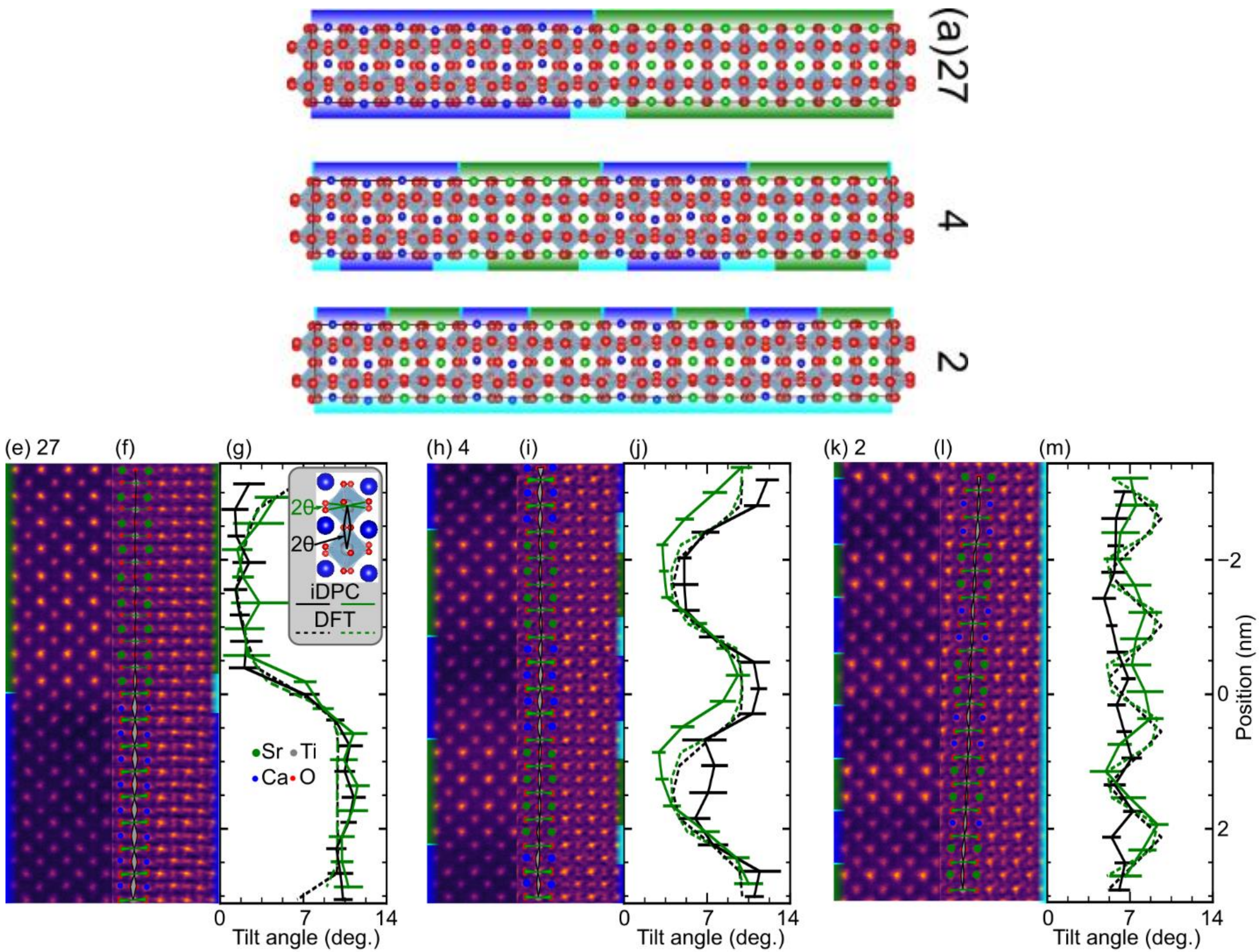




SL design to manipulate coherent phonon transport

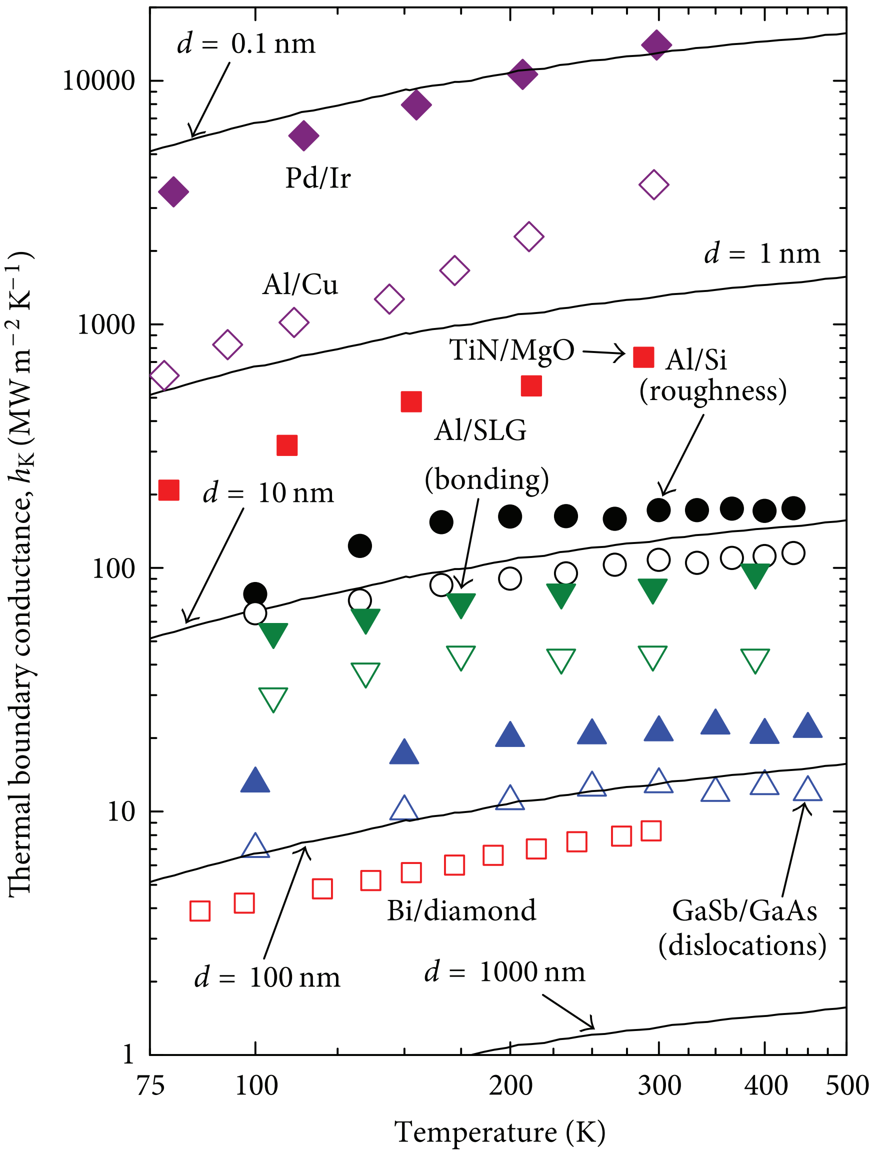
Nature Materials **13**, 168 (2013)

Experimental evidence of phonon coherence

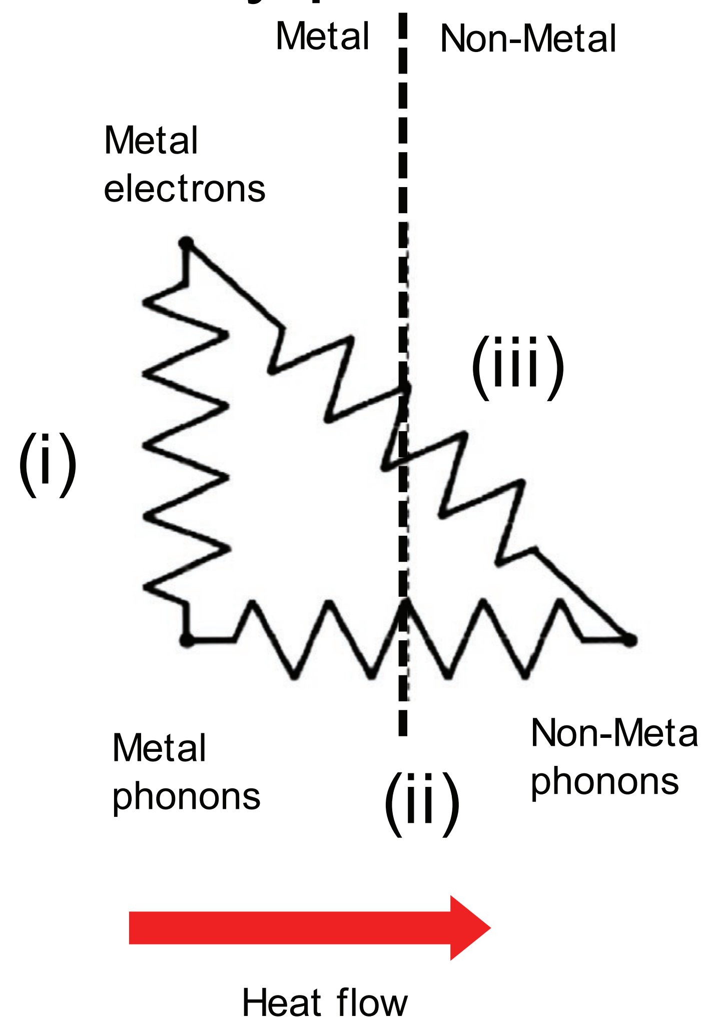


Nature **601**, 556 (2022)

Ultrahigh electron-electron TBC

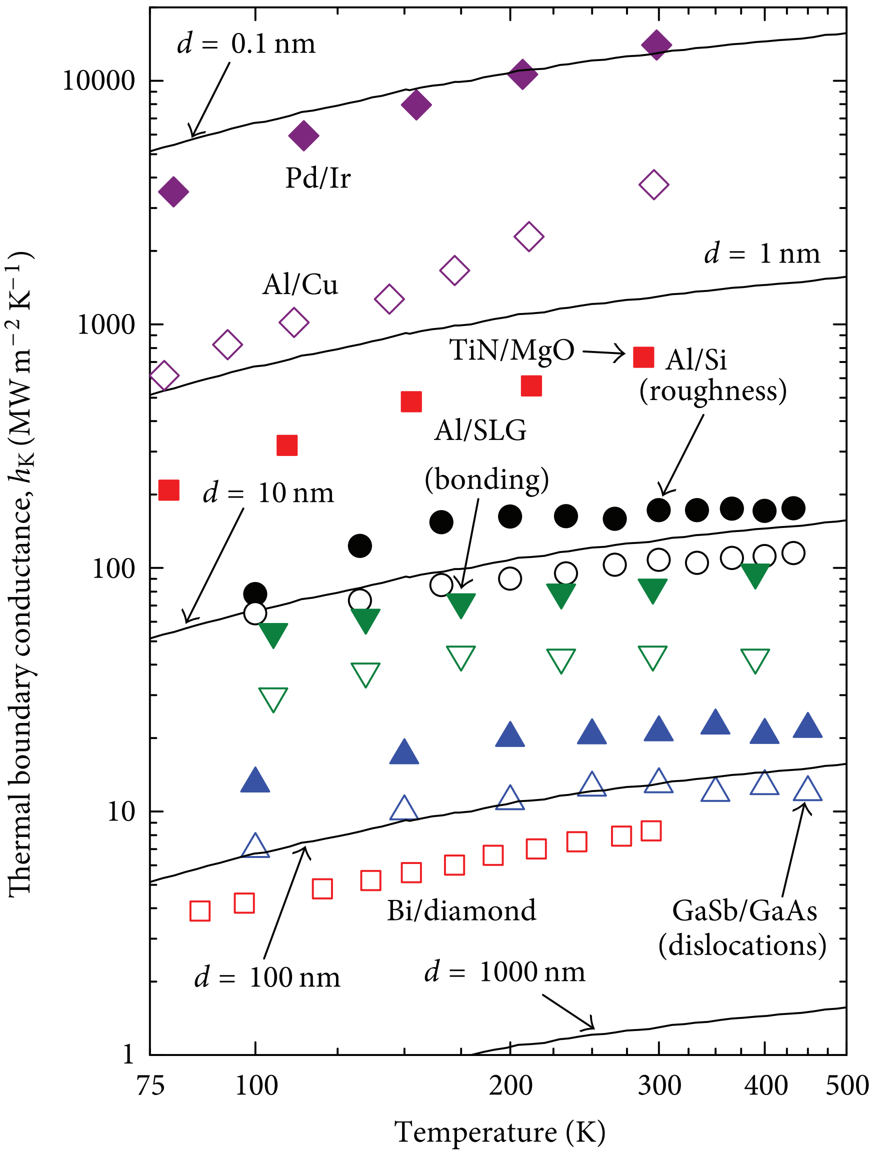


Metal/insulator interfaces dominated by phonon transport

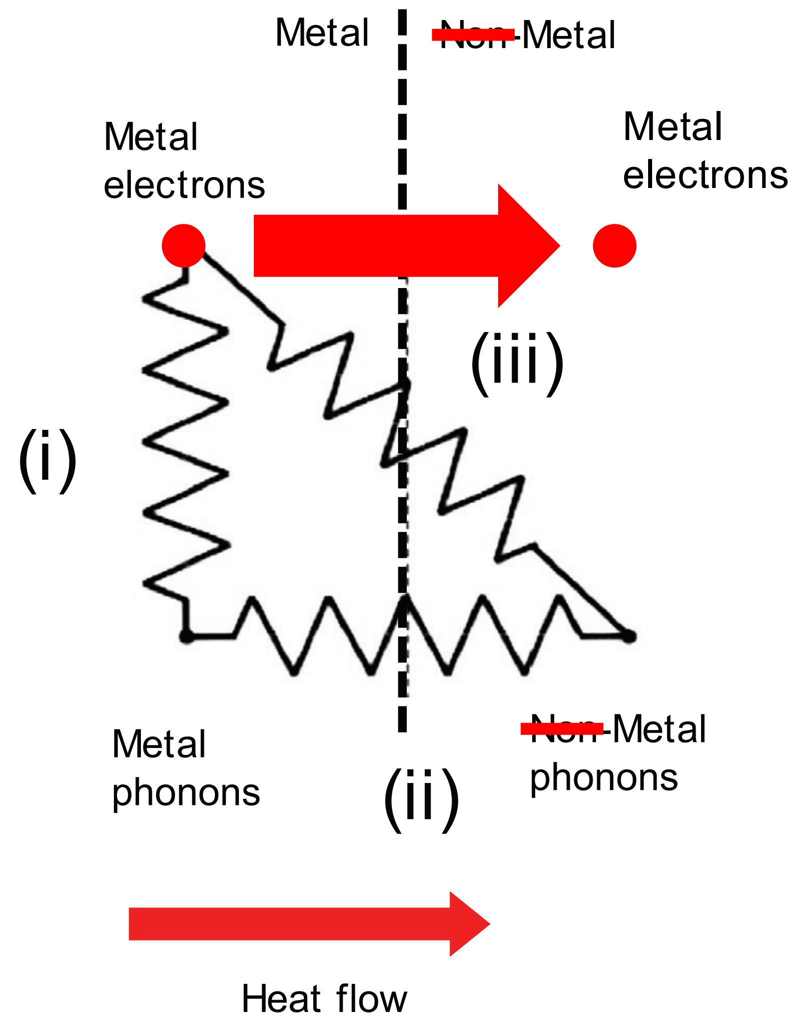


Hopkins *ISRN Mech. Eng.* **2013** 682586 (2013)

Ultrahigh electron-electron TBC

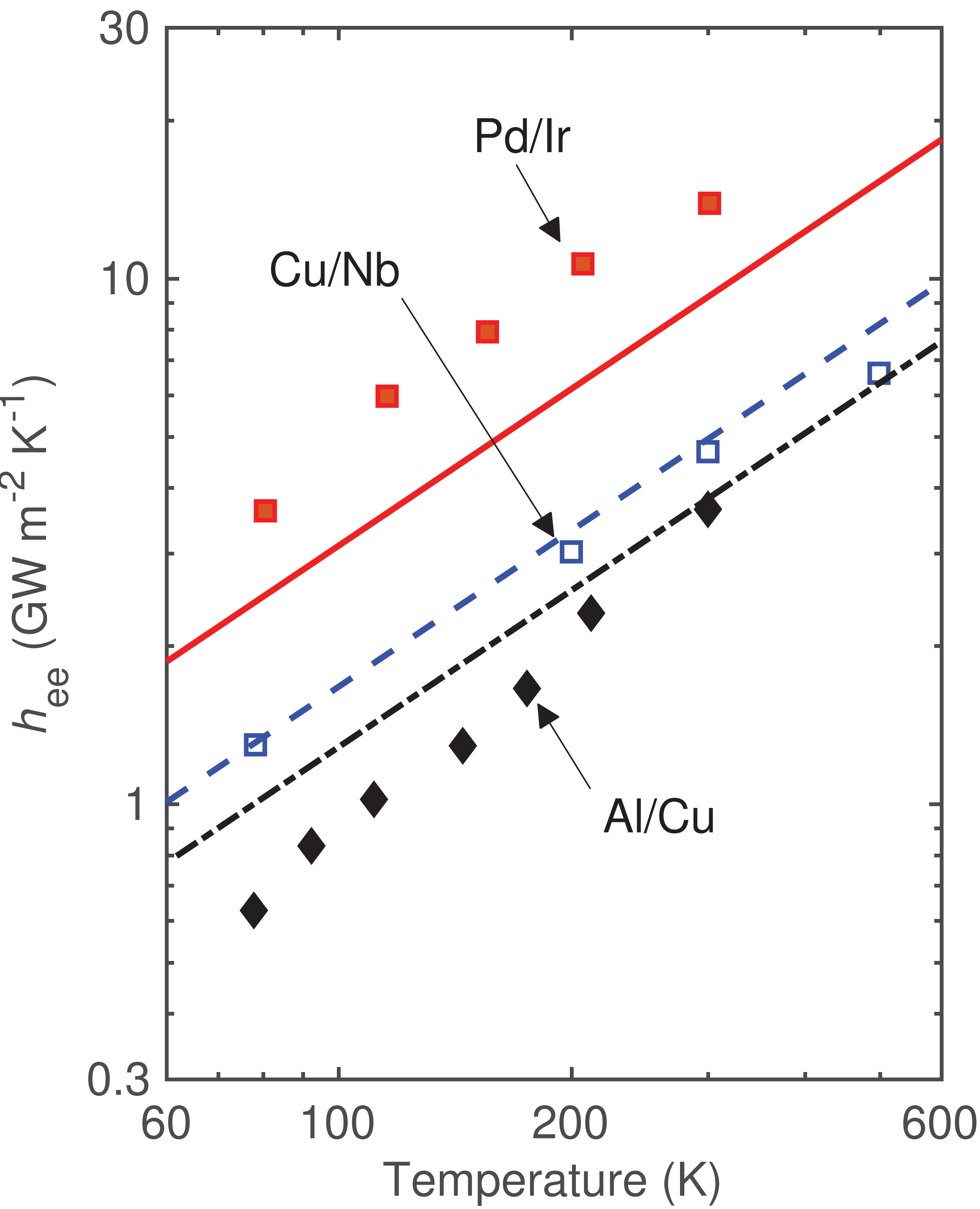


Related to DOS of electrons at Fermi surface of materials



Hopkins *ISRN Mech. Eng.* **2013** 682586 (2013)

Ultrahigh electron-electron TBC



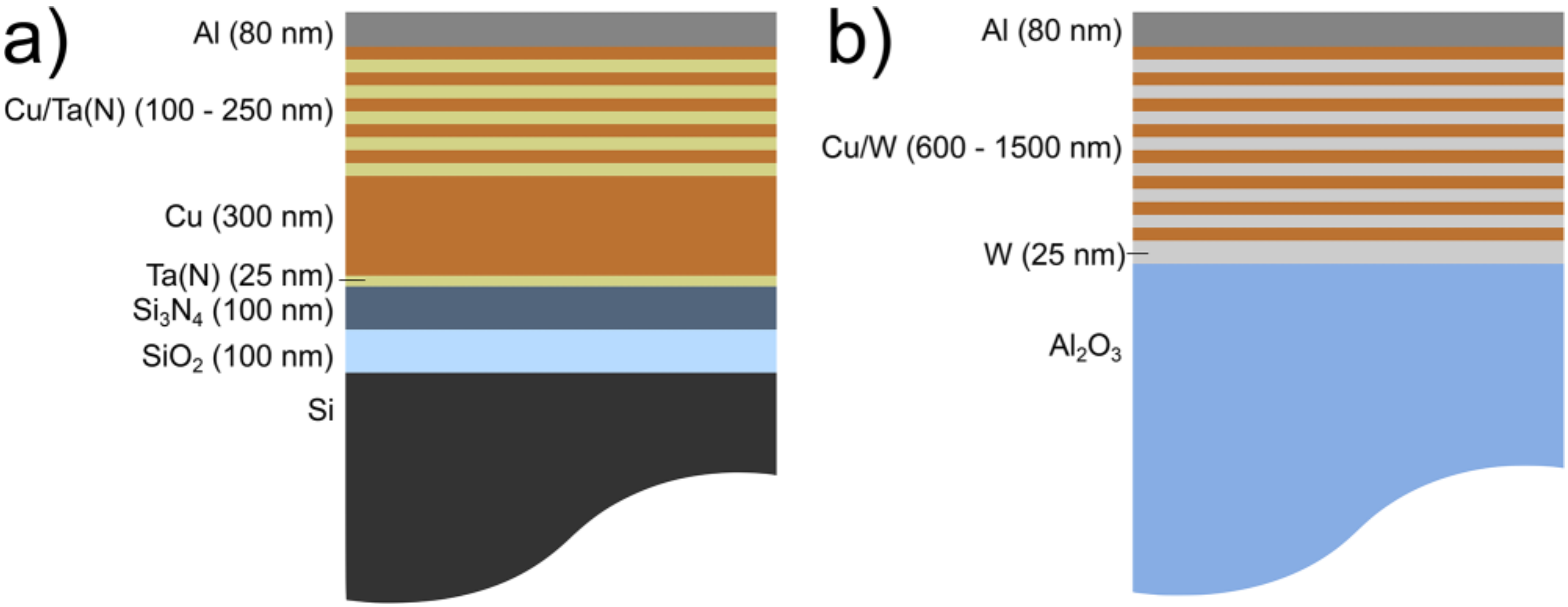
Similar to phonon-dominated conduction: materials with high electron thermal conductivities do not necessarily result in high TBC interfaces

APL **106** 093114

Metal/metal multilayers

But how do defects/structure/etc impact electron-electron thermal boundary conductance?

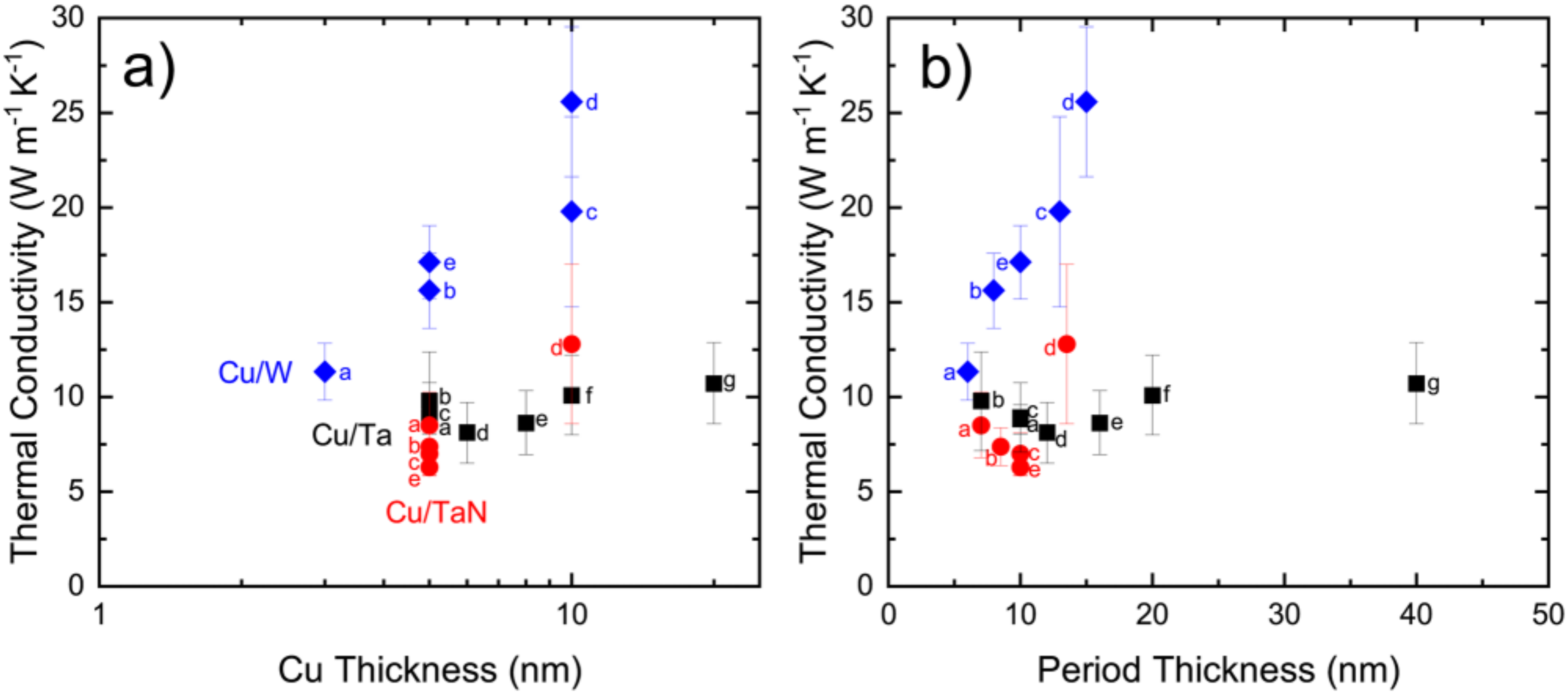
Collaboration with Claudia Cancellieri Lars Jeurgens and Giacomo Lorenzin



Cancellieri *J. Appl. Phys.* **128** 195302 (2020)

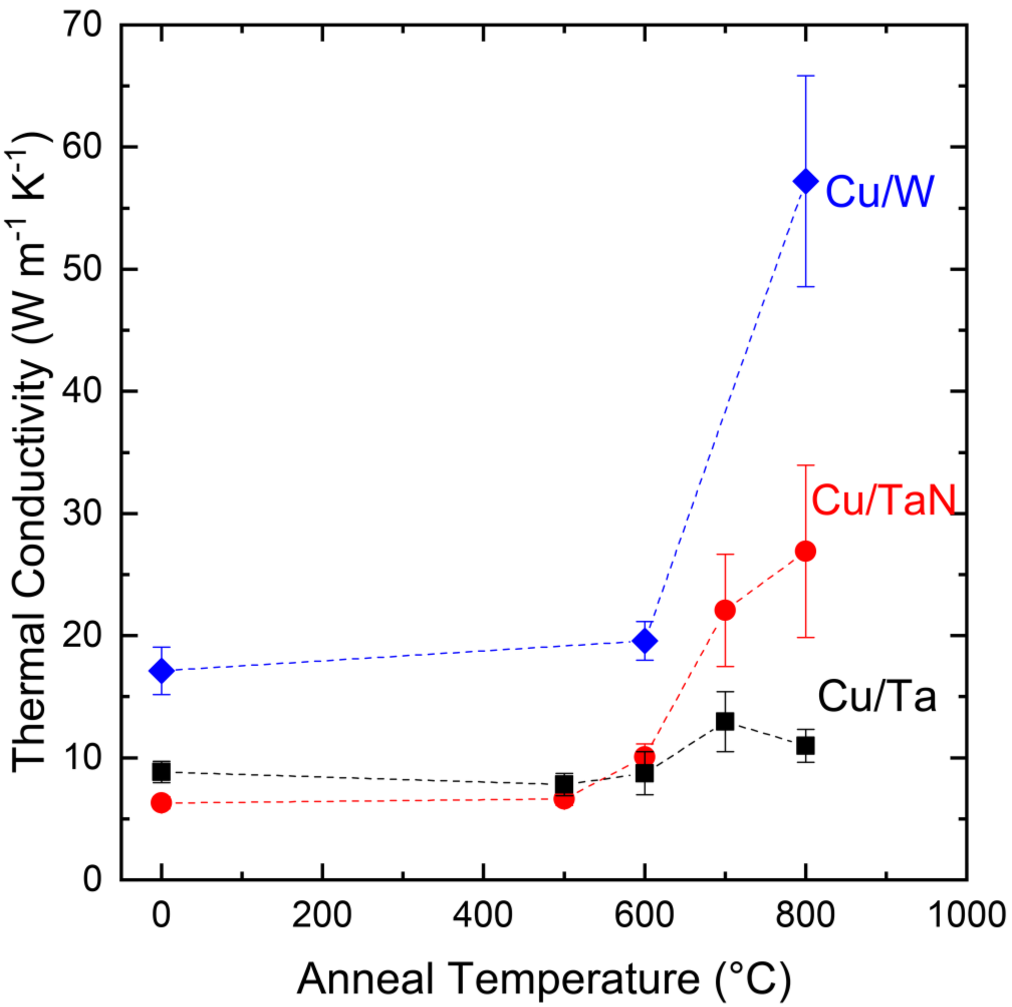
Interfacial resistances dictate metal/metal multilayers

High thermal conductors not that high anymore due to electron-driven interfacial resistances



Cancellieri *J. Appl. Phys.* **128** 195302 (2020)

Interfacial resistances dictate metal/metal multilayers

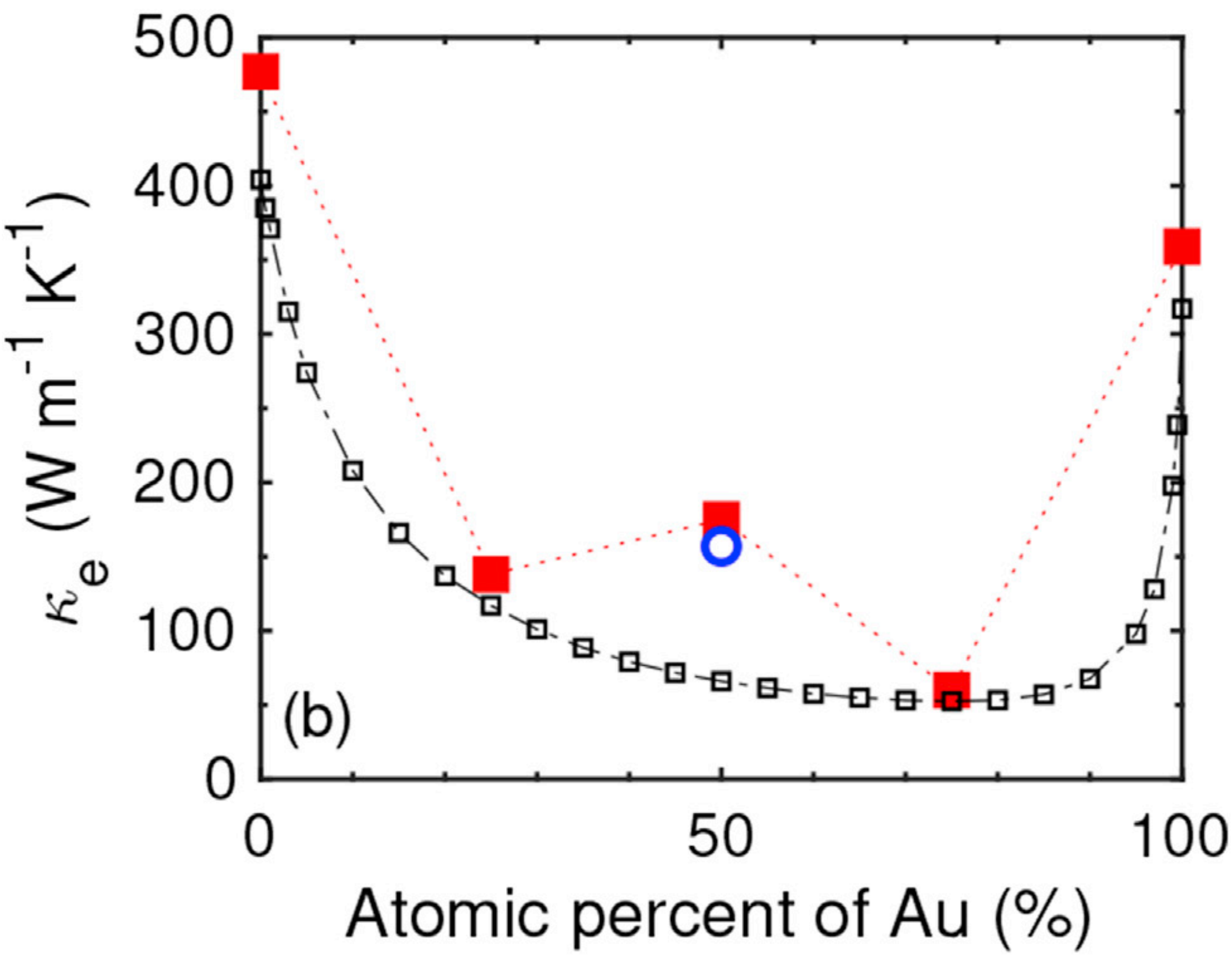


Annealing promotes diffusion, removes interface resistance, but still reduced electron conductivity due to compositions/alloy effects

Cancellieri *J. Appl. Phys.* **128** 195302 (2020)

Electron scattering with composition changes and defects decreases thermal conductivity – scattering + bandstructure

Example of Au_xCu_{1-x} alloys (computational)

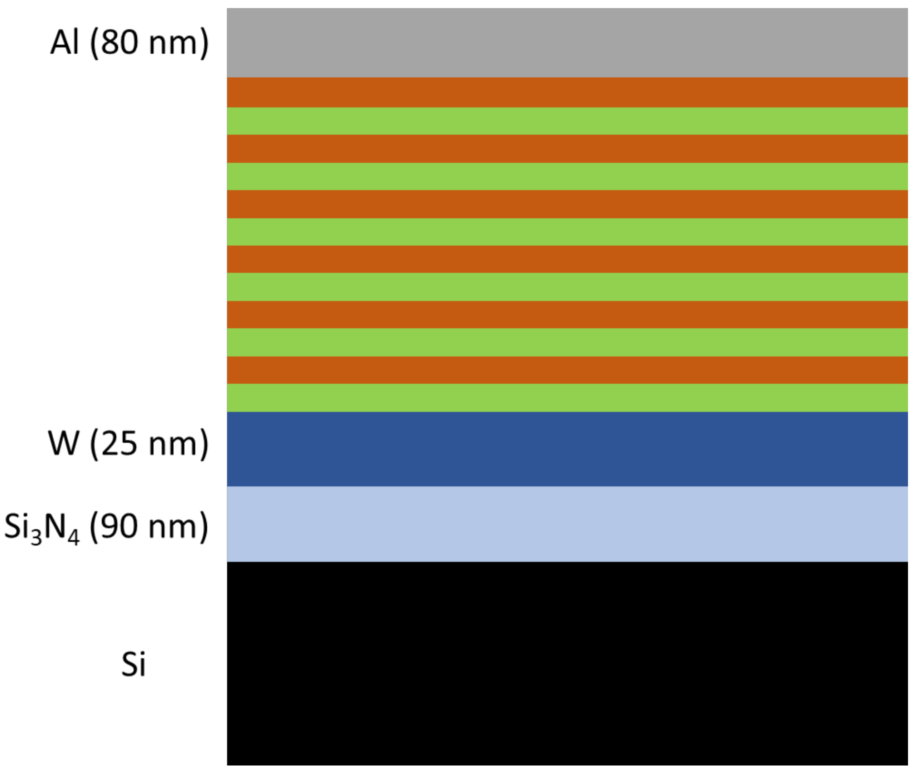


Mat. Today Phys. **12** 100175 (2020)

Can we control thermal conductivity in metal multilayers?

What we know: Metal multilayer thermal conductivity dictated by interface density and defects (compositions)

Hypothesis: Strain engineering to control structure and scattering rates



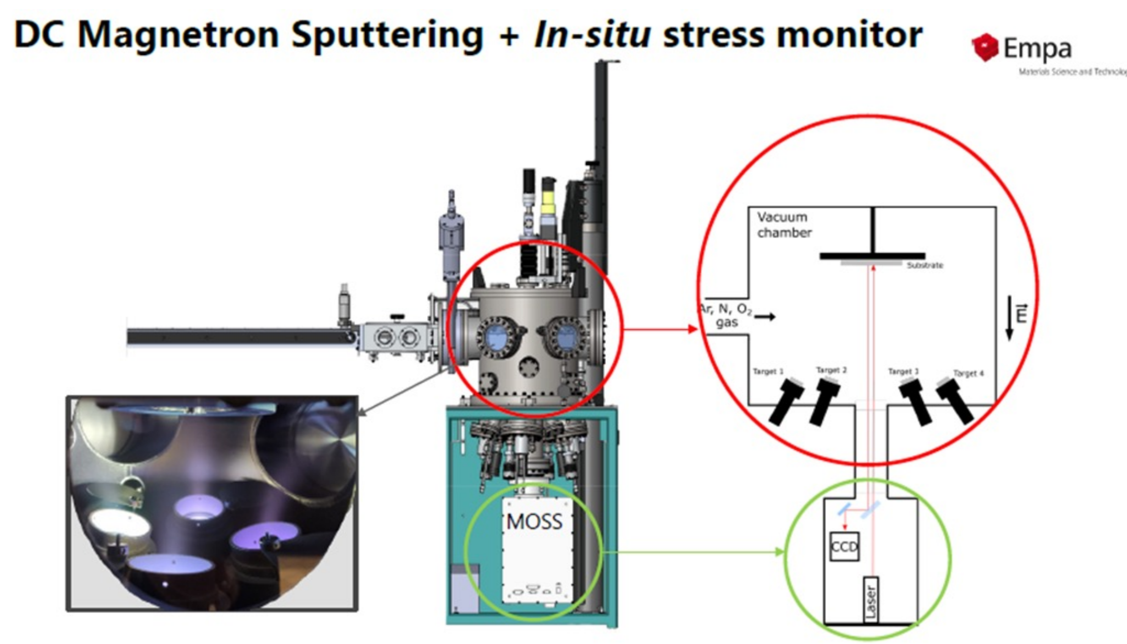
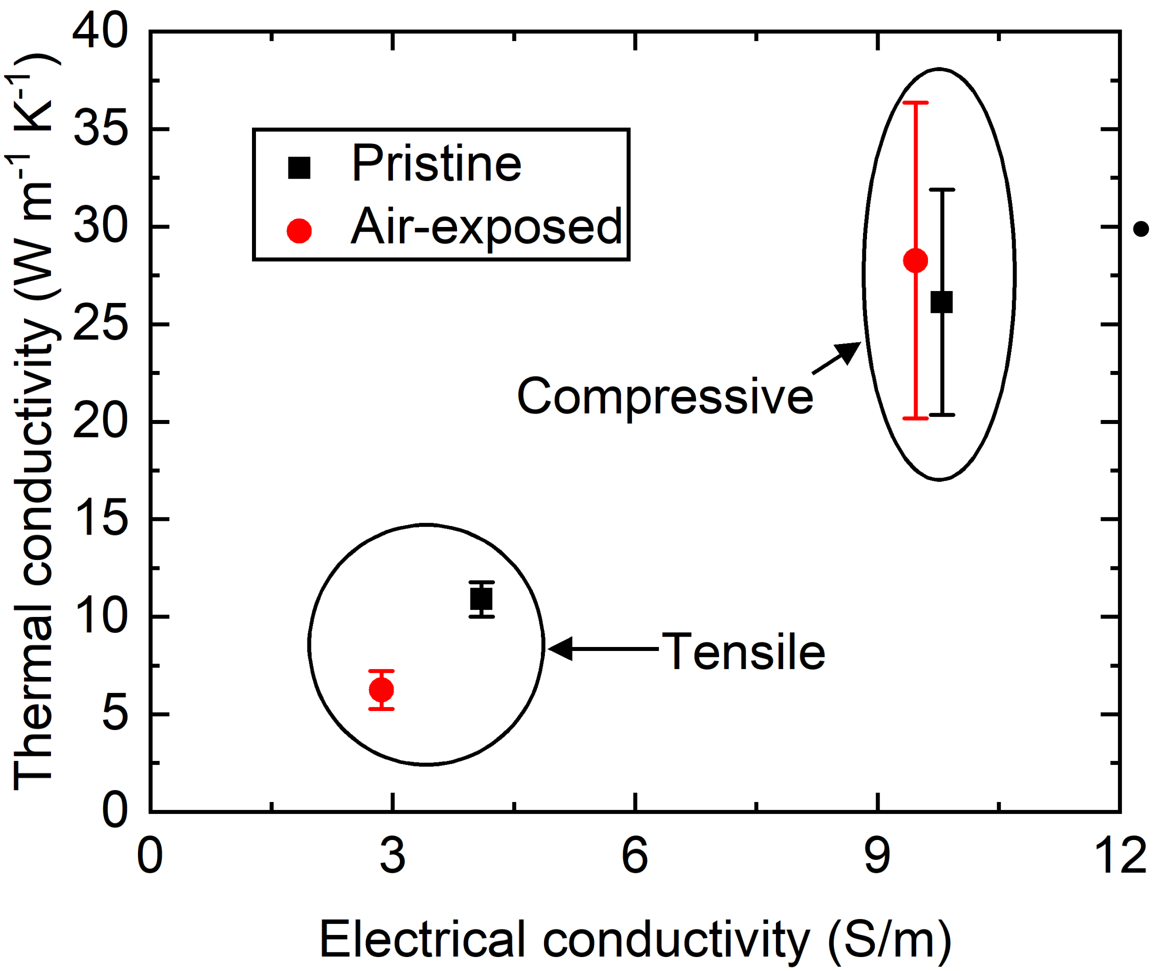


Image courtesy of C. Cancellieri and G. Lorenzin

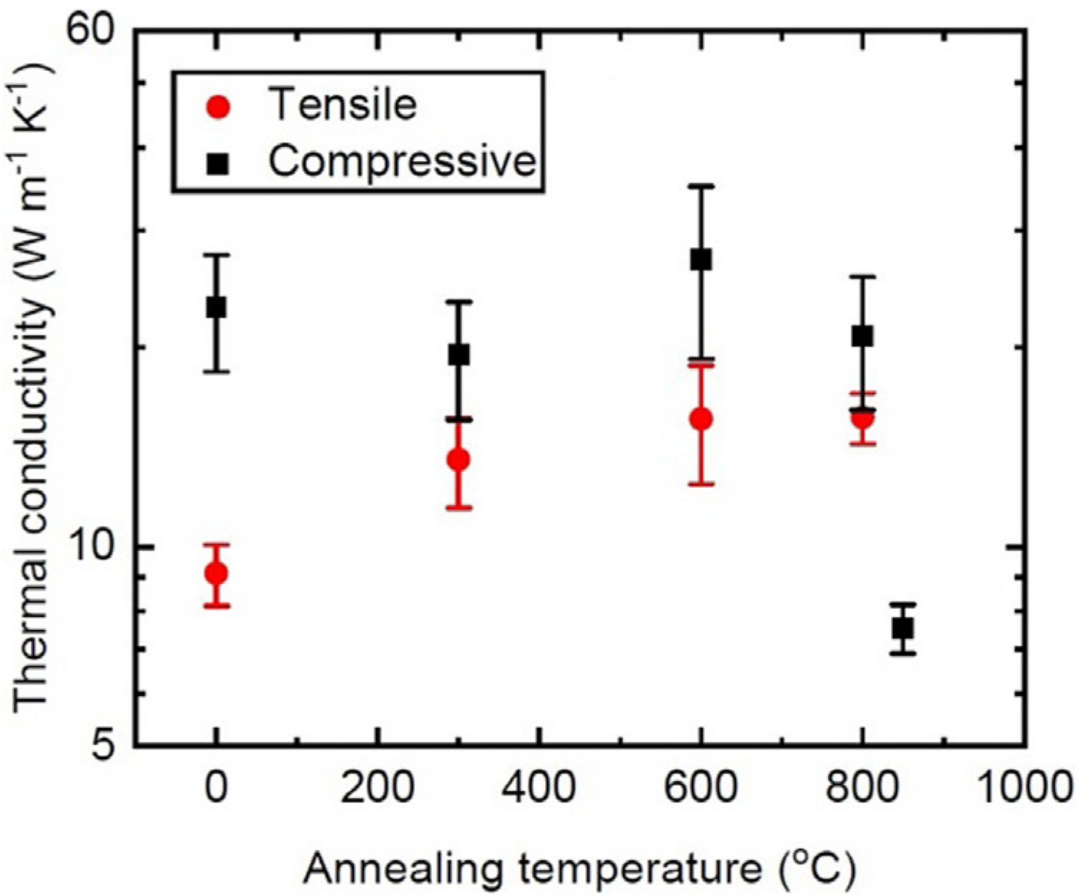
Can we control thermal conductivity in metal multilayers?



- 10 nm period Cu/W multilayers with under either compressive or tensile stress
- Structure of tensile sample more open that can increase oxygen diffusion from ambient/processing compared to compressive

Acta Mat. **240**, 118315 (2022)

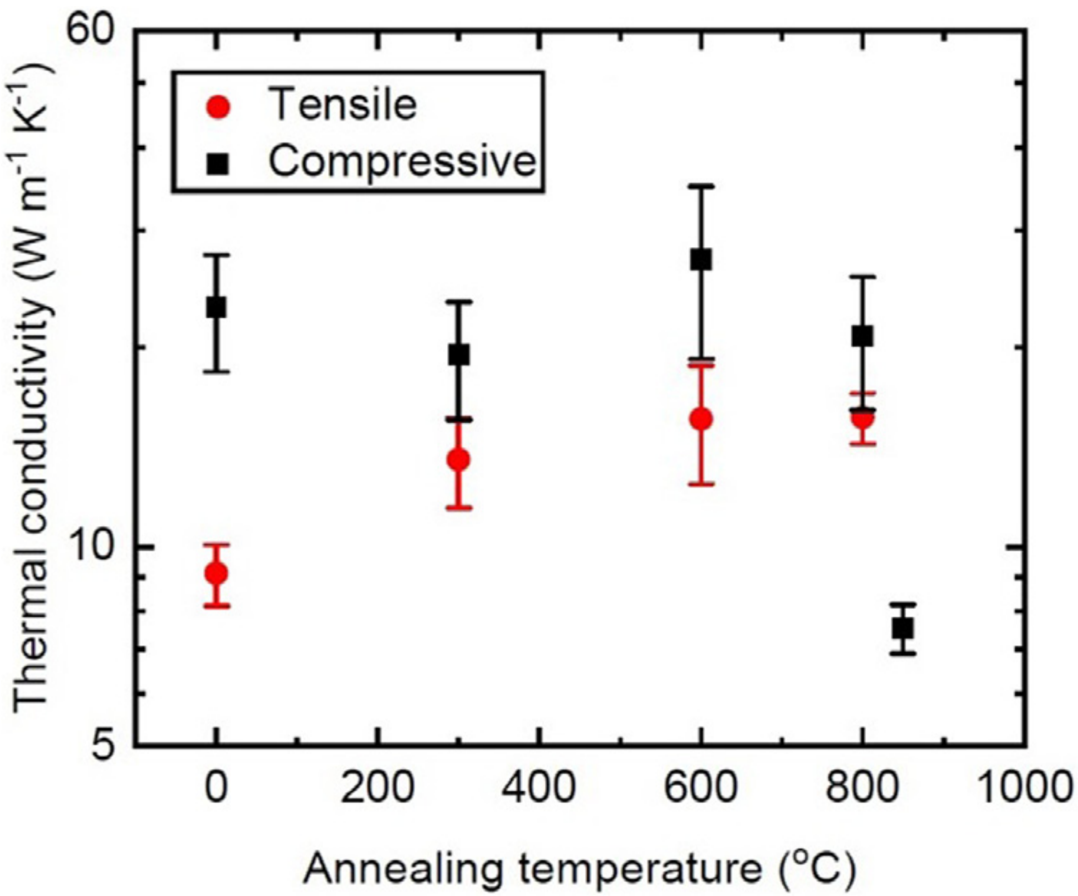
Can we control thermal conductivity in metal multilayers?

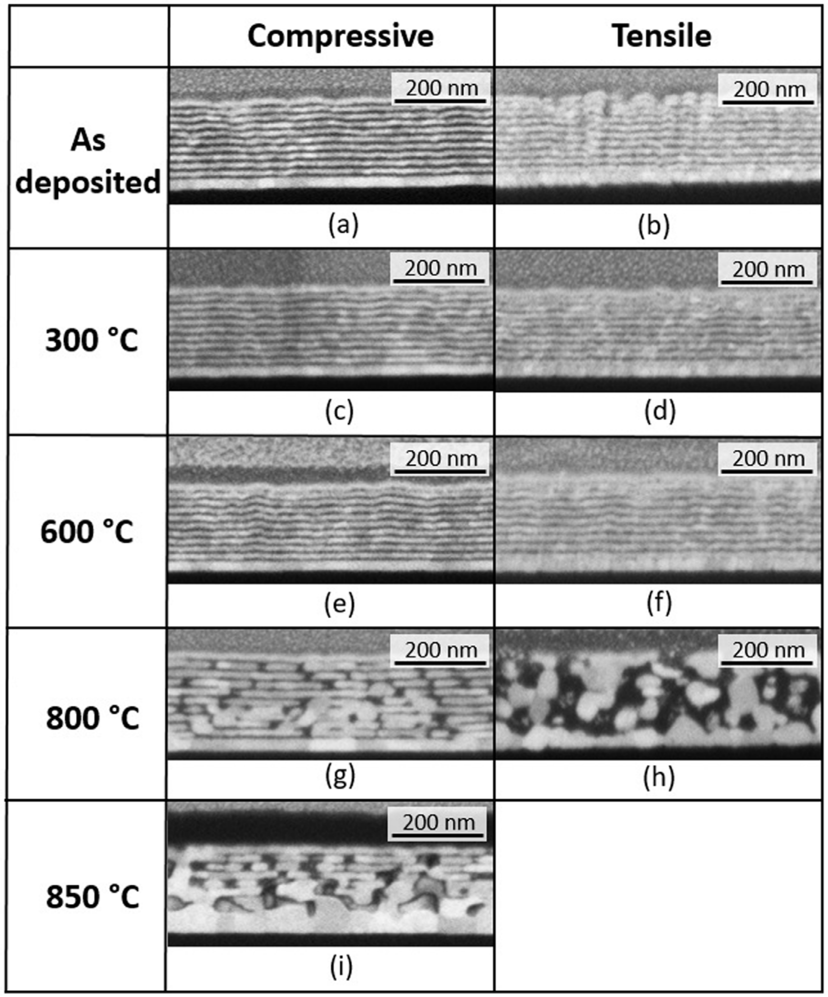


- 10 nm period Cu/W multilayers with under either compressive or tensile stress
- Structure of tensile sample more open that can increase oxygen diffusion from ambient/processing compared to compressive
- Hypothesis: both strain and oxygen defects can impact structure, quality and nanocomposite formation at high temperatures (effort ongoing)

Acta Mat. **240**, 118315 (2022)

Can we control thermal conductivity in metal multilayers?





Acta Mat. **240**, 118315 (2022)

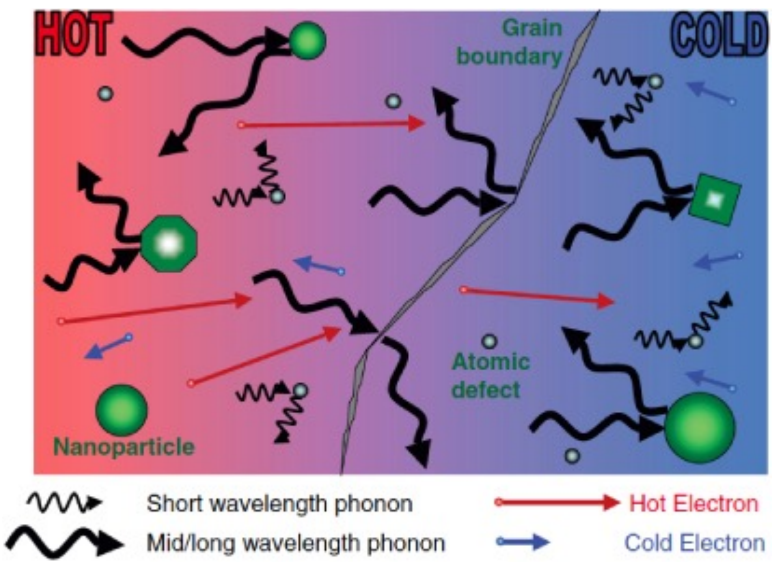
Outline

1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

Electron-phonon scattering in metals

$$\kappa = \frac{1}{3}Cv\lambda = \frac{1}{3}Cv_g^2\tau$$

$$\tau = f\left(\tau_{\text{intrinsic}}, \tau_{\text{impurity}}, \tau_{\text{boundary}}\right)$$



Adv. Mat. **22**, 3970 (2010)

$$G = \frac{\pi^2}{6}\frac{m_e C_s^2 n_e}{\tau(T_e)T_e}$$

$$G(T_e) = \frac{\pi\hbar k_B \lambda\langle\omega^2\rangle}{g(\varepsilon_F)}\int_{-\infty}^{\infty} g^2(\varepsilon)\left(-\frac{\partial f}{\partial \varepsilon}\right)d\varepsilon$$

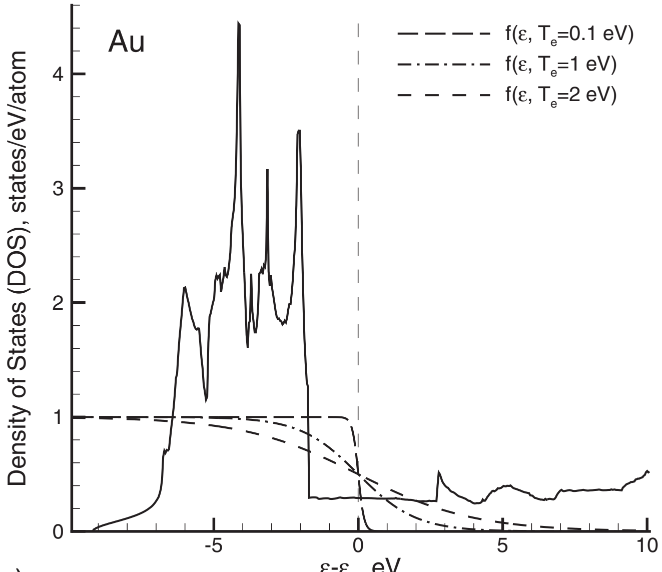
PHYSICAL REVIEW B **77**, 075133 (2008)

Electron-phonon coupling and electron heat capacity of metals under conditions of strong electron-phonon nonequilibrium

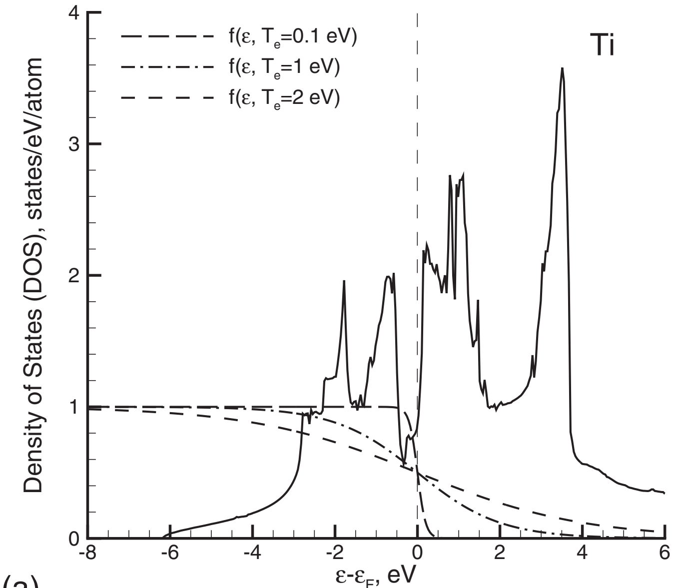
Zhibin Lin and Leonid V. Zhigilei*
Department of Materials Science and Engineering, University of Virginia, 395 McCormick Road, Charlottesville, Virginia 22904-4745, USA

Vittorio Celli
Department of Physics, University of Virginia, 382 McCormick Road, Charlottesville, Virginia 22904-4714, USA
(Received 23 August 2007; revised manuscript received 22 December 2007; published 28 February 2008)

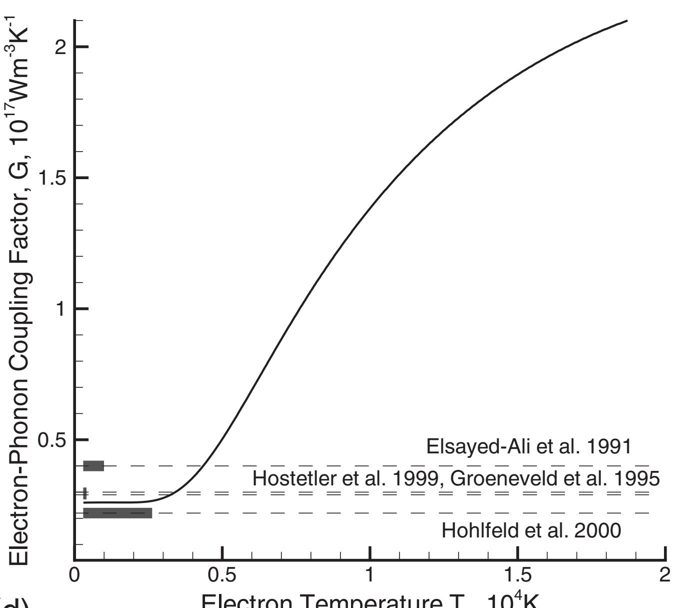
Electron-phonon coupling factor



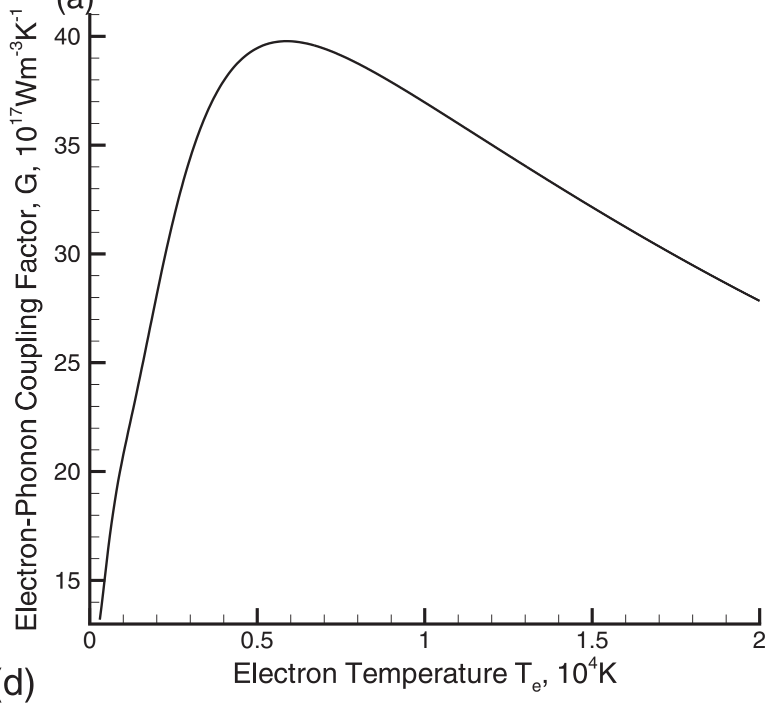
(a)



(a)

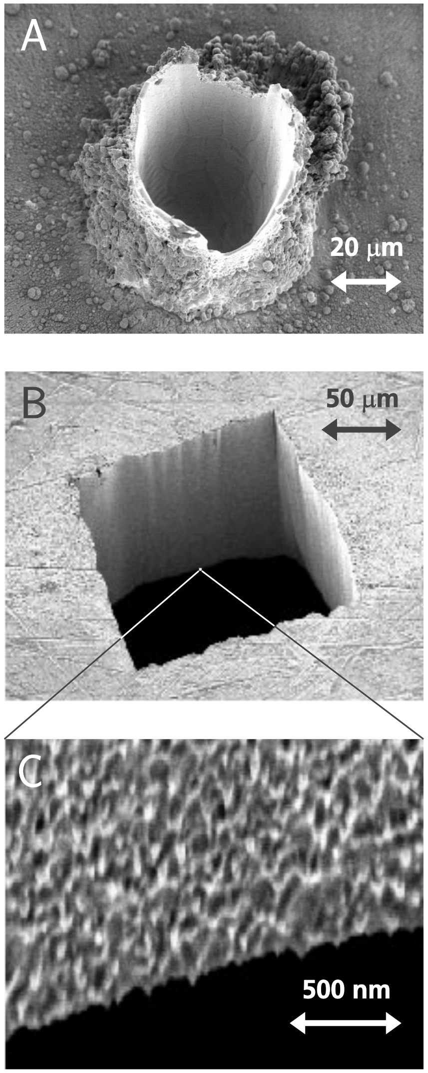


(d)



(d)

High electron temperatures out of equilibrium with lattice: Metal manufacturing, lasers processing, high power/high frequency transistors



The role of electron–phonon coupling in femtosecond laser damage of metals

S.-S. Wellershoff, J. Hohlfeld, J. Güdde, E. Matthias

Appl. Phys. A 69 [Suppl.], S99–S107 (1999)

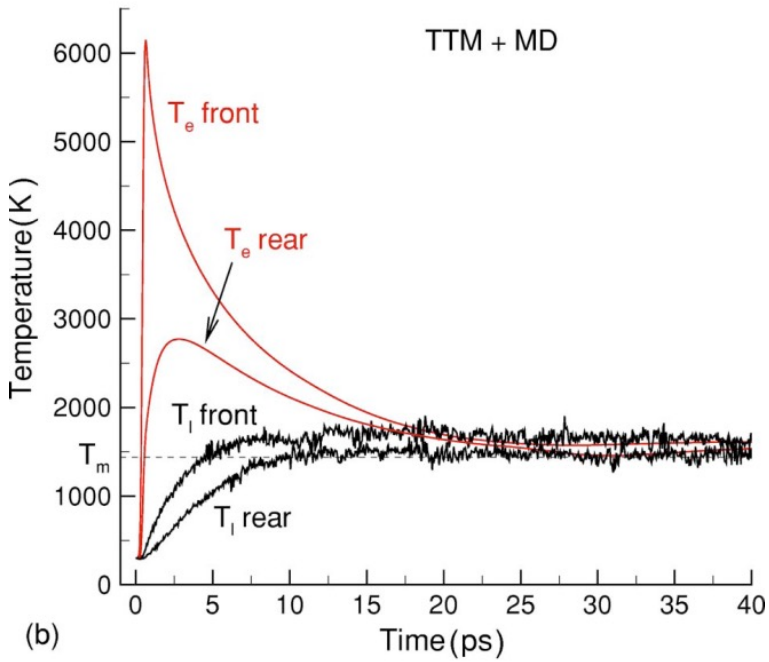
PHYSICAL REVIEW B **68**, 064114 (2003)

Combined atomistic-continuum modeling of short-pulse laser melting and disintegration of metal films

Dmitriy S. Ivanov and Leonid V. Zhigilei*

Department of Materials Science and Engineering, University of Virginia, 116 Engineer's Way, Charlottesville, Virginia 22904-4745, USA

(Received 3 March 2003; published 28 August 2003)



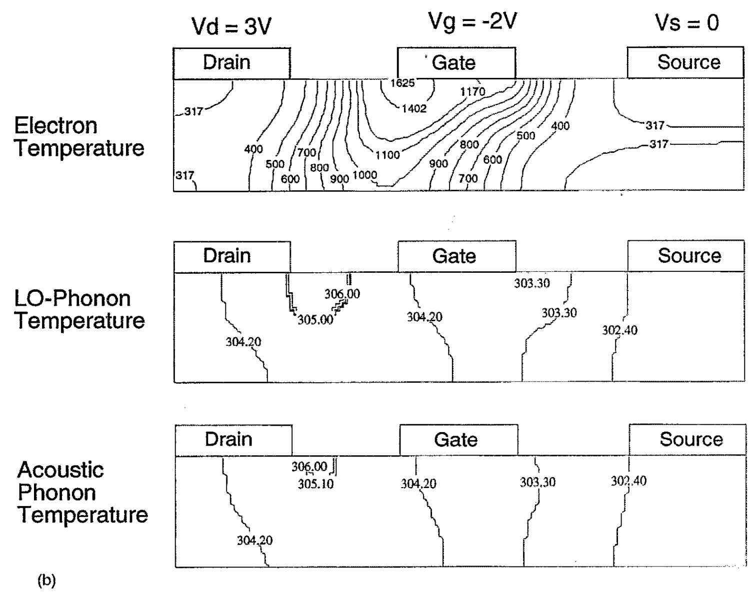
Effect of gate voltage on hot-electron and hot-phonon interaction and transport in a submicrometer transistor

A. Majumdar

Department of Mechanical and Environmental Engineering, University of California, Santa Barbara, California 93106

K. Fushinobu and K. Hijikata

Department of Mechano-Aerospace Engineering, Tokyo Institute of Technology, 2-12-1 O-Okayama, Meguro-ku, Tokyo 152, Japan



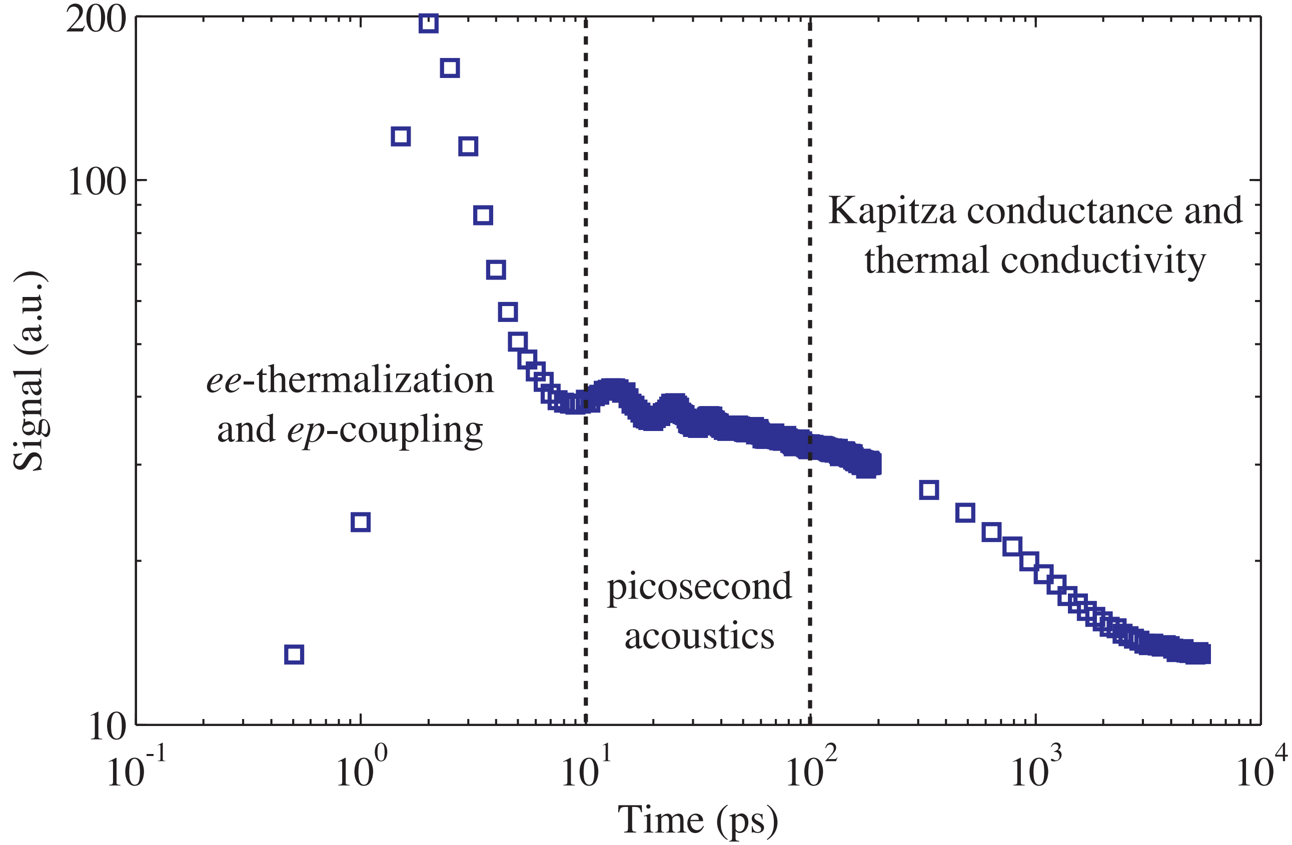
$$C_e(T = 300\,K) \approx 10^4\,\mathrm{J\,m^{-3}\,K^{-1}}$$

$$C_o(T = 300\,K) \approx 10^6\,\mathrm{J\,m^{-3}\,K^{-1}}$$

How do we measure? Let's go back a few (a lot of) slides…

Temporal regimes in TDTR

FANTASTIC temporal resolution (limited by pulse width)



Pulse absorption (~100 fs)

↓

Fermi relaxation and ballistic transport (few hundred fs)

↓

Electron-phonon coupling (a few ps)

↓

Strain propagation in film (10's of ps)

↓

Thermal diffusion (hundreds of ps to ns)

Electron thermalization and scattering (<10 ps)

Journal of Heat Transfer

APRIL 2011, Vol. 133 / **044505-1**

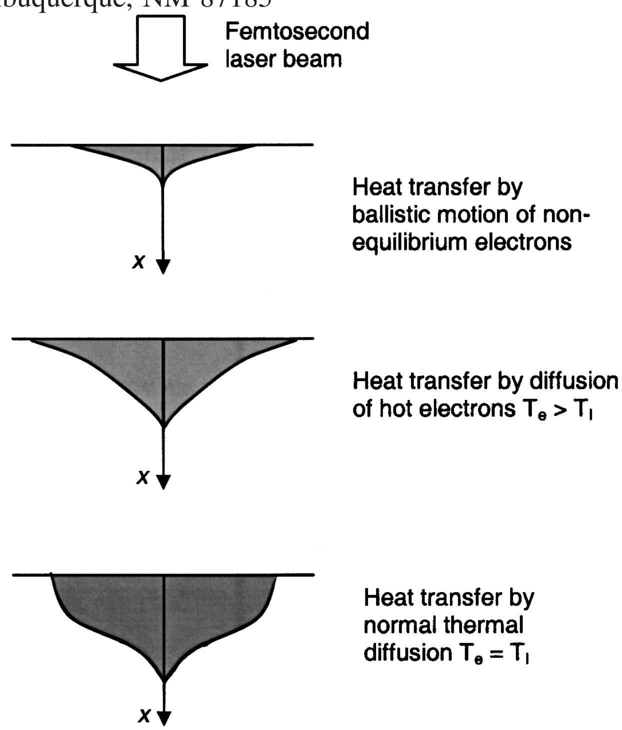
Re-examining Electron-Fermi Relaxation in Gold Films With a Nonlinear Thermoreflectance Model

Patrick E. Hopkins
e-mail: pehopki@sandia.gov

Leslie M. Phinney

Justin R. Serrano

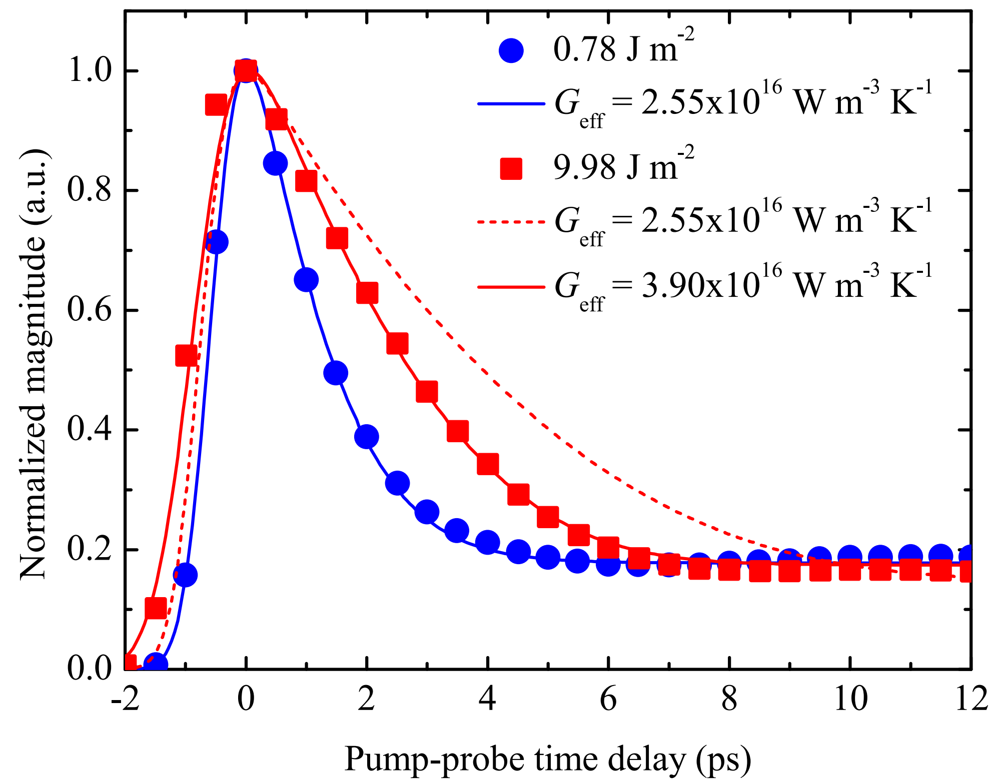
Sandia National Laboratories,
Albuquerque, NM 87185



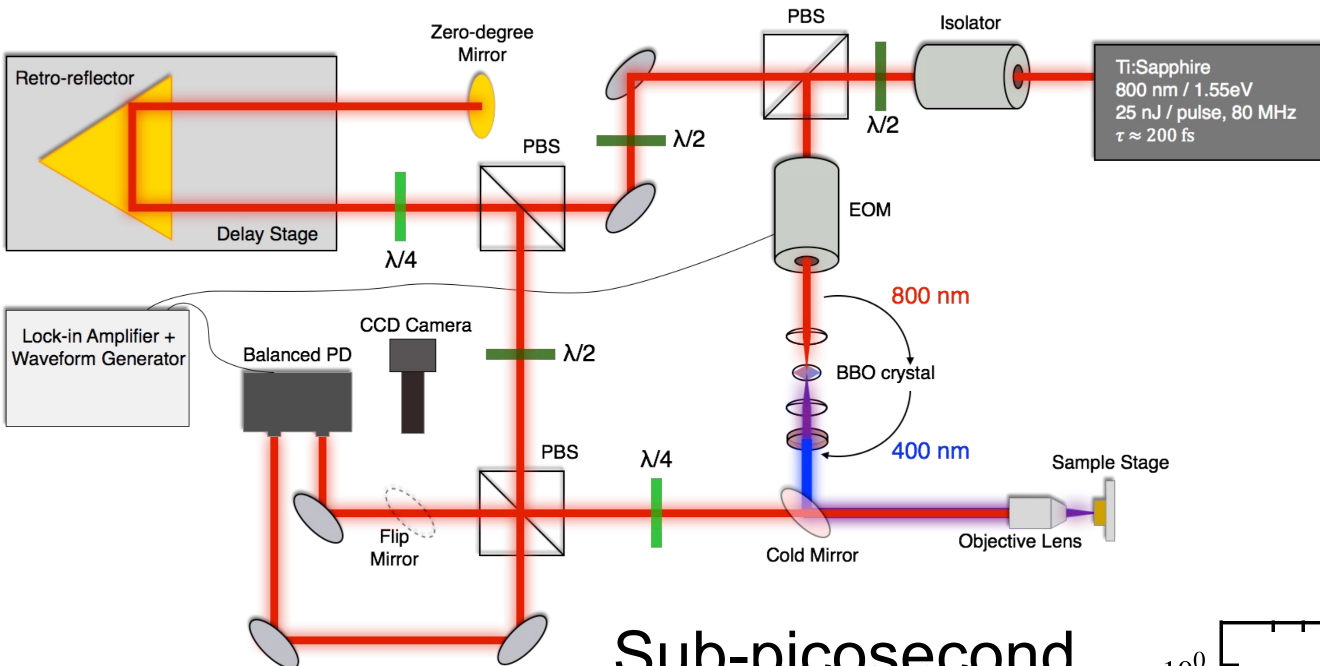
APPLIED PHYSICS LETTERS **103**, 211910 (2013)

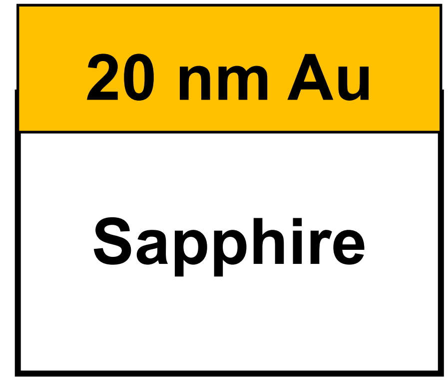
Ultrafast and steady-state laser heating effects on electron relaxation and phonon coupling mechanisms in thin gold films

Patrick E. Hopkins,[1,a)] John C. Duda,[1] Bryan Kaehr,[2,3] Xiao Wang Zhou,[4] C.-Y. Peter Yang,[4] and Reese E. Jones[4]
[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico 87106, USA*
[3]*Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA*
[4]*Sandia National Laboratories, Livermore, California 94550, USA*



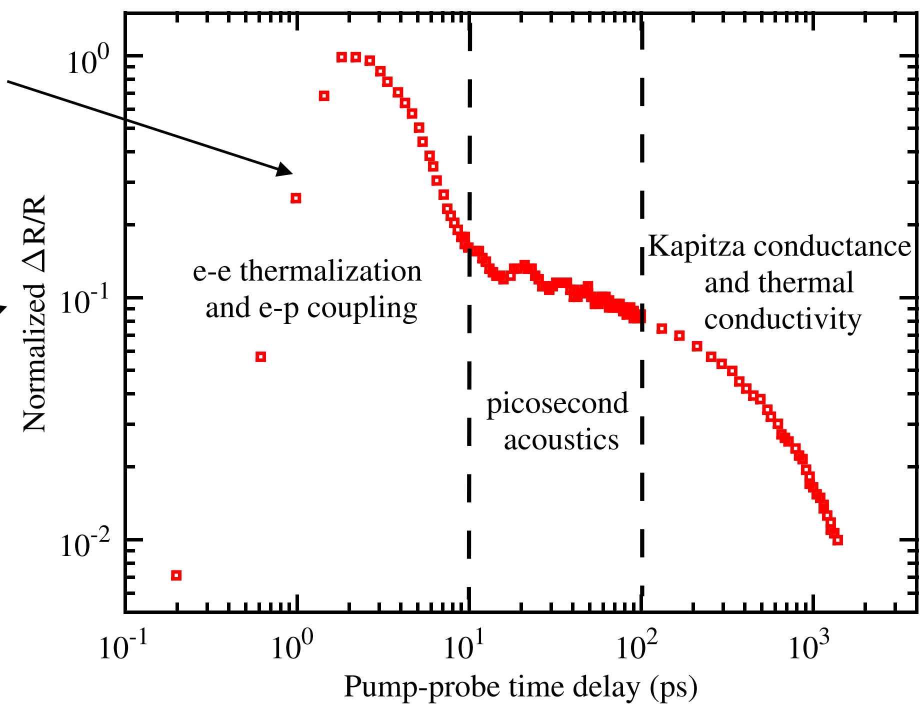
Ultrafast pump-probe to measure EP coupling





Energy no longer deposited in system

Sub-picosecond heat event

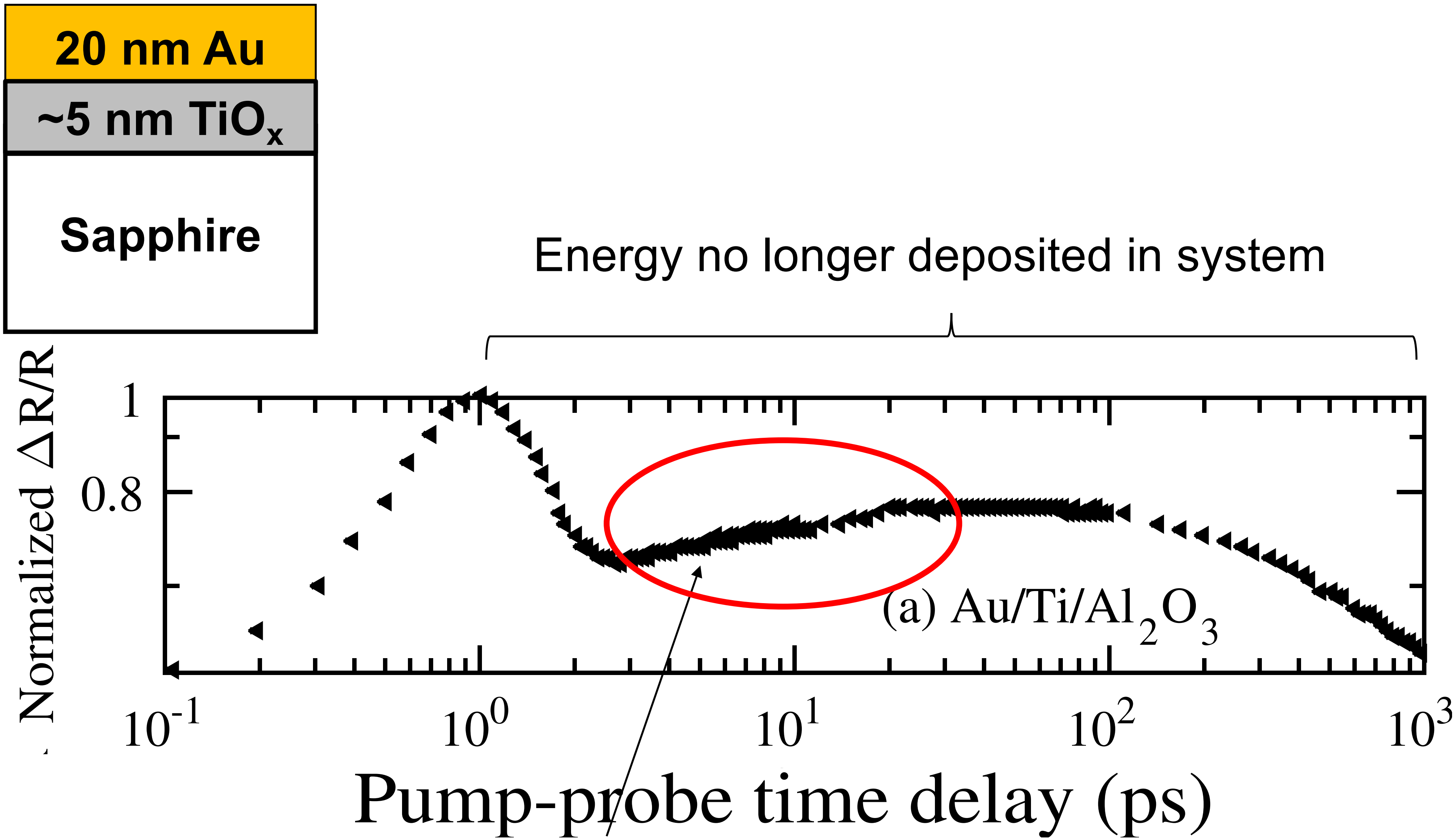


Directly related to temperature (advantage of probing metal)

ACS Photonics **5**, 4880

JAP **117**, 105105

Ultrafast pump-probe to measure EP coupling



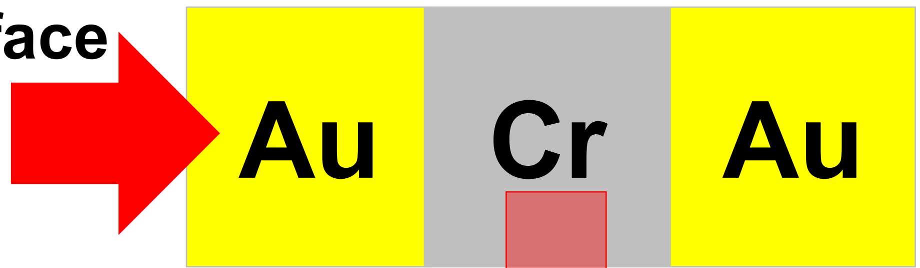
So why does temperature at surface increase when no energy is deposited in the system?

JAP **117**, 105105

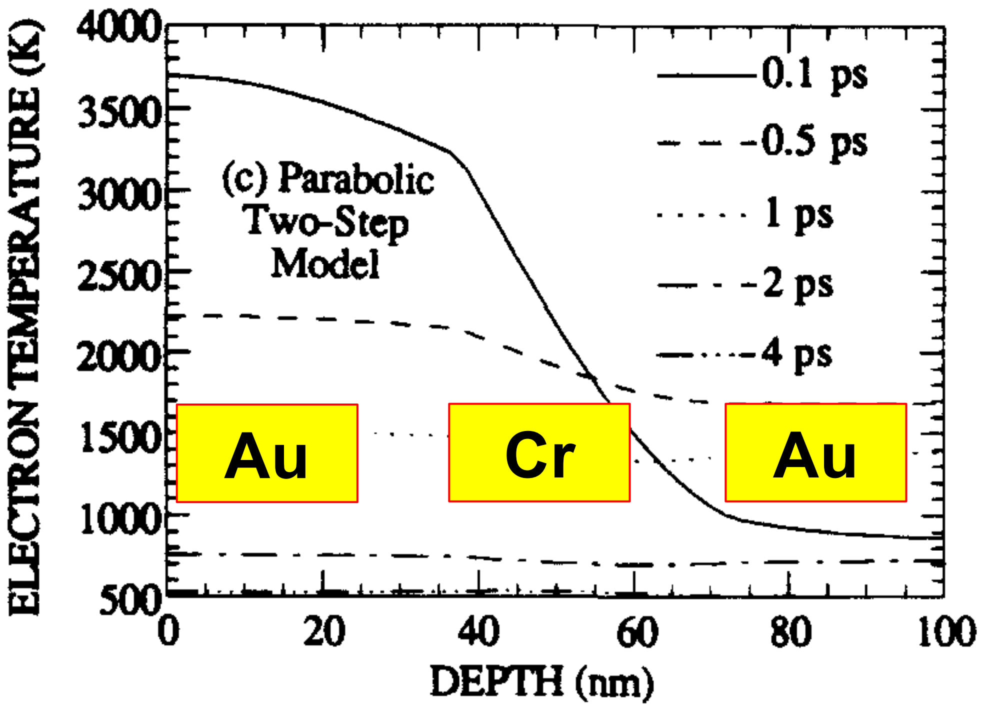
Electron-phonon interactions at metal/metal interfaces

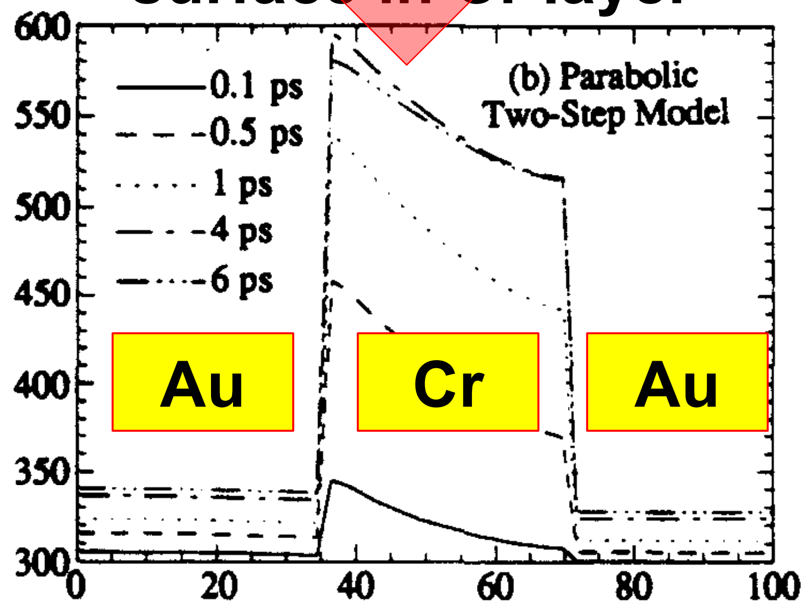
Recall seminal predictions by Tien
Qiu and Tien, *IJHMT* **37**, 2789 (1994)

Laser excites Au surface



Lattice heats up under surface in Cr layer

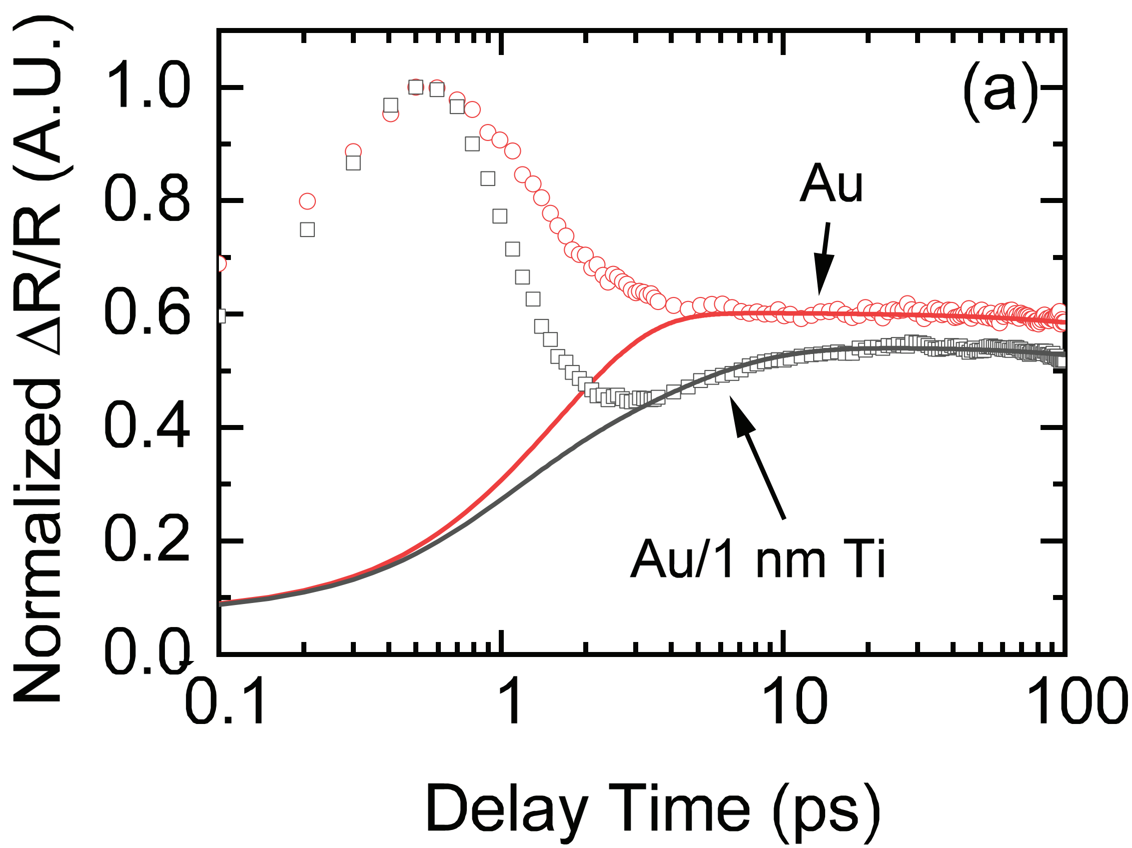


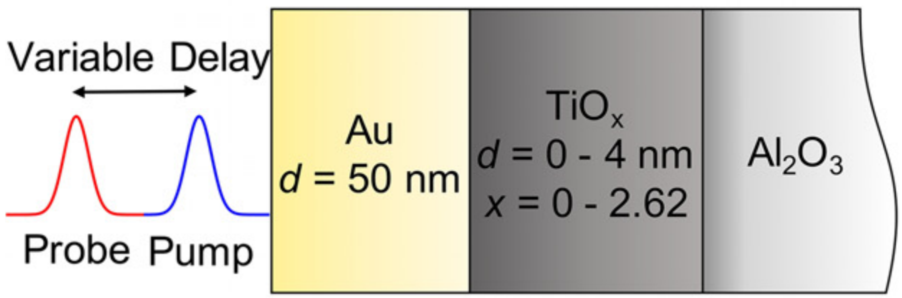


Au lattice slowly responds

Electron mediated TBC at metal/doped non-metal is the key

- Energy easily transmitted across interface when out of equilibrium with phonons
- Slowly "goes back" across the interface when diffusive
- See early observations of indirect heating by:
    - Qiu and Tien, *IJHMT* **37**, 2789 (1994)
    - Choi, Wilson and Cahill, *PRB* **89**, 064307 (2014)

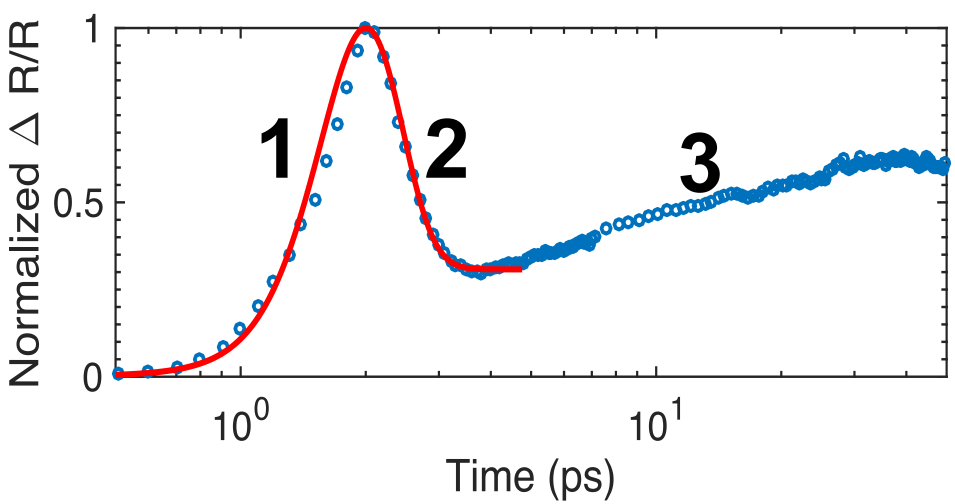


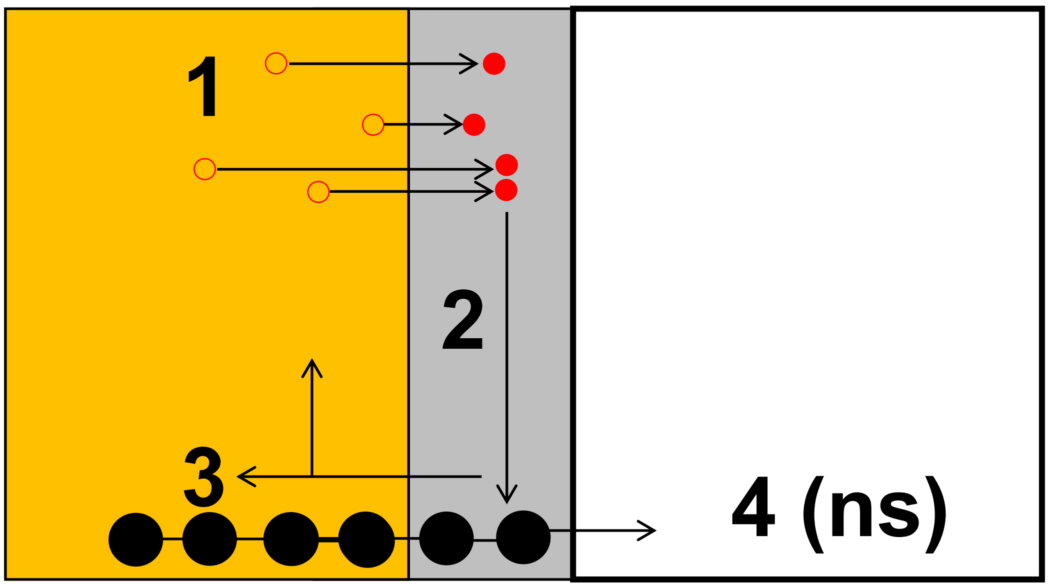


JAP **117**, 105105
APL **118**, 163503

Ballistic thermal injection

- Excited electrons in metal from pulse do not thermalize with lattice and deposit their energy to lattice in sub-surface layer
- Ballistic transport of electron energy through gold into titantium



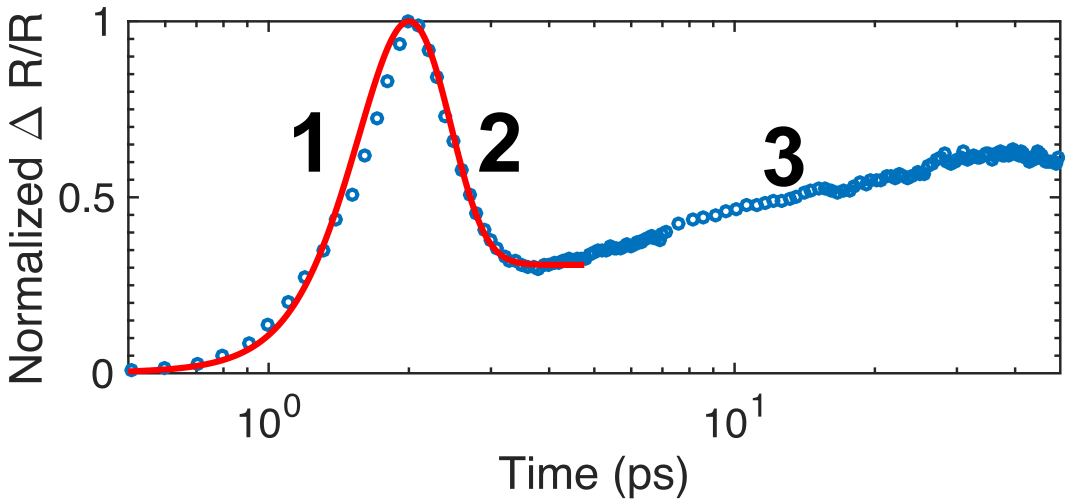


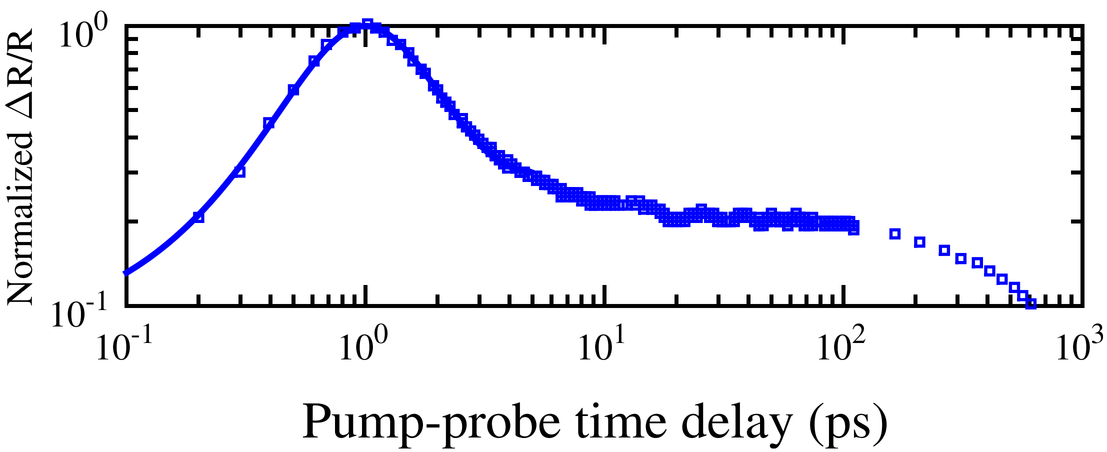
<u>When would we see this effect?</u>

1. Metal/metal or metal/non-metal interfaces with large differences in electron-phonon coupling factor
2. Films with thicknesses less than electron-phonon mean free path
3. Interfaces with very little electron-electron thermal resistance

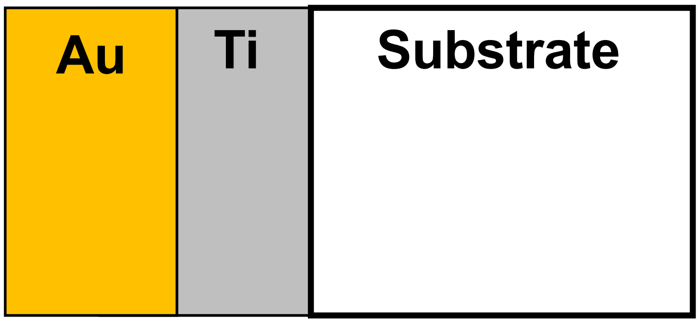
TDTR measurements of time scales of noneq. transport

Hypothesis: If Au thickness (d_{Au}) is thicker than electron-phonon mean free path (λ_{ep}), nonequilibrium at interface will be negligible and "back heating" (time regime 3) will not be observed

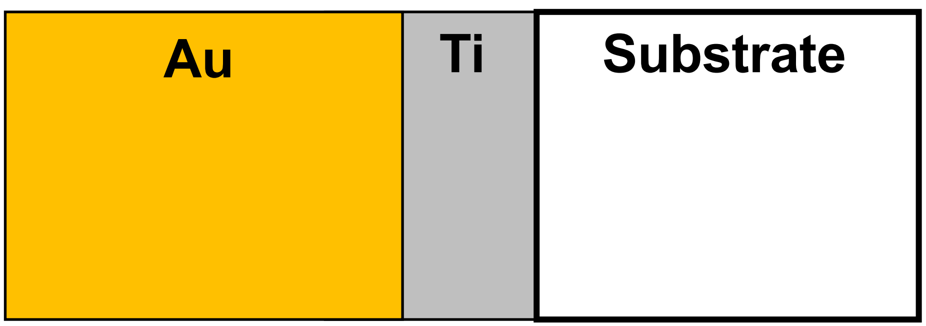




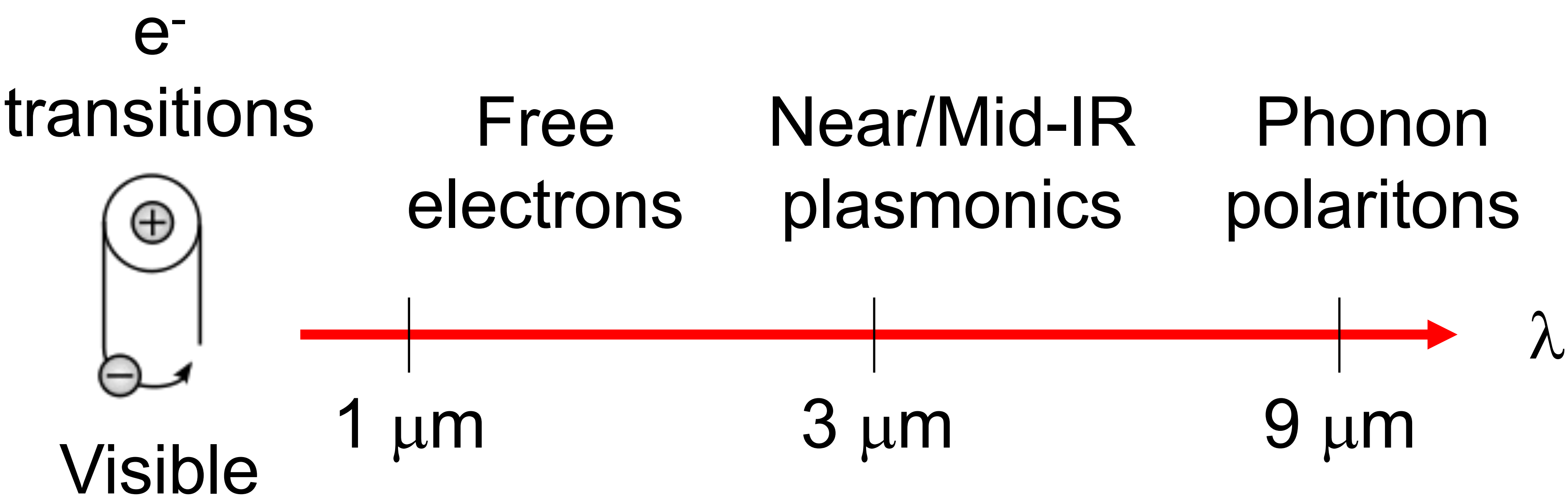
$$d_{\mathrm{Au}} < \lambda_{ep}$$



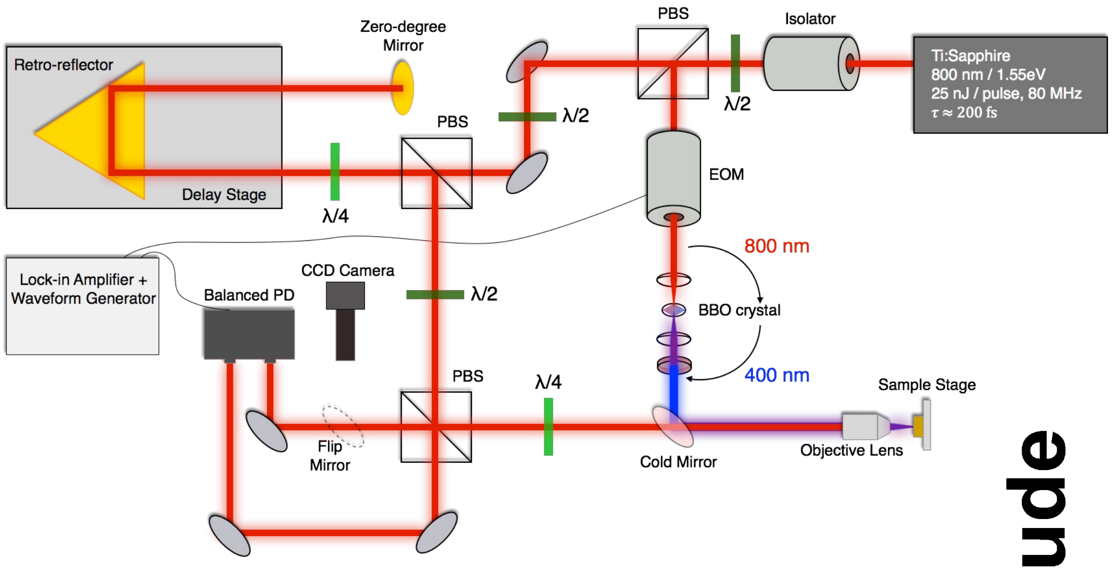
$$d_{\mathrm{Au}} > \lambda_{ep}$$

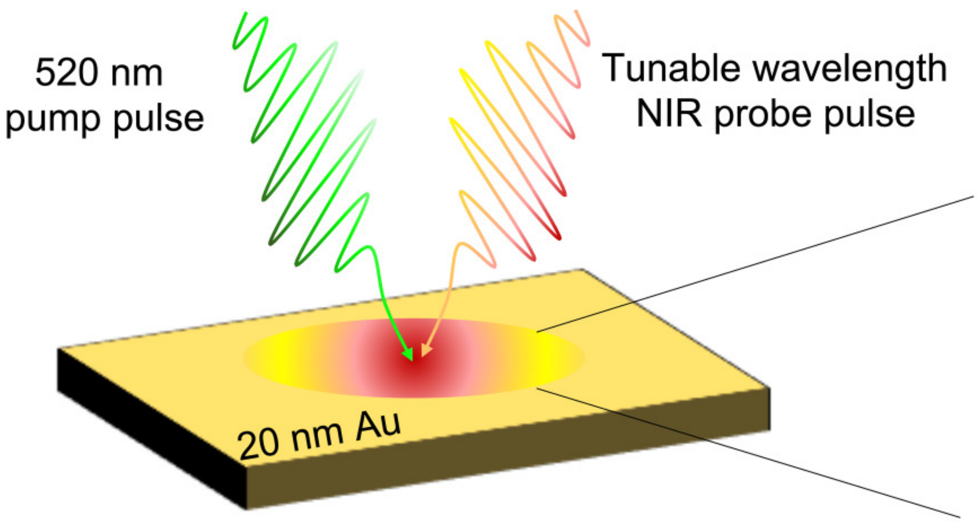


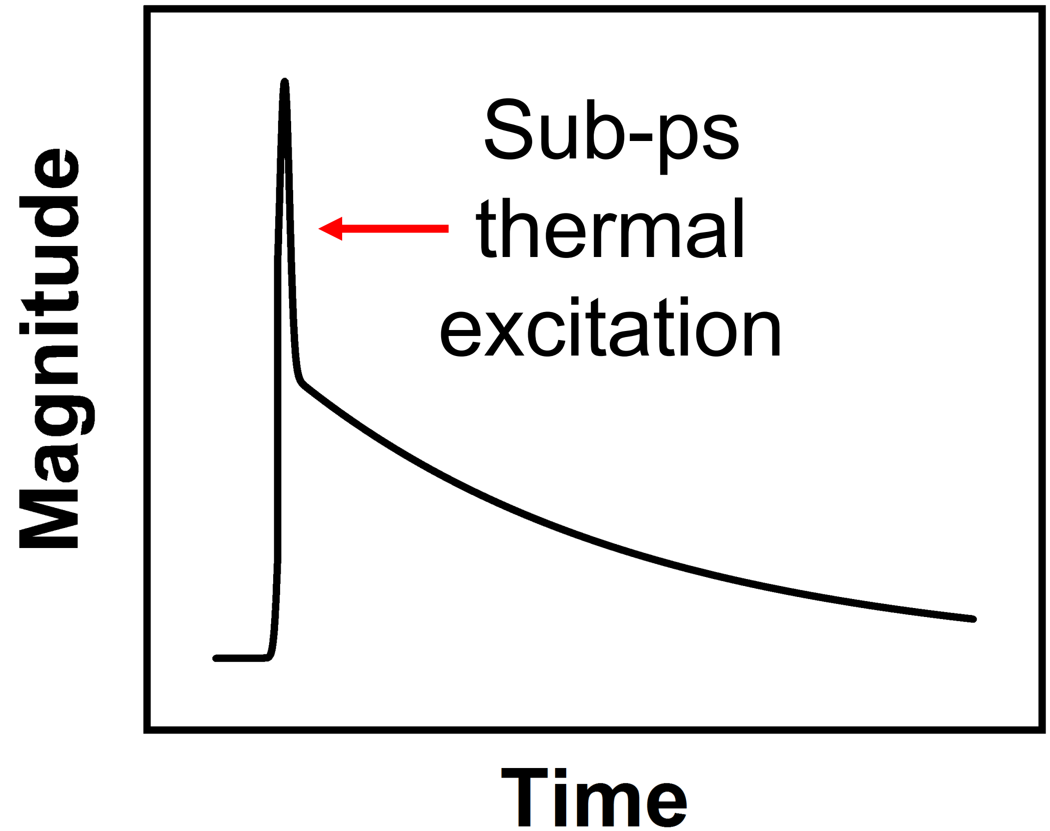
Pumping with heat, probing in IR away from e^- transitions



Measuring thermal lifetimes/scattering rates….in the IR





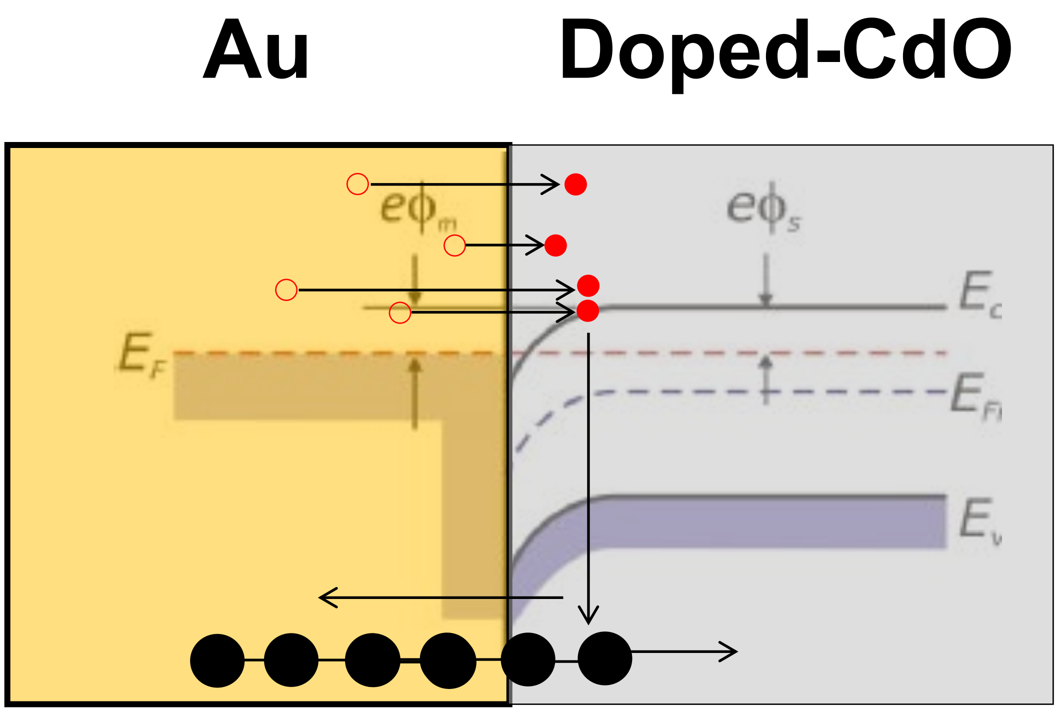


Picoseconds to nanoseconds

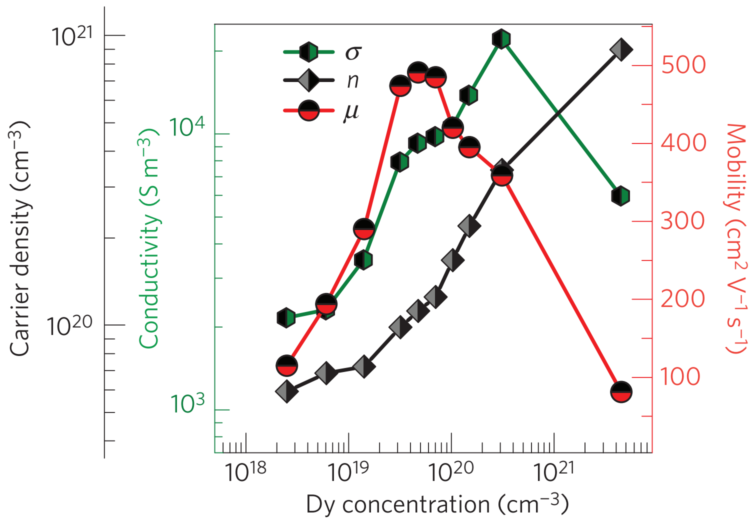
Nonequilibrium at metal/doped non-metal interfaces

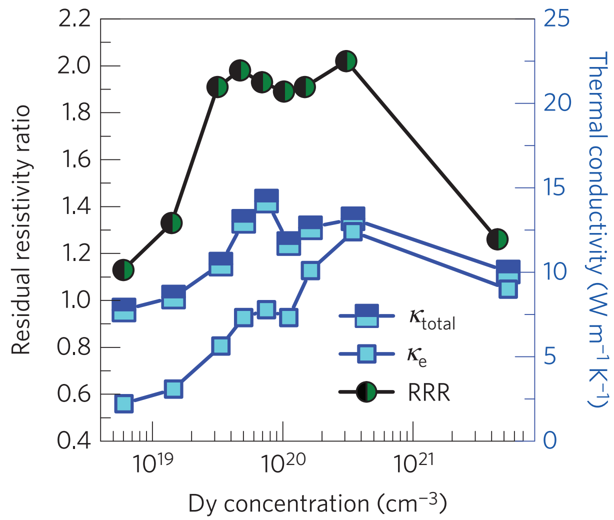
- Consider ohmic contact between metal and doped non-metal
- Vary carrier concentration in non-metal

- Electron injection from Au to interfacial layer must occur to observe "back heating" effect
- Will not occur when interface is insulating

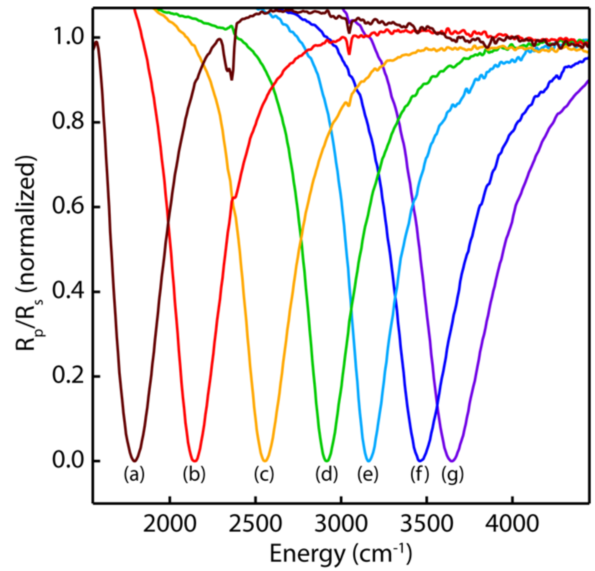


CdO – a gateway for mid-IR plasmonics





Nat. Mat. **14**, 414
Appl. Phys. Lett. **108**, 021901



ACS Photonics **4**, 1885

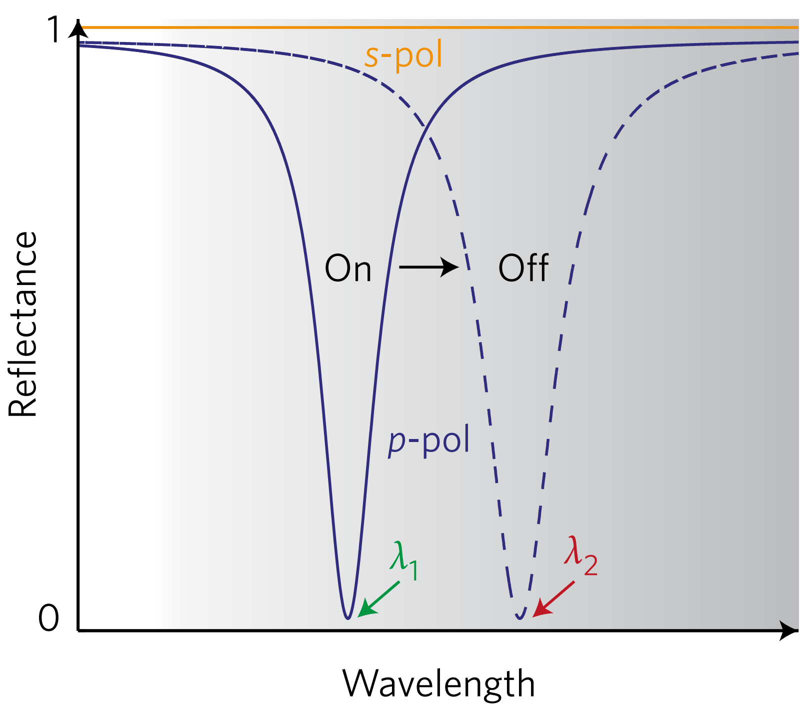
- Large electron mobility in CdO results in large electronic thermal conductivity
- Doping concentration tunes electronic conductivity and IR absorption

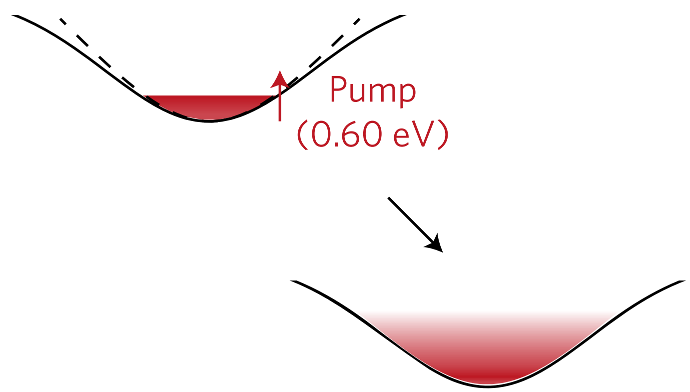
Carrier scattering and relaxation drives optical properties

Dielectric function

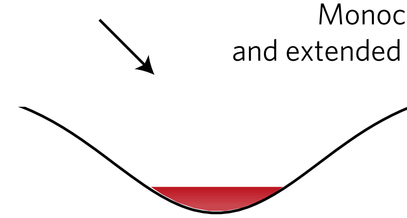
Reflectivity $R \propto \varepsilon \propto \tau$ Relaxation time ~ Scattering time

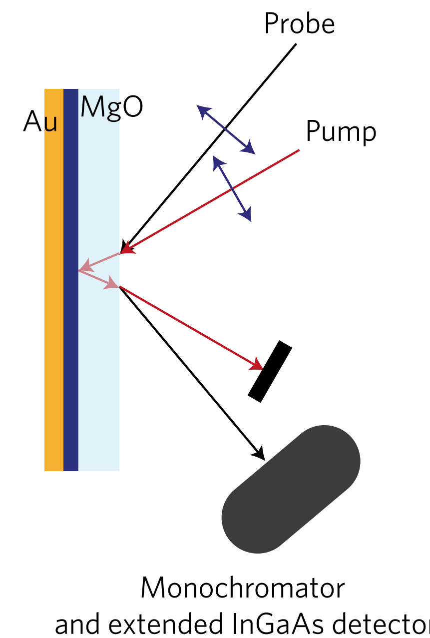
Example: Near IR polarization switching and absorption modulation in thin film CdO





$$\omega_{\text{plasma}} \propto \sqrt{\frac{n_e}{m^*}}$$

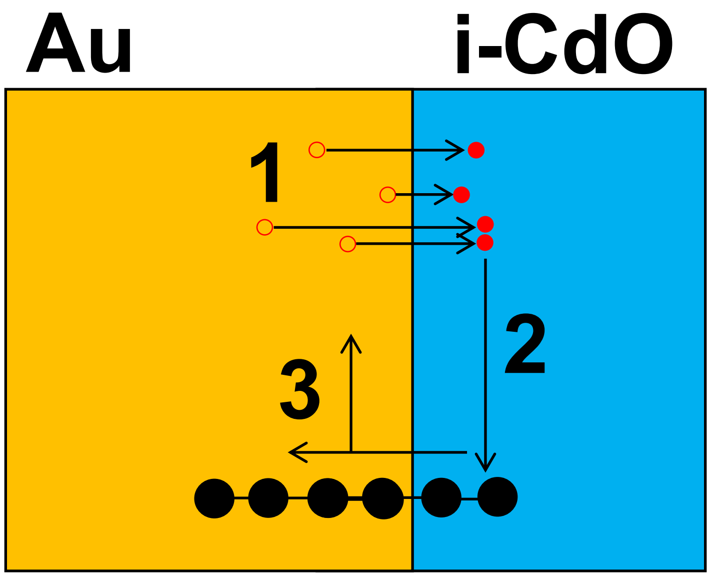


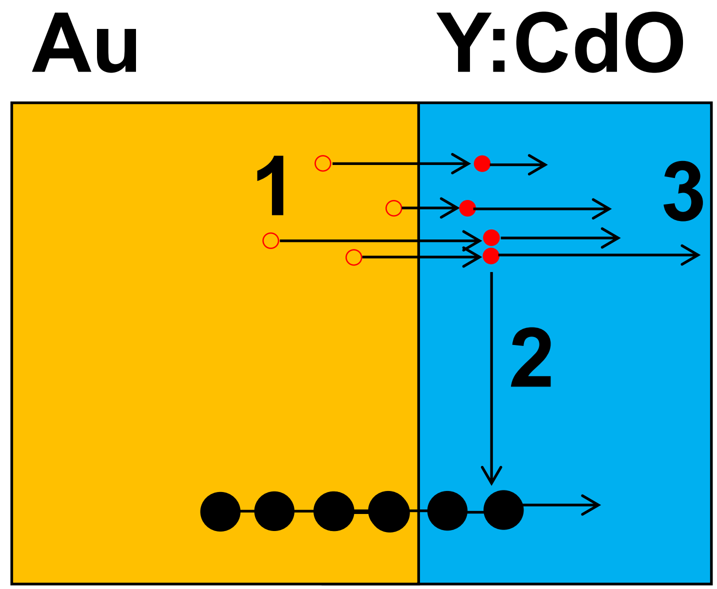


Nature Photonics **11**, 390

Nonequilibrium processes at Au/CdO interfaces

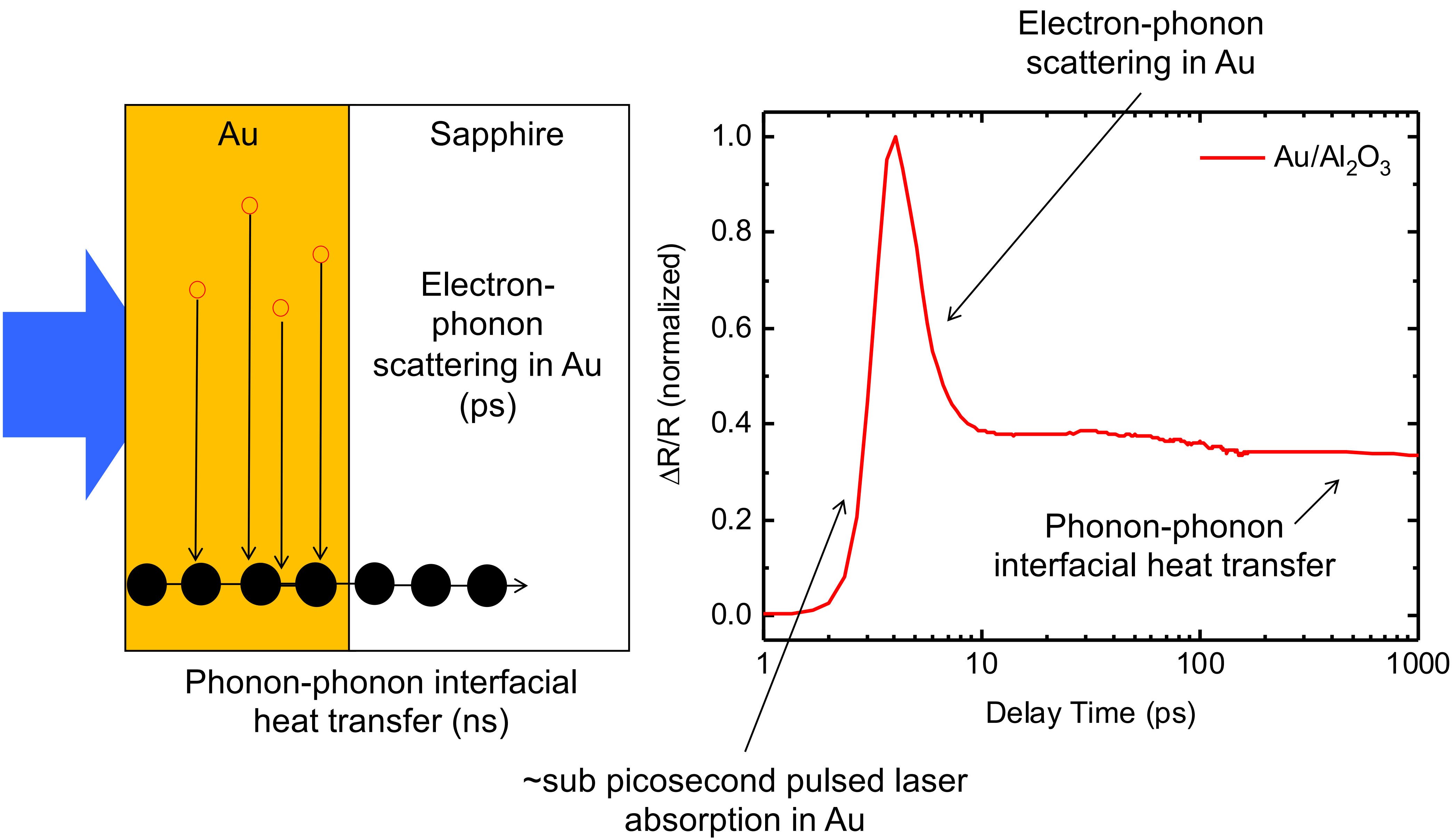
Doping will control electron-electron TBC and electron thermal conductivity in CdO, vary "back heating"





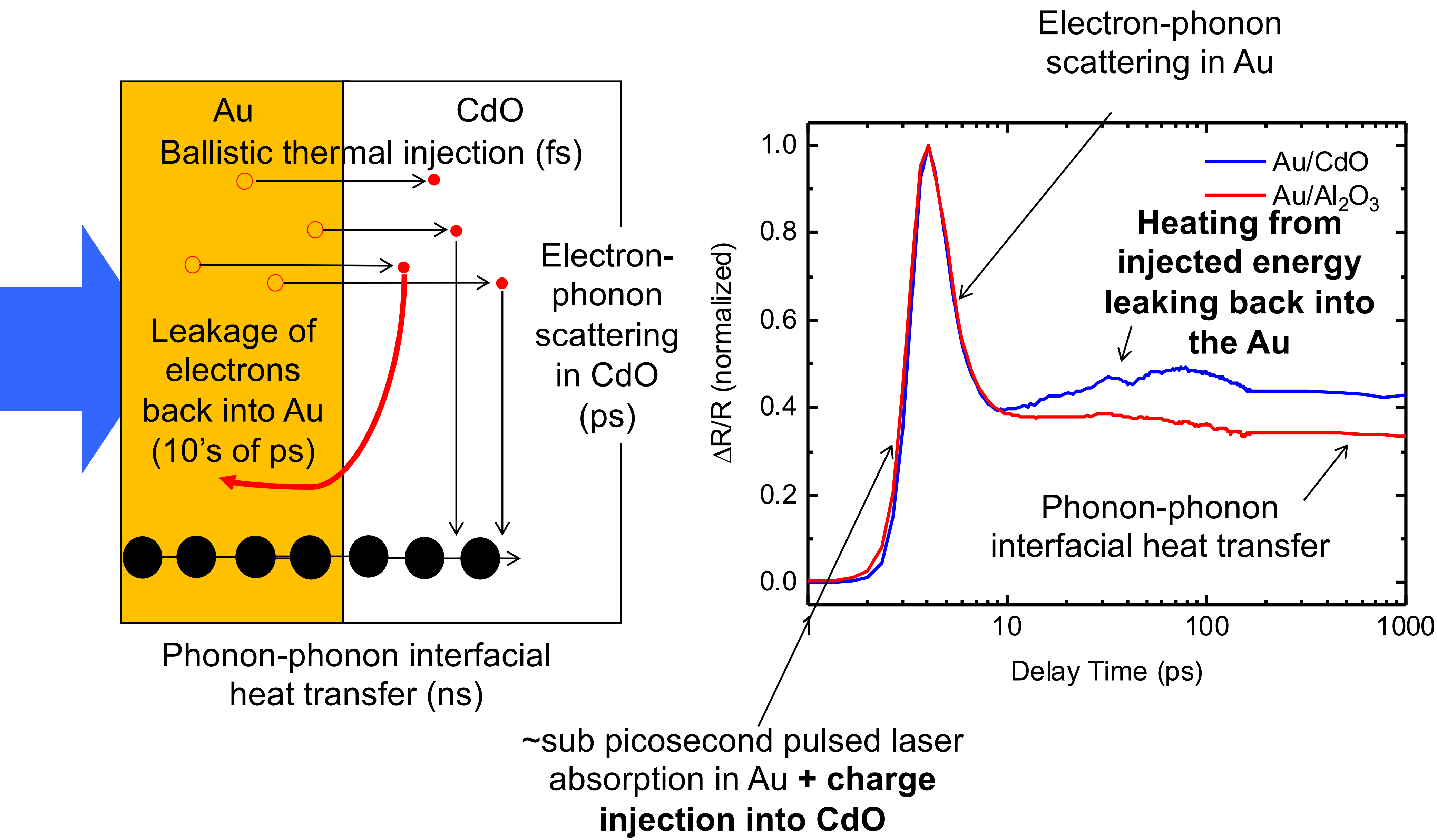
Tomko *et al. Nature Nano.* **16**, 47 (2021)

Nonequilibrium processes at Au/CdO interfaces



Tomko *et al. Nature Nano.* **16**, 47 (2021)

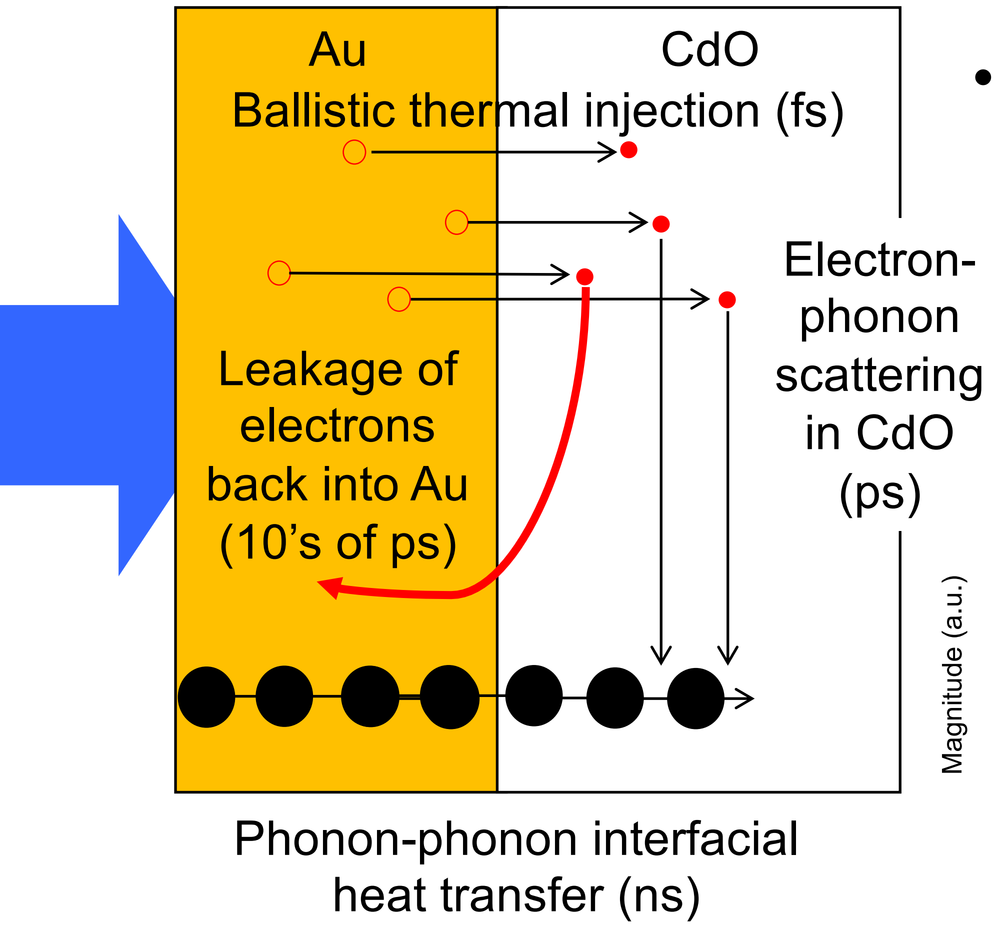
Nonequilibrium processes at Au/CdO interfaces

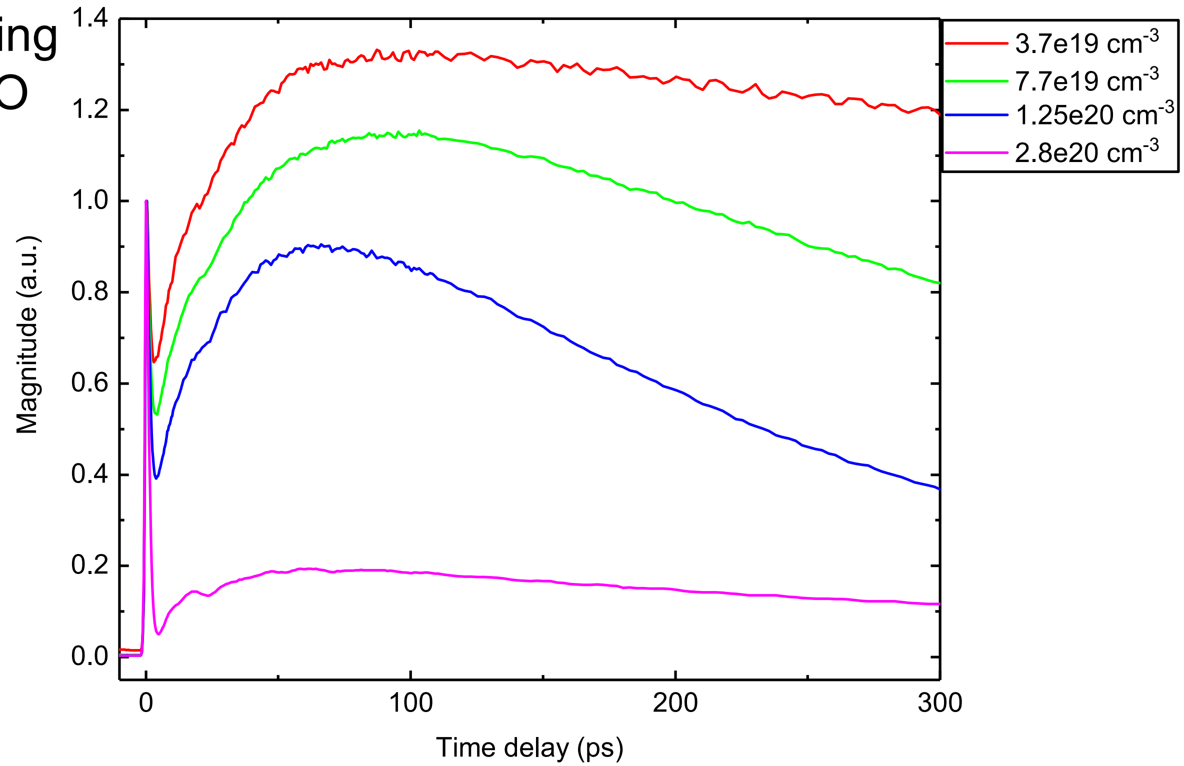


Tomko *et al. Nature Nano.* **16**, 47 (2021)

Nonequilibrium processes at Au/CdO interfaces

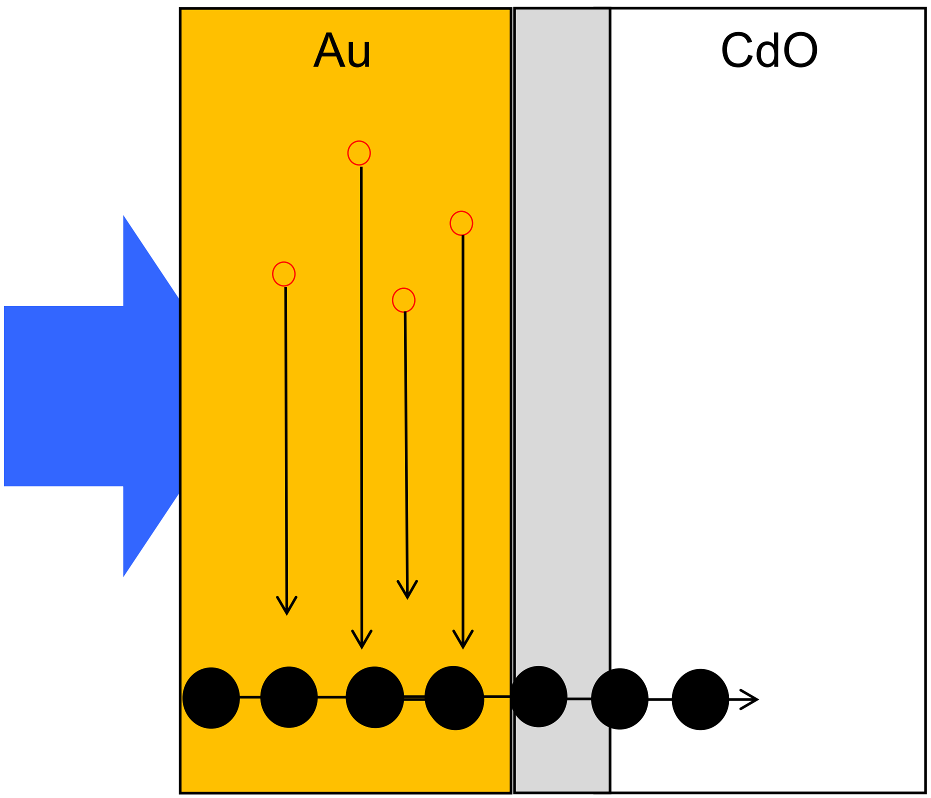
- Doping of CdO dictates amount of "electron leakage" back into Au
- Lower resistivity + higher e-ph coupling in CdO, less electronic back heating into gold contact





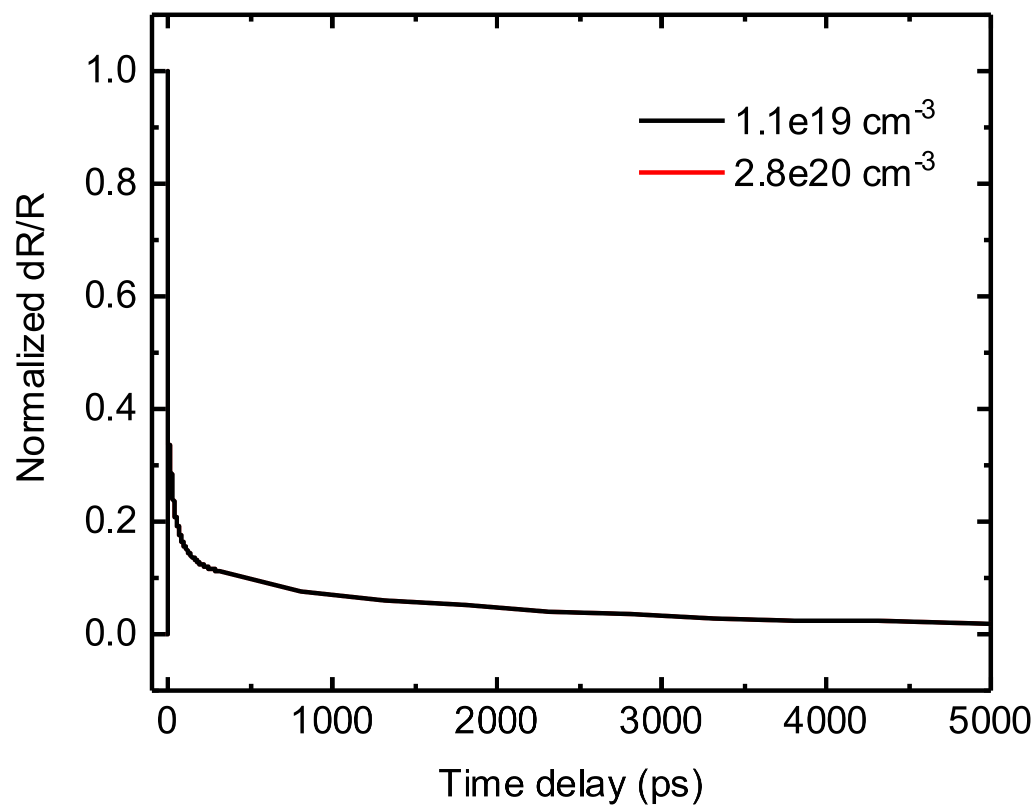
Tomko *et al. Nature Nano.* **16**, 47 (2021)

Nonequilibrium processes at Au/CdO interfaces



- Transparent buffer layer stops ballistic electrons, but allows light to transmit

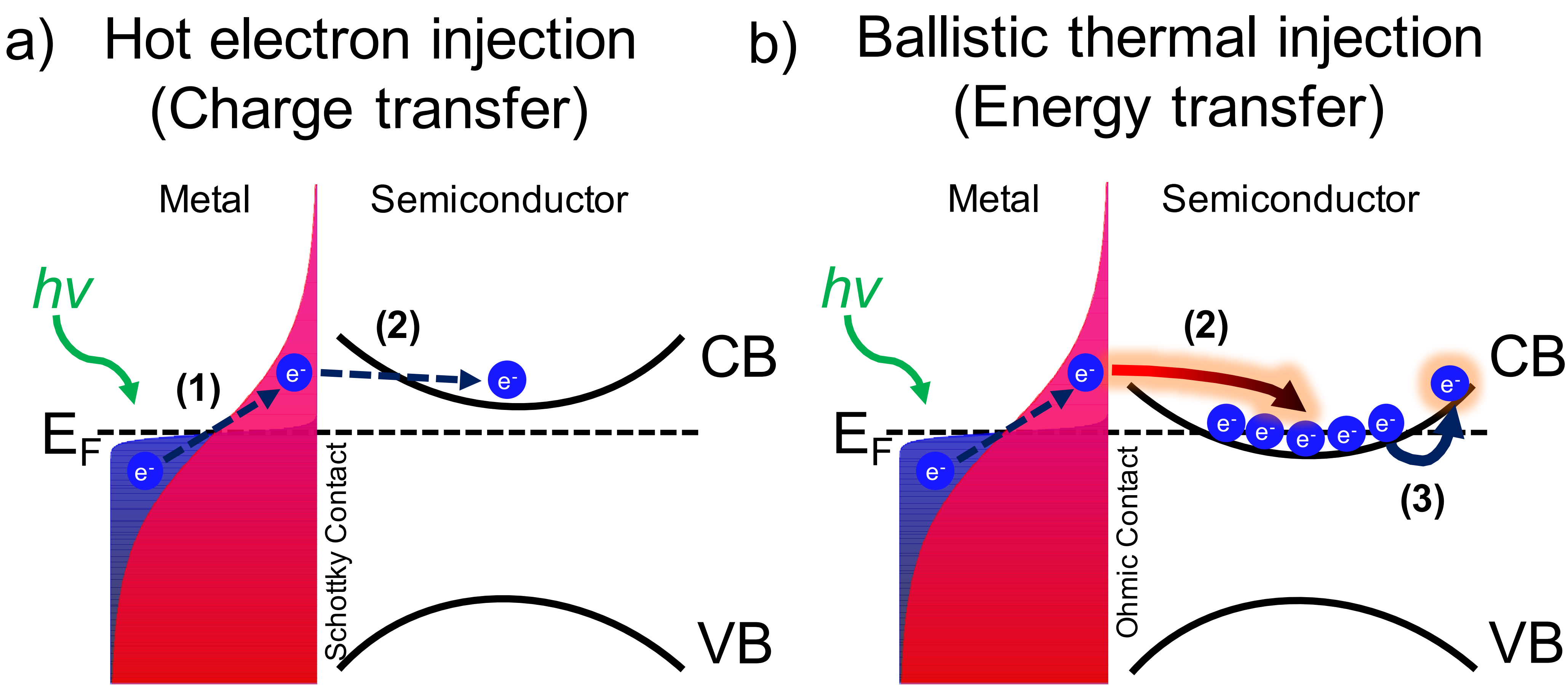
- No back-heating observed for any dopant concentration!



Tomko *et al. Nature Nano.* **16**, 47 (2021)

Ballistic thermal injection

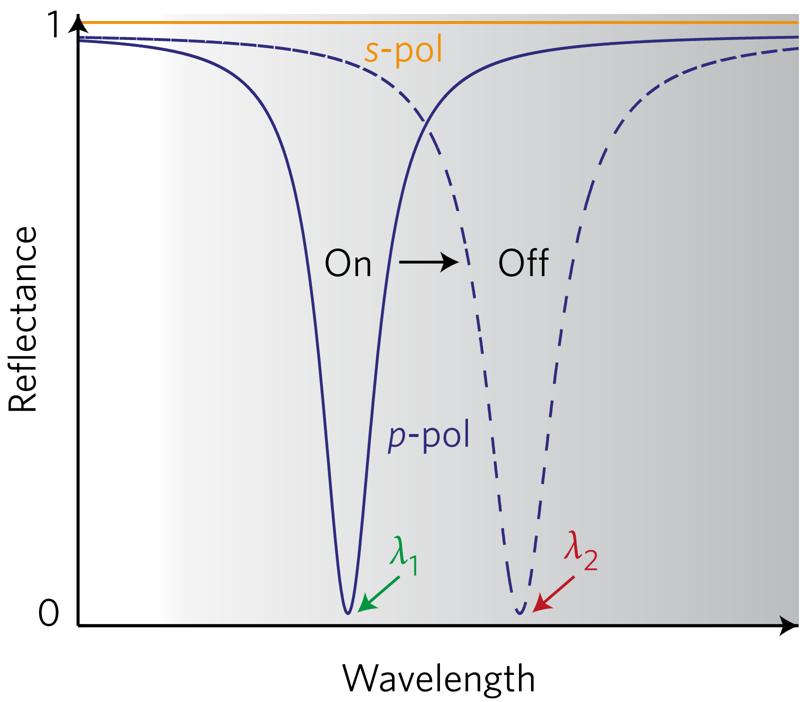
- Can enable a “transient thermal diode” effect
- Energy easily transmitted across interface when traveling ballistically
- Slowly “goes back” across the interface when diffusive
- Is this just hot electron injection (charge)?
  - Too slow of process
  - Can further rule this out by monitoring CdO plasmon response

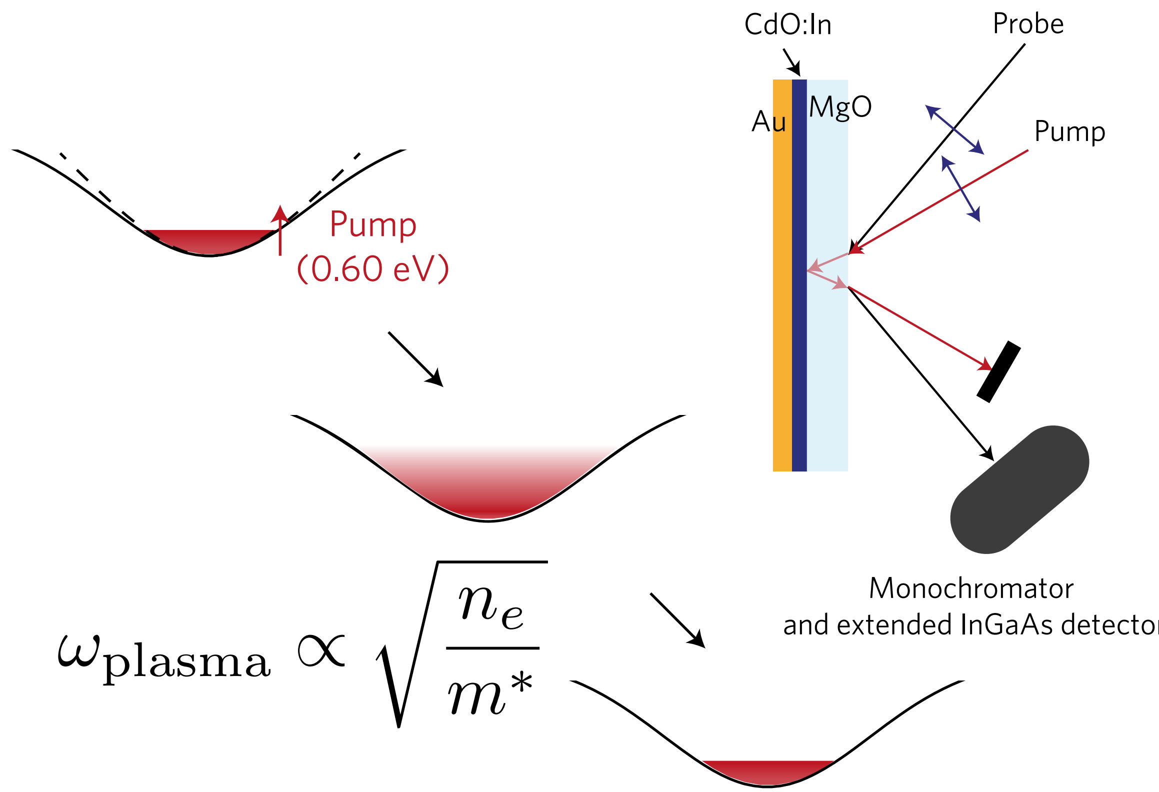


Carrier scattering and relaxation drives optical properties

Recall earlier example

- Changing in carrier density via short pulse absorption can modulate plasmon resonance in CdO
- Can we modulate and control plasmon lifetimes with heat?

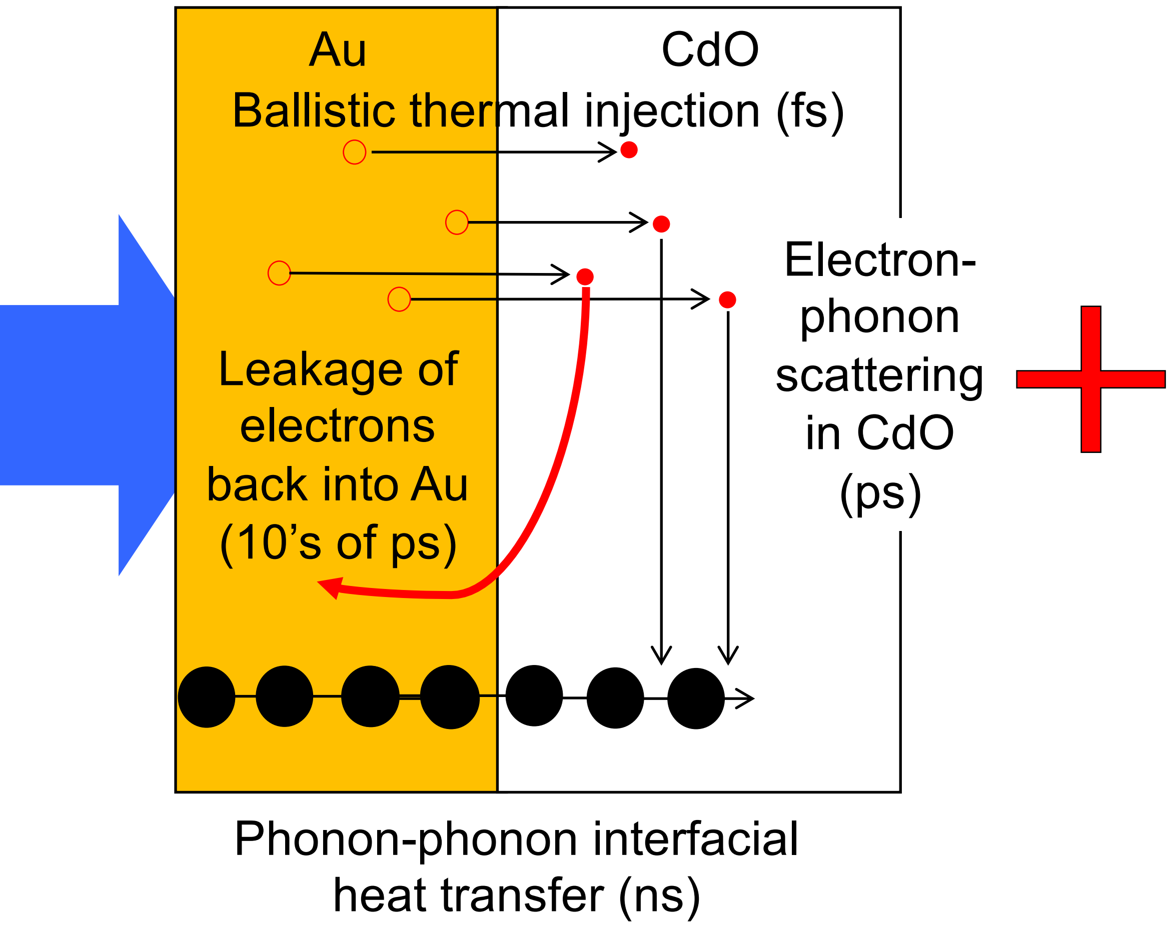


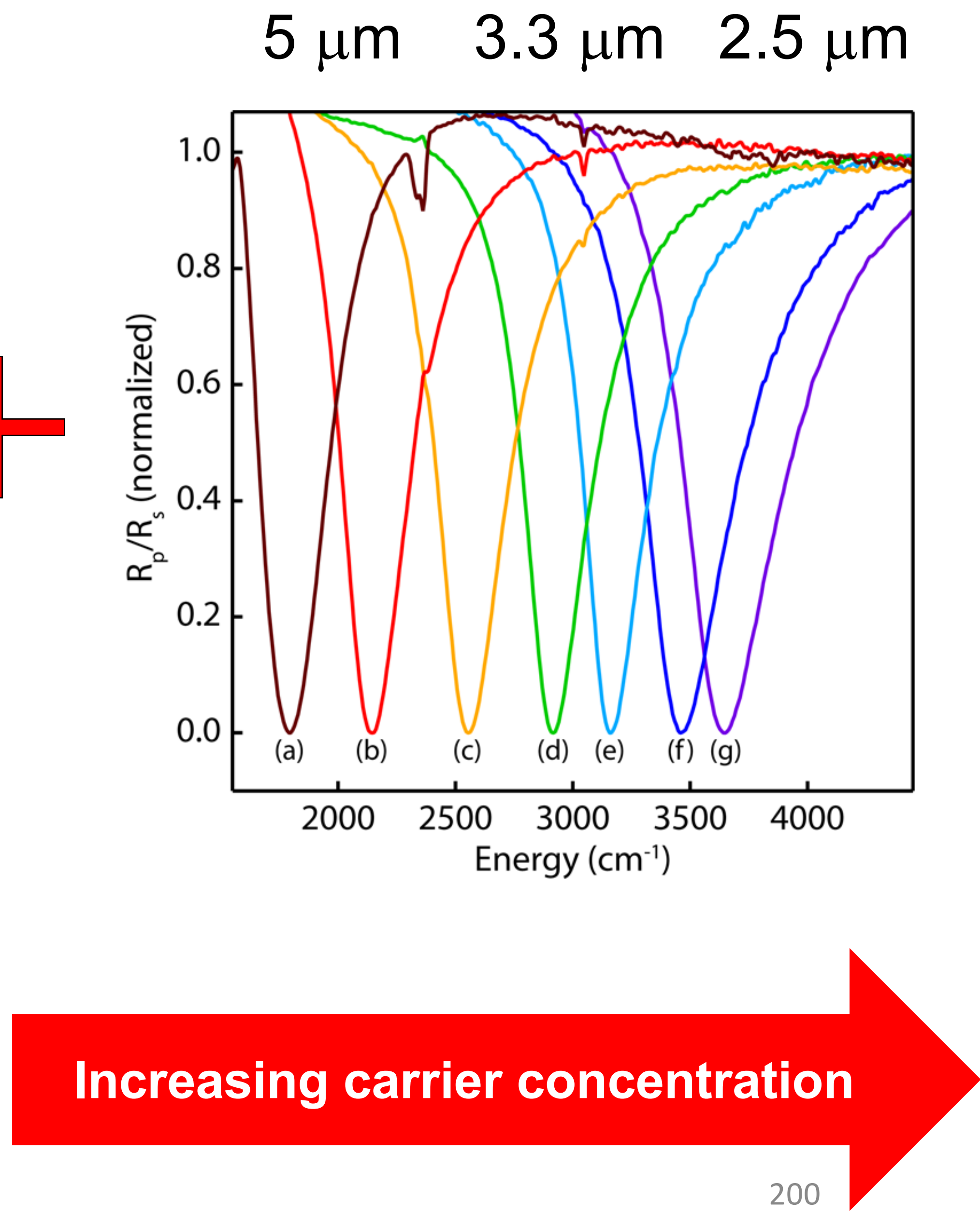


$$\omega_{\mathrm{plasma}} \propto \sqrt{\frac{n_e}{m^*}}$$

Nature Photonics **11**, 390

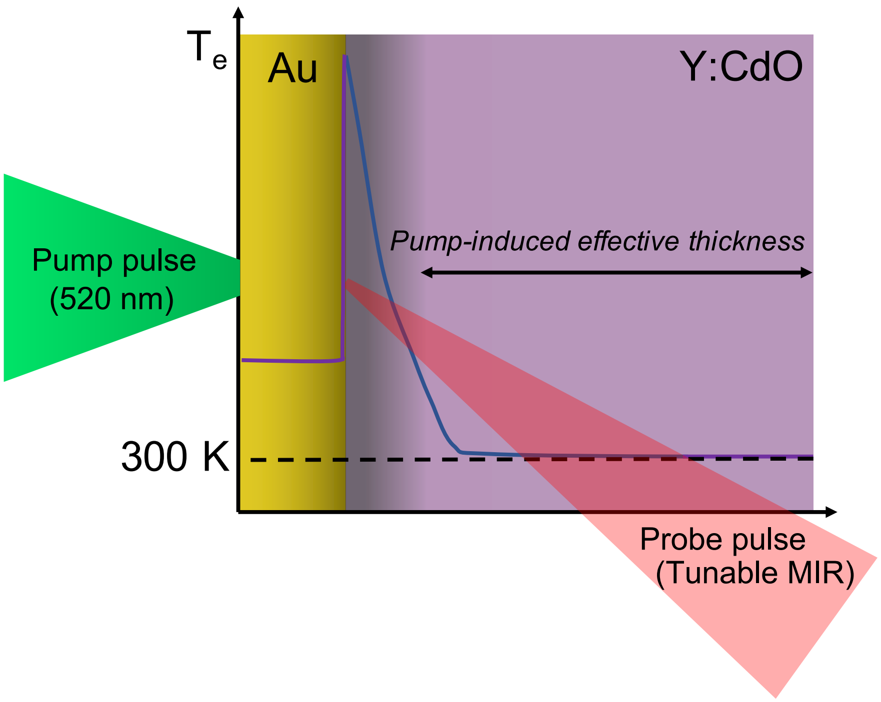
Nonequilibrium electrons to control CdO plasmons



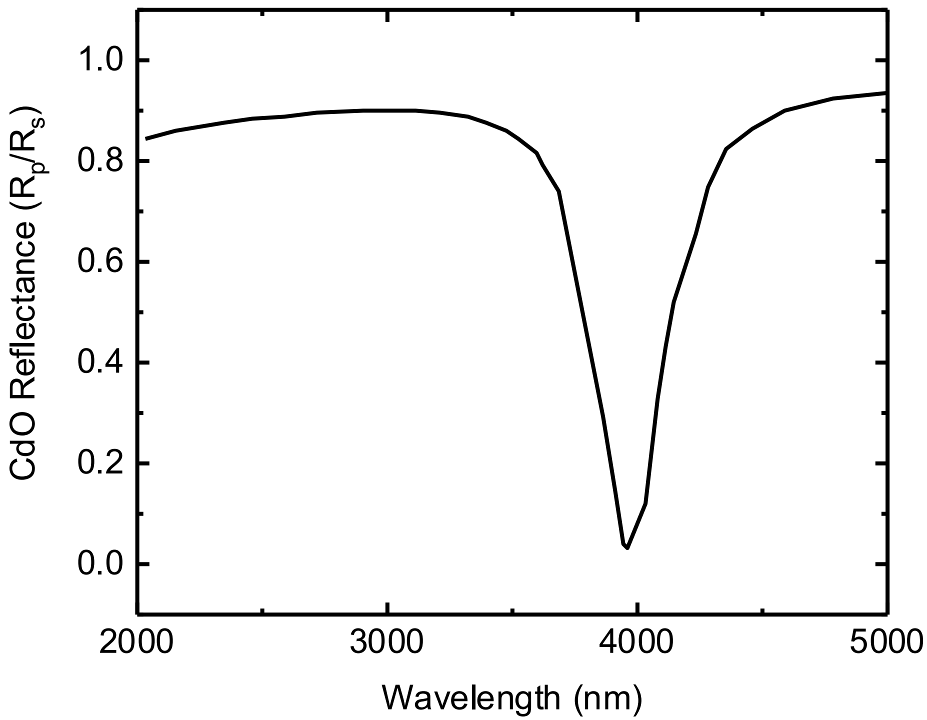


Pump electrons in Au, probe plasmon in CdO

How is the IR plasmon response of CdO impacted by ballistic thermal injection?



Au/CdO/sapphire absorption response

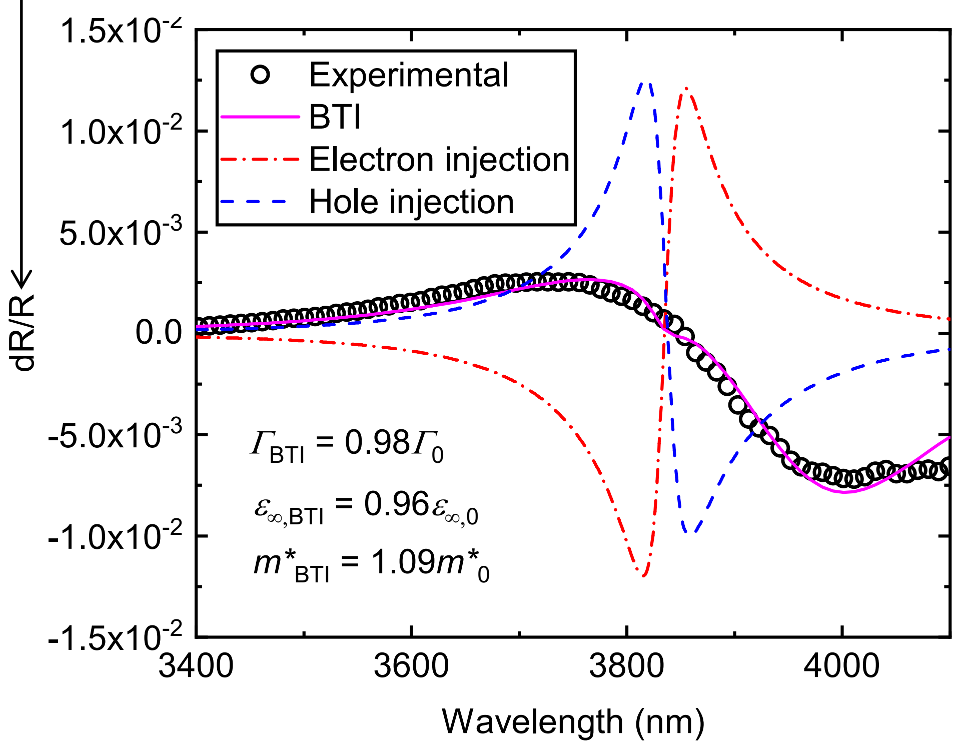


Tomko *et al. Nature Nano.* **16**, 47 (2021)

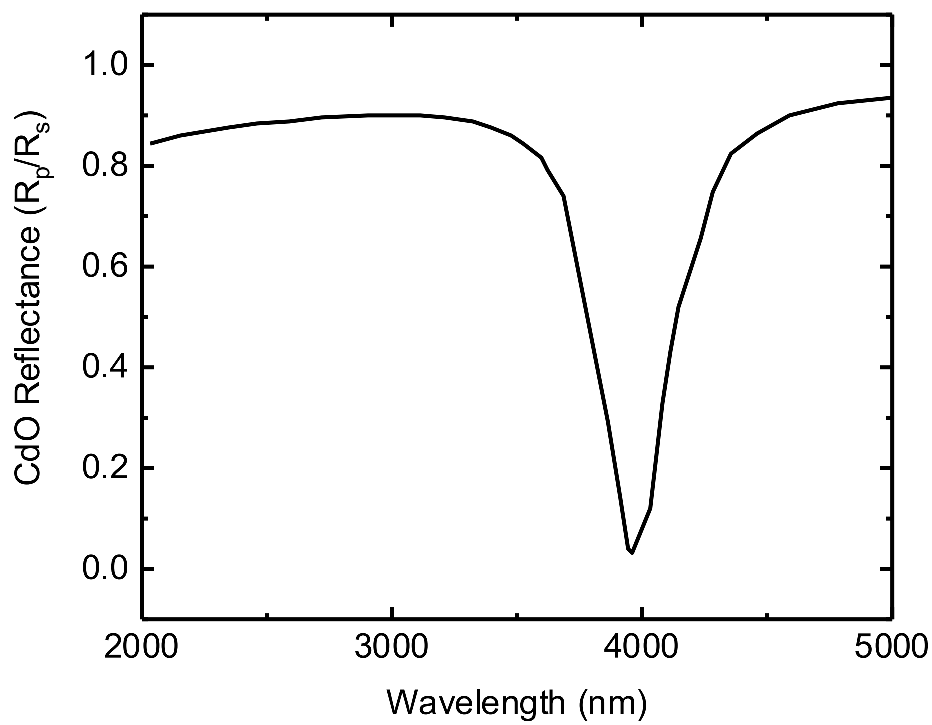
Pump electrons in Au, probe plasmon in CdO

Asymmetric red shift in ENZ plasmon mode due to BTI

Note we are measuring *change* in reflectivity, dR



Au/CdO/sapphire absorption response

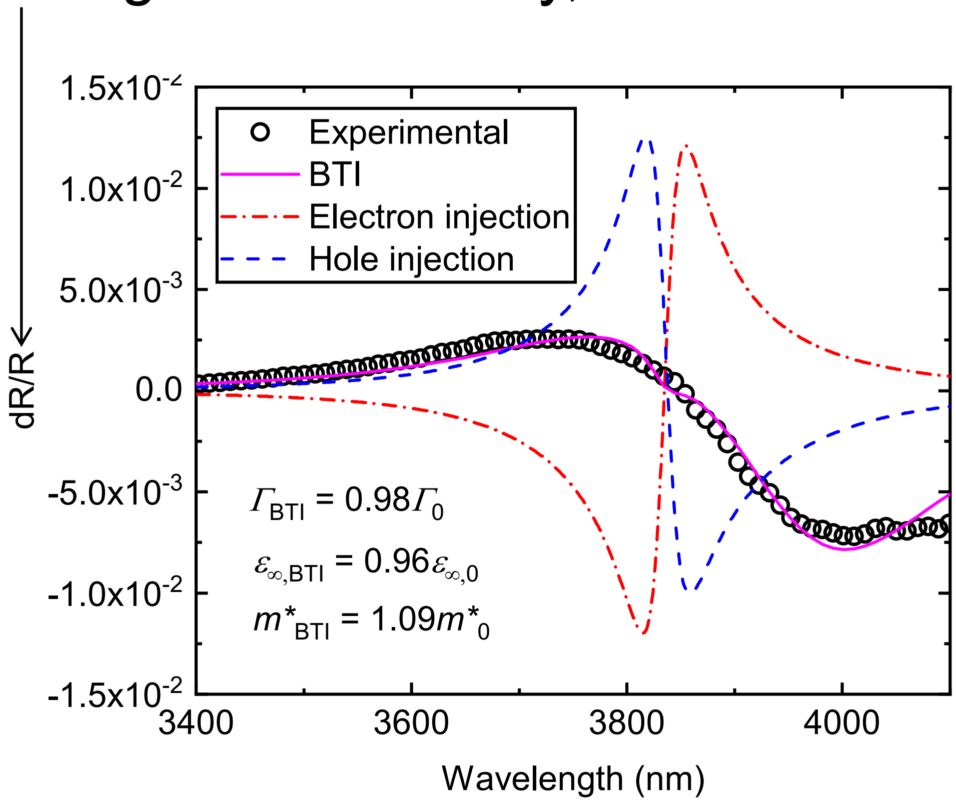


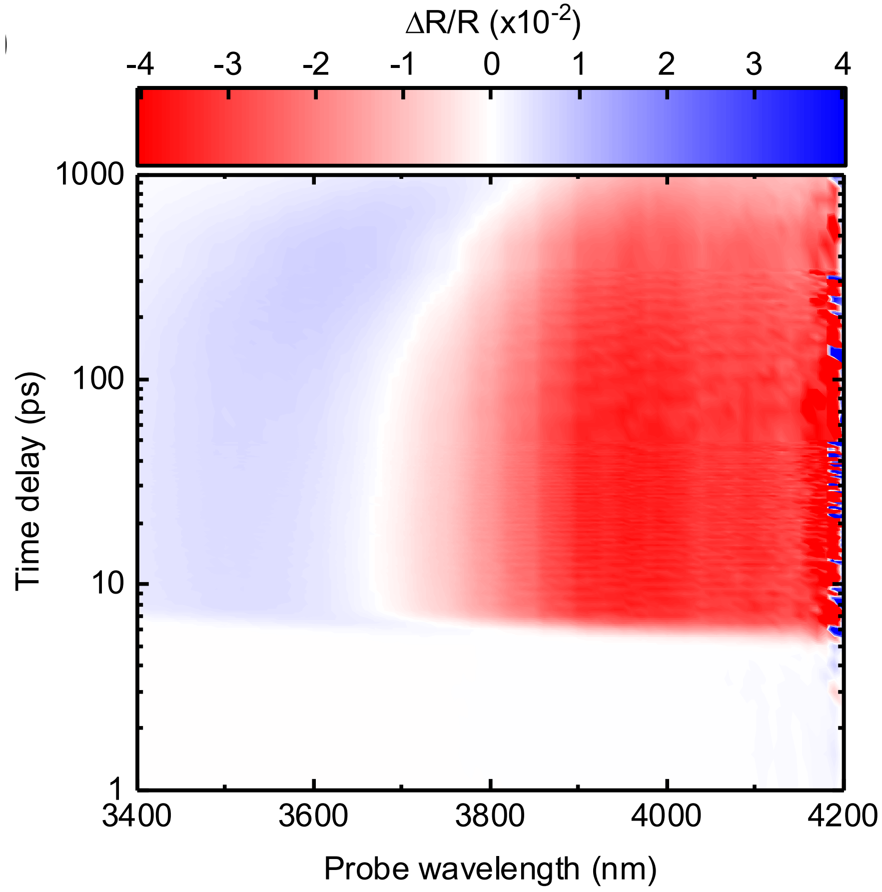
Tomko *et al. Nature Nano.* **16**, 47 (2021)

Pump electrons in Au, probe plasmon in CdO

Asymmetric red shift in ENZ plasmon mode due to BTI

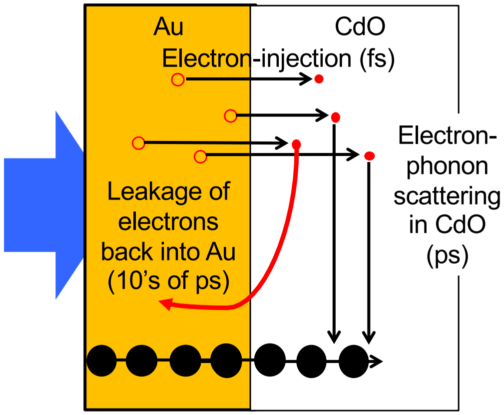
Note we are measuring *change* in reflectivity, dR



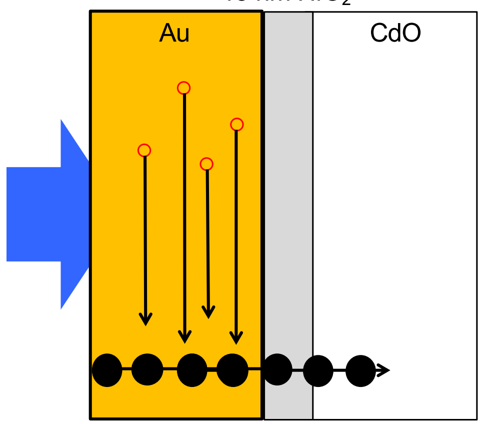


Tomko *et al. Nature Nano.* **16**, 47 (2021)

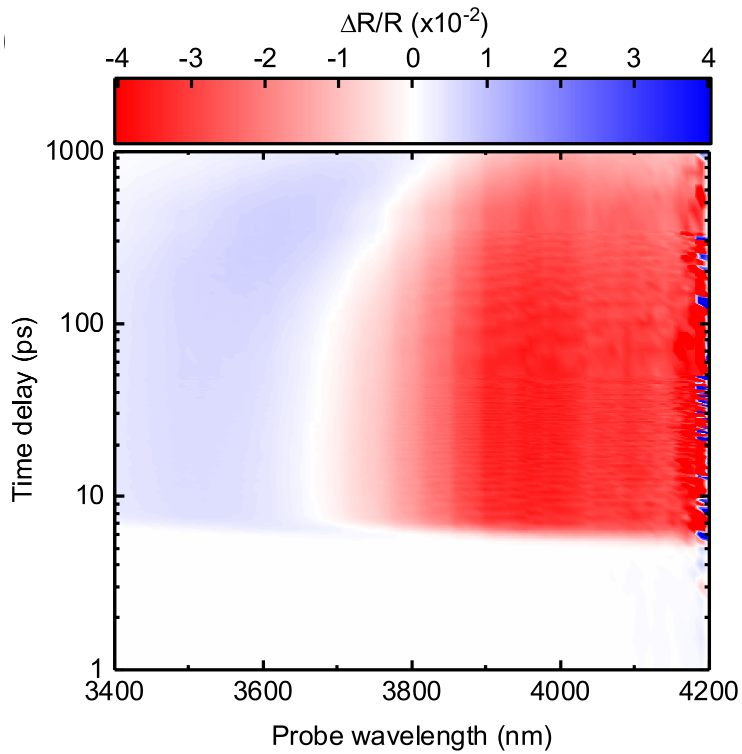
And it's not an optical artifact



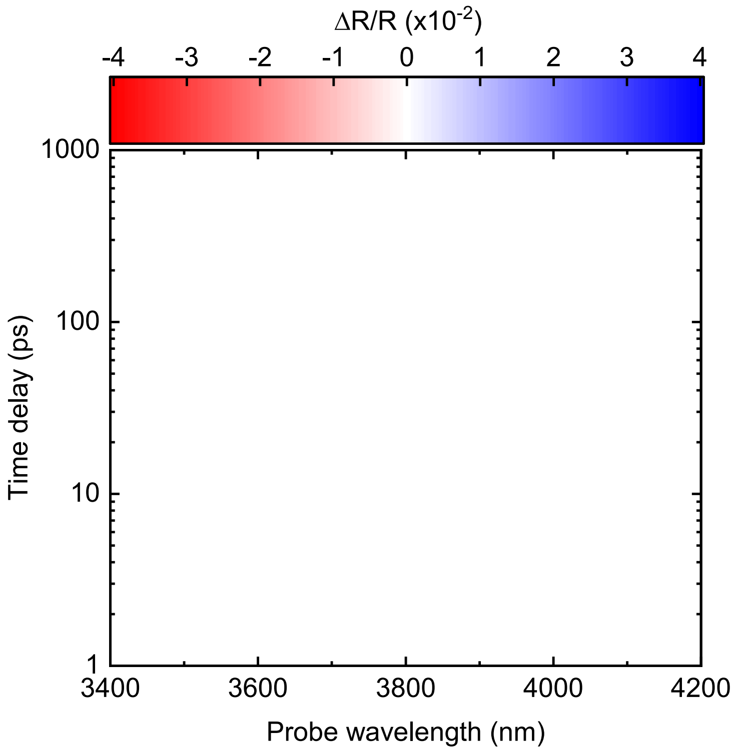
15 nm HfO_2 layer prevents any electron energy from moving from Au to CdO, resulting in no measurable response



Au/CdO



Au/HfO_2/CdO (same scale)



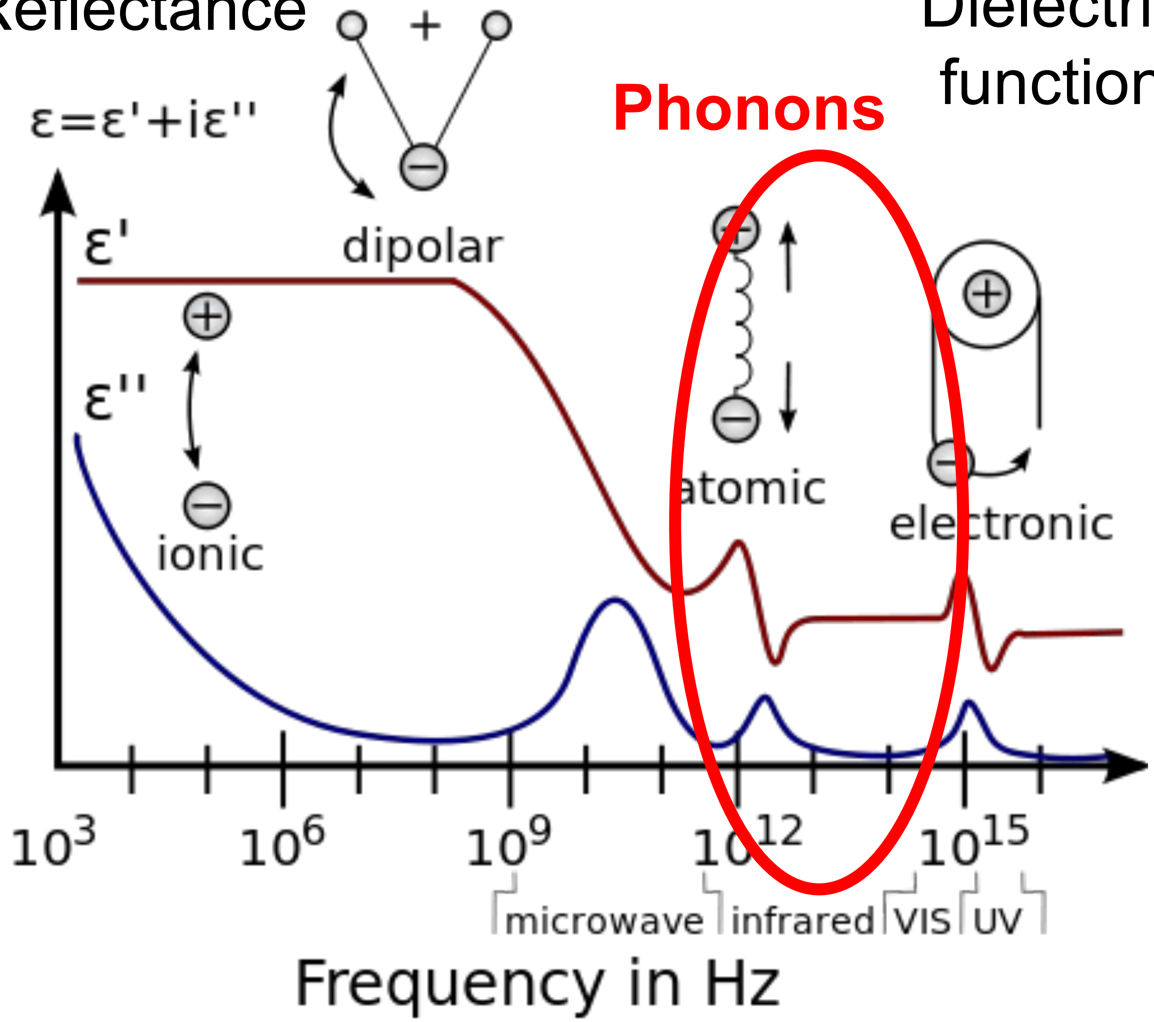
Measuring thermal lifetimes/scattering rates

Absorption

$$\Delta R \propto \Delta\alpha \propto \Delta\varepsilon\,(T, \tau)$$

Reflectance

Dielectric function



"Permittivity" Wikipedia

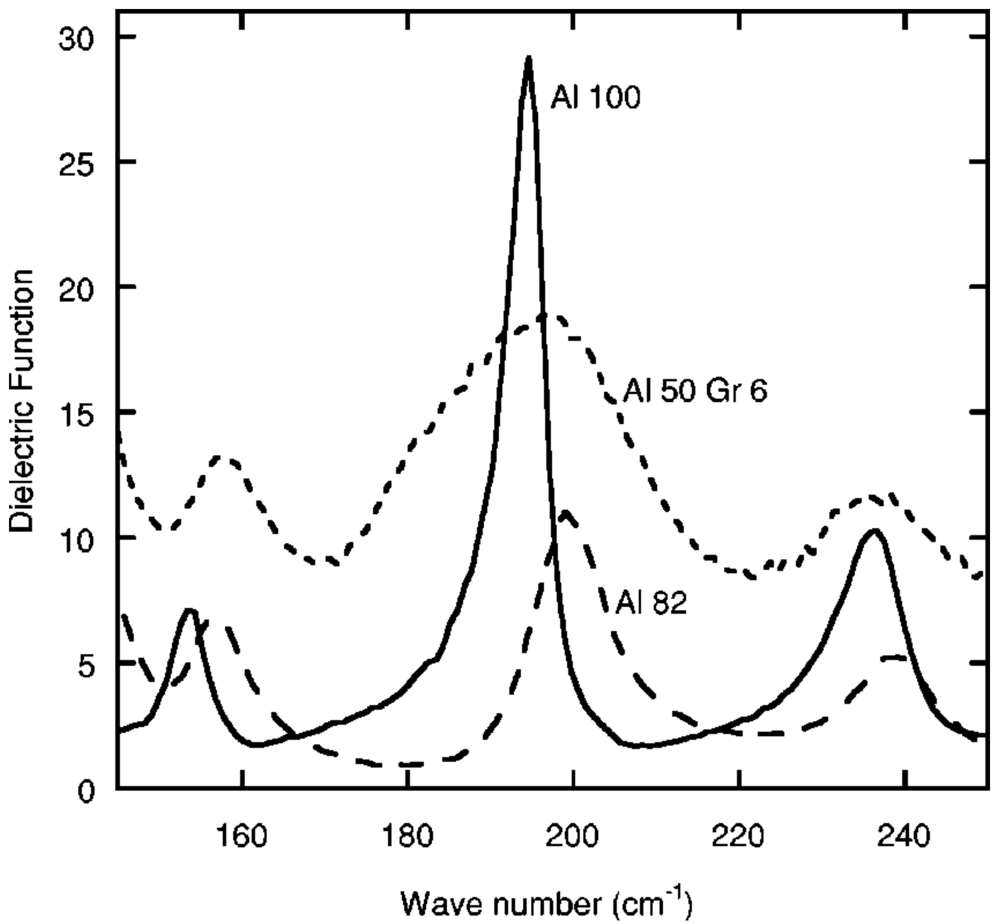
Spectral measurements of phonon lifetimes

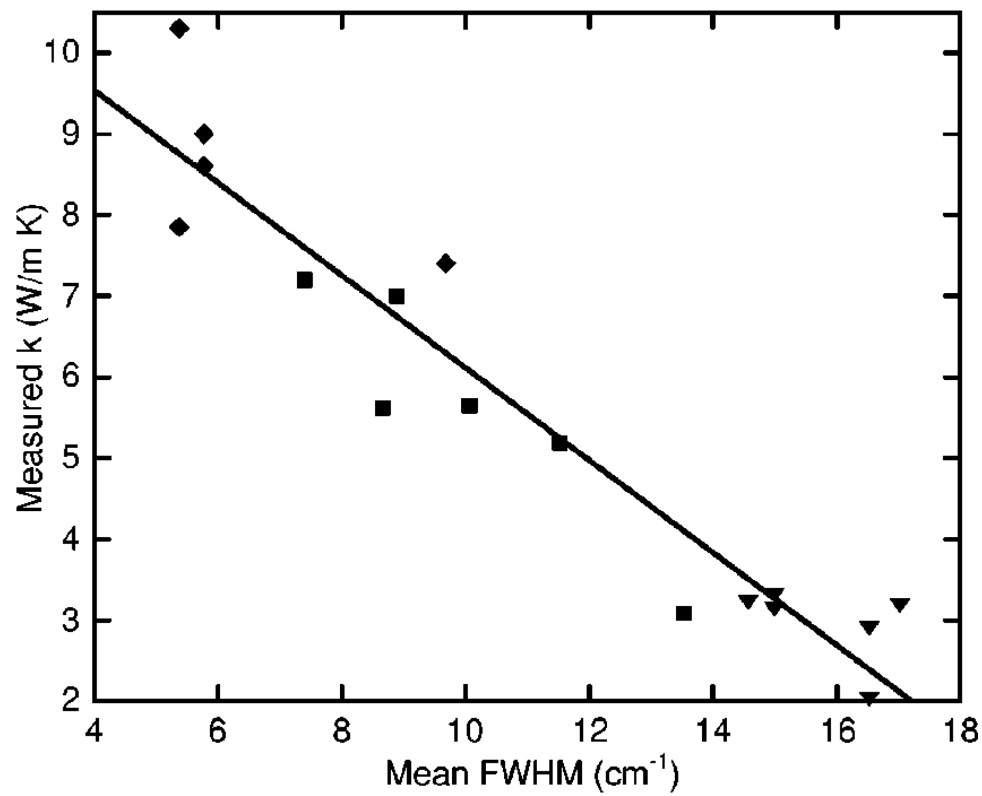
Absorption

$$\Delta R \propto \Delta\alpha \propto \Delta\varepsilon\,(T,\tau)$$

Reflectance

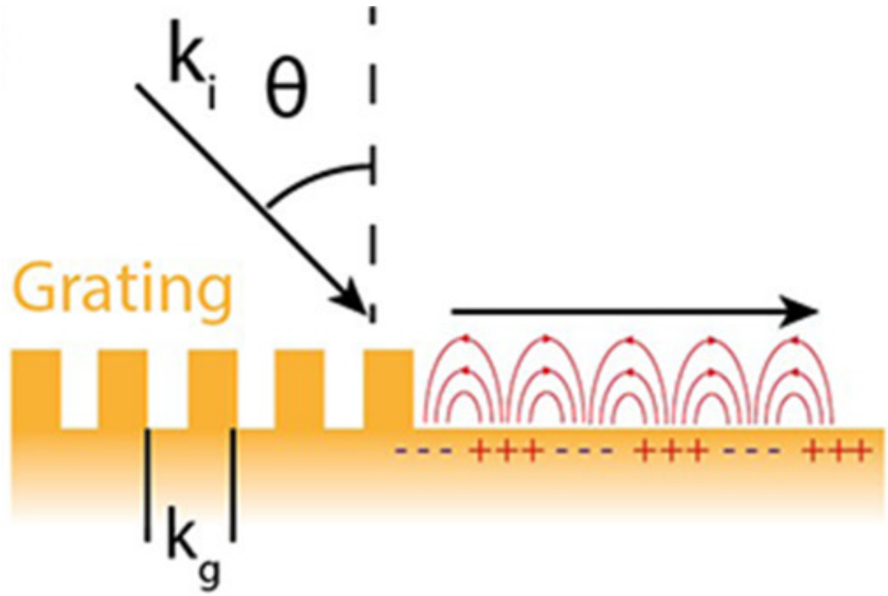
Dielectric function



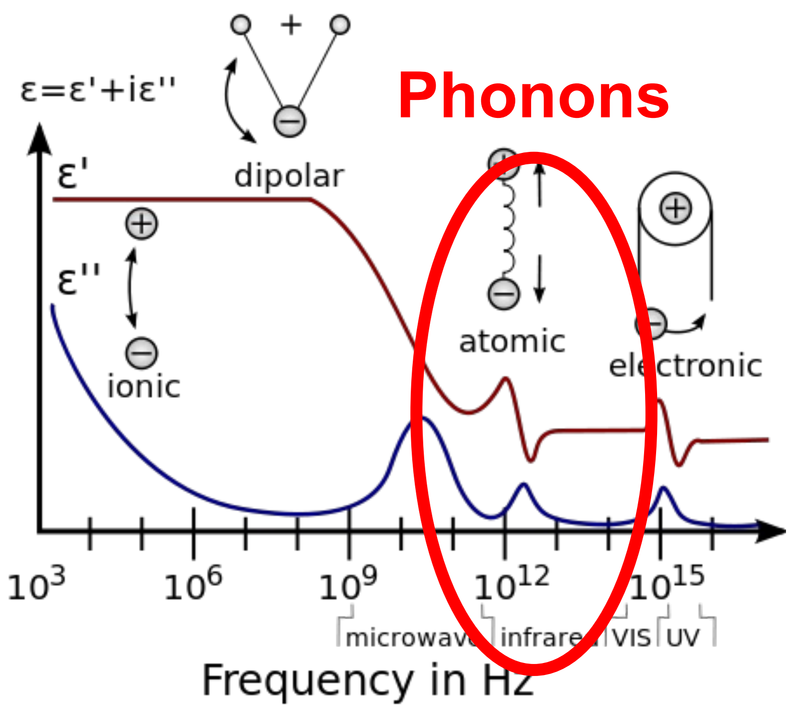


Giesting and Hofmeister, *PRB* **65**, 144305

Phonon-polaritons: enhancing heat transfer?



J. Appl. Phys. **125**, 191102 (2019)

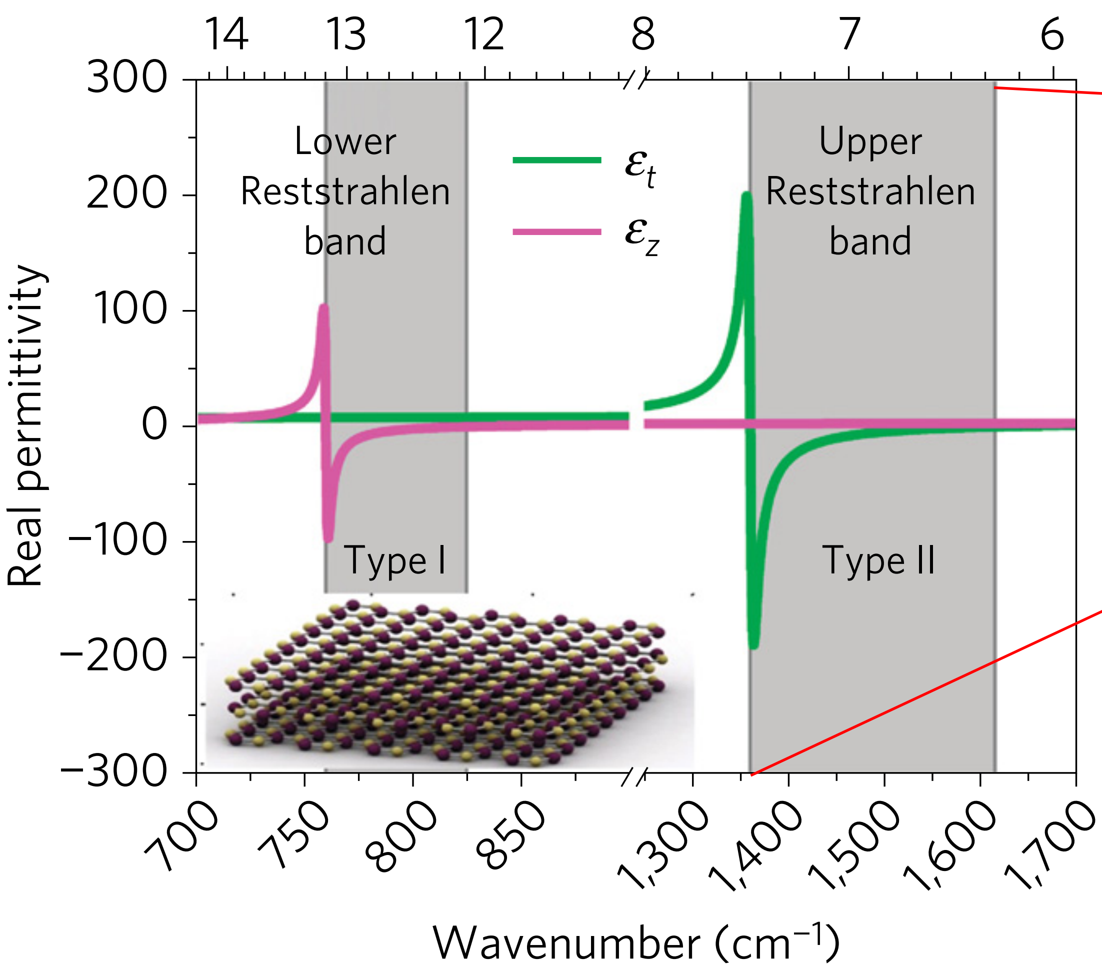


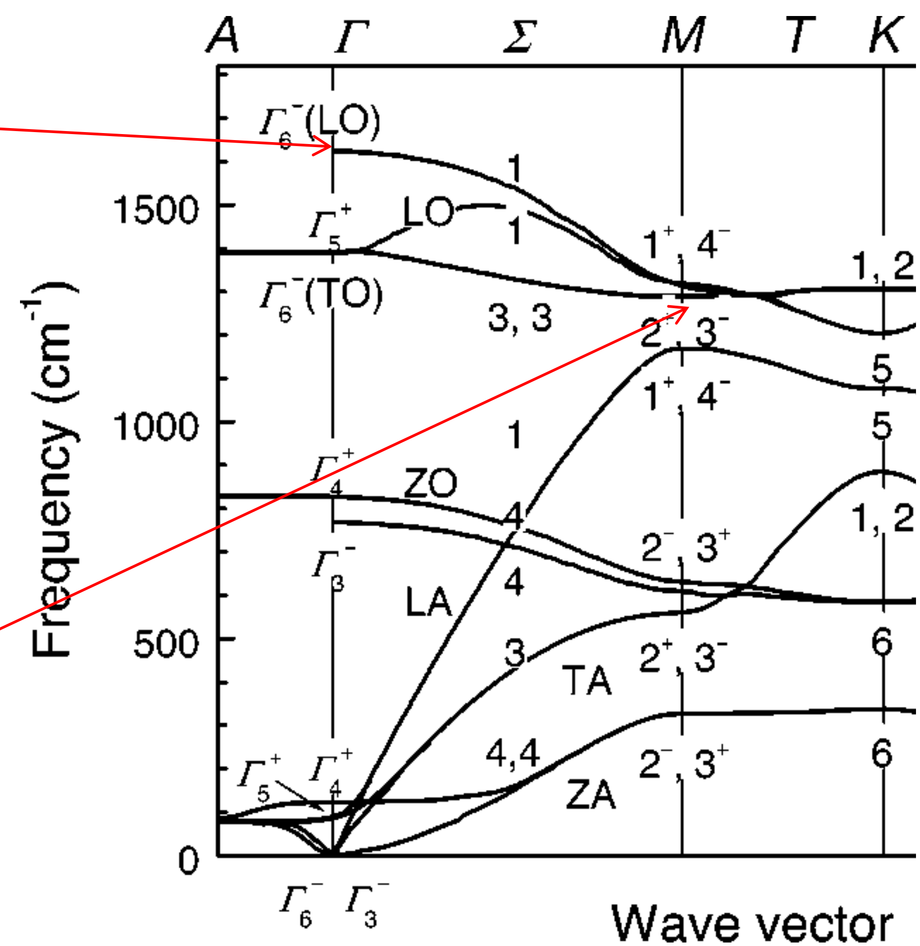
- Light couples with electric dipole creating quasiparticle
- At IR wavelengths: Phonon polariton!
- Quasiparticle can propagate at ~1% of the speed of light!

$$\kappa = \frac{1}{3} C v \lambda = \frac{1}{3} C v_g^2 \tau$$

Spectral measurements of polariton lifetimes

- Phonon-polaritons (PhP) in h-BN exist in Reststrahlen band bounded by optical phonon energies at Γ-point



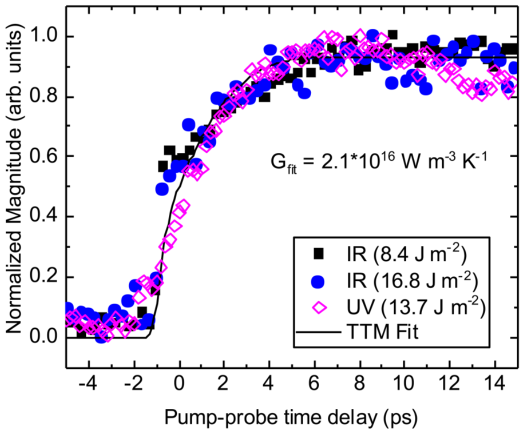


Review Article: *Nature Mat.* **16**, 182

PRB **71**, 205201

Can we modulate and control PhP lifetimes with heat?

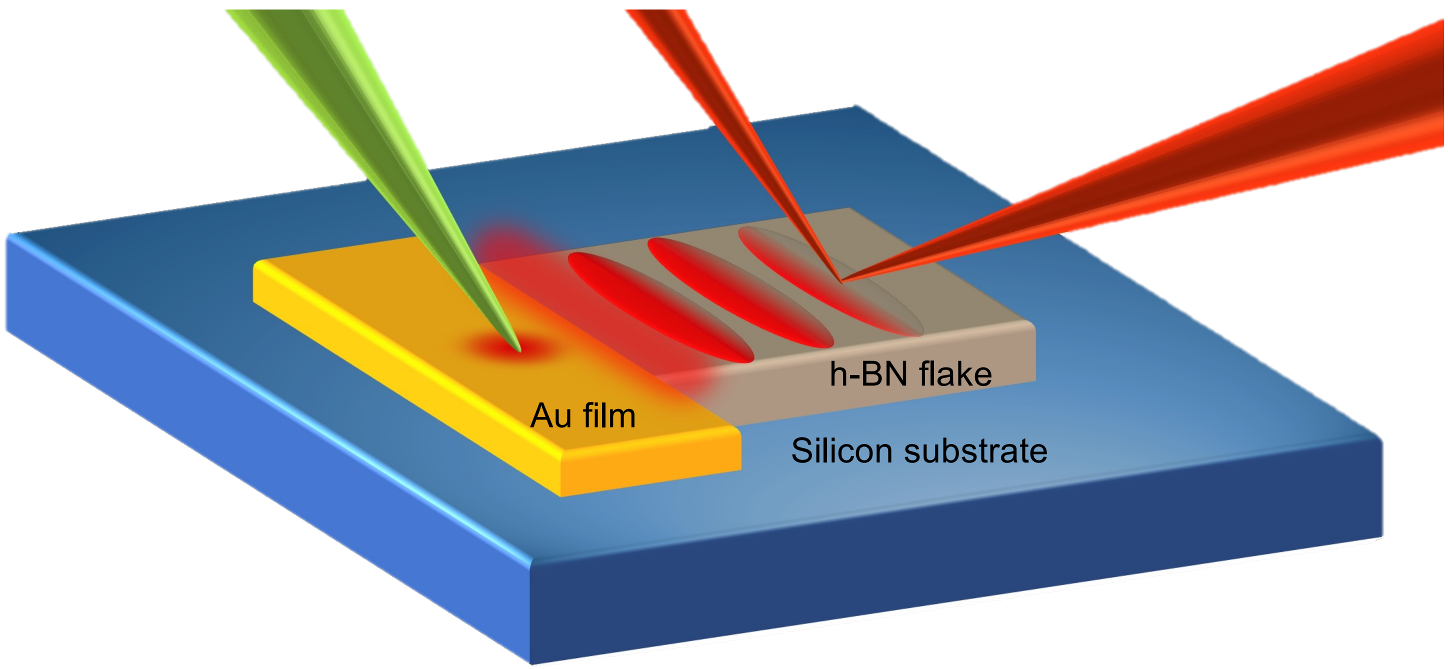
- **Can we modulate and control PhP lifetimes with heat?**



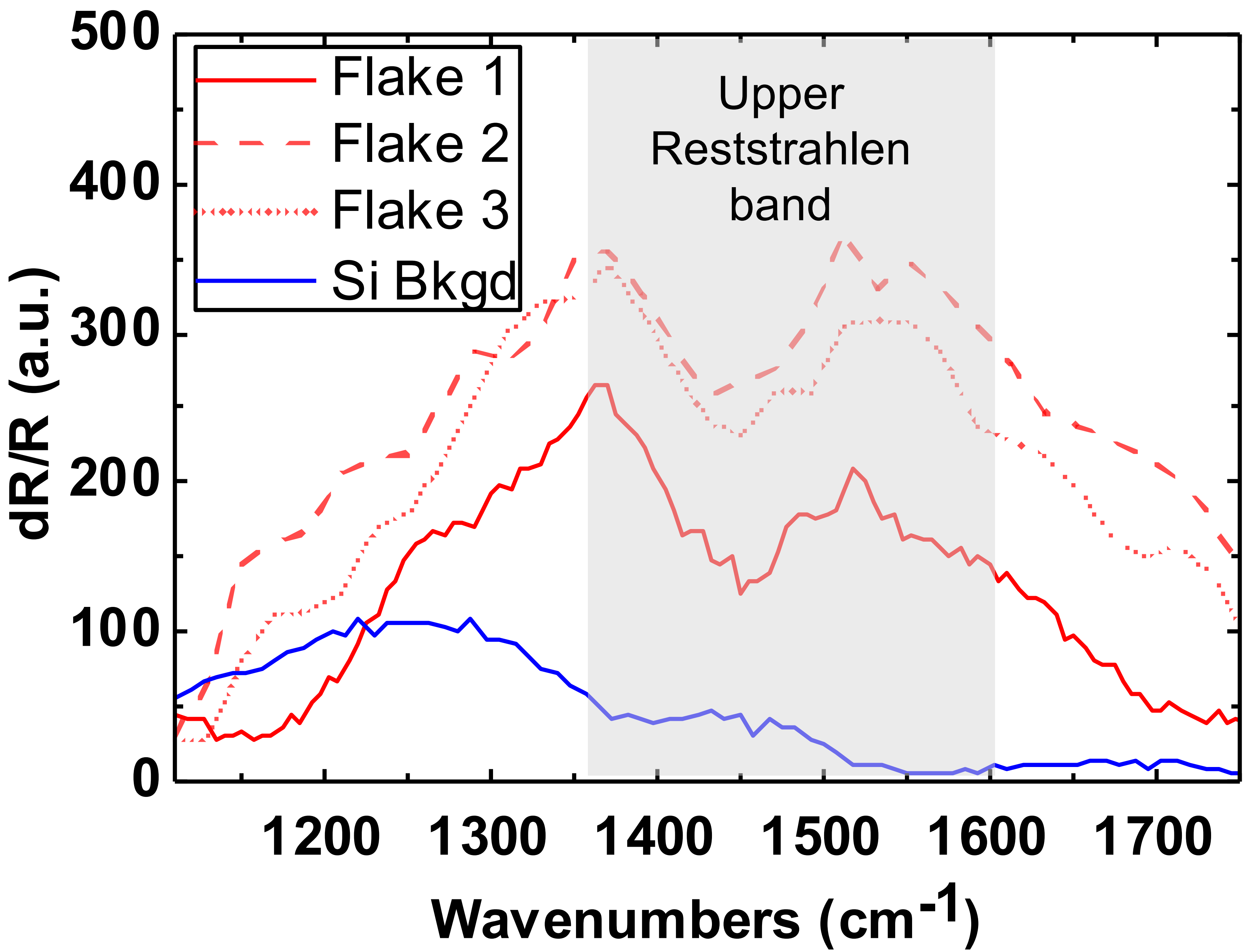
520 nm pump heats Au film in < 5 ps

- MIR time delayed probe of ΔR across upper Reststrahlen band
- How does heat from Au indirectly change PhP in h-BN?

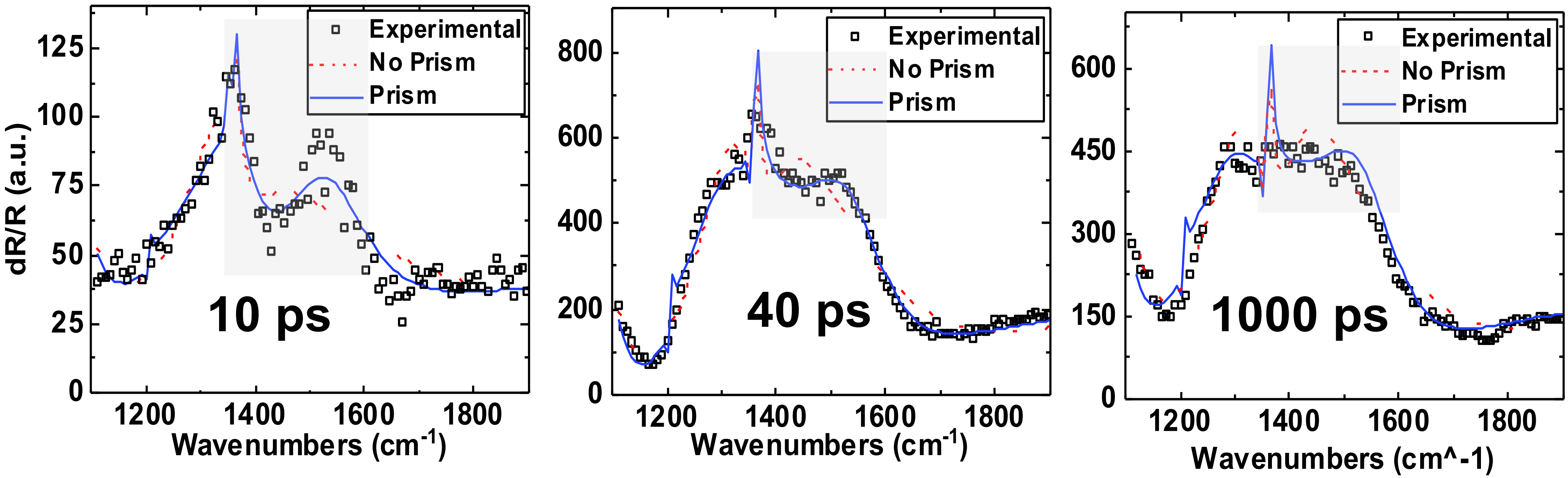
Will Hutchins
Daniel Hirt



Can we modulate and control PhP lifetimes with heat?



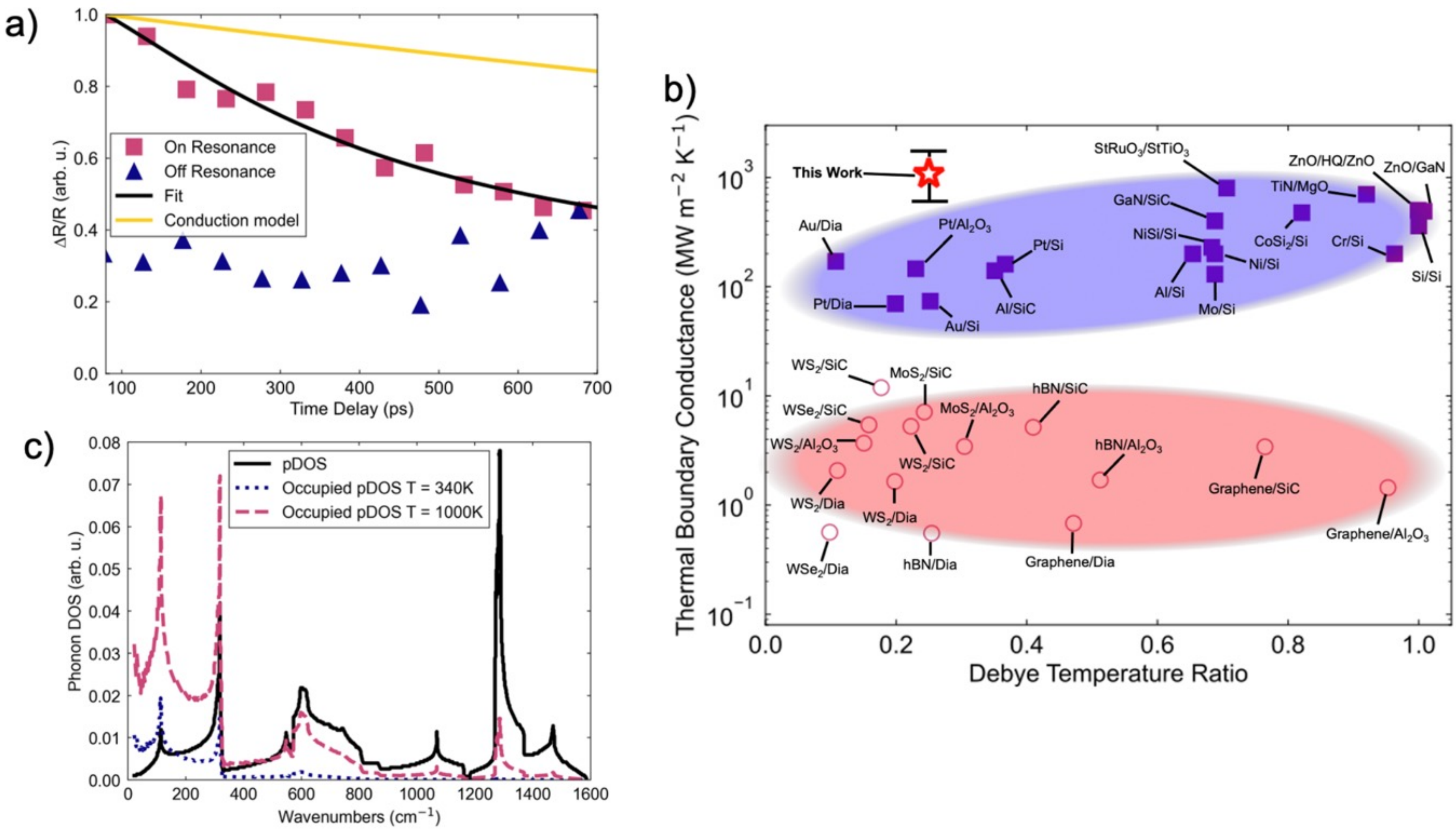
Can we modulate and control PhP lifetimes with heat? Yes



- **Short times (10 ps), gold radiatively couples to PhPs in h-BN**
  - Too short time for phonon TBC causing optical phonon heating
  - Reststrahlen band too high energy for thermalized distribution from phonon conduction
- **Radiatively excited PhP in h-BN decays into thermalized optical phonon distribution**
  - PhP sinks heat from Au via radiation faster than other optical phonon modes from conduction
- **Conductive processes at long time, MIR probes optical phonon reflectivity changes due to Au heating**

Enhanced heat sinking with PhP coupling

- Temperatures of PhP modes decay an order of magnitude faster than non-PhP modes
- Order of magnitude enhance thermal conductance away from hot spot (Au/h-BN thermal boundary conductance increase)



Implication: Radiation heat sinking?? Radiation detection??

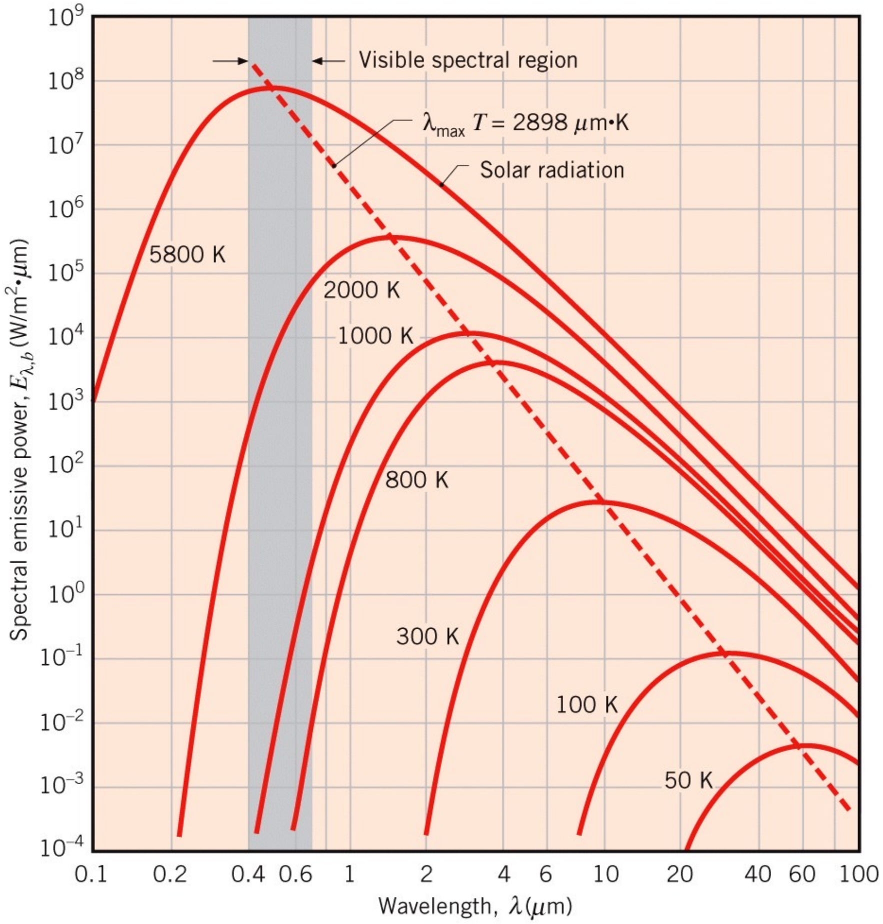
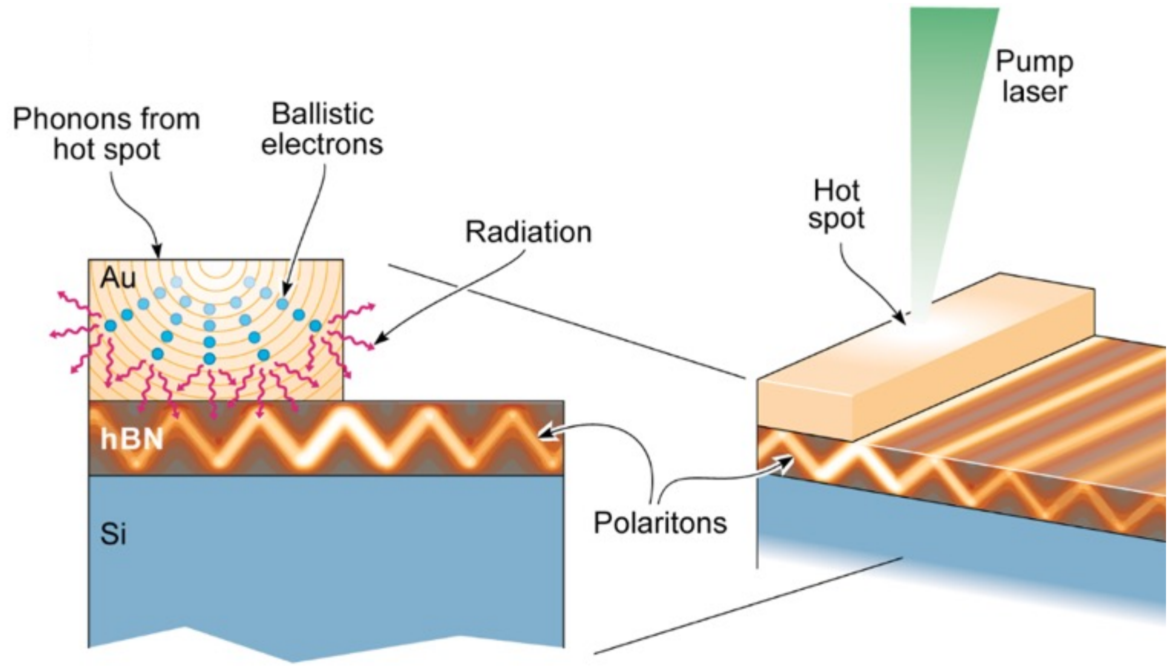


Figure 12.12

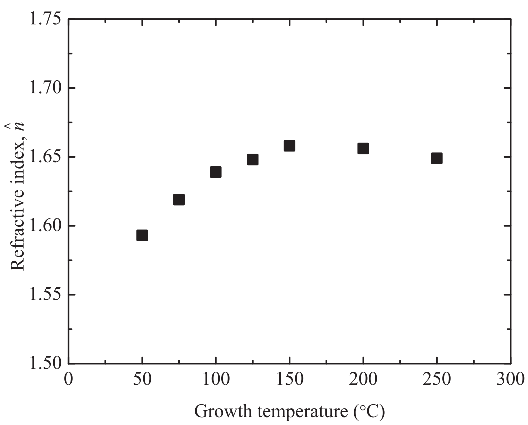


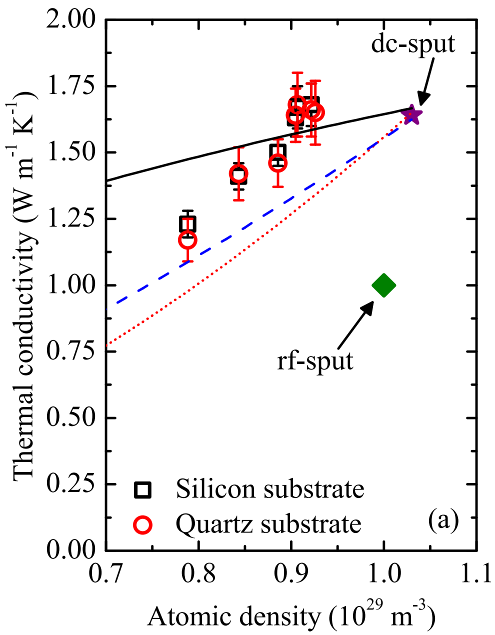
Fundamentals of Heat and Mass Transfer

Outline

1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

Deposition/processing effects on thermal conductivity





Deposition temperature (°C)	Amorphous film thickness (nm)	Index of refraction	Atomic densities (10^{28} m^{-3})	Al_2O_3 thermal conductivity (W m^{-1} K^{-1})
50	≤ 20	1.60	8.02	1.28 ± 0.13
50	50.6	1.60	8.02	1.35 ± 0.21
200	≤ 20	1.66	9.29	1.88 ± 0.19
200	59.7	1.66	9.29	1.87 ± 0.26
300	50.5	1.71	9.48	2.13 ± 0.15
300	96.6	1.71	9.48	2.02 ± 0.07
300	202.2	1.71	9.48	1.98 ± 0.06

APPLIED PHYSICS LETTERS **104**, 253107 (2014)

Density dependence of the room temperature thermal conductivity of atomic layer deposition-grown amorphous alumina (Al_2O_3)

Caroline S. Gorham,[1] John T. Gaskins,[1] Gregory N. Parsons,[2] Mark D. Losego,[2] and Patrick E. Hopkins[1,a)]
[1]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, USA*

ECS Journal of Solid State Science and Technology, **6** (10) N189-N208 (2017)

Review—Investigation and Review of the Thermal, Mechanical, Electrical, Optical, and Structural Properties of Atomic Layer Deposited High-k Dielectrics: Beryllium Oxide, Aluminum Oxide, Hafnium Oxide, and Aluminum Nitride

John T. Gaskins,[a] Patrick E. Hopkins,[a,b,c,z] Devin R. Merrill,[d,e] Sage R. Bauers,[d] Erik Hadland,[d] David C. Johnson,[d,z] Donghyi Koh,[e,f] Jung Hwan Yum,[f] Sanjay Banerjee,[f,*] Bradley J. Nordell,[g] Michelle M. Paquette,[g] Anthony N. Caruso,[g] William A. Lanford,[h] Patrick Henry,[e] Liza Ross,[e] Han Li,[e] Liyi Li,[e] Marc French,[e] Antonio M. Rudolph,[e] and Sean W. King[e,*,z]

Thin Solid Films 650 (2018) 71–77

Contents lists available at ScienceDirect

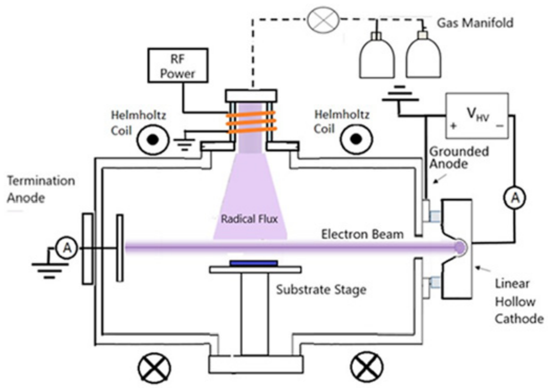
Thin Solid Films

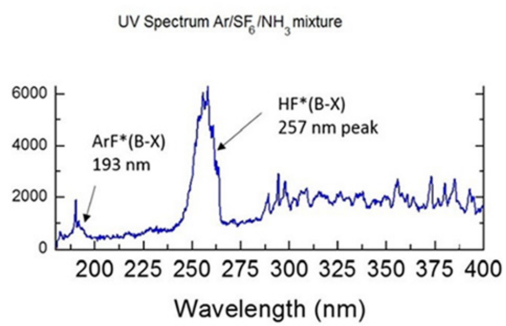
journal homepage: www.elsevier.com/locate/tsf

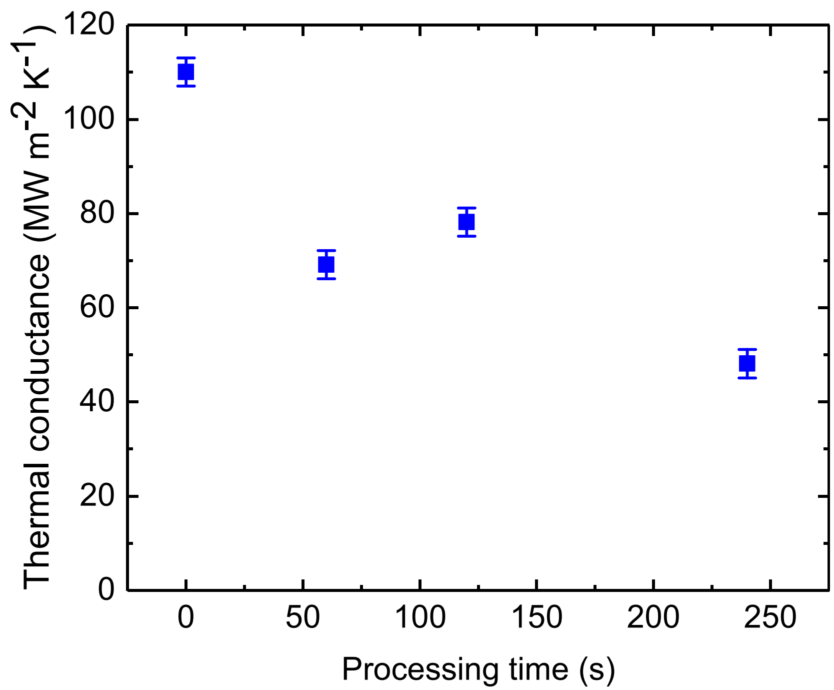
Density and size effects on the thermal conductivity of atomic layer deposited TiO_2 and Al_2O_3 thin films

Mallory E. DeCoster[a], Kelsey E. Meyer[b], Brandon D. Piercy[c], John T. Gaskins[a], Brian F. Donovan[d], Ashutosh Giri[a], Nicholas A. Strnad[e,f], Daniel M. Potrepka[g], Adam A. Wilson[h], Mark D. Losego[c], Patrick E. Hopkins[a,b,i,*]

Deposition/processing effects on thermal conductivity







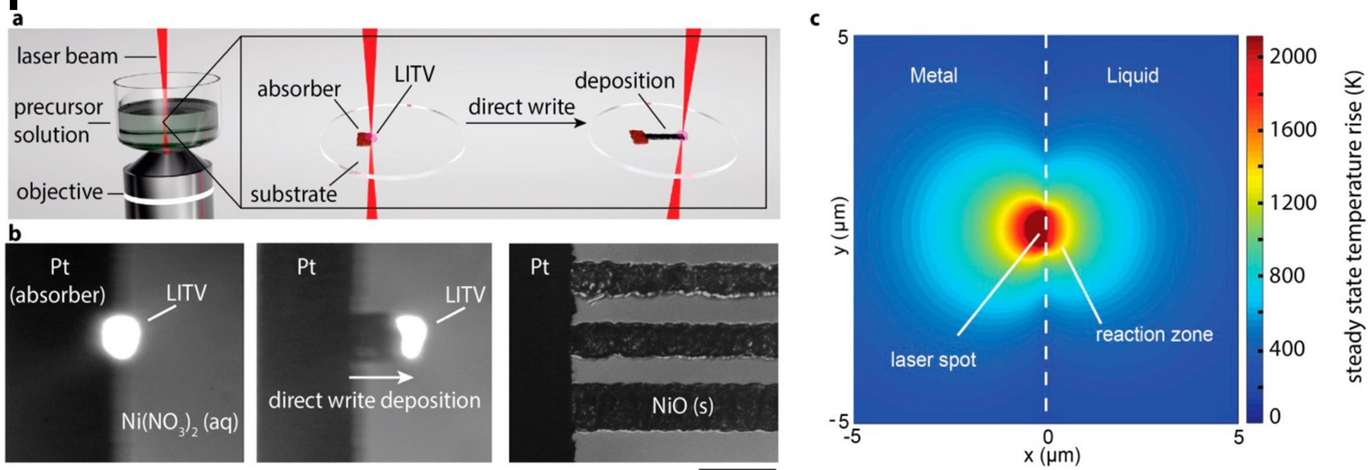
Thermal conductance of aluminum oxy-fluoride passivation layers

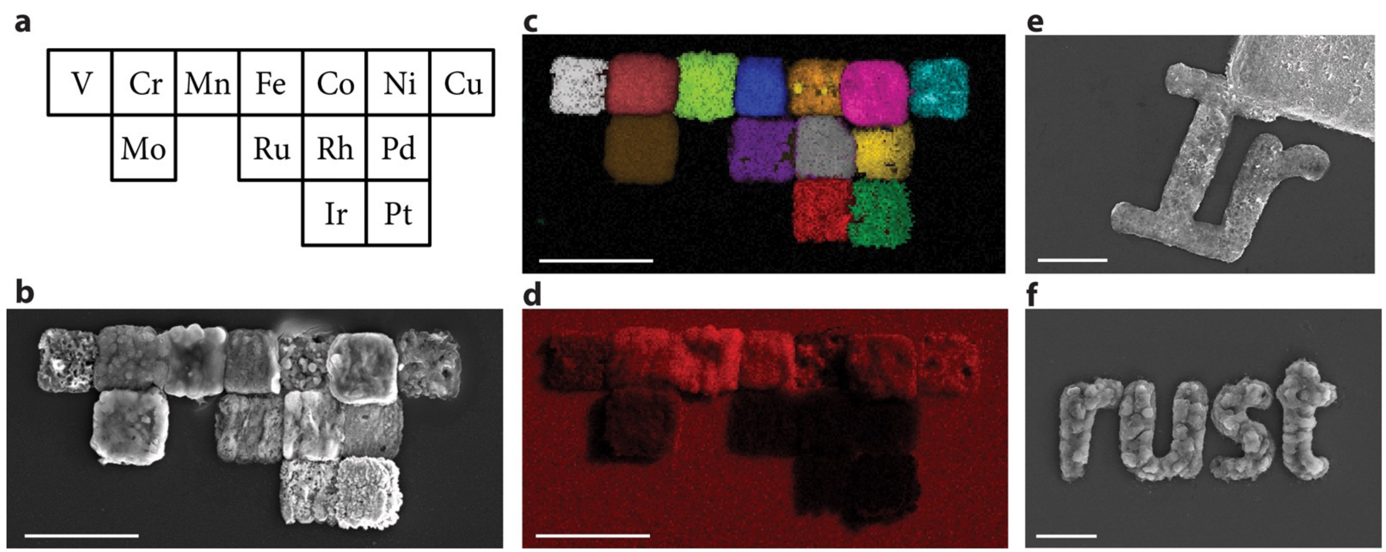
Cite as: Appl. Phys. Lett. **115**, 191901 (2019); doi: 10.1063/1.5120028
Submitted: 16 July 2019 · Accepted: 18 October 2019 ·
Published Online: 4 November 2019

View Online Export Citation CrossMark

John A. Tomko,[1] David R. Boris,[2] Samantha G. Rosenberg,[3,a)] Scott G. Walton,[2] and Patrick E. Hopkins[1,4,5,b)]

Laser-based direct writing: a thermally driven process



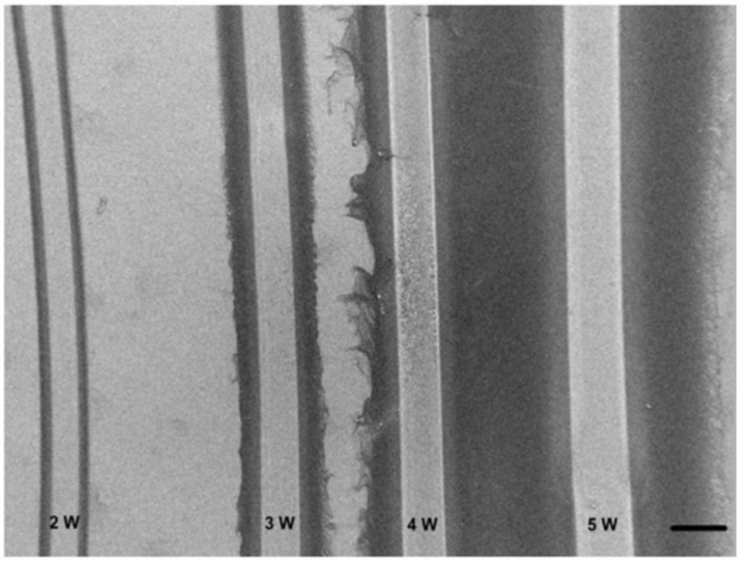


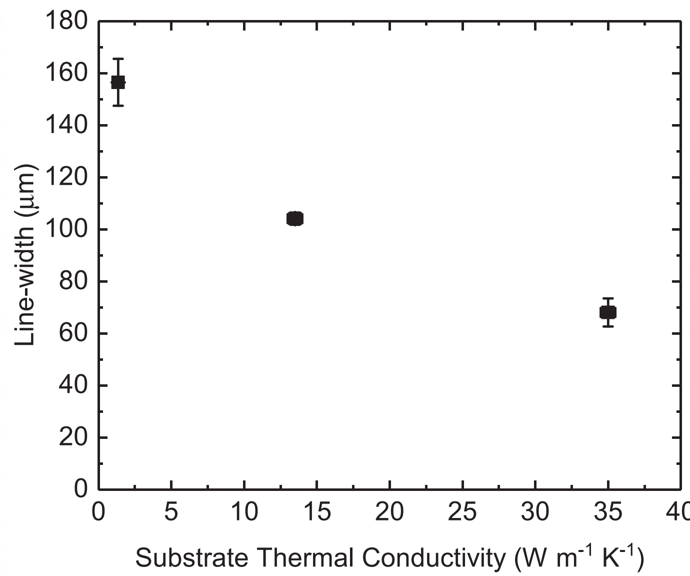
ACS APPLIED MATERIALS & INTERFACES
Letter
www.acsami.org

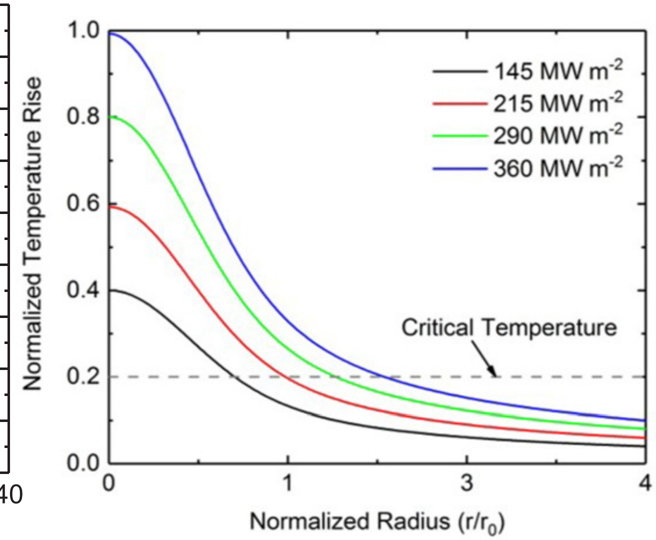
Using Laser-Induced Thermal Voxels to Pattern Diverse Materials at the Solid–Liquid Interface

Lauren D. Zarzar,[*,†] B. S. Swartzentruber,[‡] Brian F. Donovan,[§] Patrick E. Hopkins,[§] and Bryan Kaehr[*,⊥,#]

Thermal conductivity of substrate can limit direct write





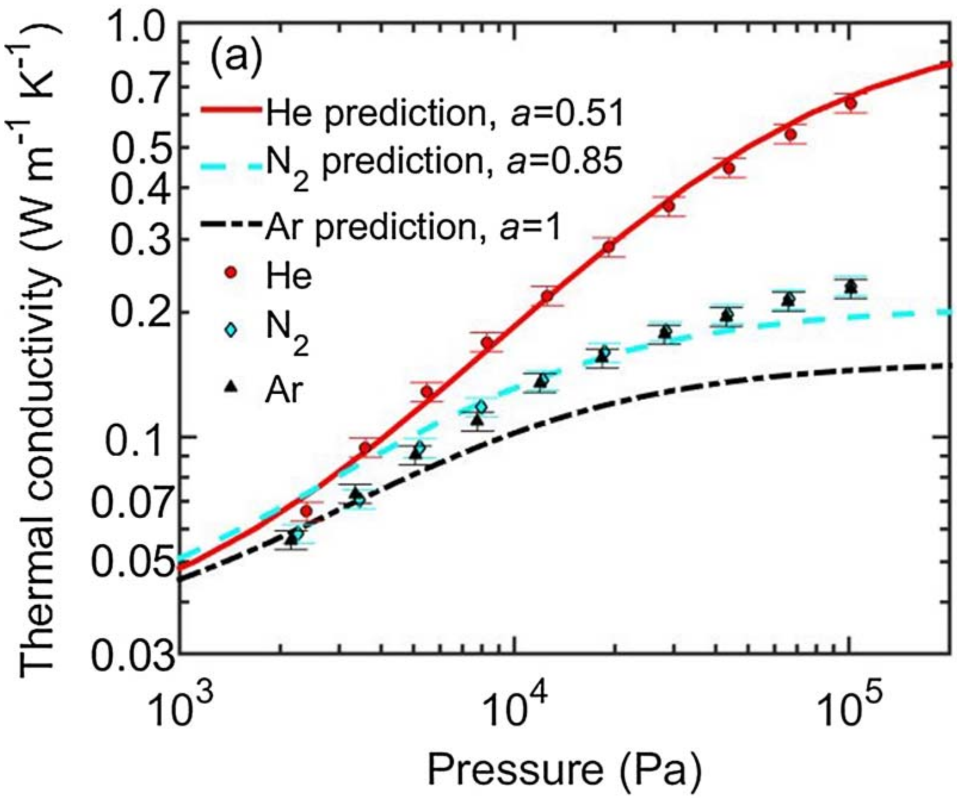


APPLIED PHYSICS LETTERS **112**, 051906 (2018)

Substrate thermal conductivity controls the ability to manufacture microstructures via laser-induced direct write

John A. Tomko,[1] David H. Olson,[2] Jeffrey L. Braun,[2] Andrew P. Kelliher,[1] Bryan Kaehr,[3] and Patrick E. Hopkins[1,2,4,a)]
[1]*Department of Materials Science and Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[2]*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*
[3]*Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA*
[4]*Department of Physics, University of Virginia, Charlottesville, Virginia 22904, USA*

Gas pressure increases thermal conductivity of AM powders



Additive Manufacturing 21 (2018) 201–208

Contents lists available at ScienceDirect

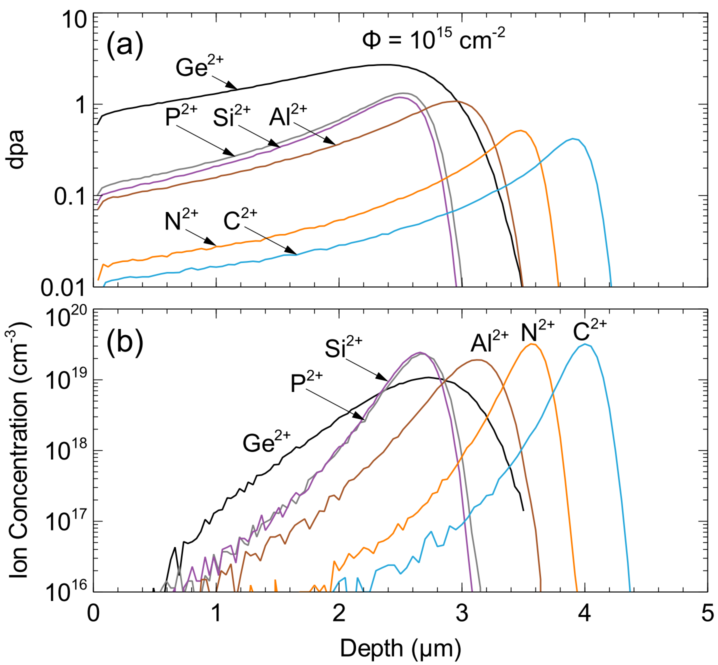
Additive Manufacturing

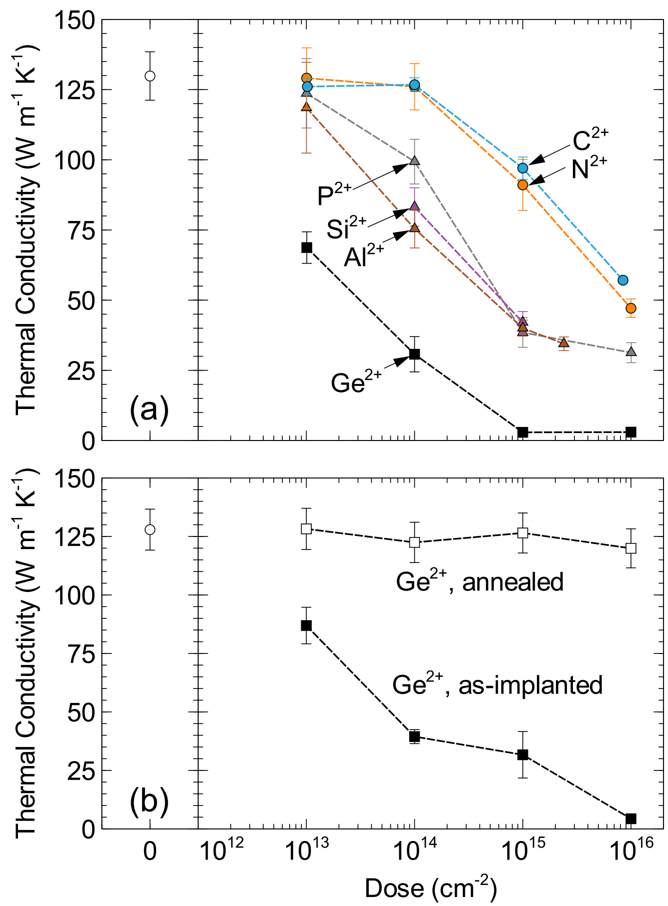
journal homepage: www.elsevier.com/locate/addma

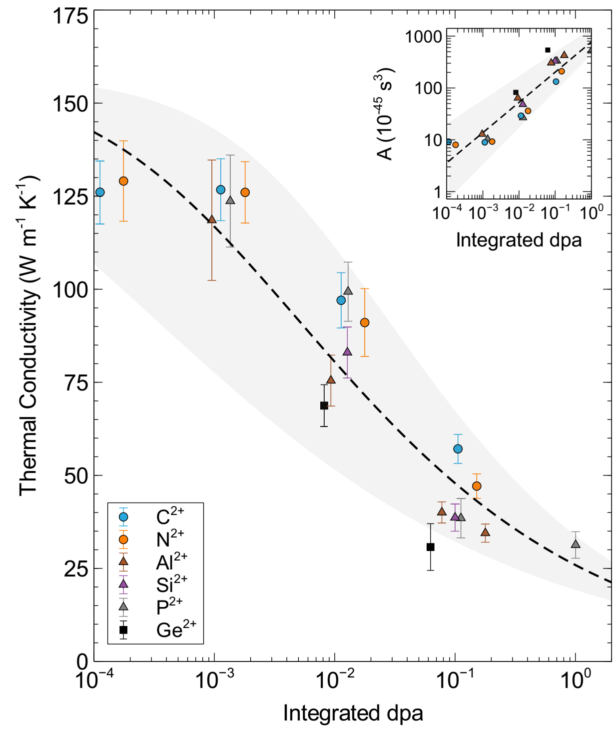
Thermal conductivity of metal powders for powder bed additive manufacturing

Lien Chin Wei[a], Lili E. Ehrlich[a], Matthew J. Powell-Palm[a], Colt Montgomery[a], Jack Beuth[a,b], Jonathan A. Malen[a,b,*]

Ion implantation decreased thermal conductivity of materials





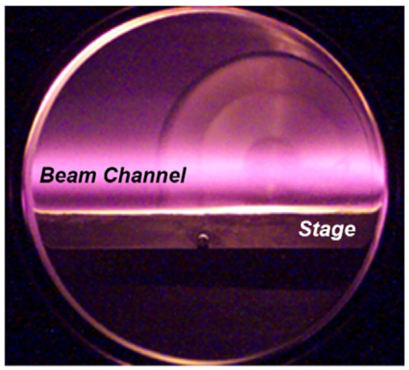


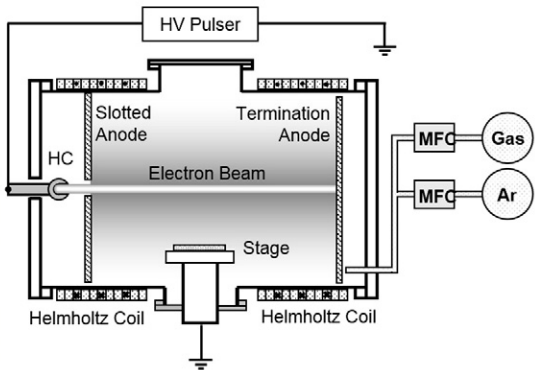
PHYSICAL REVIEW B **104**, 134306 (2021)

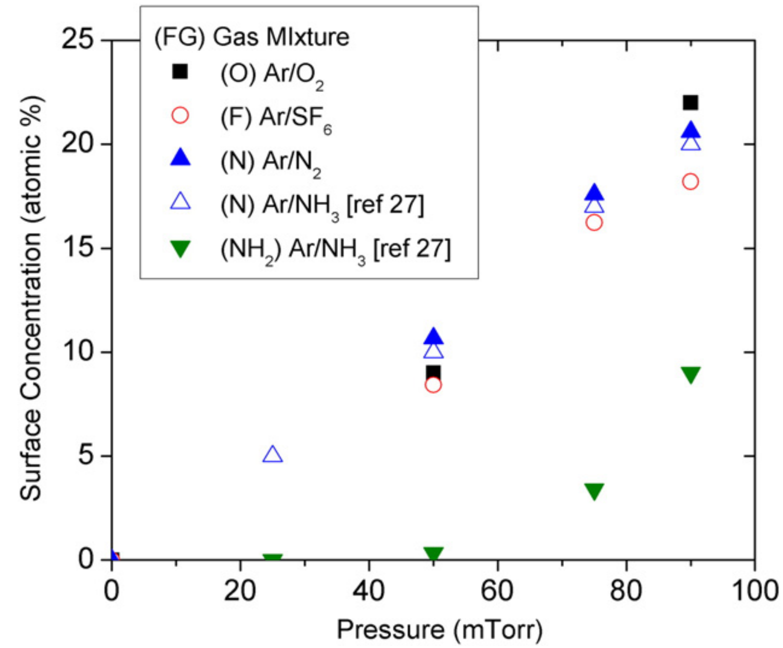
Reductions in the thermal conductivity of irradiated silicon governed by displacement damage

Ethan A. Scott [1,2] Khalid Hattar [2] Eric J. Lang [2] Kiumars Aryana [1] John T. Gaskins [1,3] and Patrick E. Hopkins [1,4,5,*]

Plasma processing impacts thermal resistance at interfaces







Surface & Coatings Technology 314 (2017) 148–154

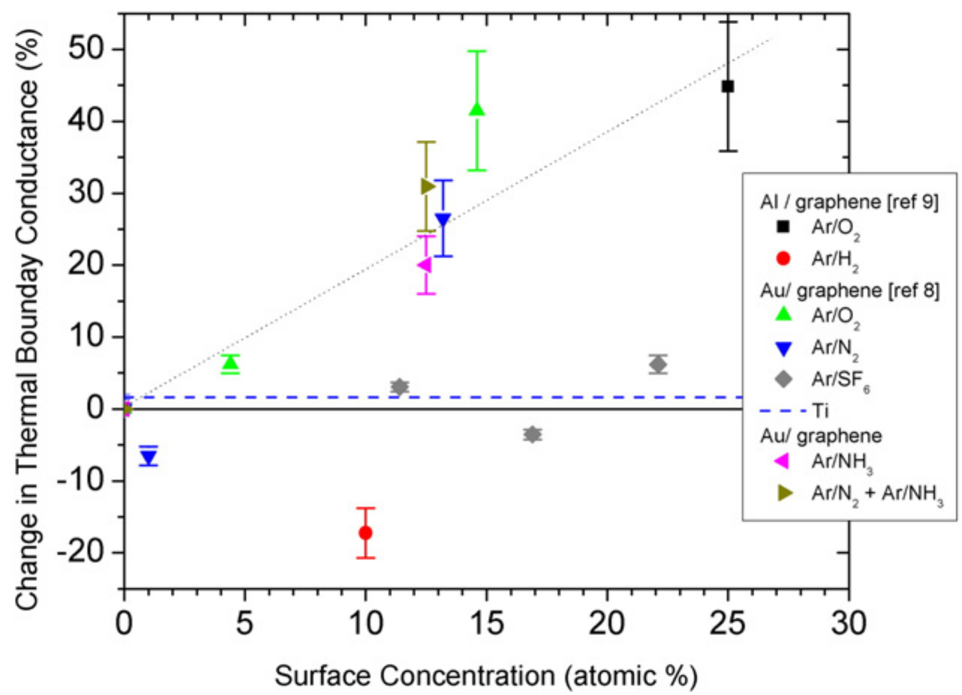
Contents lists available at ScienceDirect

Surface & Coatings Technology

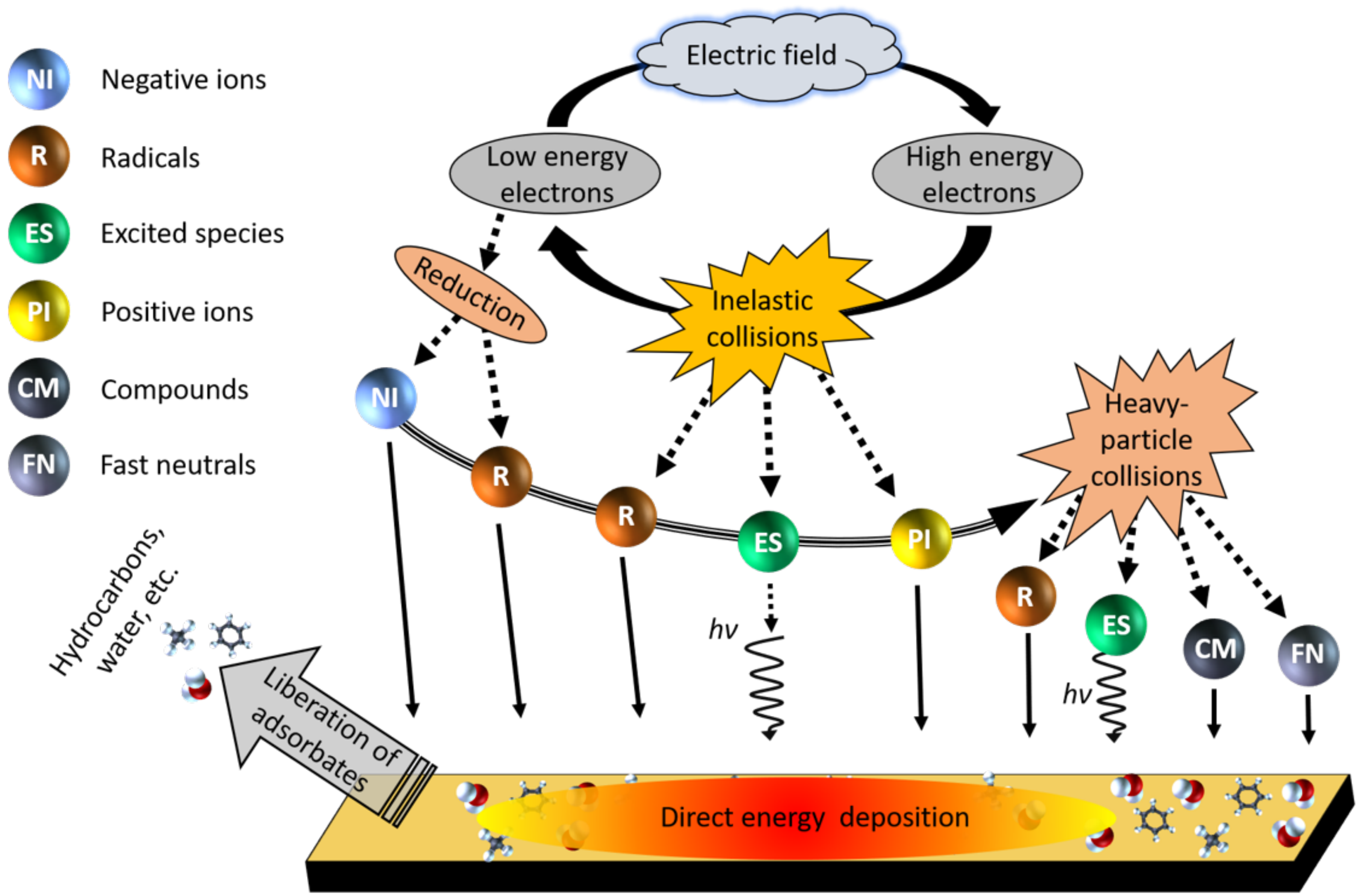
journal homepage: www.elsevier.com/locate/surfcoat

Plasma-based chemical functionalization of graphene to control the thermal transport at graphene-metal interfaces

S.G. Walton [a,*], B.M. Foley [b], S.C. Hernández [a], D.R. Boris [a], M. Baraket [a], J.C. Duda [b], J.T. Robinson [c], P.E. Hopkins [b]



Plasma surface interactions

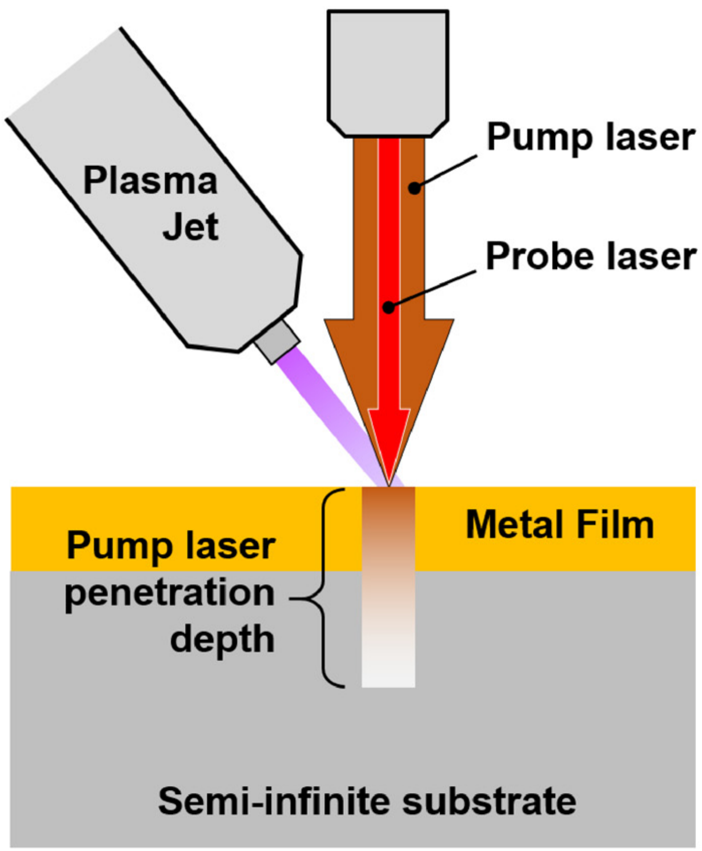


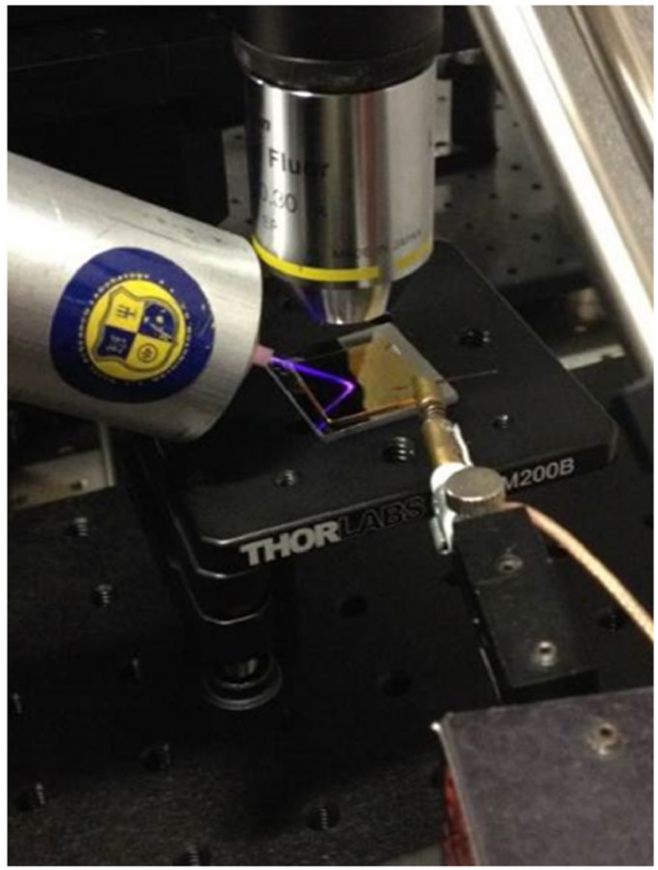
"Plasma-induced surface cooling," *Nature Comm.* **13**, 2623 (2022)

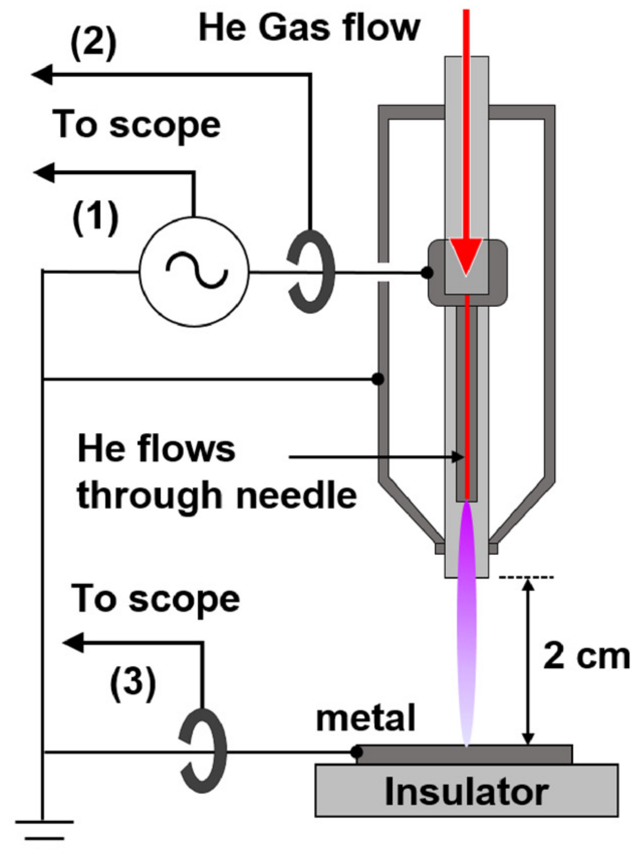
Thermoreflectance for plasma diagnostics

"Plasma-surface interactions in atmospheric pressure plasmas: *In situ* measurements of electron heating in materials"

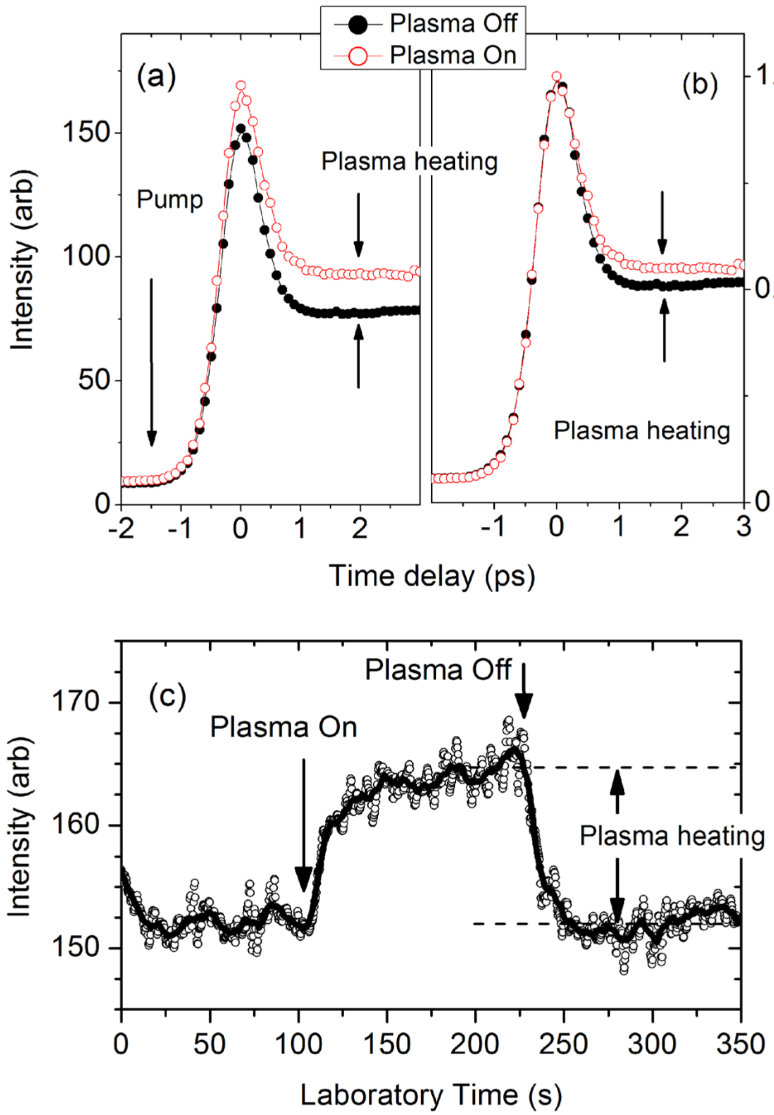
J. Appl. Phys. **124**, 043301 (**Editor's Pick**)

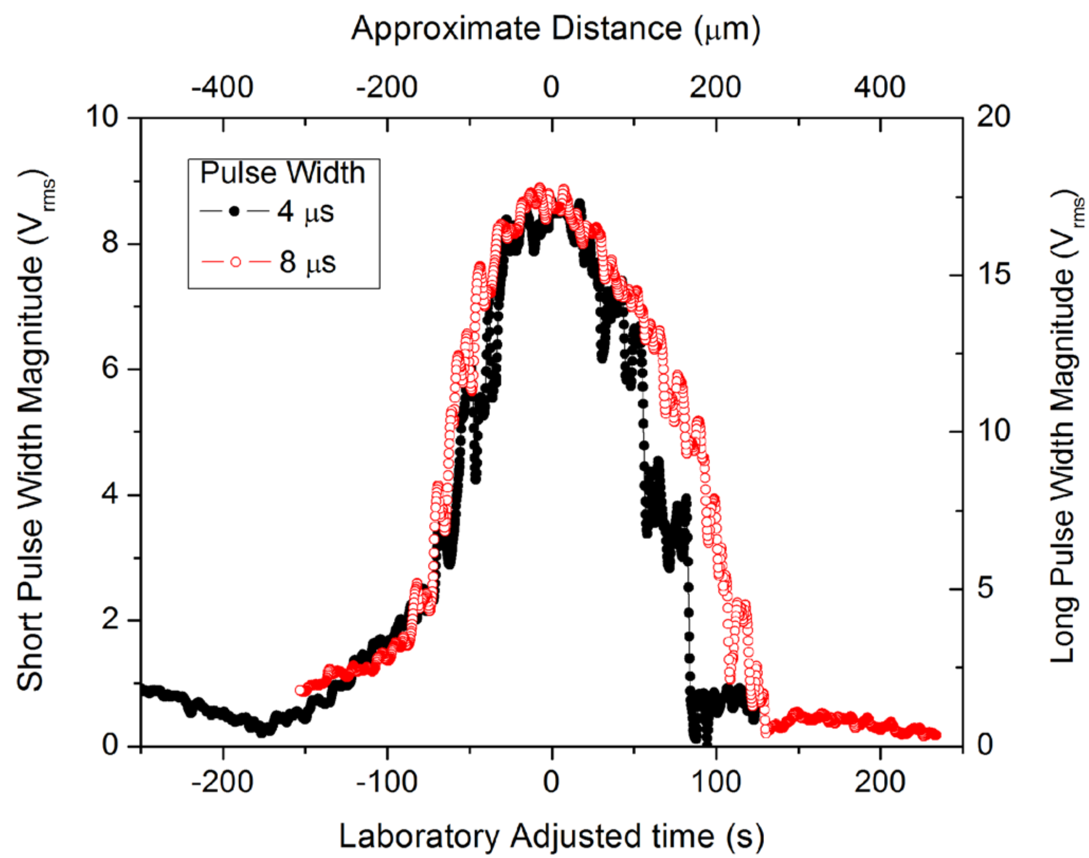






Thermoreflectance for plasma diagnostics

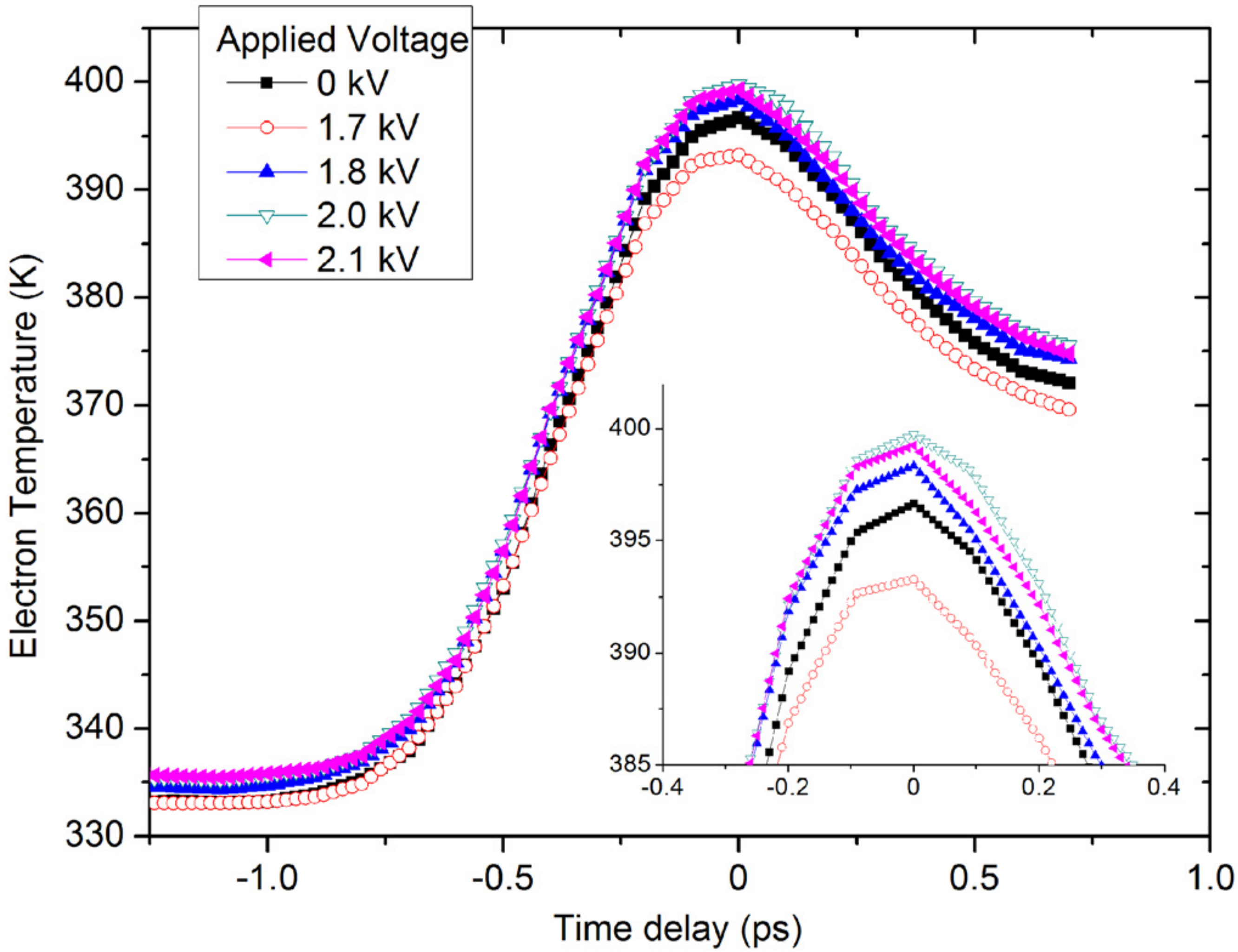




"Plasma-surface interactions in atmospheric pressure plasmas: *In situ* measurements of electron heating in materials"

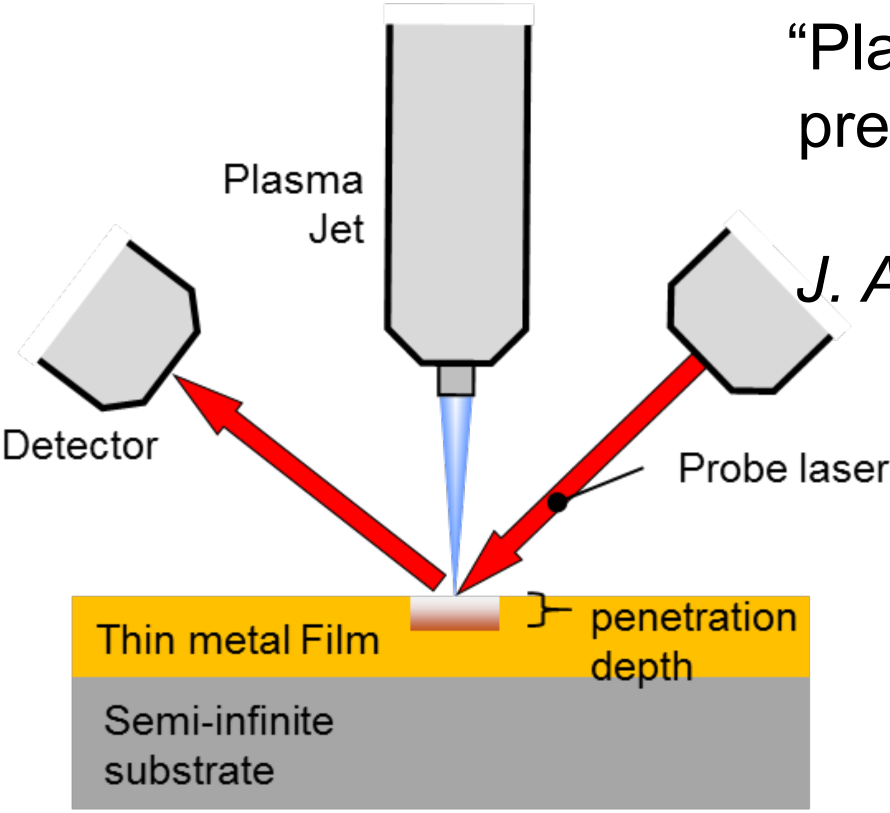
J. Appl. Phys. **124**, 043301 (**Editor's Pick**)

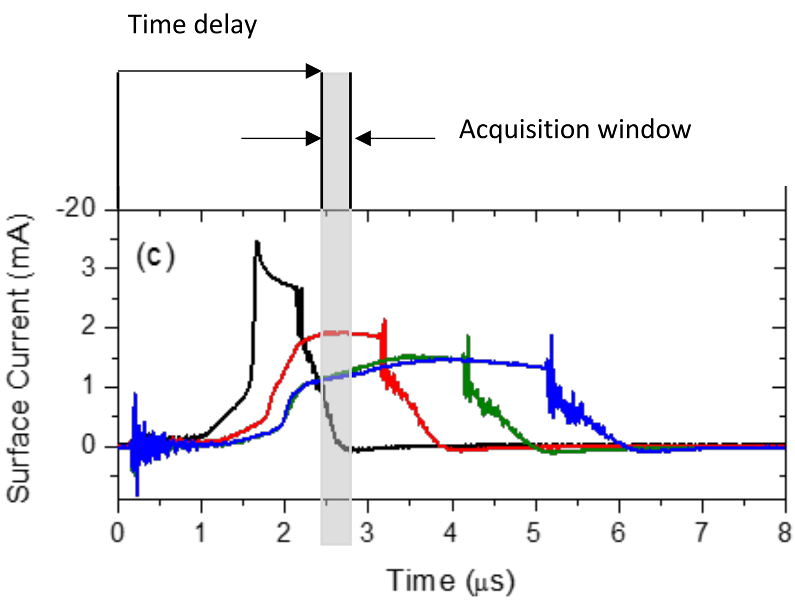
Thermoreflectance for plasma diagnostics



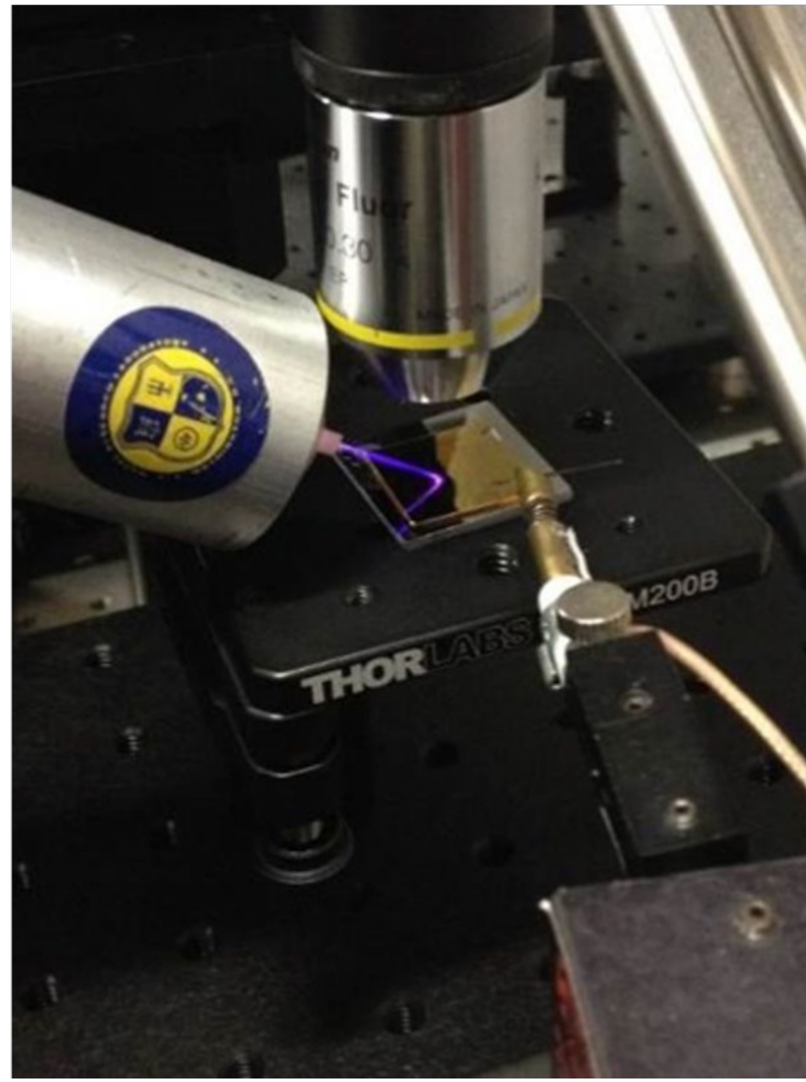
“Plasma-surface interactions in atmospheric pressure plasmas: *In situ* measurements of electron heating in materials”
J. Appl. Phys. **124**, 043301 (**Editor’s Pick**)

Temporally resolved Thermoreflectance for plasma diagnostics





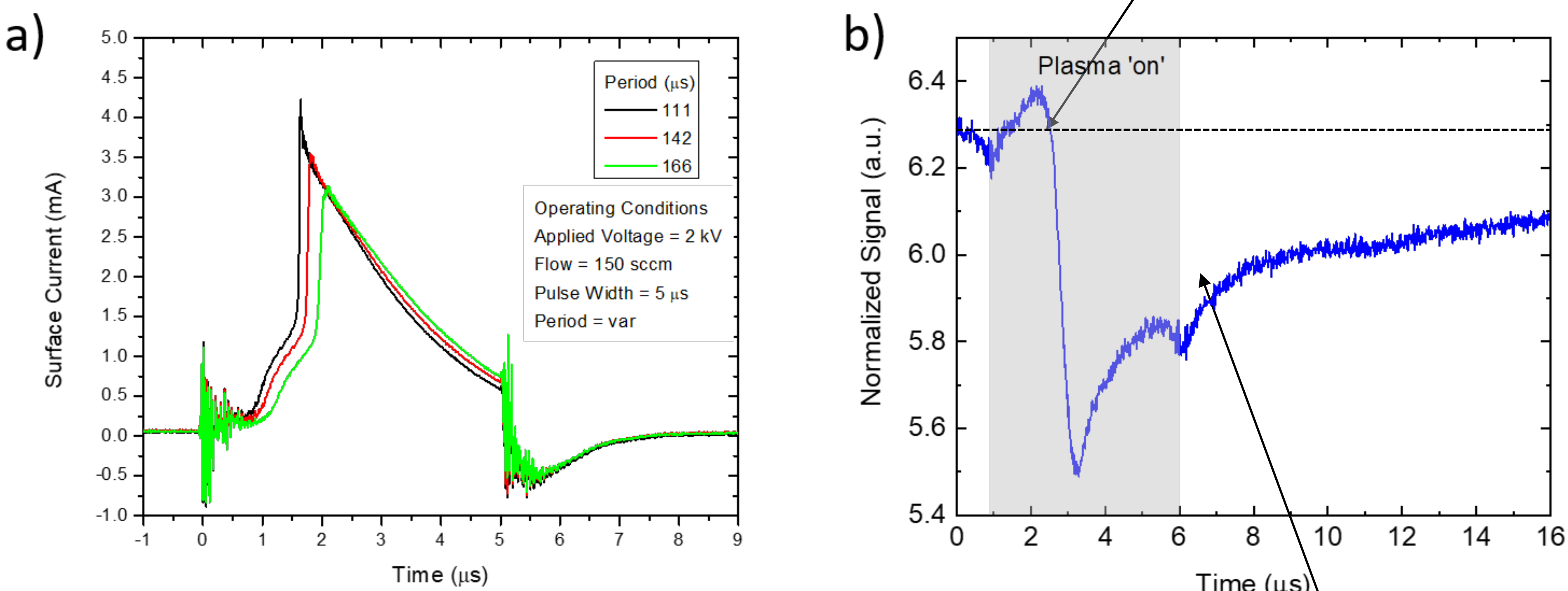
"Plasma-surface interactions in atmospheric pressure plasmas: *In situ* measurements of electron heating in materials"
J. Appl. Phys. **124**, 043301 (**Editor's Pick**)



Tracking surface temperature during plasma jet irradiation

Surface current temporal profile for different DC pulse widths

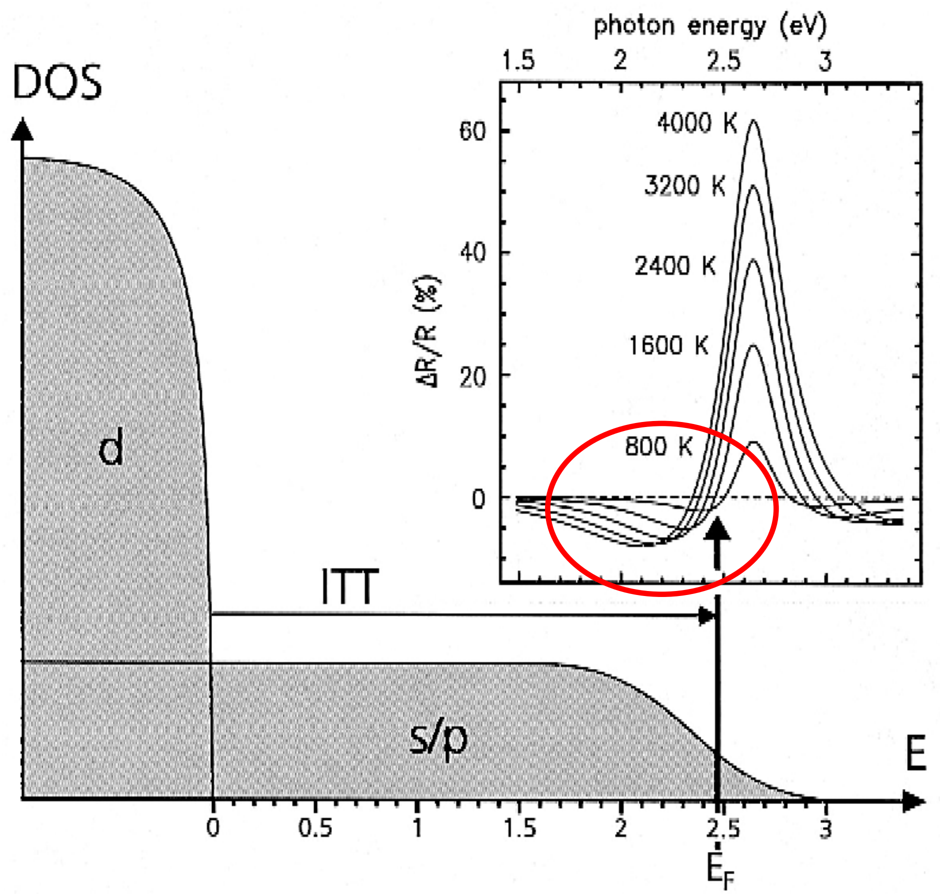
Note sign inflection….why?



"Accommodation" from neutrals?

"Plasma-induced surface cooling," *Nature Comm.* **13**, 2623 (2022)

Thermoreflectance of gold



ELSEVIER

Chemical Physics 251 (2000) 237–258

Chemical Physics

www.elsevier.nl/locate/chemphys

Electron and lattice dynamics following optical excitation of metals

J. Hohlfeld, S.-S. Wellershoff, J. Güdde, U. Conrad, V. Jähnke, E. Matthias

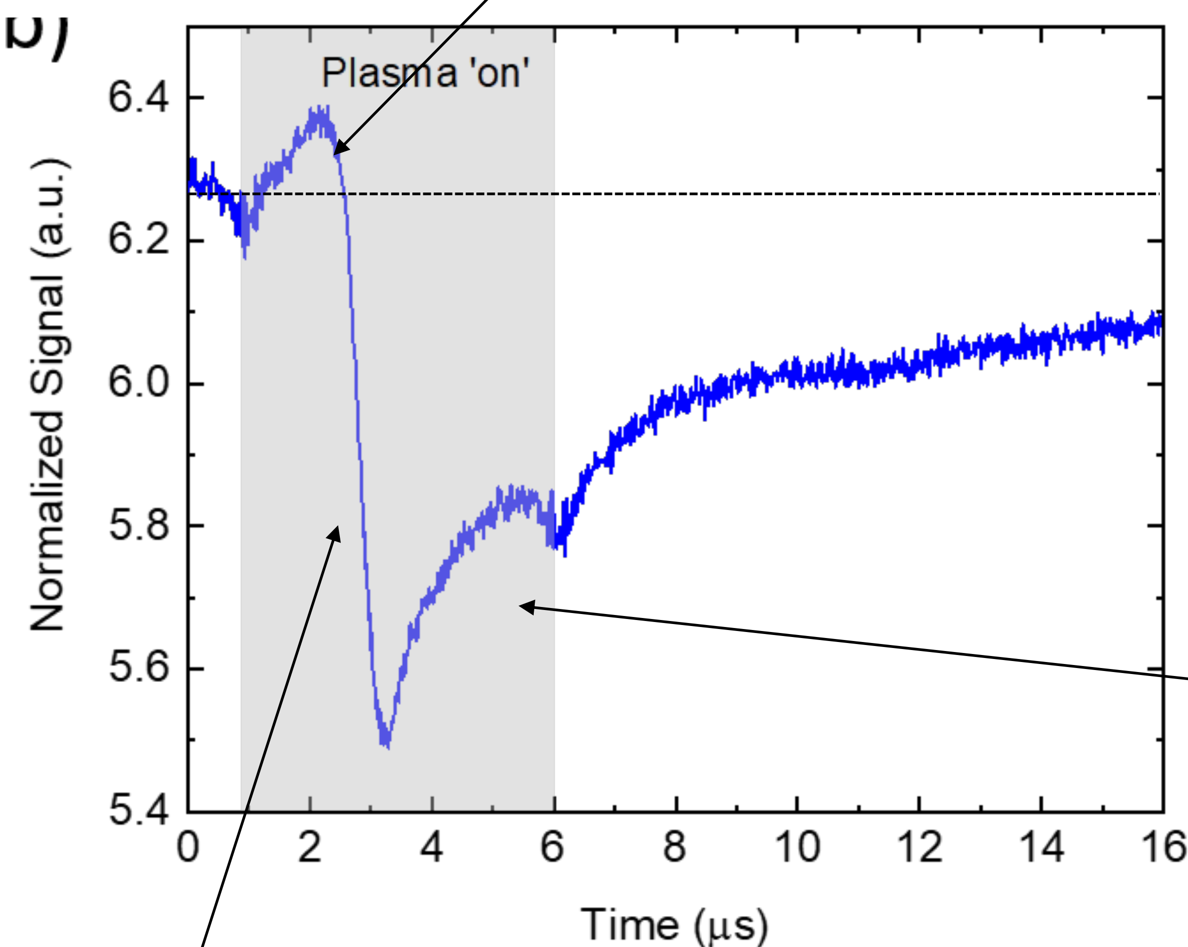
Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

Received 13 May 1999

- Probe energies below d-band to Fermi level transition (ITT)
  - Increase ΔT
  - Decrease $\Delta R/R$

Plasma cooling of surface

Plasma cooling: either mass removal or electron ejection from high eV photons (can not rule out either)



Cooling of surface from decrease in surface current or pulse turning off

-ΔR = increase in temperature from energy transfer to gold

"Plasma-induced surface cooling," *Nature Comm.* **13**, 2623 (2022)

What we covered today

1. What makes a high and low thermal conductivity material – an electron and phonon nanoscale perspective
2. Thermal conductivity of thin films: how film dimensional and growth conditions can lead to interfaces and defects that scatter electrons and phonons, thus reducing the thermal conductivity of materials
3. Thermal conductivity measurements: thin film methods
4. Thermal boundary resistance: coherent and incoherent heat transfer across interfaces in nanostructures
5. Coupled nonequilibrium heat transfer: Energy coupling among electron, phonons and photons including ultrafast laser pulse effects
6. Heat transfer in materials during synthesis and manufacturing, including plasma-material interactions during deposition and laser-based manufacturing

Society of Vacuum Coaters Education Program in conjunction with the 66th Annual Technical Conference

Tutorial Course: M-230

Nanoscale Heat Transfer in Thin Films and Interfaces

Instructor: Patrick E. Hopkins
Chief Science Officer, Co-Founder, *Laser Thermal, Inc*.
patrick@laserthermal.com
https://laserthermal.com/
Whitney Stone Professor of Engineering, *University of Virginia*
phopkins@virginia.edu
https://patrickehopkins.com/

Monday, May 8, 2023

SVC tutorial offerings and descriptions along with details of the SVC Webinar and On-Site Course programs can be found on the SVC Web Site: www.svc.org