



SCHOOL of ENGINEERING & APPLIED SCIENCE
UNIVERSITY of VIRGINIA

Thin film testing

DARPA MATRIX Workshop

12/10/2015



Patrick E. Hopkins

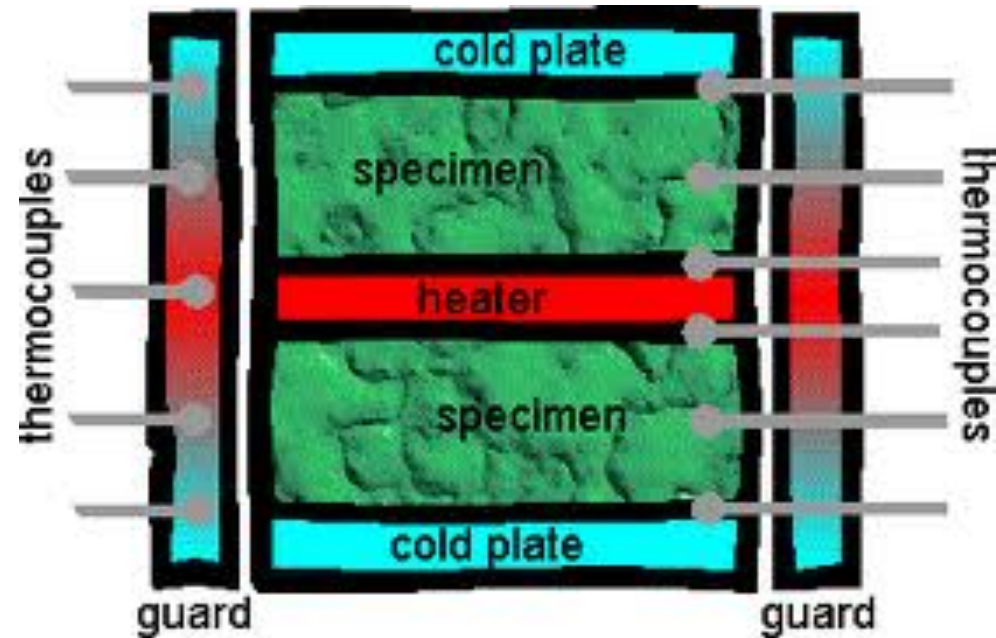
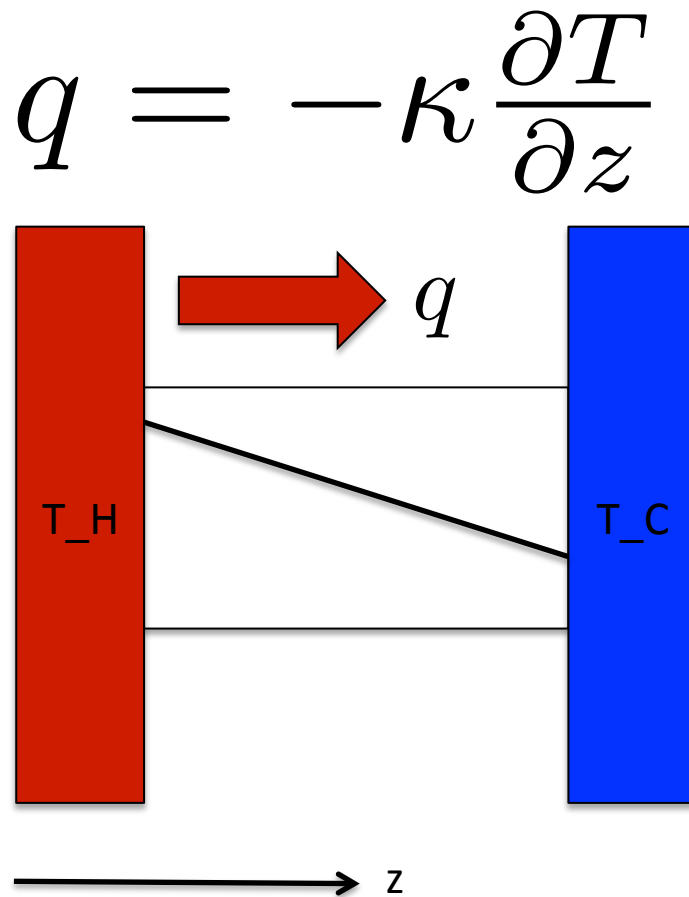
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Overview: Methods for testing κ of thin films

- **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**
 - **TDTR**
 - **FDTR**
 - **Limitations and potentials....**

Steady state measurements of κ in bulk systems

“Guarded hot plate”



Steady state measurements of κ in nanosystems

Utilize ability to “nano pattern” electrical contacts and circuits

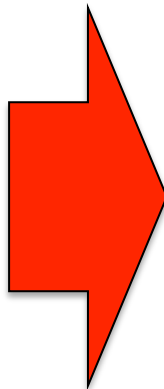
Fourier Law

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

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

Joule heating

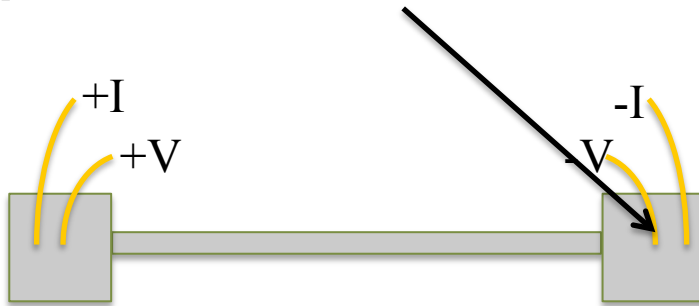
$$q = I^2 R$$


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

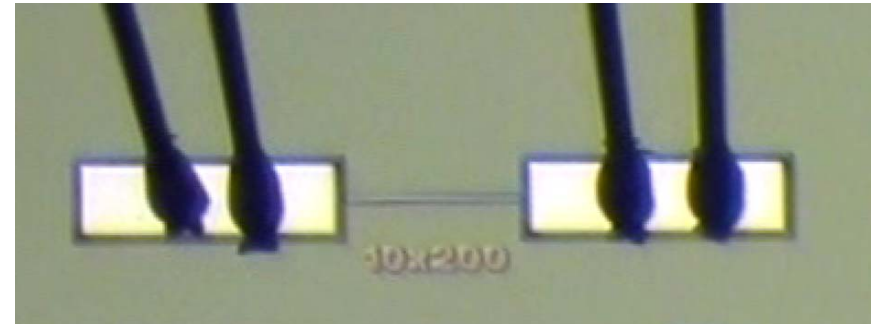
Measured resistance is related to thermal conductivity (and a lot of other known quantities)

Steady state measurements of κ in nanosystems

Electrical/thermal contact resistances are inherently present in measurements



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



How do you make these contacts in a nanosystem??

Lithography/metal processing

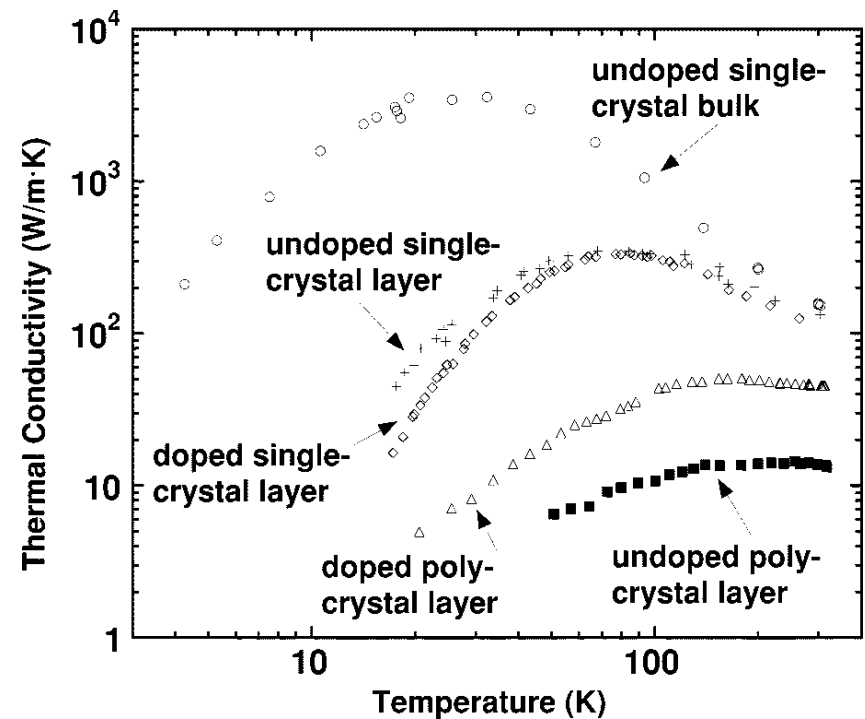
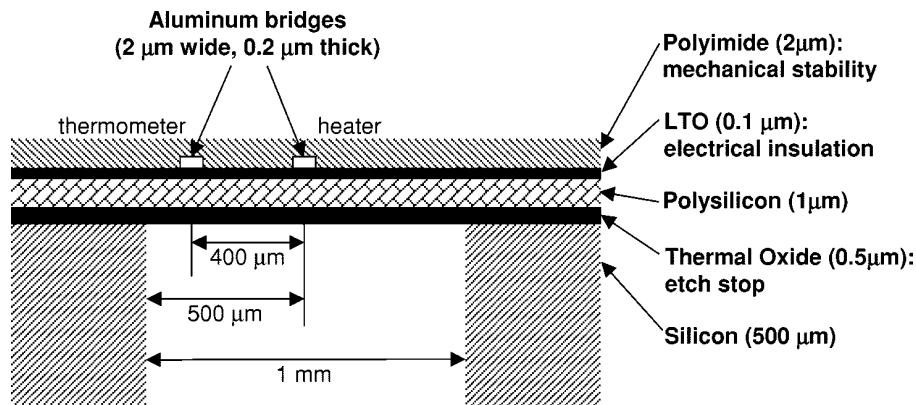
P. E. Hopkins and L. M. Phinney. Thermal conductivity measurements on polycrystalline silicon micro-bridges using the 3w technique. *Journal of Heat Transfer*, 131:043201, 2009.

Steady state measurements of κ in nanosystems

In-plane thermal conductivity measurements of Si thin films

Thermal Conductivity of Doped Polysilicon Layers

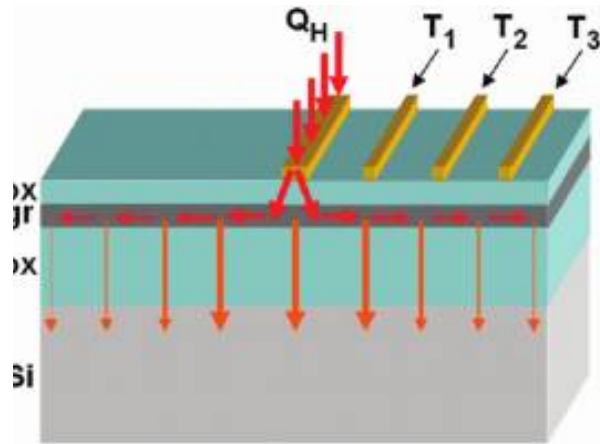
Angela D. McConnell, Srinivasan Uma, *Member, IEEE*, and Kenneth E. Goodson, *Associate Member, IEEE*



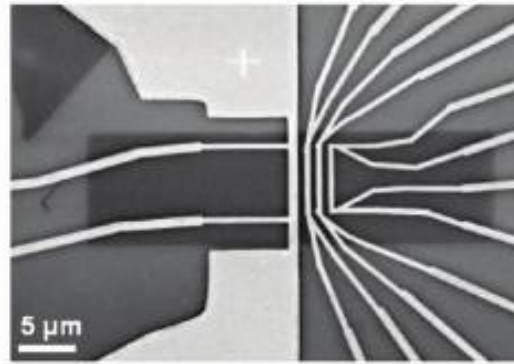
A. D. McConnell, S. Uma, and K. E. Goodson. Thermal conductivity of doped polysilicon layers. **Journal of Microelectromechanical Systems**, 10:360–369, 2001.

Steady state measurements of κ in nanosystems

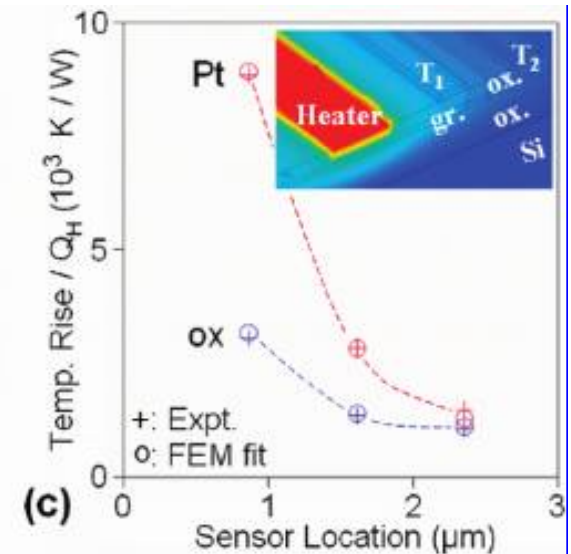
In-plane thermal conductivity of “atomically thin” film (i.e., graphene)



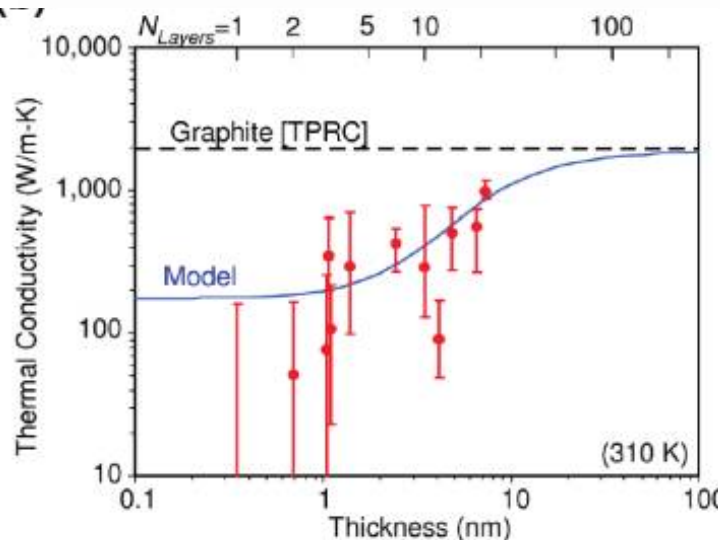
(a)



(b)



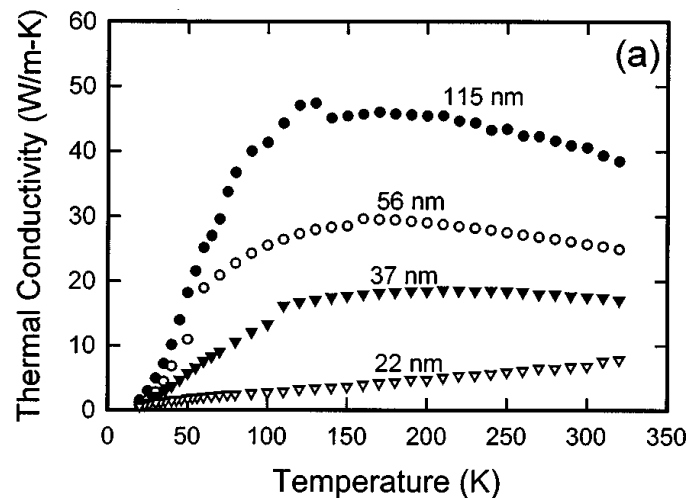
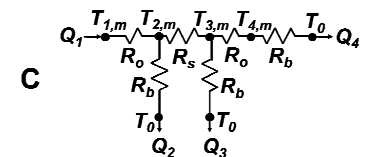
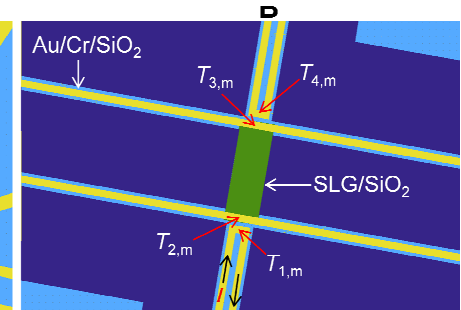
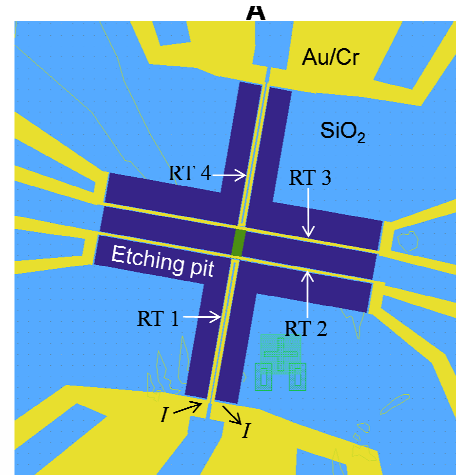
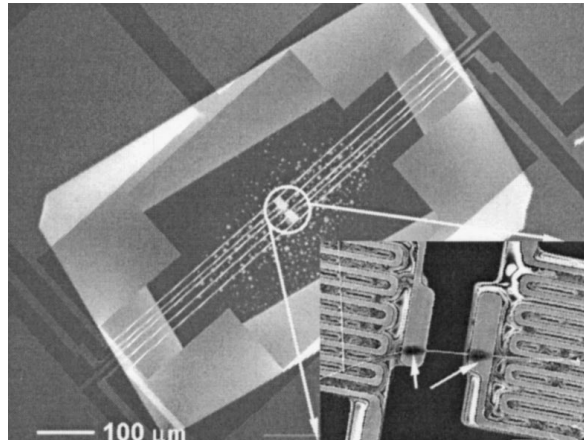
(c)



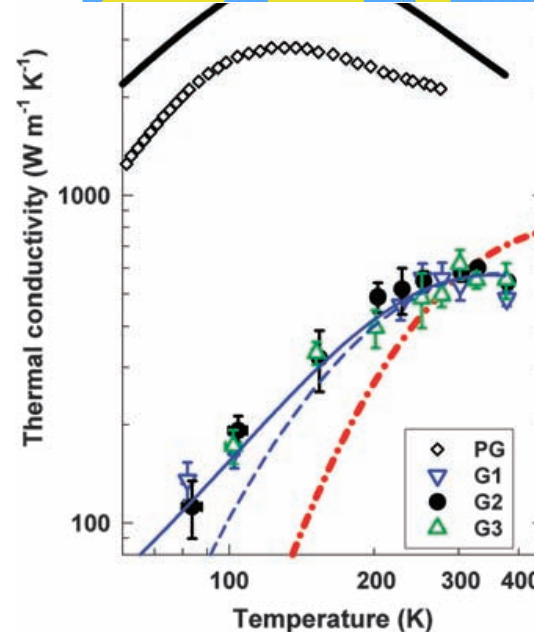
W. Jang, Z. Chen, W. Bao, C. N. Lau, and C. Dames.
Thickness-dependent thermal conductivity of encased
graphene and ultrathin graphene. **Nano Letters**,
10:3909–3913, 2010.

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.

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 - TDTR
 - FDTR
 - Limitations and potentials....

Steady state vs. transient measurements of κ

Steady state = The Fourier Law

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

Heat capacity
enters the
picture

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

Transient = The Heat Equation

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

The source term
can make a
difference

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

Transient measurements of κ

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

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

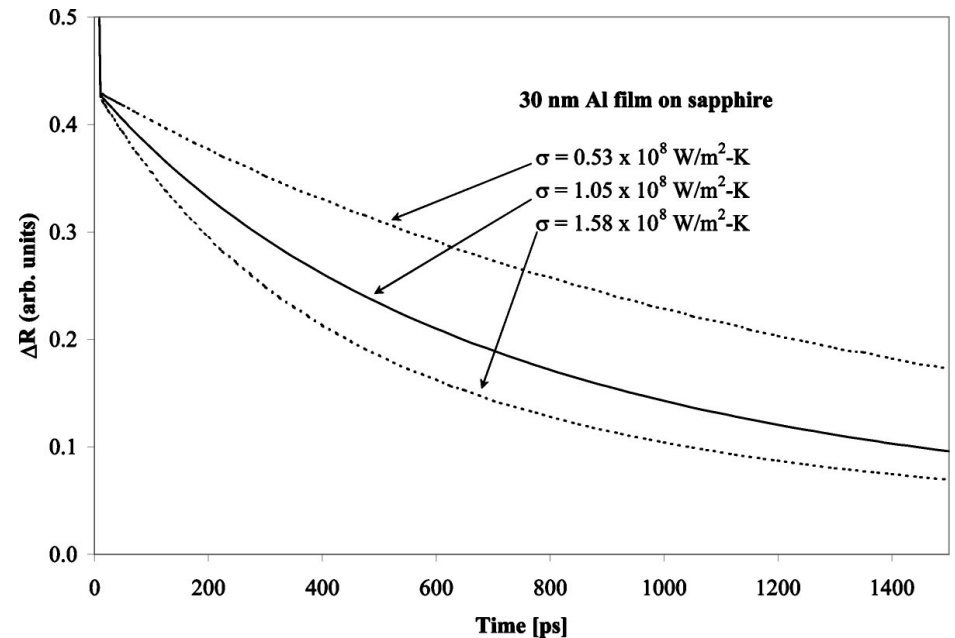


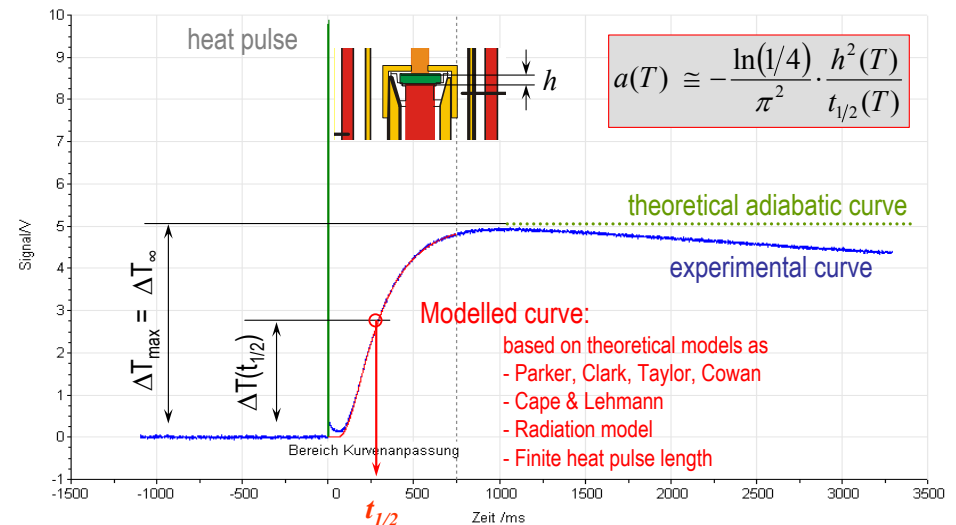
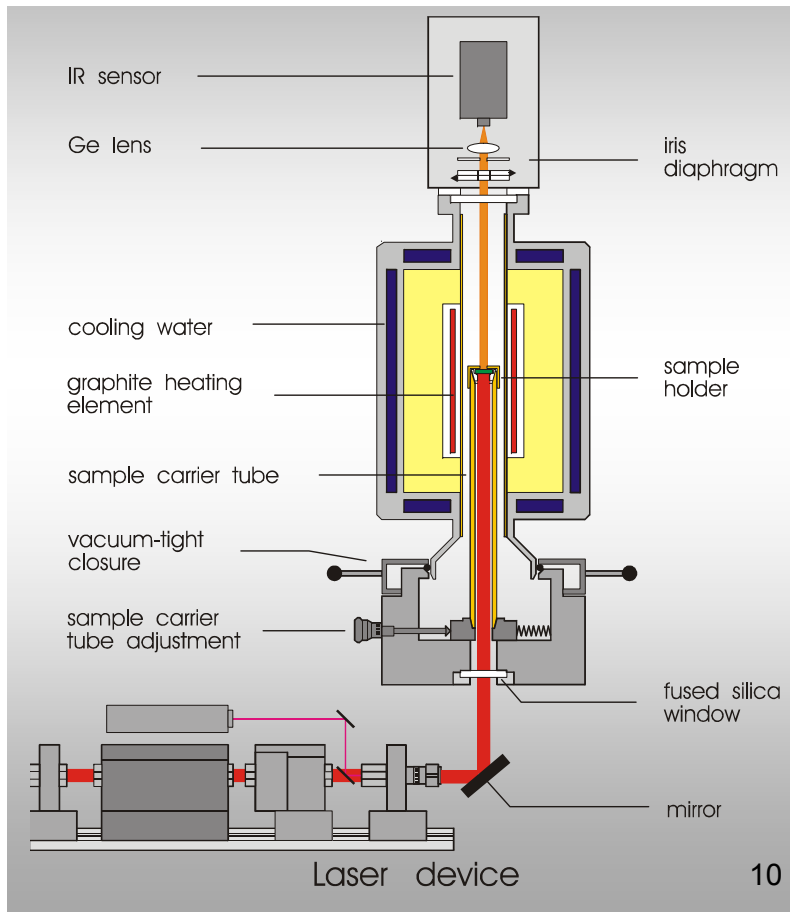
Fig. 2 Modeled thermal response of 30 nm Al film on a sapphire substrate with $\sigma = 1.05 \times 10^8 \text{ W/m}^2 \text{ K}$. The dotted lines are the thermal response for the same film with $\pm 50\%$ change in σ .

How are these transient decays measured?

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.

Transient measurements of κ in bulk systems

Laser Flash



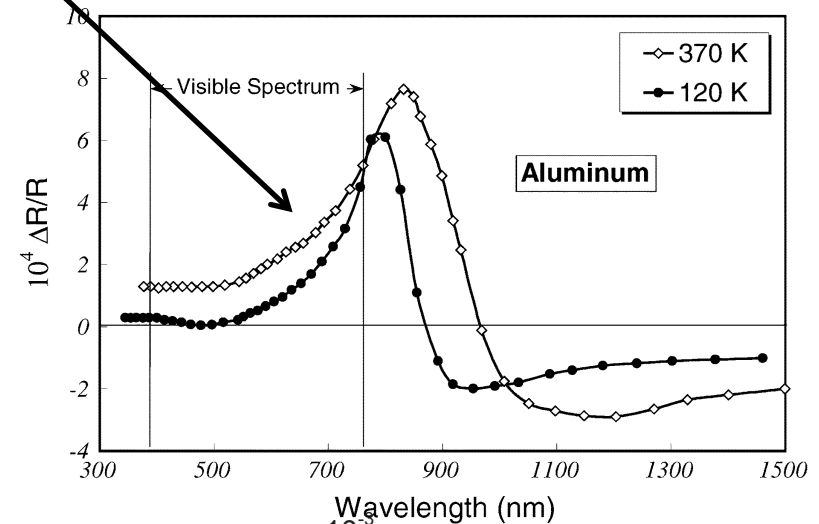
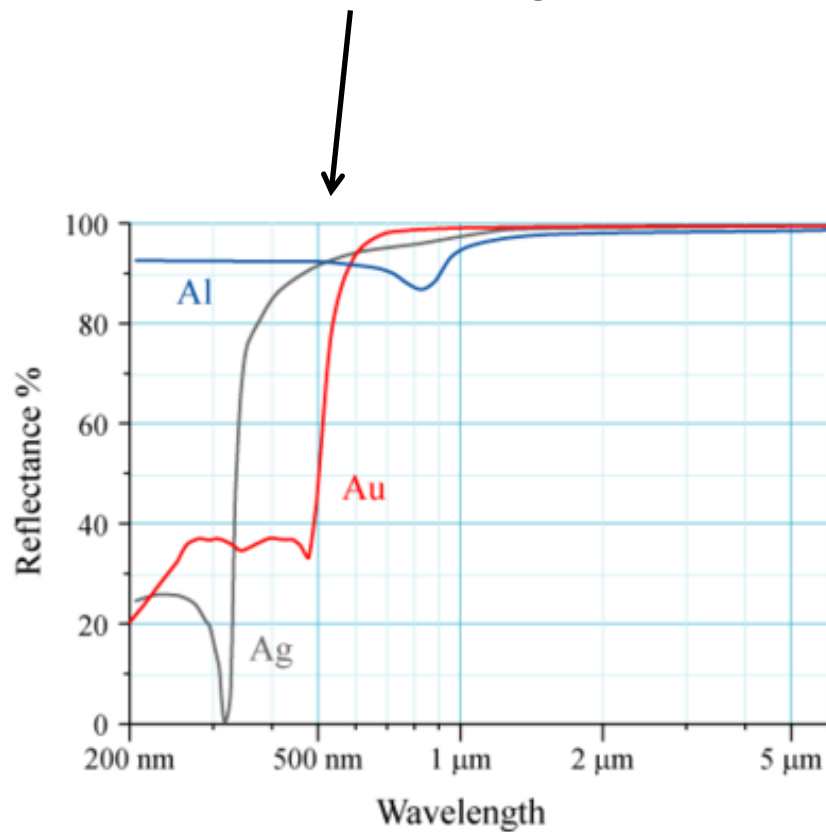
- Very dependent on surface emissivity
- Terrible sensitivity in nanosystems

Transient measurements of κ in nanosystems

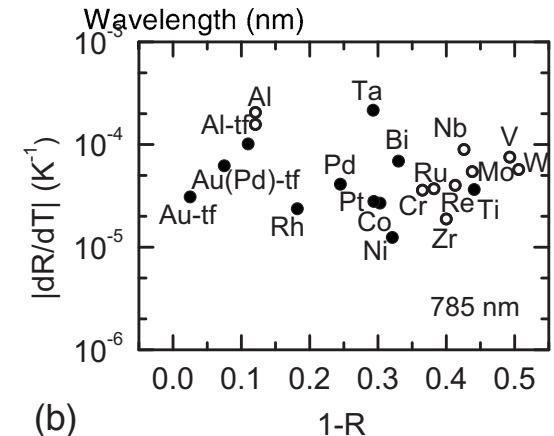
Turn to optics....Thermoreflectivity

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

Reflectivity vs. Thermoreflectivity



Y. Wang, J. Y. Park, Y. K. Koh, and D. G. Cahill. Thermoreflectance of metal transducers for time-domain thermoreflectance. **Journal of Applied Physics**, 108:043507, 2010.

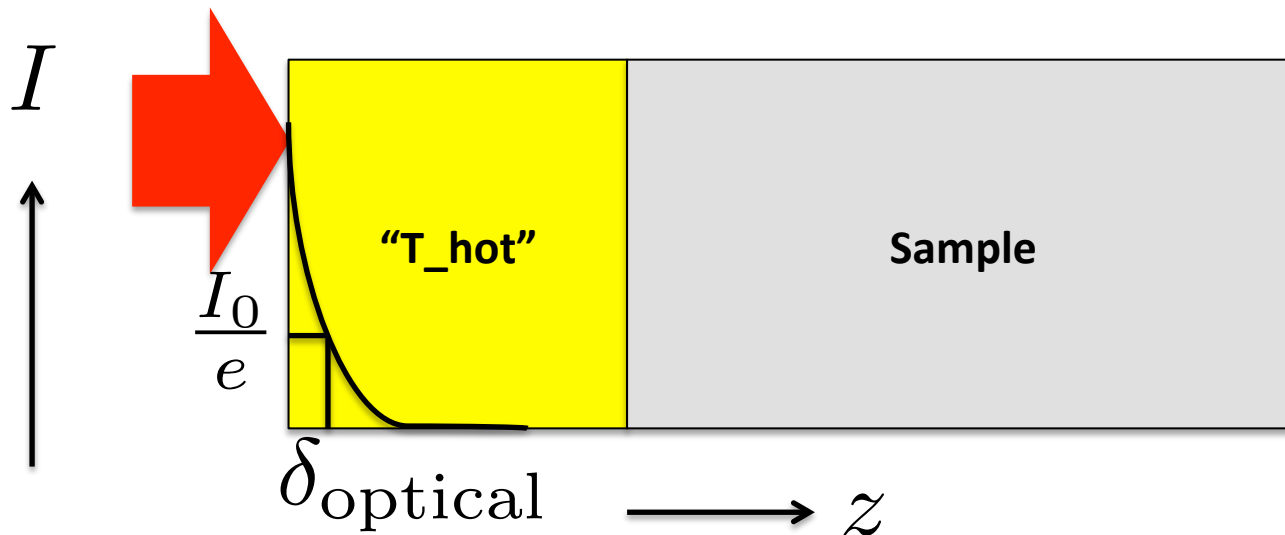


(b)

What's the advantage? Optical absorption of metals

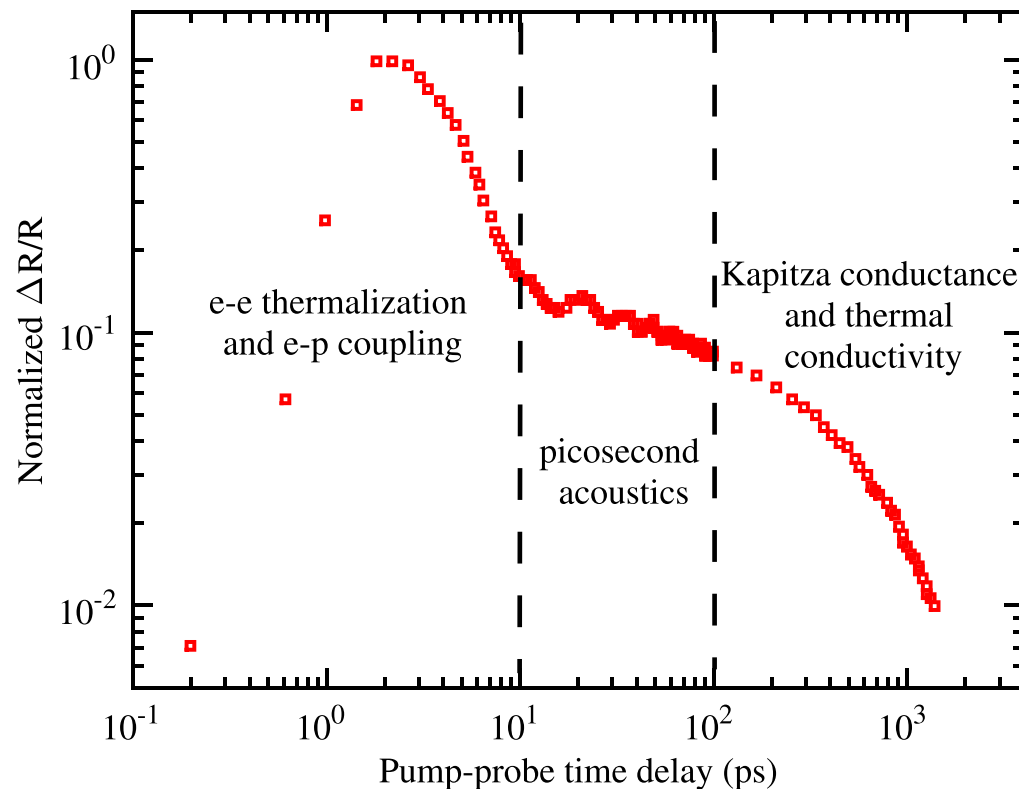
- Use a metal film as a opto-thermal transducer
- Spatial resolution limited by optical penetration depth of metal
- Absorbed light in metal converted to heat so metal is “thermometer”

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



Let's also include temporal resolution

- Transient Thermoreflectance (TTR)
- Sub-picosecond laser pulses
- **Rule of thumb: ~ 100 ps for heat to propagate through a 100 nm Al film**



R. Cheaito, C. S. Gorham, A. Misra, K. Hattar, and P. E. Hopkins. Thermal conductivity measurements via time-domain thermoreflectance for the characterization of radiation induced damage. **Journal of Materials Research**, **30:1403–1412**, 2015.

A. Giri, J. T. Gaskins, B. F. Donovan, C. Szejewski, R. J. Warzoha, M. A. Rodriguez, J. Ihlefeld, and P. E. Hopkins. Mechanisms of nonequilibrium electron-phonon coupling and thermal conductance at interfaces. **Journal of Applied Physics**, **117(10):105105**, 2015.

Transient measurements of κ in nanosystems

Temporal decay of single laser pulse can be used to measure κ of thin films

Pump-probe: nanosecond pump

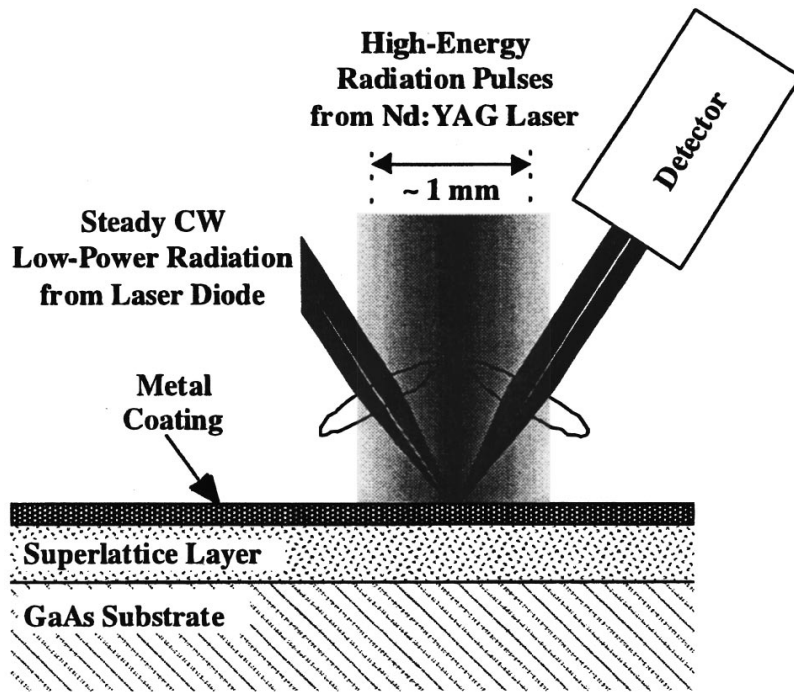


FIG. 1. The thermoreflectance method for measuring the vertical thermal resistance of $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ superlattice layers.

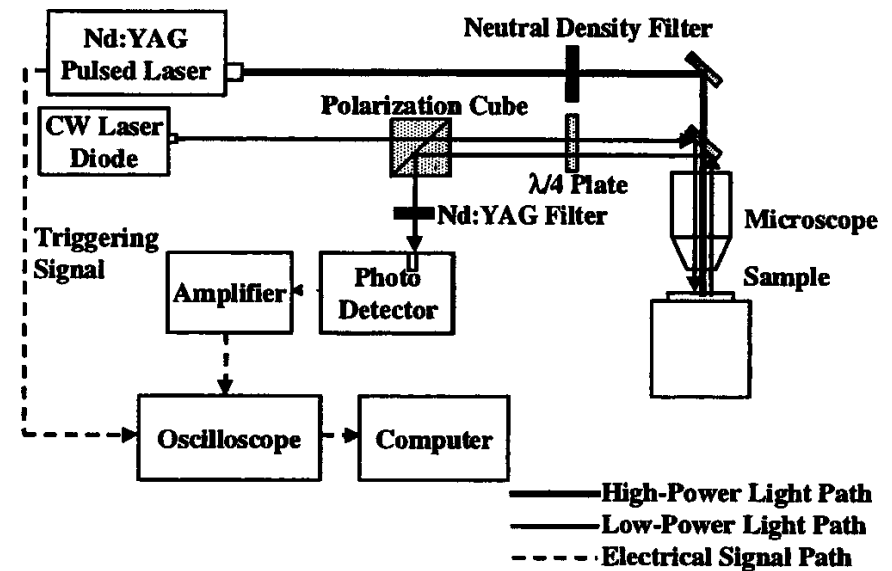


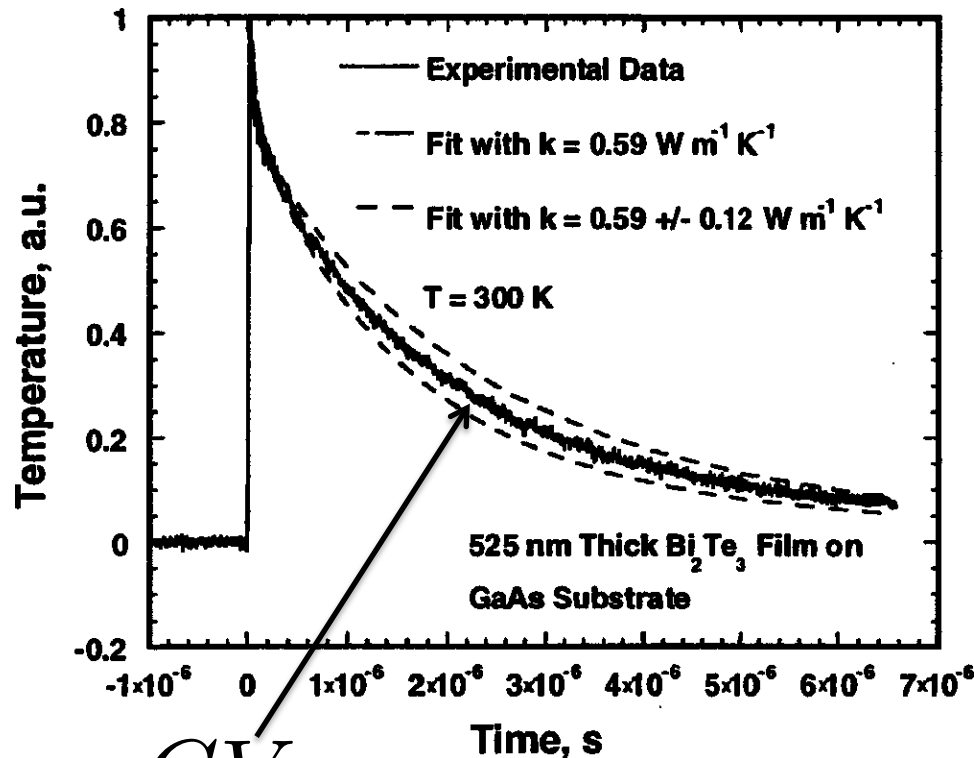
FIG. 2. Diagram describing the paths for radiation and electrical signals in the experimental setup.

M. N. Touzelbaev, P. Zhou, R. Venkatasubramanian, and K. E. Goodson. Thermal characterization of $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ superlattices. *Journal of Applied Physics*, 90:763–767, 2001.

Transient measurements of κ in nanosystems

Temporal decay of single laser pulse can be used to measure κ of thin films

Pump-probe: nanosecond pump



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

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

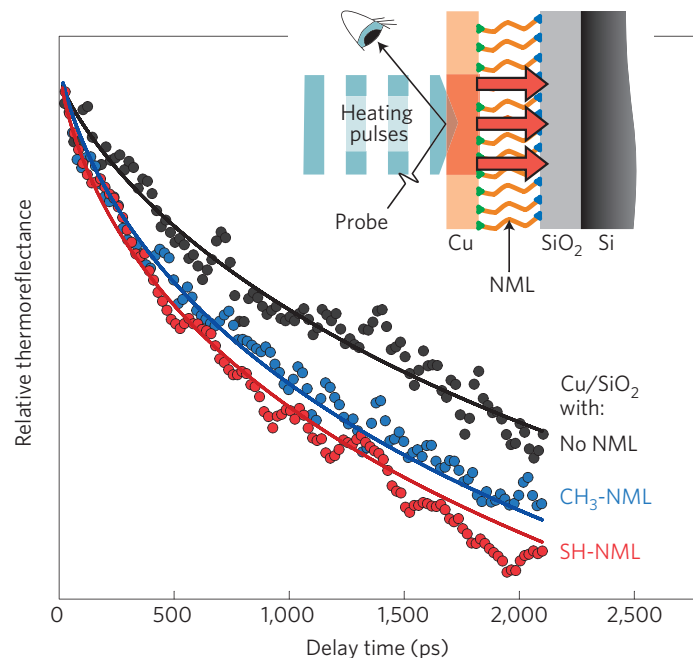
$$\delta = \sqrt{\frac{\tau \kappa}{C}}$$

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

M. N. Touzelbaev, P. Zhou, R. Venkatasubramanian, and K. E. Goodson. Thermal characterization of $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ superlattices. *Journal of Applied Physics*, 90:763–767, 2001.

Temporal decay of single laser pulse can be used to measure κ of thin films

- What “thermal resistor” dominates temporal decay?
- Turn to modulated heating....



P. J. O'Brien, S. Shenogin, J. Liu, P. K. Chow, D. Laurencin, P. H. Mutin, M. Yamaguchi, P. Keblinski, and G. Ramanath. Bonding-induced thermal conductance enhancement at inorganic heterointerfaces using nanomolecular monolayers. **Nature Materials**, 12:118–122, 2013.

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 - FDTR
 - TDTR

Steady state vs. transient vs. periodic measurements of κ

Steady state = The Fourier Law

Transient = The Heat Equation

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

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

Heat capacity
enters the
picture

The source term
can make a
difference

$q(t)$ vs. $q(t, \omega)$

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**

What separates “**frequency domain**”
measurements from everything else????

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

Thermal penetration depth

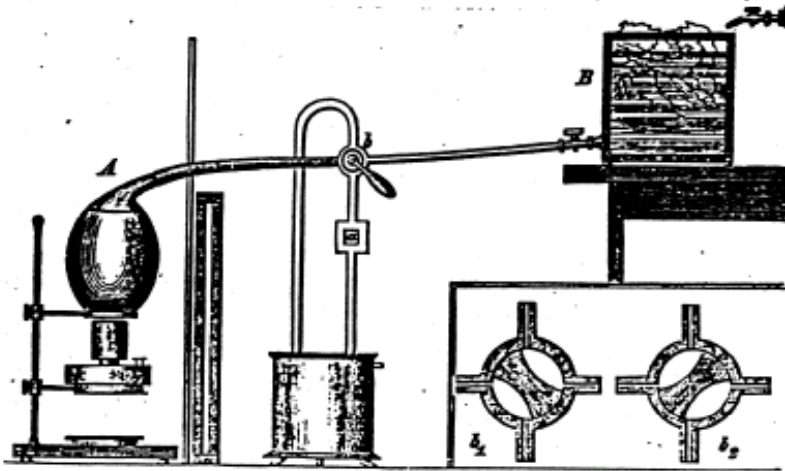
Ångström method

Used fixed temperature
boundary conditions

$$T(x=0) = 0^\circ\text{C} \quad 0 < t < \Gamma/2$$

$$T(x=0) = 100^\circ\text{C} \quad \Gamma/2 < t < \Gamma$$

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 Ångström method

JOURNAL OF APPLIED PHYSICS

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15 FEBRUARY 2004

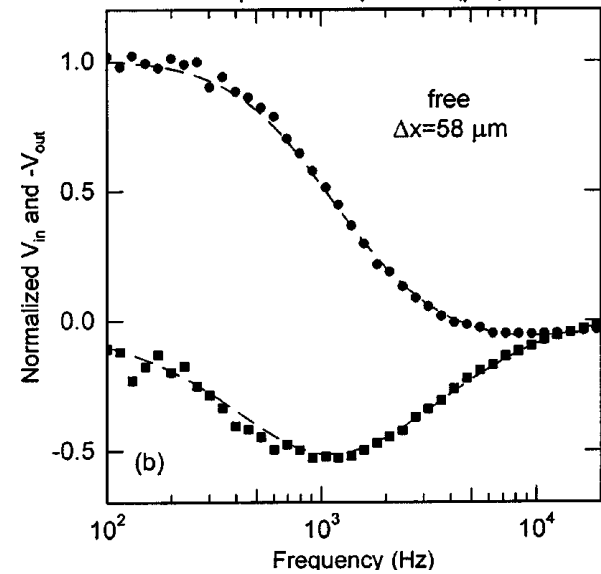
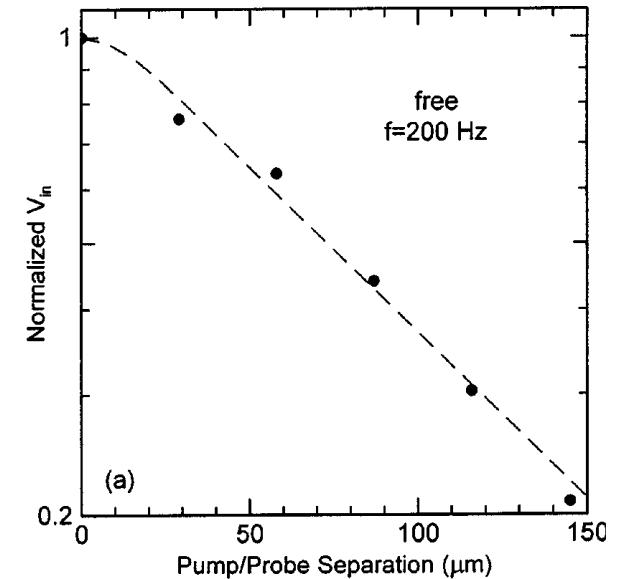
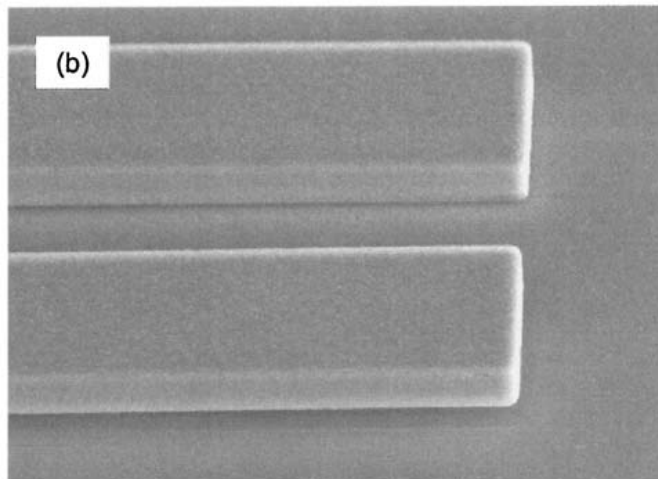
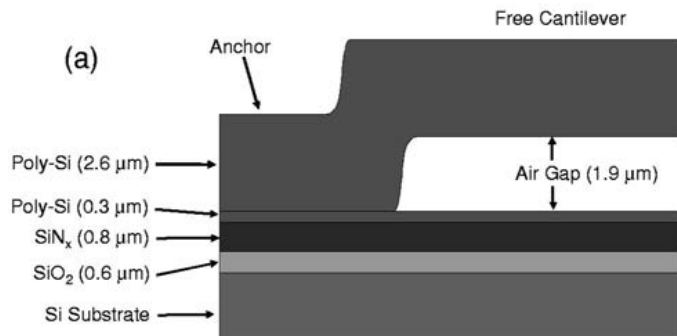
Thermal contact conductance of adhered microcantilevers

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

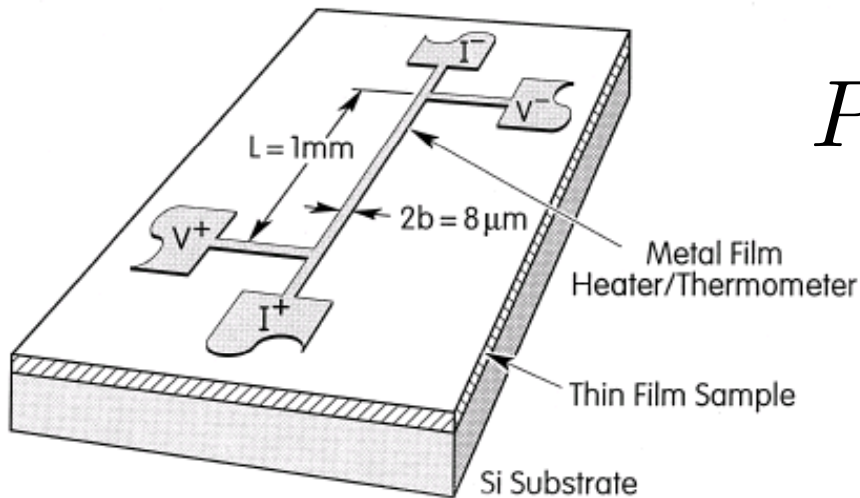


Frequency-domain measurements of κ in thin films

3ω technique

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

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



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

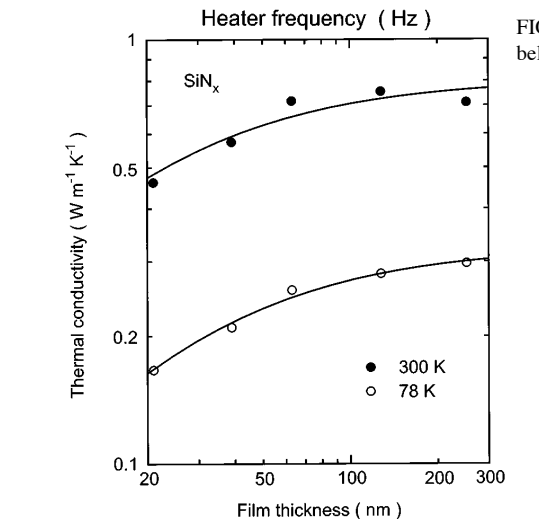
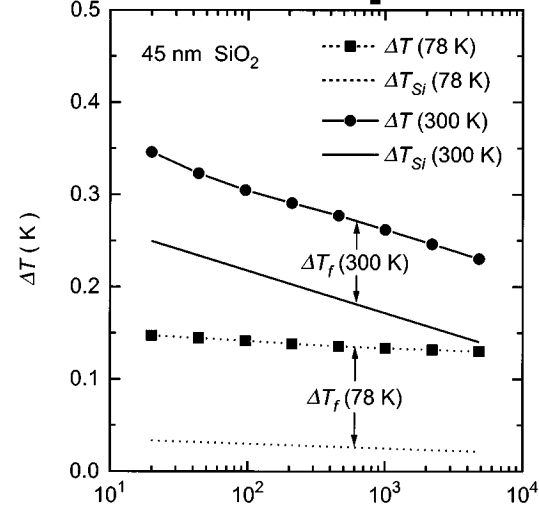
$$\Delta R \propto \exp[i2\omega t]$$

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

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

Frequency-domain measurements of κ in thin films

3 ω technique – been used extensively for thin films



S. M. Lee and D. G. Cahill. Heat transport in thin dielectric films. **Journal of Applied Physics**, 81(6):2590– 2595, 1997.

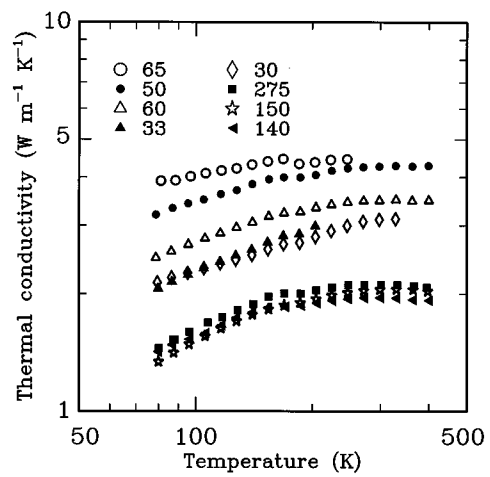
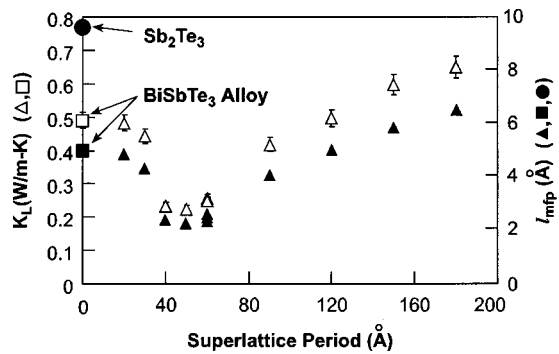
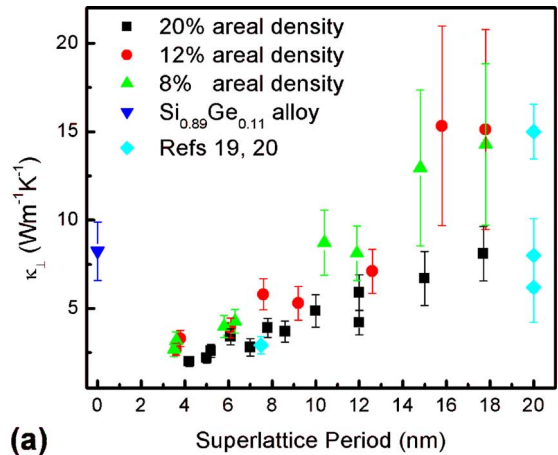
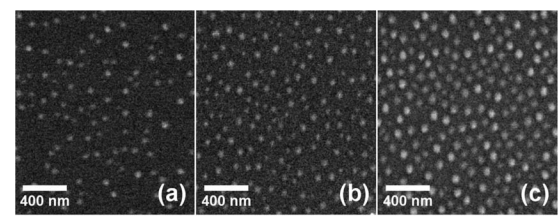


FIG. 2. Thermal conductivity of Si-Ge superlattices. Each symbol is labelled by the superlattice period L measured in Å.



R. Venkatasubramanian. Lattice thermal conductivity reduction and phonon localizationlike behavior in superlattice structures. **Physical Review B**, 61:3091–3097, 2000.

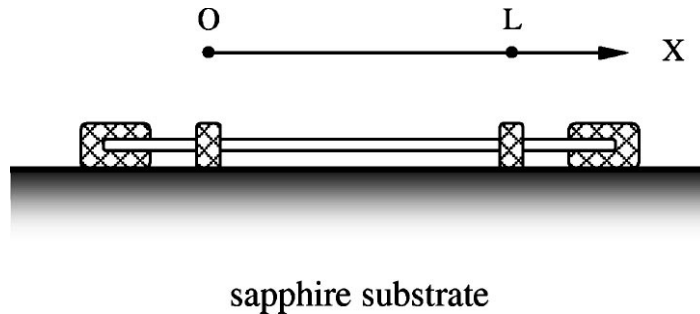
S. M. Lee, D. G. Cahill, and R. Venkatasubramanian. Thermal conductivity of Si-Ge superlattices. **Applied Physics Letters**, 70(22): 2957–2959, 1997.



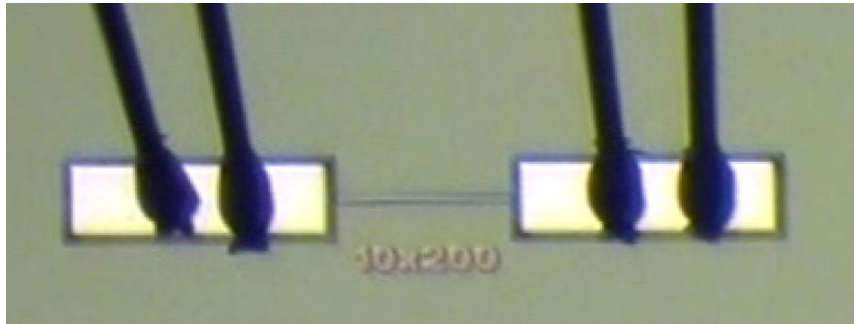
M. L. Lee and R. Venkatasubramanian. Effect of nanodot areal density and period on thermal conductivity in SiGe/Si nanodot superlattices. **Applied Physics Letters**, 92:053112, 2008.

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.

Recent review of electrical resistivity-based thermal conductivity measurement techniques for nanosystems

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 electrothermal methods. **Annual Review of Heat Transfer**, 16:7–49, 2013.

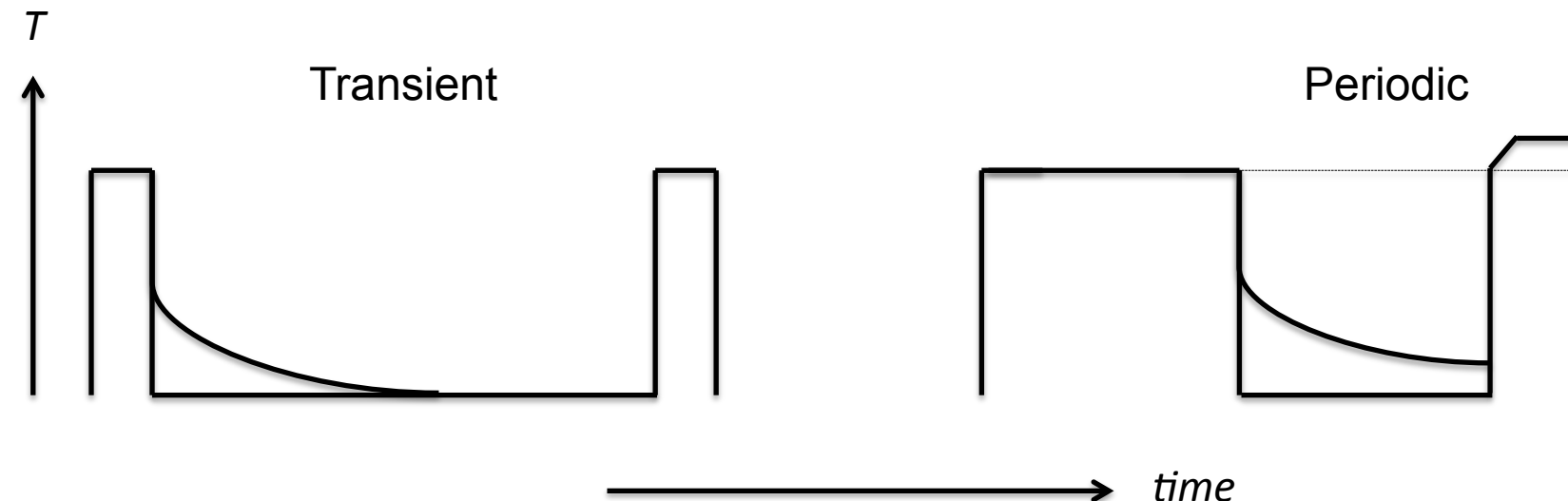
What about optical periodic heating techniques?

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

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!!!!

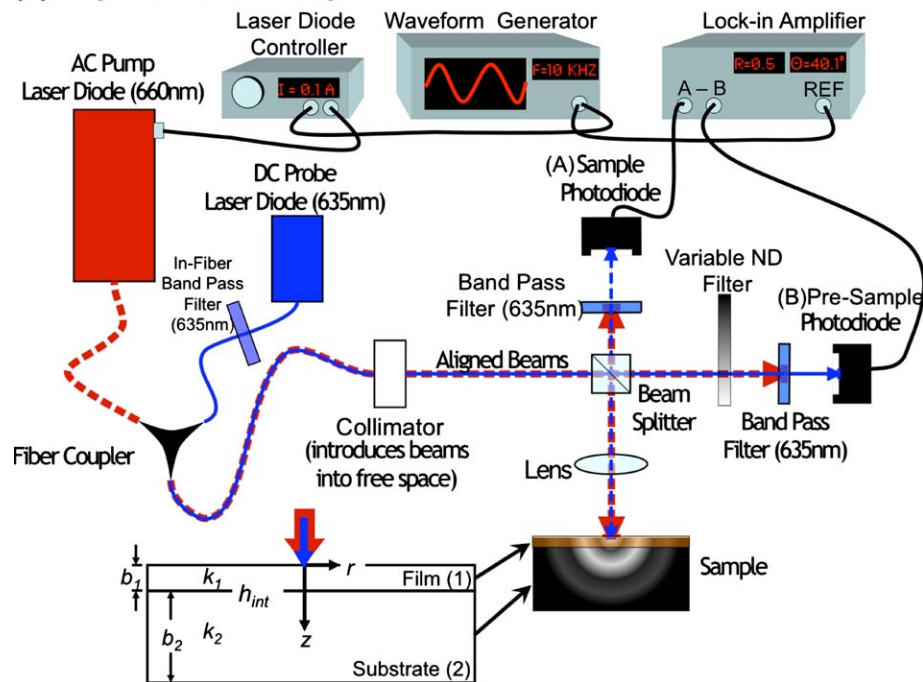
NEED HEATING EVENT TO BE “FELT” BY NEXT HEATING EVENT



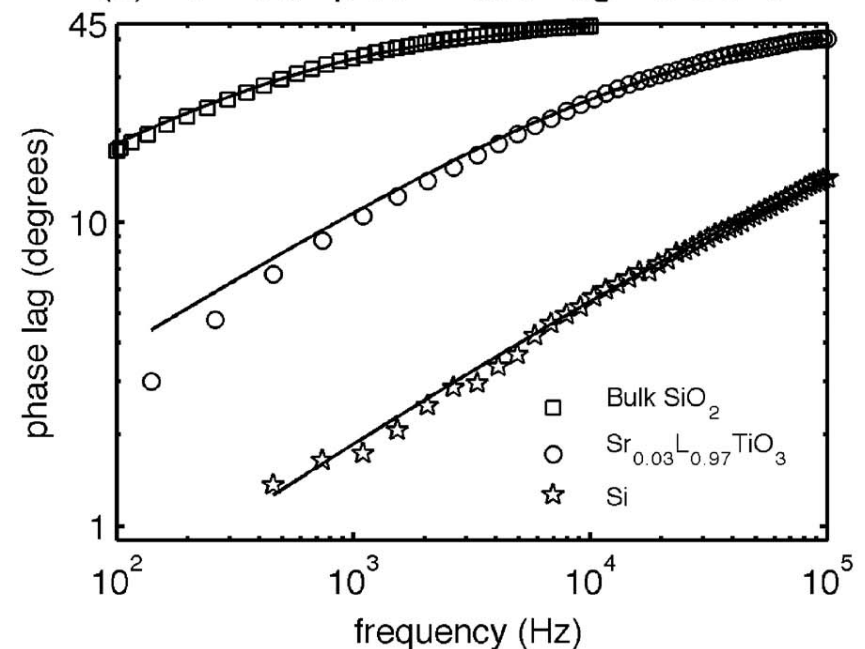
Frequency-domain measurements of κ in thin films

Continuous wave frequency domain thermoreflectance (FDTR)

(a) Experimental Setup



(a) Bulk Samples Phase Lag Data and Fit

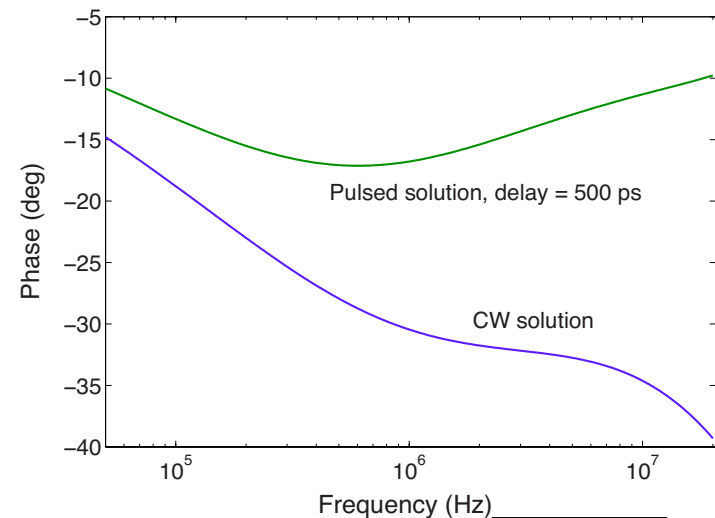
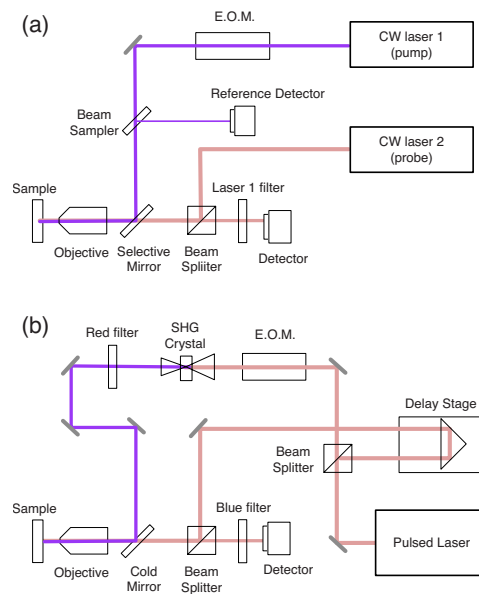


J. A. Malen, K. Baheti, T. Tong, Y. Zhao, J. A. Hudgings, and A. Majumdar. Optical measurement of thermal conductivity using fiber aligned frequency domain thermoreflectance. **Journal of Heat Transfer**, 133(8):081601, 2011.

Frequency-domain measurements of κ in thin films

Continuous wave vs. pulsed frequency domain thermoreflectance (FDTR)

Different responses, and can resolve different thermal properties



$$\delta_{\text{CW}} = \sqrt{\frac{\kappa}{\pi f C}}$$

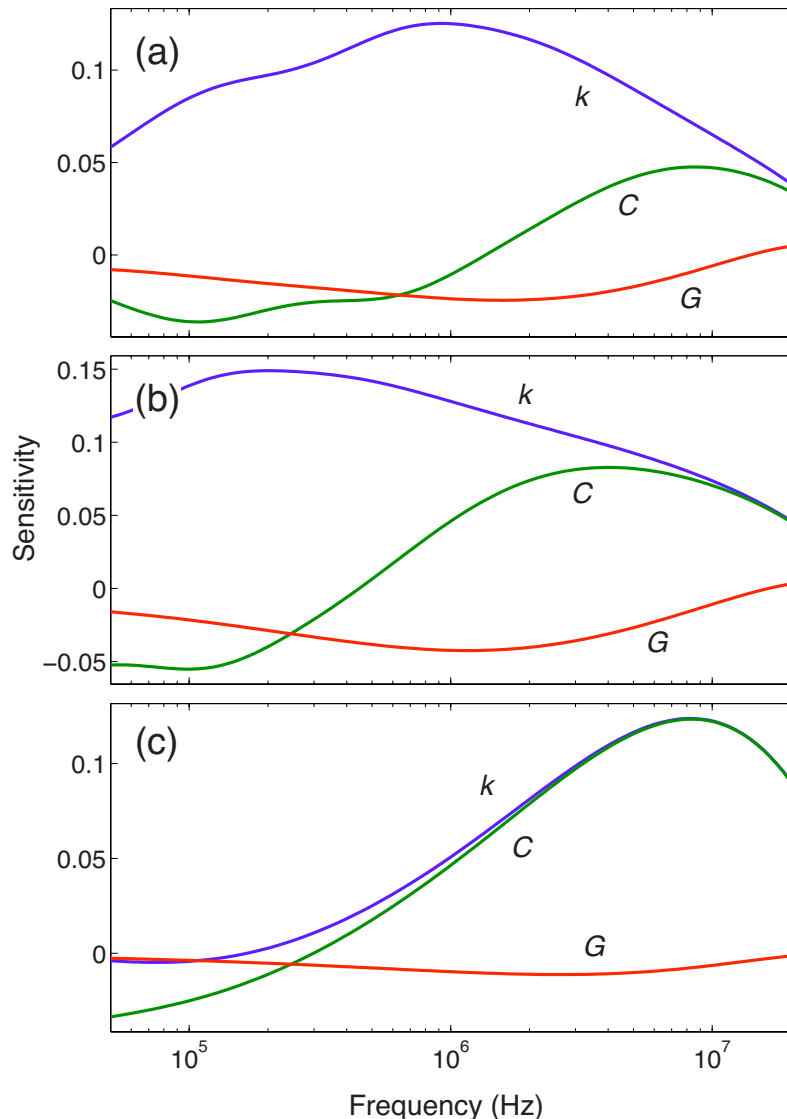
$$\delta_{\text{pulsed}} \propto \sqrt{\tau \frac{\kappa}{C}}$$

Advantage
Optical techniques can operate at higher modulation frequencies (smaller δ , better resolution to “nano”)

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.

Frequency-domain measurements of κ in thin films

FDTR with pulsed lasers



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

Silicon

**Where are the diffusivity
and effusivity regimes?**

Sapphire

**In what materials can you
accurately 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.

Frequency-domain measurements of κ in thin films

FDTR with pulsed lasers Can also measure in-plane thermal conductivity

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

Characterization of thin metal films via frequency-domain thermorefectance

Aaron J. Schmidt,^{1,2,a)} Ramez Cheaito,² and Matteo Chiesa²

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

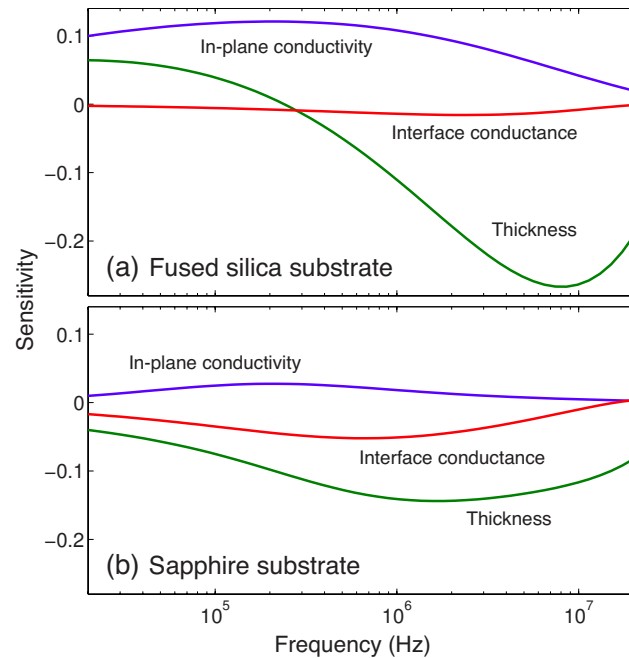


FIG. 5. (Color online) (a) The sensitivity parameter, Eq. (5), for in-plane thermal conductivity, metal-substrate boundary conductance, and metal thickness, for a sample consisting of an 80 nm film of Au deposited on a fused silica substrate. (b) The same sensitivities, calculated for a sapphire substrate.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

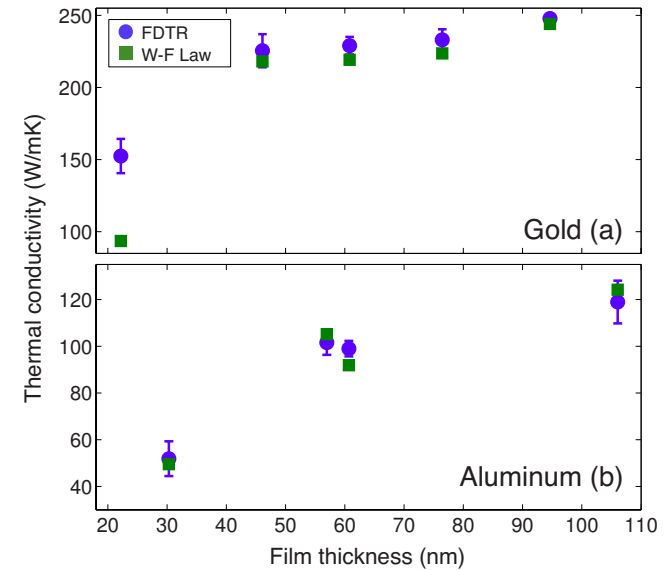
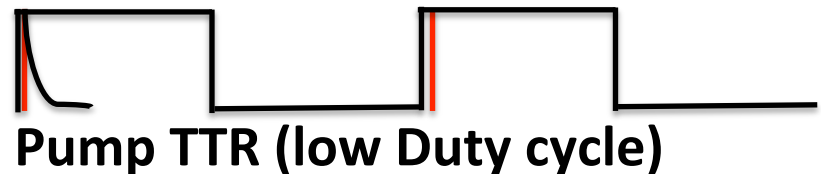
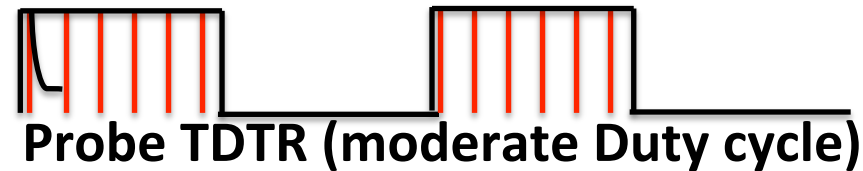
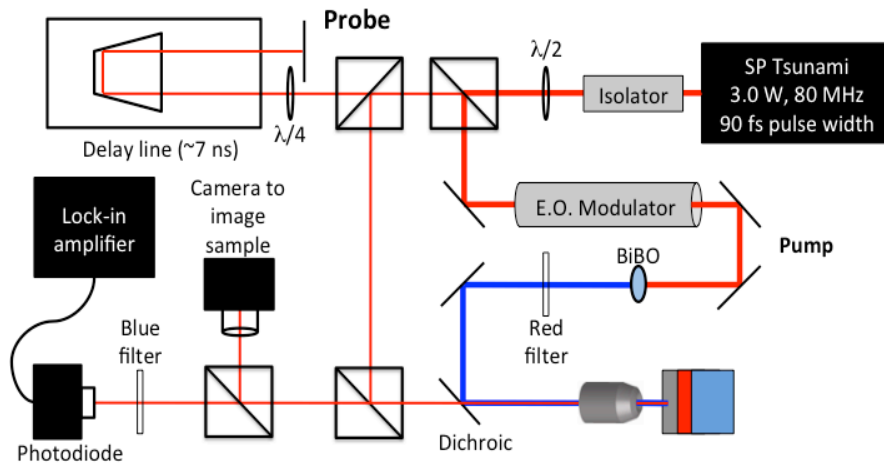


FIG. 4. (Color online) Thermal conductivity data obtained for (a) Au and (b) Al films on fused silica substrates. Circles are values obtained with the FDTR method, while the squares are values computed from electrical conductivity measurements using the WF law.

Time domain thermoreflectance (TDTR)

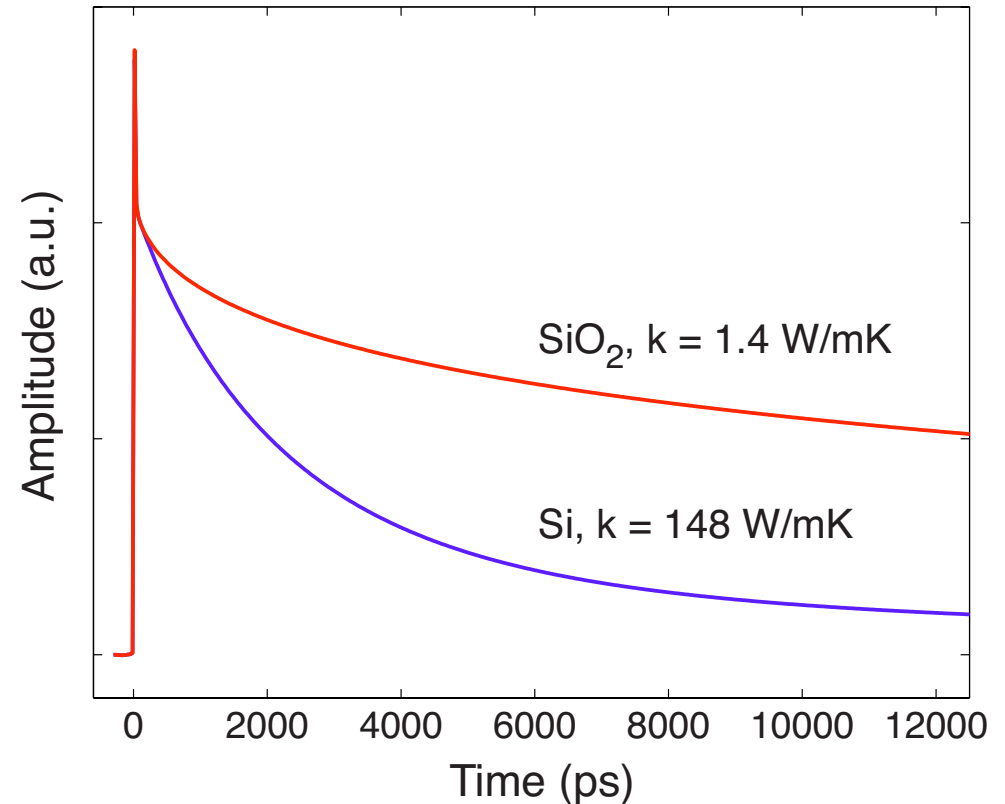
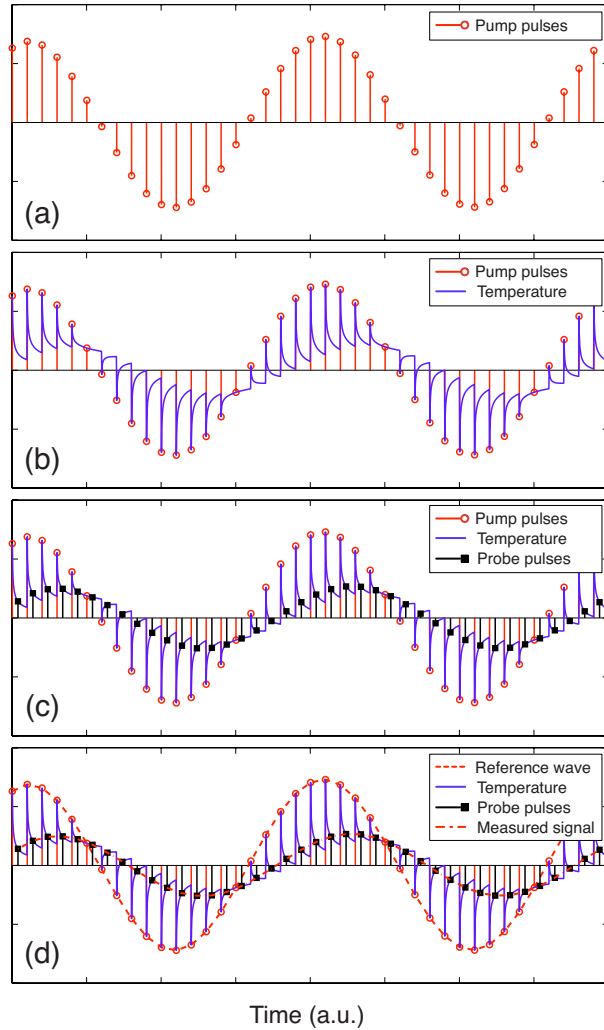
Can we achieve transient and periodic thermometry in nanosystems?

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.

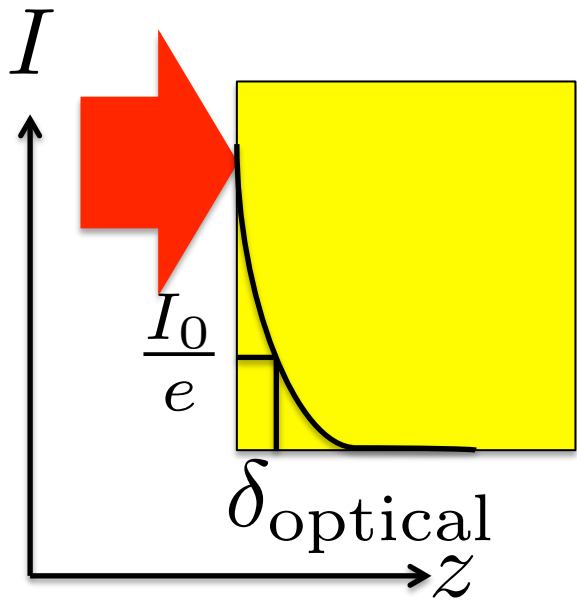
Time domain thermoreflectance (TDTR)



A. J. Schmidt, X. Chen, and G. Chen. Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance. **Review of Scientific Instruments**, **79**:114902, 2008.

Spatial regimes in TDTR

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



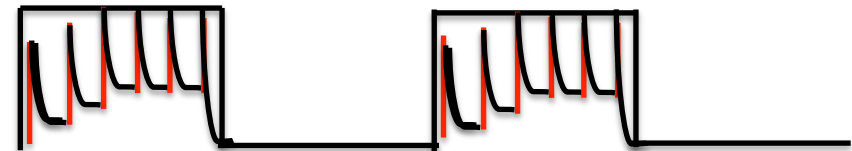
$$\delta_{\text{optical}} = \frac{\lambda}{4\pi k}$$

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)



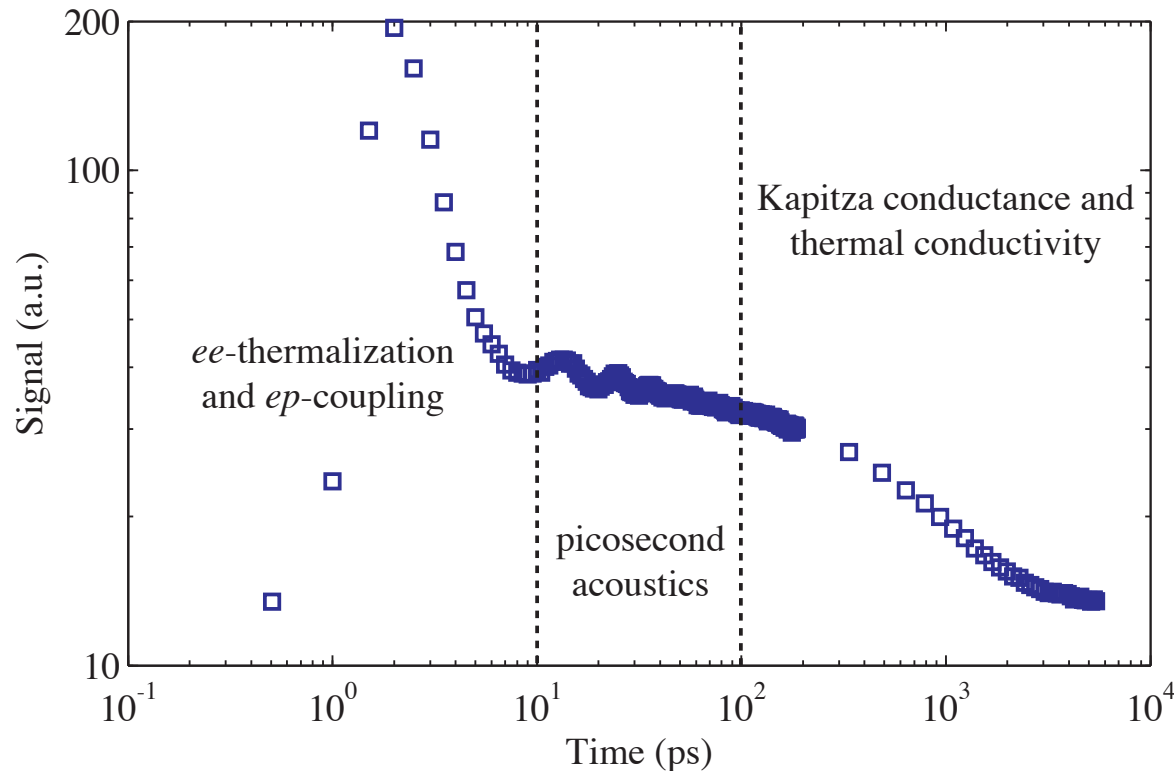
Accumulation leads to **MODULATED** heating event

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

Time domain thermoreflectance (TDTR)

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)



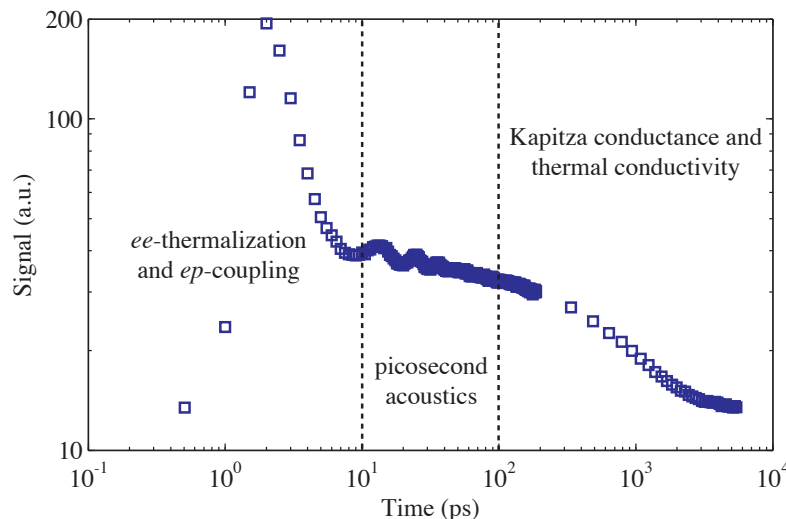
Thermal diffusion
(hundreds of ps to ns)

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

Time domain thermorefectance (TDTR)

Some TDTR References

- Cahill, "Analysis of heat flow in layered structures for time-domain thermorefectance," *Review of Scientific Instruments* **75**, 5119 (2004)
- Schmidt *et al.*, "Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermorefectance," *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 thermorefectance," *Journal of Heat Transfer* **133**, 061601 (2011)
- Schmidt, "Pump-probe thermorefectance," *Annual Review of Heat Transfer* **16**, 159 (2013)



So what can we can measure with 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

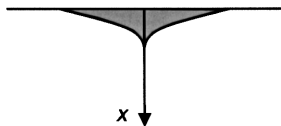
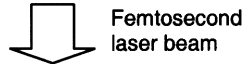
Patrick E. Hopkins

e-mail: pehopki@sandia.gov

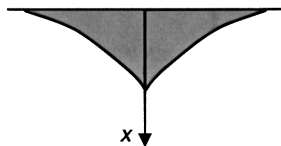
Leslie M. Phinney

Justin R. Serrano

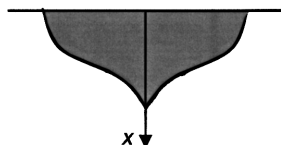
Sandia National Laboratories,
Albuquerque, NM 87185



Heat transfer by
ballistic motion of non-
equilibrium electrons



Heat transfer by diffusion
of hot electrons $T_e > T_l$



Heat transfer by
normal thermal
diffusion $T_e = T_l$

APPLIED PHYSICS LETTERS 103, 211910 (2013)



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

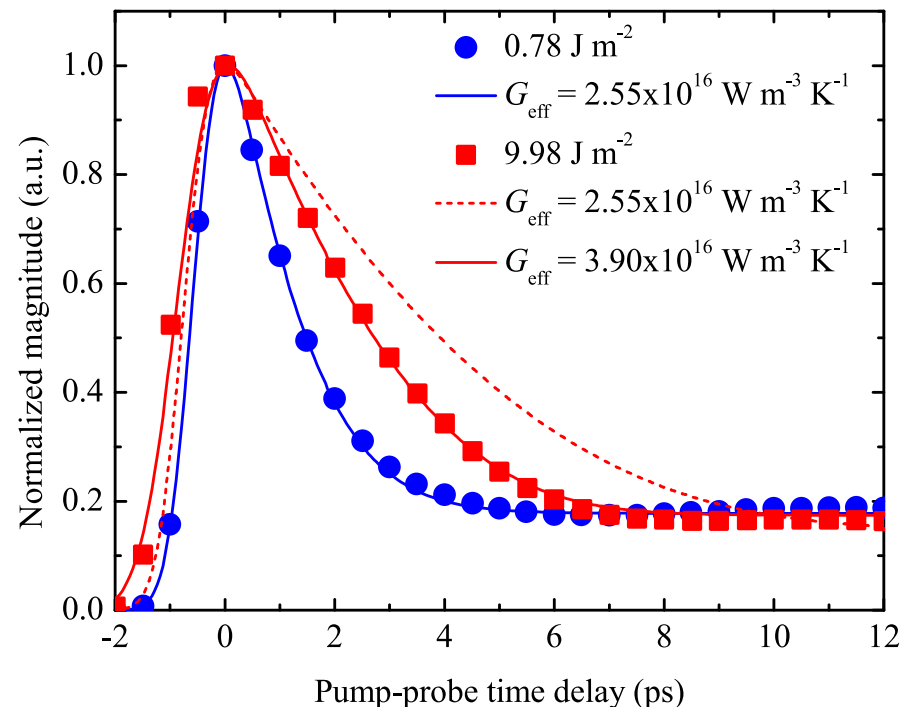
Patrick E. Hopkins,^{1,a)} John C. Duda,¹ Bryan Kaehr,^{2,3} Xiao Wang Zhou,⁴ C.-Y. Peter Yang,⁴ and Reese E. Jones⁴

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

²Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico 87106, USA

³Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA

⁴Sandia National Laboratories, Livermore, California 94550, USA



Electron-phonon scattering at interfaces

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



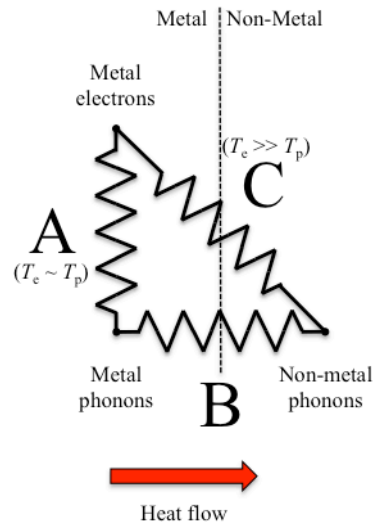
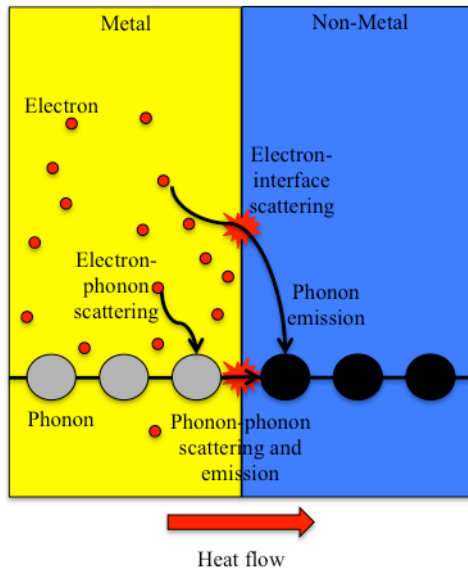
Mechanisms of nonequilibrium electron-phonon coupling and thermal conductance at interfaces

Ashutosh Giri,¹ John T. Gaskins,¹ Brian F. Donovan,¹ Chester Szejewski,¹ Ronald J. Warzoha,² Mark A. Rodriguez,³ Jon Ihlefeld,³ and Patrick E. Hopkins^{1,a)}

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

²Department of Mechanical Engineering, United States Naval Academy, Annapolis, Maryland 21401, USA

³Sandia National Laboratories, Albuquerque, New Mexico 87123, USA

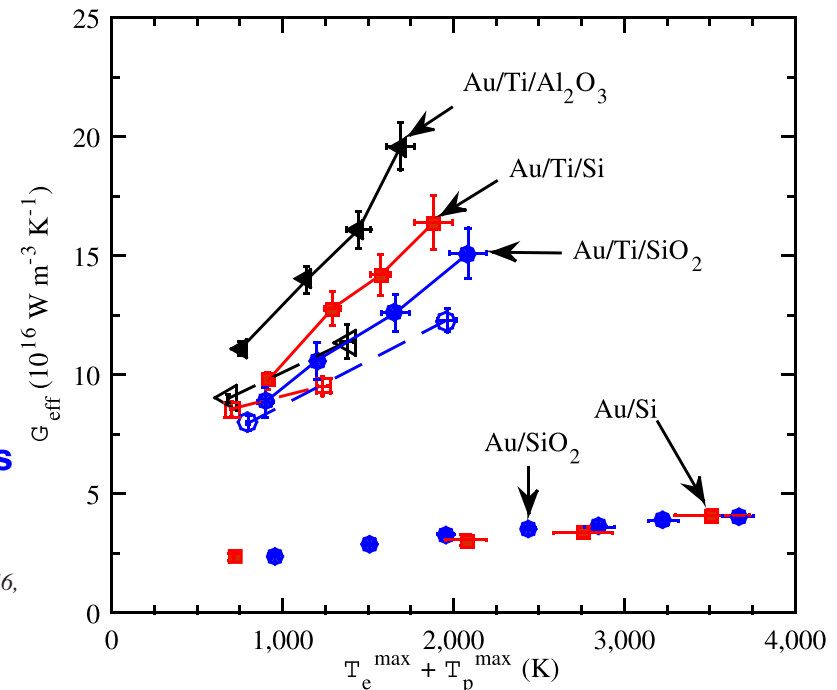


JOURNAL OF APPLIED PHYSICS **105**, 023710 (2009)

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

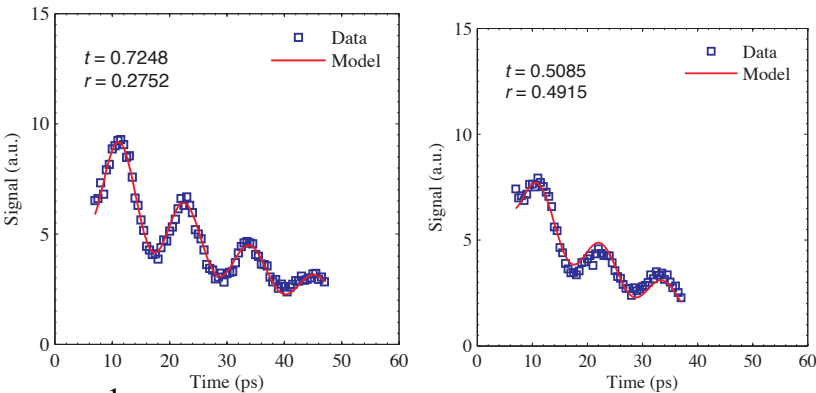
Patrick E. Hopkins,^{a)} Jared L. Kassebaum, and Pamela M. Norris

Department of Mechanical and Aerospace Engineering, University of Virginia, P.O. Box 400746, Charlottesville, Virginia 22904-4746, USA



Time domain thermoreflectance (TDTR)

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)}

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

²Sandia National Laboratories, Livermore, California 94550, USA

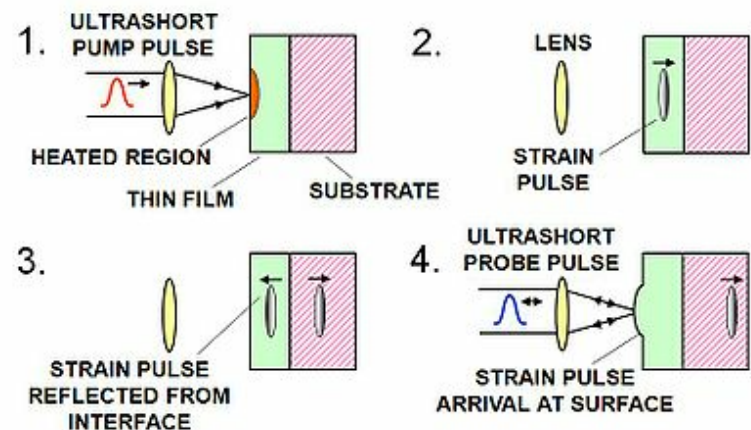
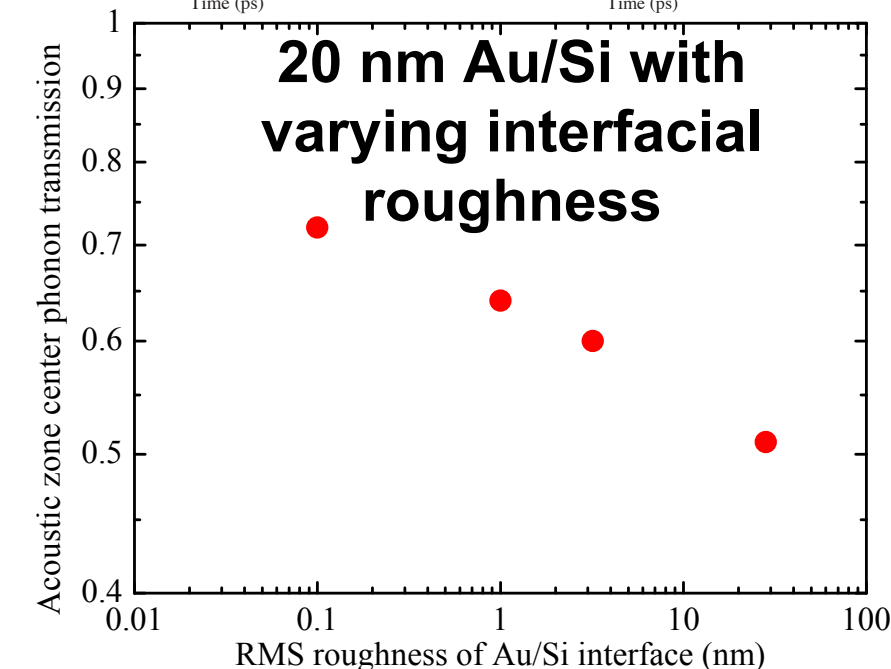


Image from Wikipedia: http://en.wikipedia.org/wiki/Picosecond_ultrasonics

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

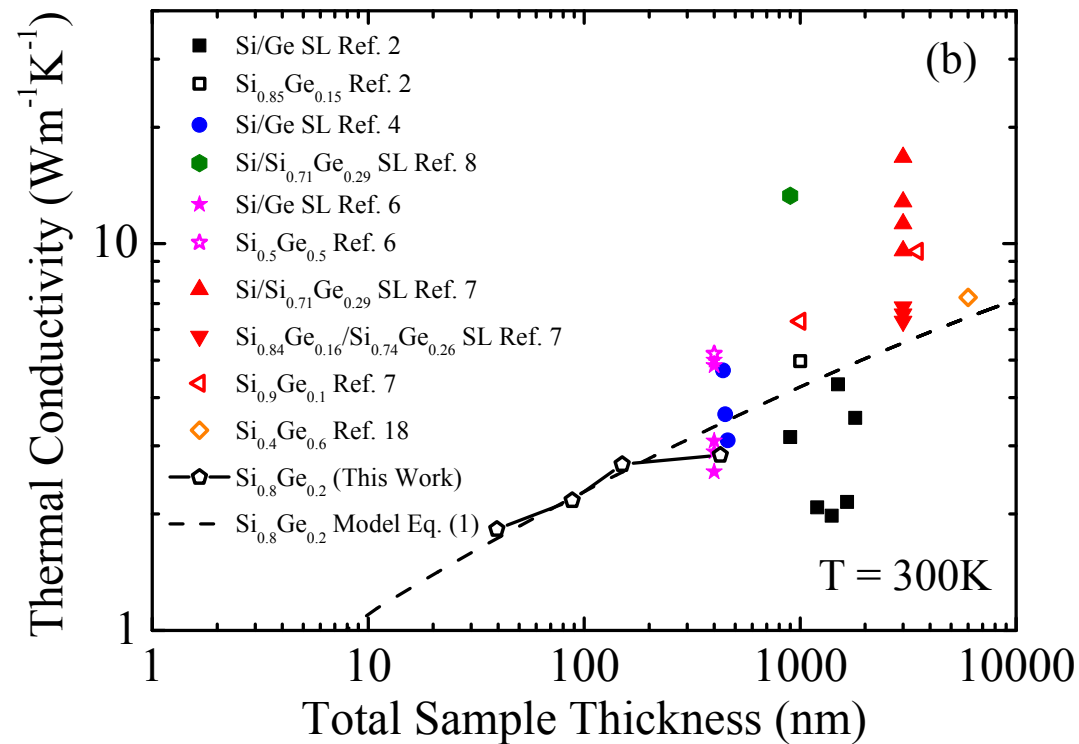
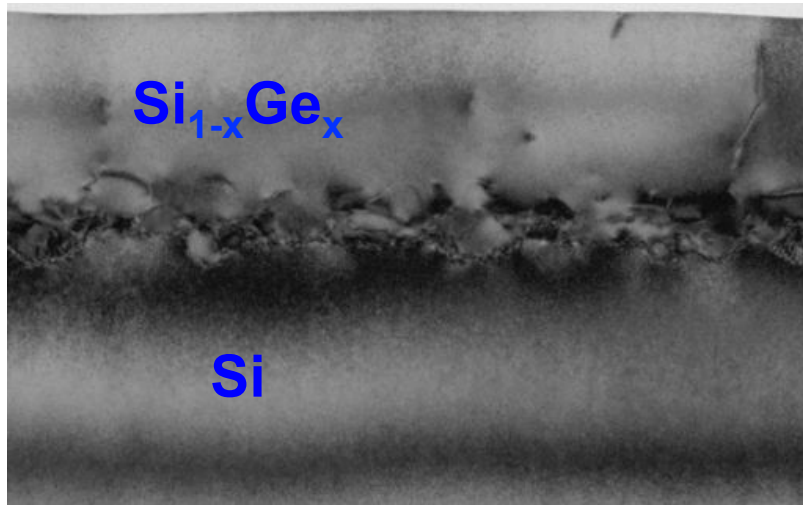
Ramez Cheaito,¹ John C. Duda,^{1,2} Thomas E. Beechem,² Khalid Hattar,² Jon F. Ihlefeld,² Douglas L. Medlin,³ Mark A. Rodriguez,² Michael J. Campion,^{2,4} Edward S. Piekos,² and Patrick E. Hopkins^{1,*}

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†}

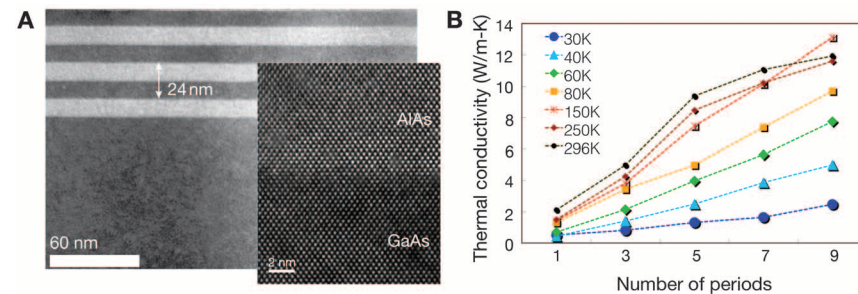
LETTERS

PUBLISHED ONLINE: 8 DECEMBER 2013 | DOI: 10.1038/NMAT3826

nature
materials

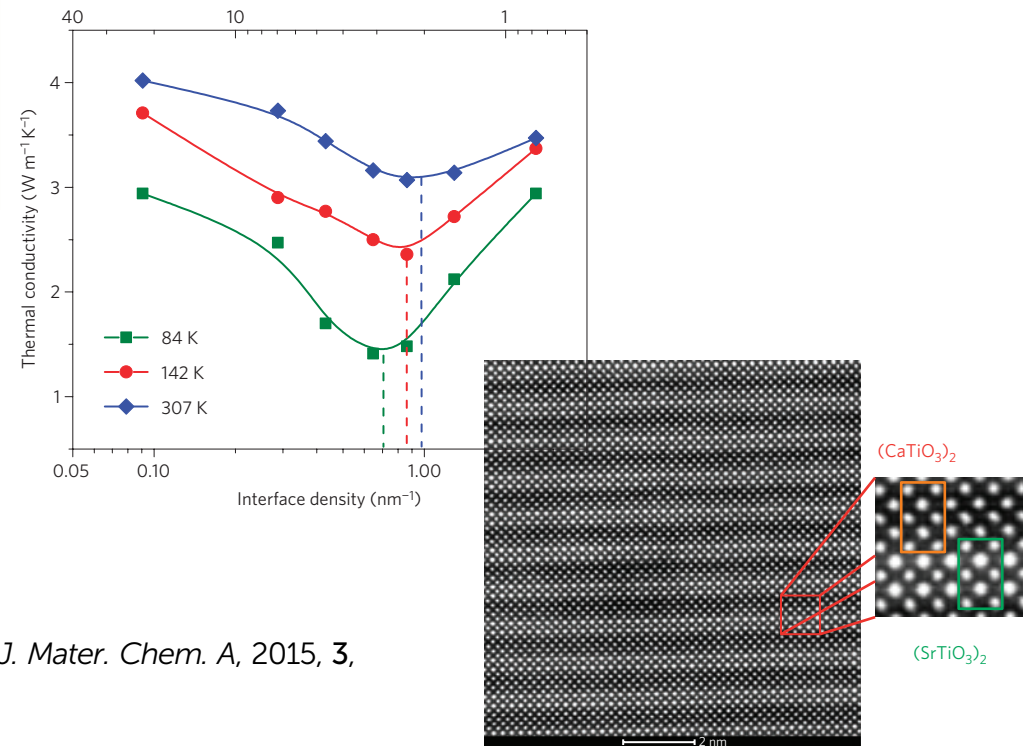
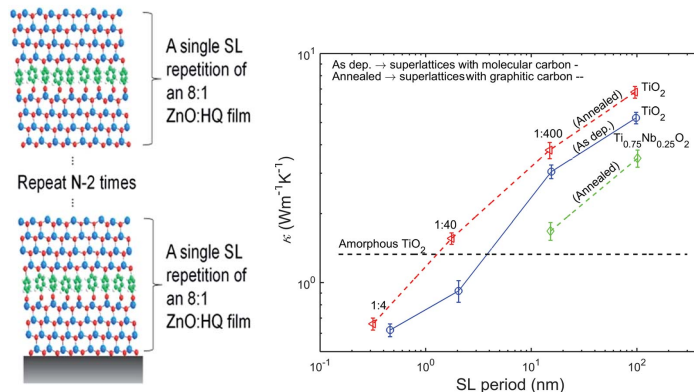
Crossover from incoherent to coherent phonon scattering in epitaxial oxide superlattices

Jayakanth Ravichandran^{1,2,†}, Ajay K. Yadav^{2,3}, Ramez Cheaito^{4,‡}, Pim B. Rossen³, Arsen Soukiasian⁵, S. J. Suresha², John C. Duda⁴, Brian M. Foley⁴, Che-Hui Lee⁵, Ye Zhu⁶, Arthur W. Lichtenberger⁷, Joel E. Moore^{2,8}, David A. Muller^{6,9}, Darrell G. Schlom^{5,9}, Patrick E. Hopkins⁴, Arun Majumdar¹⁰, Ramamoorthy Ramesh^{1,2,3,8,11*} and Mark A. Zurbuchen^{12,13,14*}



Ultra-low thermal conductivity in TiO₂: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

Nanocomposite films and structures (100's ps – ns)

Thin Solid Films 520 (2012) 6109–6117

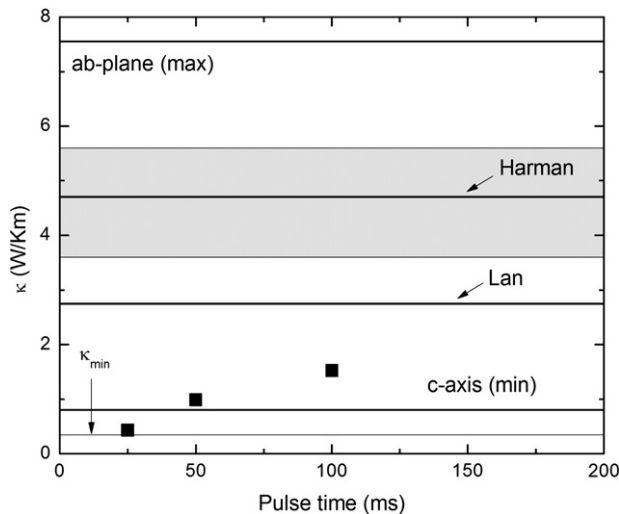
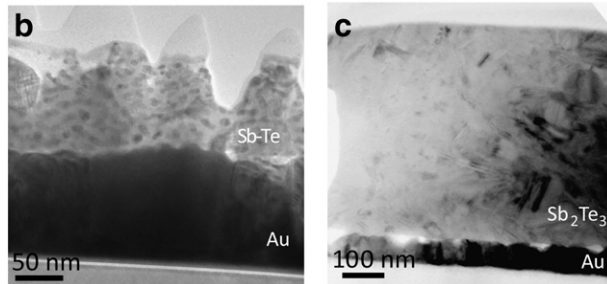
Electrodeposition and characterization of nano-crystalline antimony telluride thin films

J.L. Lensch-Falk^a, D. Banga^a, P.E. Hopkins^{b,c}, D.B. Robinson^a, V. Stavila^a, P.A. Sharma^b, D.L. Medlin^{a,*}

^a Sandia National Laboratories, Livermore, CA 94550, USA

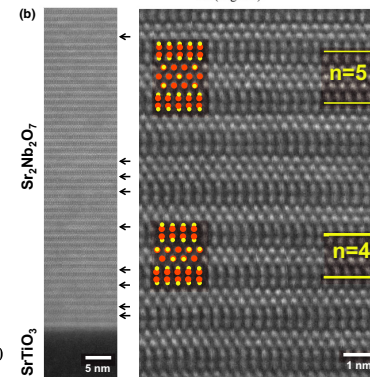
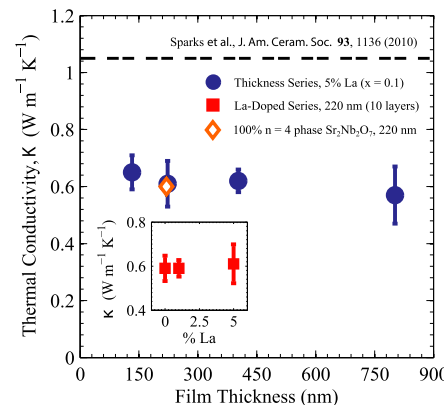
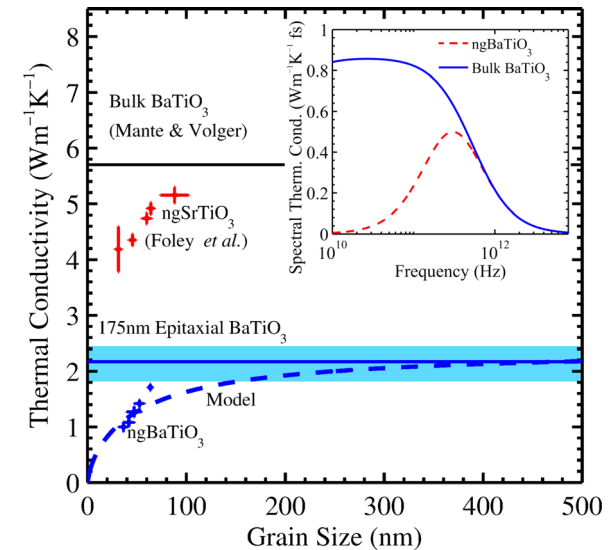
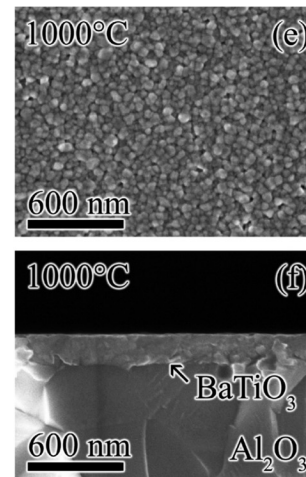
^b Sandia National Laboratories, Albuquerque, NM 87185, USA

^c Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, VA 22904, USA



Spectral phonon scattering effects on the thermal conductivity of nano-grained barium titanate

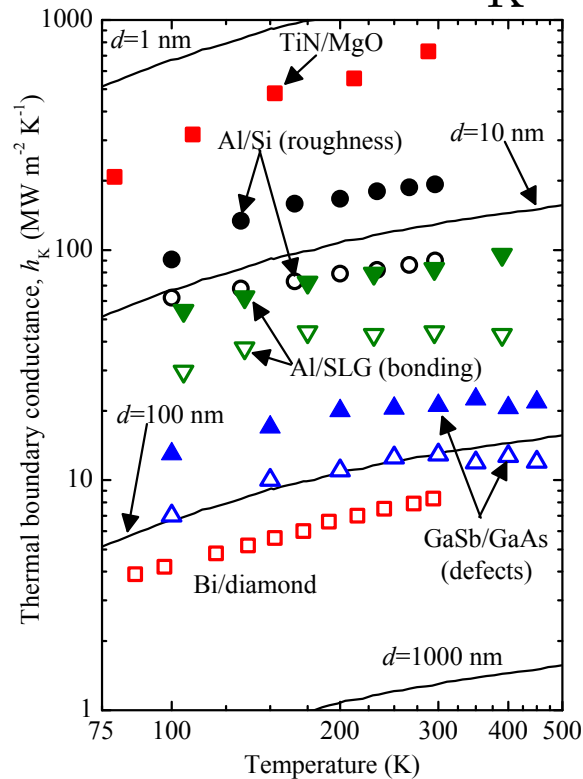
Brian F. Donovan,¹ Brian M. Foley,² Jon F. Ihlefeld,^{3,a)} Jon-Paul Maria,⁴ and Patrick E. Hopkins^{2,b)}



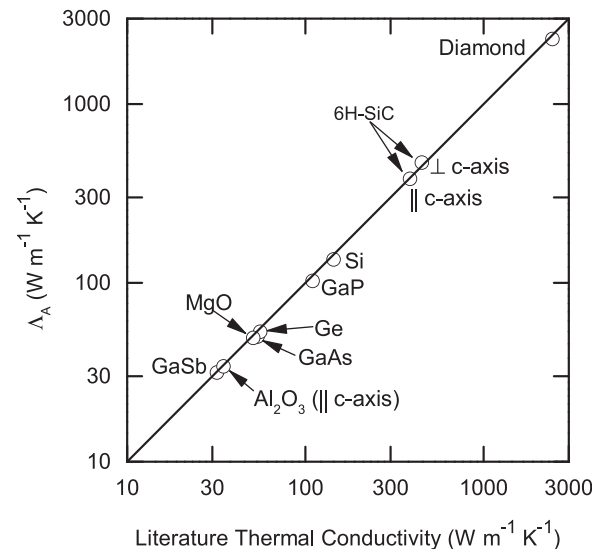
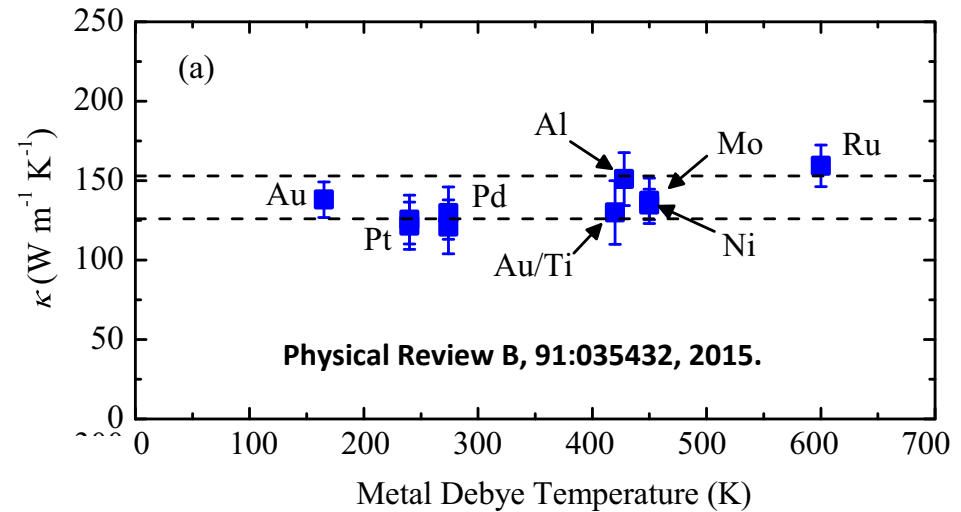
Journal of the American Ceramic Society, 98:624–628, 2015

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

$$q_{\text{int}} = h_K \Delta T = \frac{1}{R_K \Delta T}$$



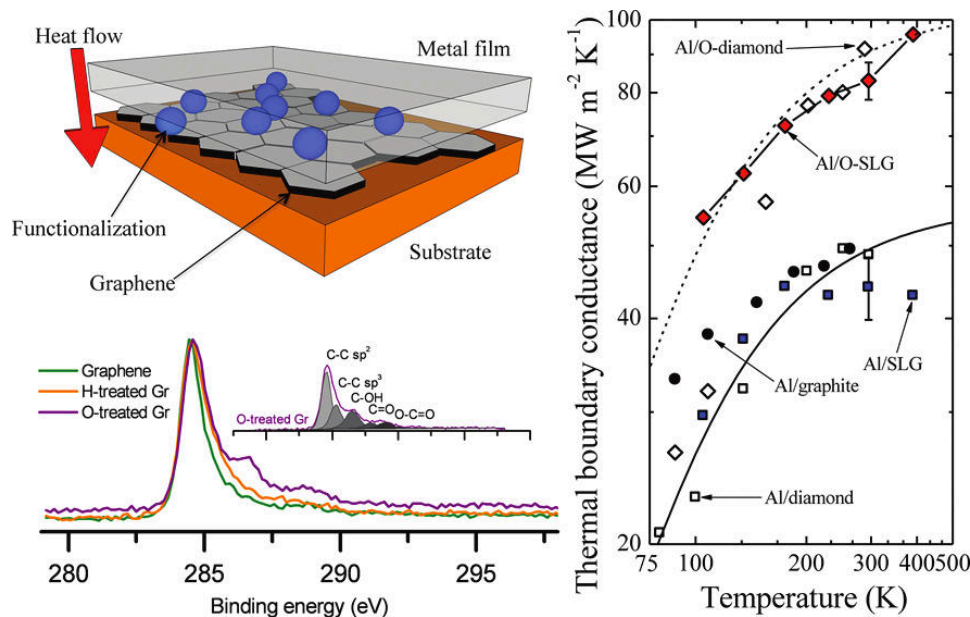
Hopkins, "Thermal transport across solid interfaces with nanoscale imperfections: Effects of roughness, disorder, dislocations and bonding on thermal boundary conductance," *ISRN Mechanical Engineering* 2013, 682586 (2013)



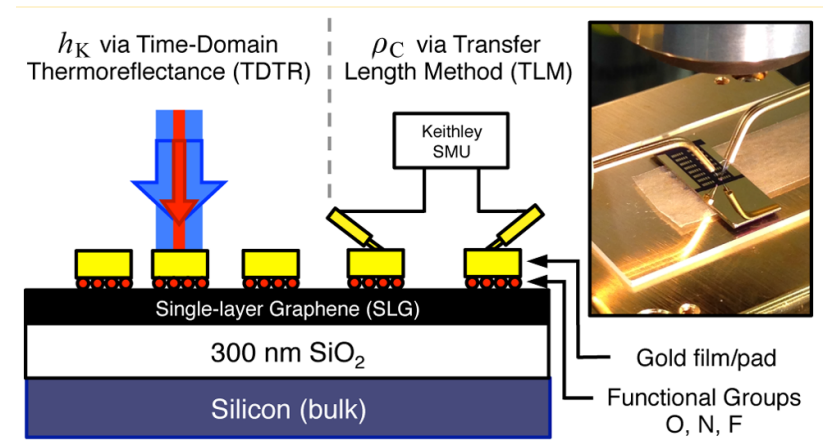
Applied Physics Letters, 107(20):203112, 2015

Time domain thermorefectance (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. Robinson, 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. Hernández, 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.

Time domain thermoreflectance (TDTR)

2D materials (e.g., graphene and their contacts)
Can also do this with FDTR by going to high modulation frequencies....and spatial mapping!

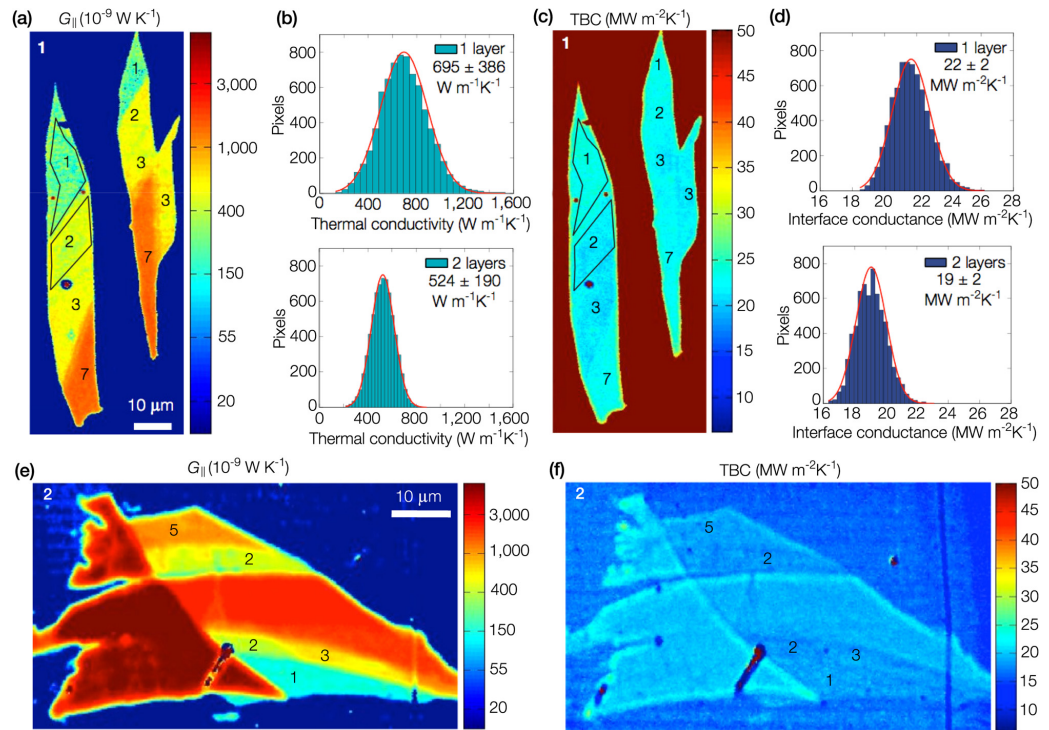
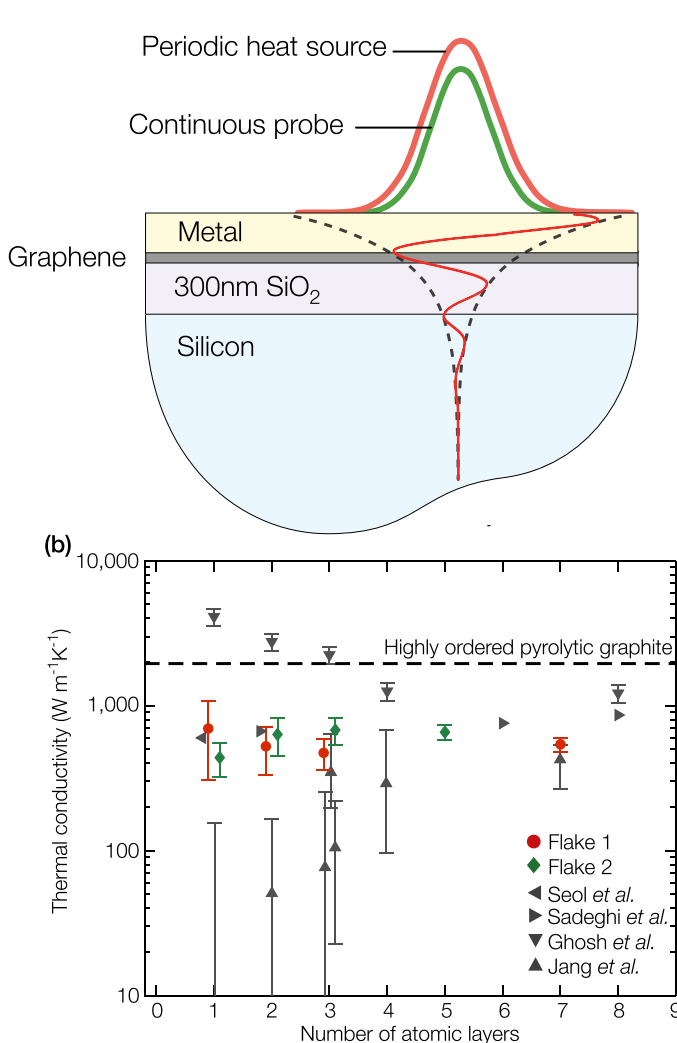


FIG. 8. Thermal conductance maps of the two samples. (a) $G_{||}$ map of flake 1. (b) Histograms of the thermal conductivity for single-layer graphene and bi-layer graphene analyzed from the polygons in (a). (c) TBC map of flake 1. (d) Histograms of TBC of single-layer graphene and bi-layer graphene, analyzed from the polygons in (c). (e) $G_{||}$ map of flake 2. (f) TBC map of flake 2. The upper limit of the color bars for (c) is set at 50 MW m⁻² K⁻¹ to highlight the graphene flake, although the measured value of TBC for Au/Ti/SiO₂ for this sample was closer to 100 MW m⁻² K⁻¹. The solid red lines in (b) and (d) are normal distribution fits.

J. Yang, E. Ziade, C. Maragliano, R. Crowder, X. Wang, M. Stefancich, M. Chiesa, A. K. Swan, and A. J. Schmidt. Thermal conductance imaging of graphene contacts. **Journal of Applied Physics**, 116(2):023515, 2014.

Time domain thermorefectance (TDTR)

Highly porous/non-conformal films/liquids

Minimum thermal conductivity considerations in aerogel thin films

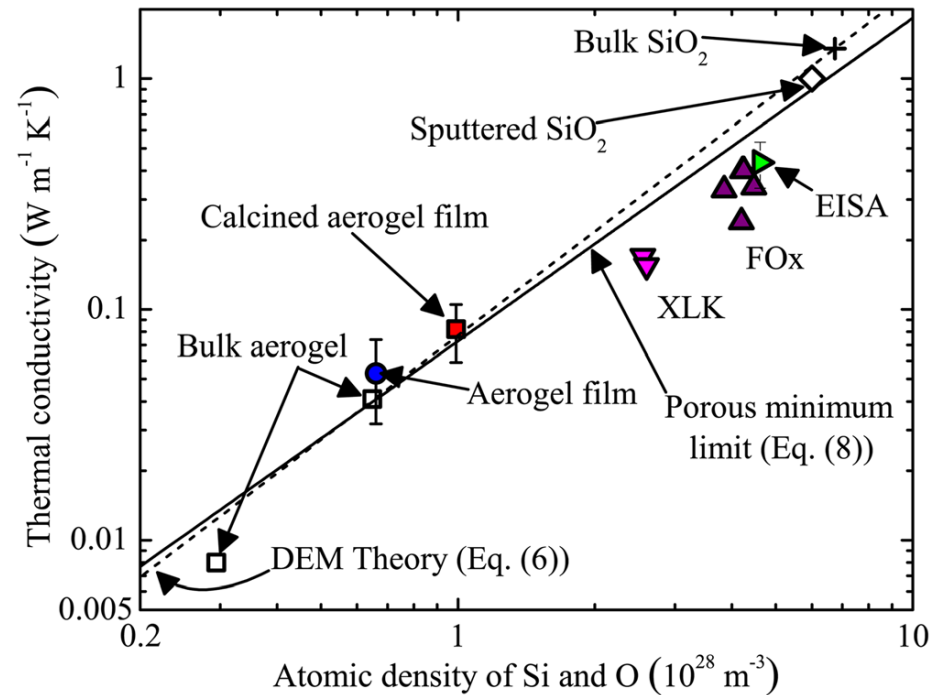
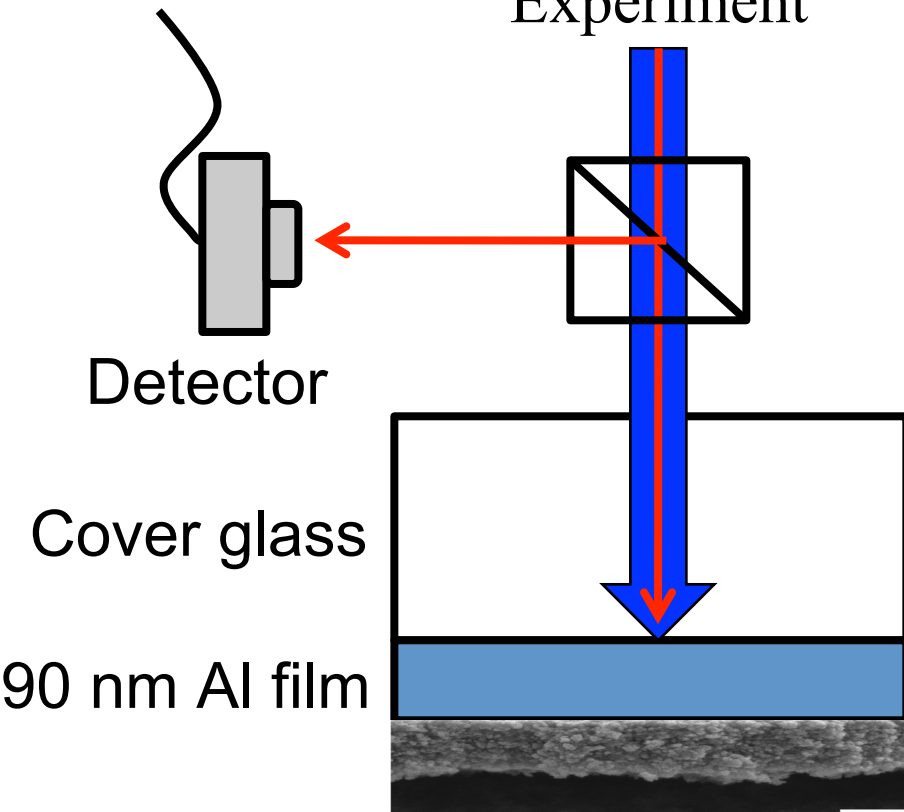
Patrick E. Hopkins,^{1,a)} Bryan Kaehr,^{2,3} Edward S. Piekos,² Darren Dunphy,³ and C. Jeffrey Brinker^{2,3}

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

²Sandia National Laboratories, Albuquerque, New Mexico 87123, USA

³Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA

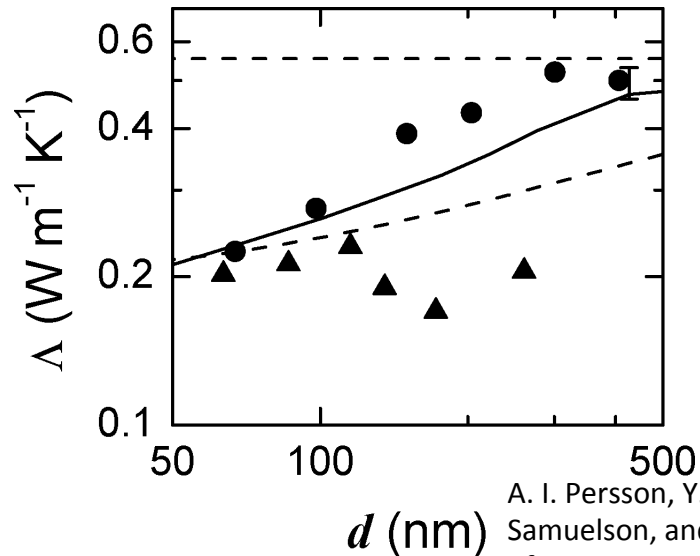
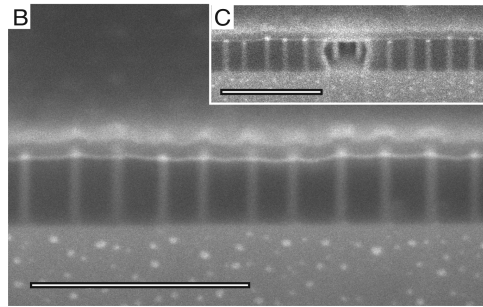
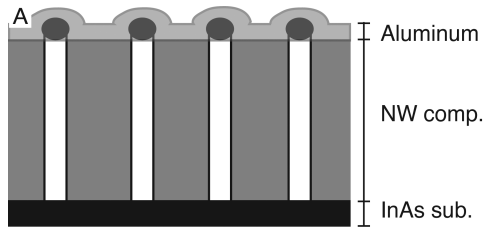
Experiment



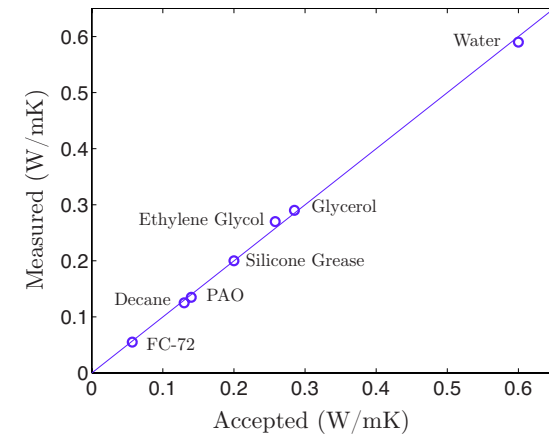
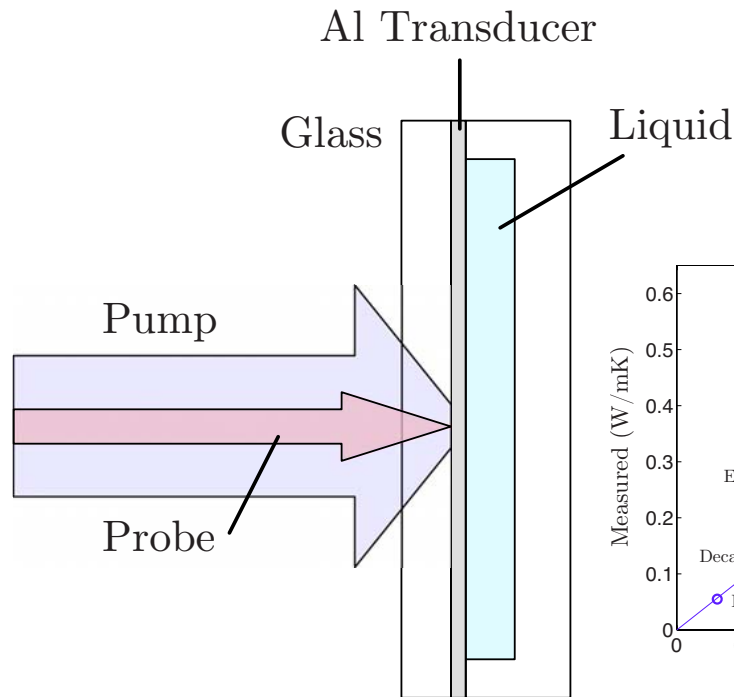
Porous or non-conformal films

Time domain thermoreflectance (TDTR)

Highly porous/non-conformal films/liquids



A. I. Persson, Y. K. Koh, D. G. Cahill, L. Samuelson, and H. Linke. Thermal conductance of InAs nanowire composites. **Nano Letters**, 9:4484–4488, 2009.



A. Schmidt, M. Chiesa, X. Chen, and G. Chen. An optical pump-probe technique for measuring the thermal conductivity of liquids. *Review of Scientific Instruments*, 79:064902, 2008.

Time domain thermoreflectance (TDTR)

“Depth profiling” of buried interfaces

APPLIED PHYSICS LETTERS **98**, 161913 (2011)

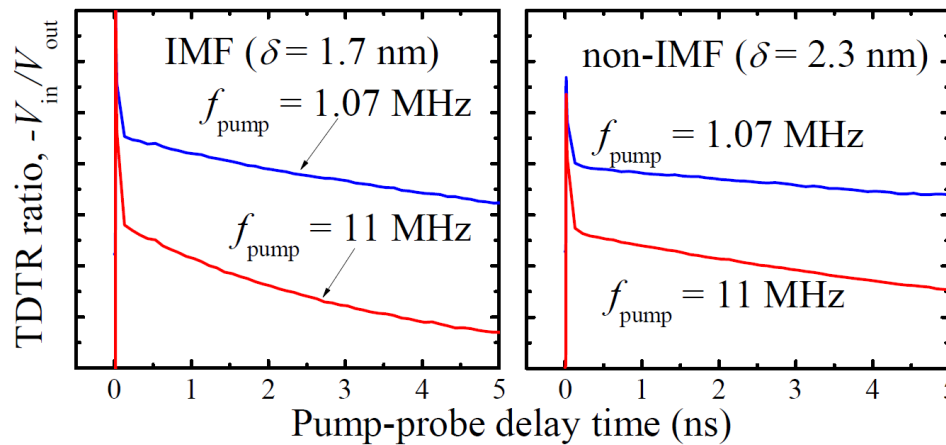
Effect of dislocation density on thermal boundary conductance across GaSb/GaAs interfaces

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 Balakrishnan³

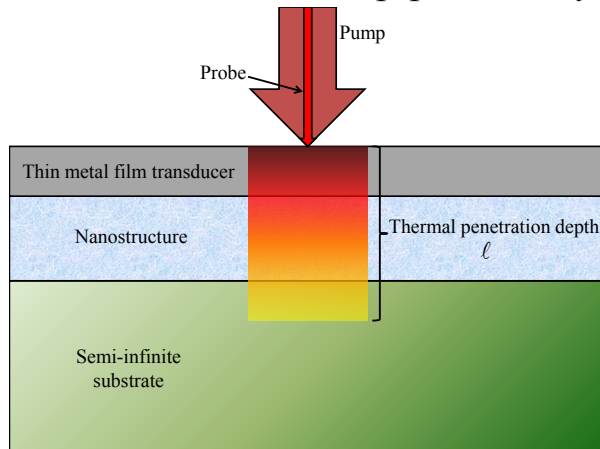
¹Sandia National Laboratories, Albuquerque, New Mexico 87185, USA

²Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA

³Department of Electrical and Computer Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA



Increase how “deep” you probe in TDTR by decreasing the frequency



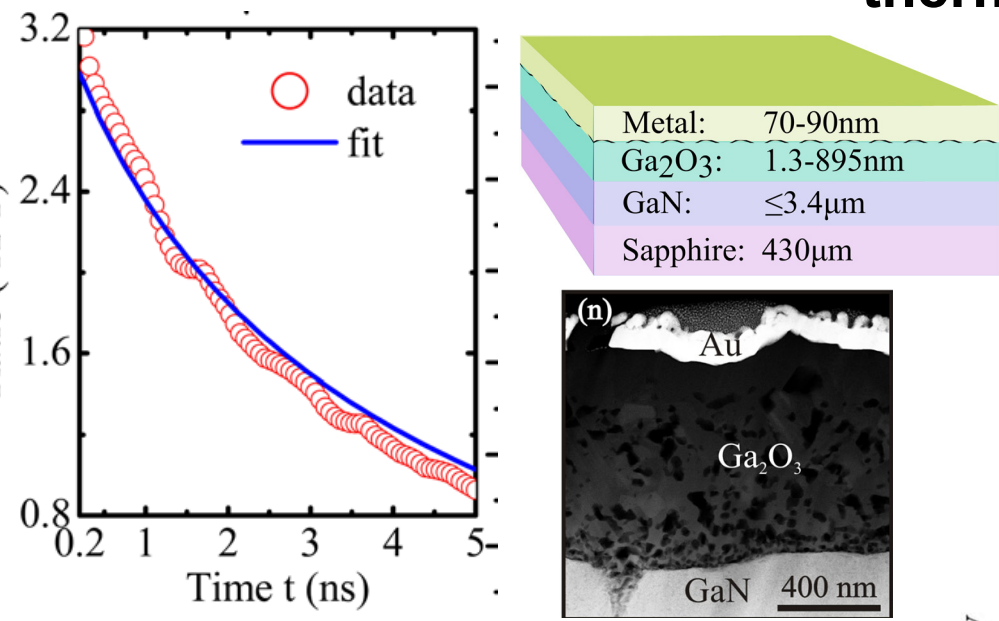
h_K – Al/GaSb ($f = 11$ MHz)

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

h_K – GaSb/GaAs ($f = 1.07$ MHz)

Limitations – TDTR (and other thermoreflectance techniques)

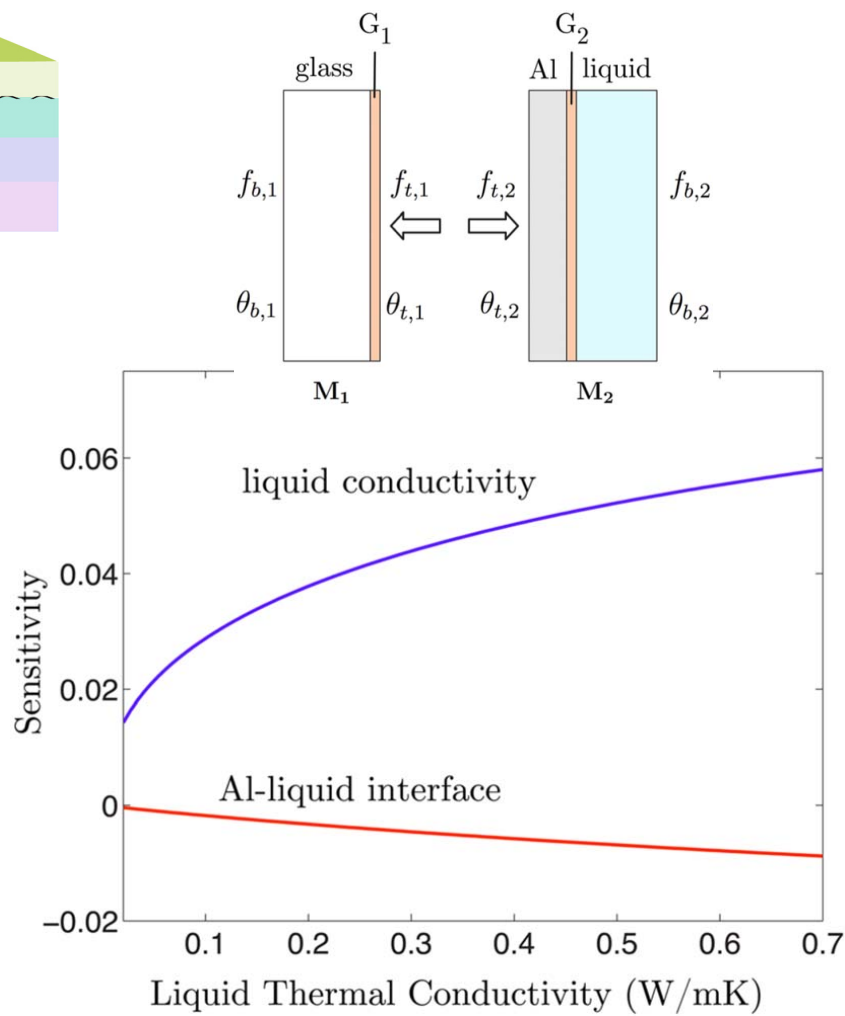
Rough surfaces (~>40 nm RMS-“ish”)



C. J. Szejewski, N. C. Creange, K. Sun, A. Giri, B. F. Donovan, C. Constantin, and P. E. Hopkins. Size effects in the thermal conductivity of gallium oxide ($\beta\text{-Ga}_2\text{O}_3$) films grown via open-atmosphere annealing of gallium nitride. **Journal of Applied Physics**, 117(8):084308, 2015.

A. Schmidt, M. Chiesa, X. Chen, and G. Chen. An optical pump-probe technique for measuring the thermal conductivity of liquids. **Review of Scientific Instruments**, 79:064902, 2008.

Thermal boundary conductance across interfaces adjacent to low thermal effusivity solids (e.g., liquids)



Thanks!

What we covered

- **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**
 - **TDTR**
 - **FDTR**
 - **Limitations and potentials....**

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