



**SCHOOL *of* ENGINEERING & APPLIED SCIENCE  
UNIVERSITY *of* VIRGINIA**

# Thin film testing

## DARPA MATRIX Workshop

### 12/10/2015



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[patrickehopkins.com](http://patrickehopkins.com)

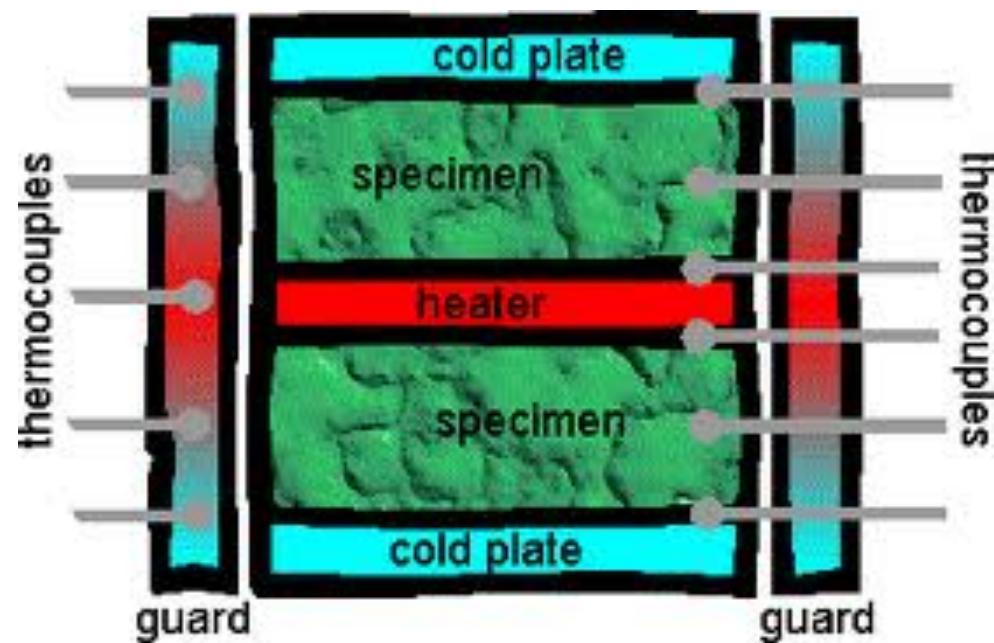
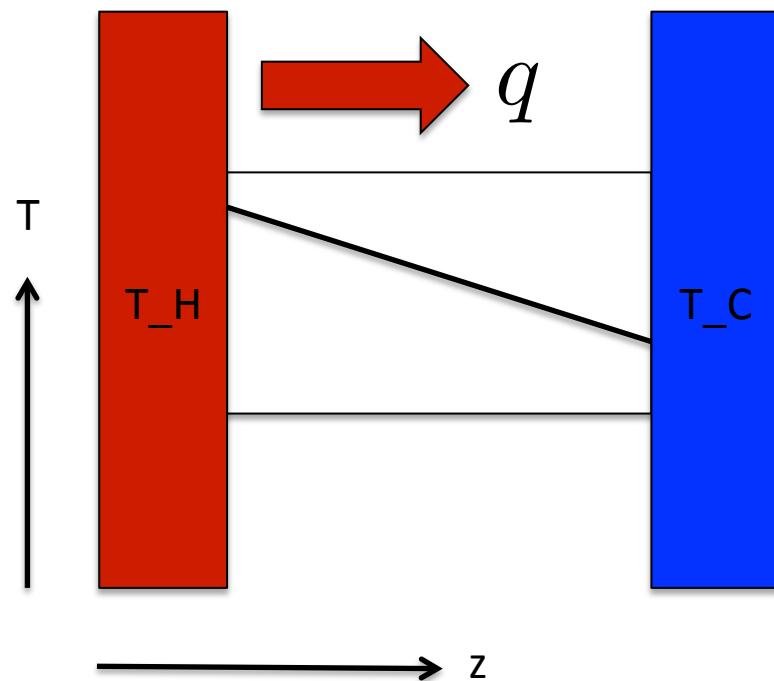
# Overview: Methods for testing $\kappa$ of thin films

- **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”)**
  - $3\omega$
  - **Thermoreflectance-based techniques**
    - TDTR
    - FDTR
    - Limitations and potentials....

# Steady state measurements of $\kappa$ in bulk systems

## “Guarded hot plate”

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



# Steady state measurements of $\kappa$ in nanosystems

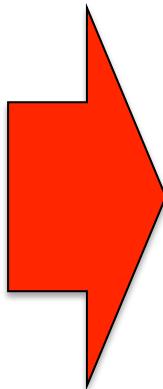
Utilize ability to “nano pattern” electrical contacts and circuits

Fourier Law

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

Joule heating

$$q = I^2 R$$



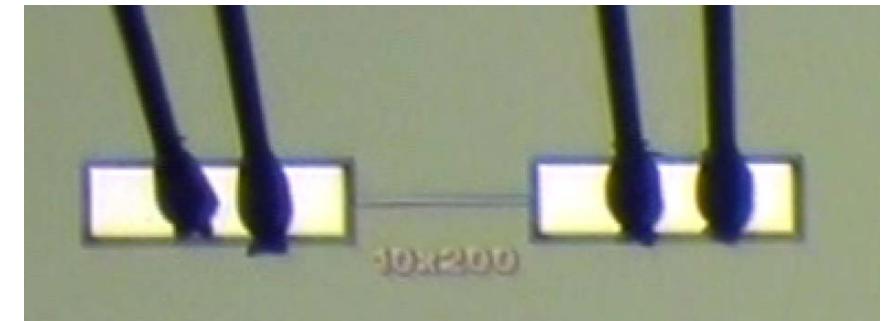
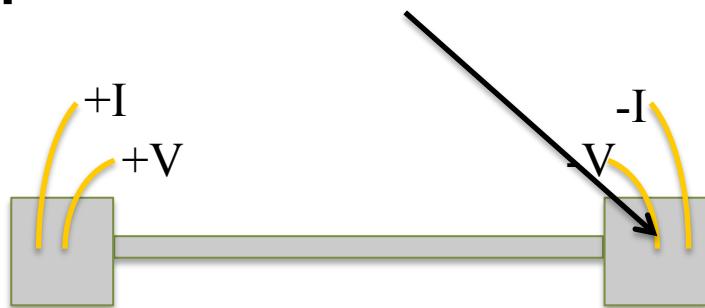
$$\kappa V \frac{\partial^2 T}{\partial x^2} + I^2 R(T) = 0$$

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

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

# Steady state measurements of $\kappa$ in nanosystems

**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???**

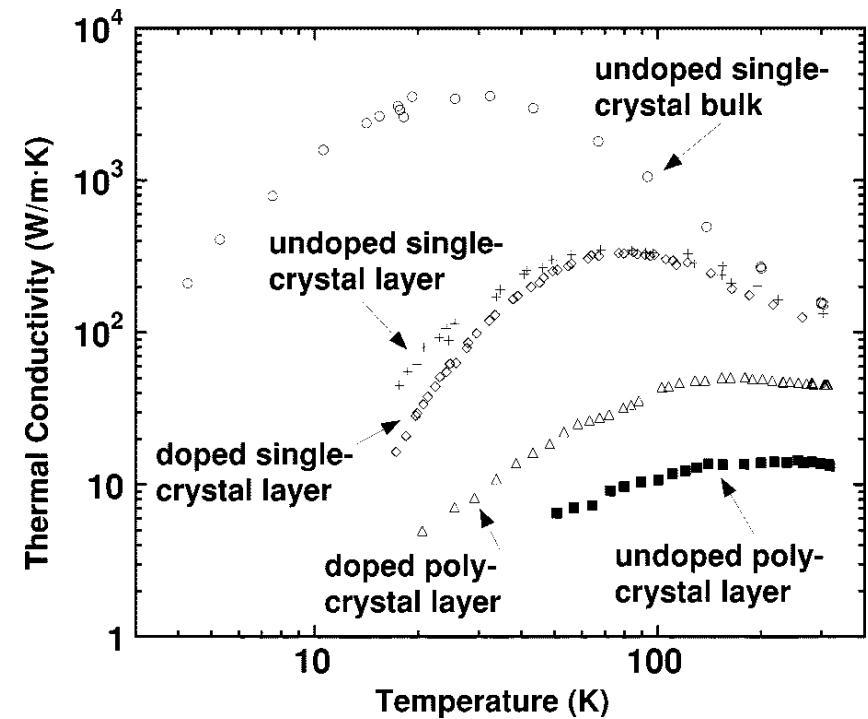
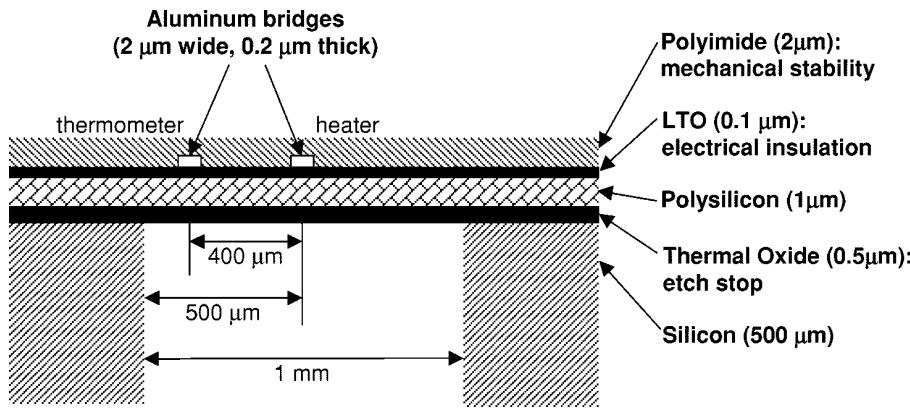
*Lithography/metal processing*

# Steady state measurements of $\kappa$ in nanosystems

## In-plane thermal conductivity measurements of Si thin films

### Thermal Conductivity of Doped Polysilicon Layers

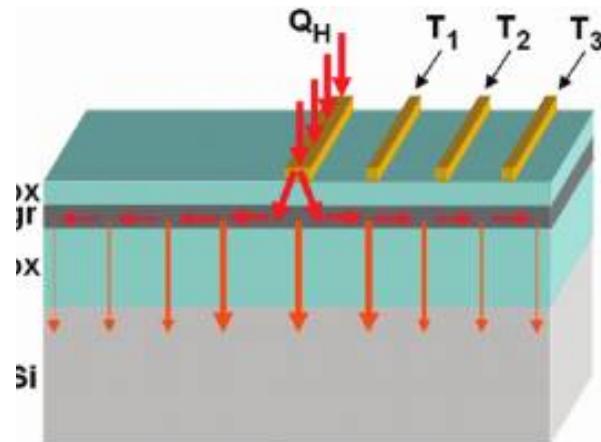
Angela D. McConnell, Srinivasan Uma, *Member, IEEE*, and Kenneth E. Goodson, *Associate Member, IEEE*



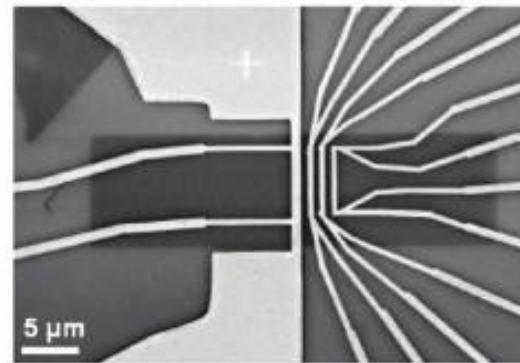
A. D. McConnell, S. Uma, and K. E. Goodson. Thermal conductivity of doped polysilicon layers. *Journal of Microelectromechanical Systems*, 10:360–369, 2001.

# Steady state measurements of $\kappa$ in nanosystems

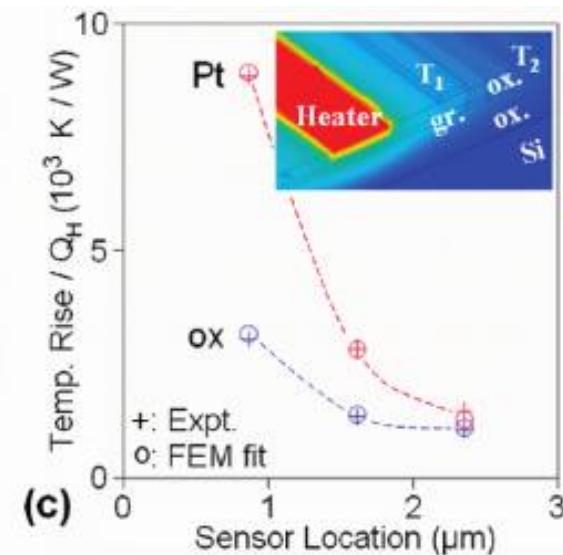
## In-plane thermal conductivity of “atomically thin” film (i.e., graphene)



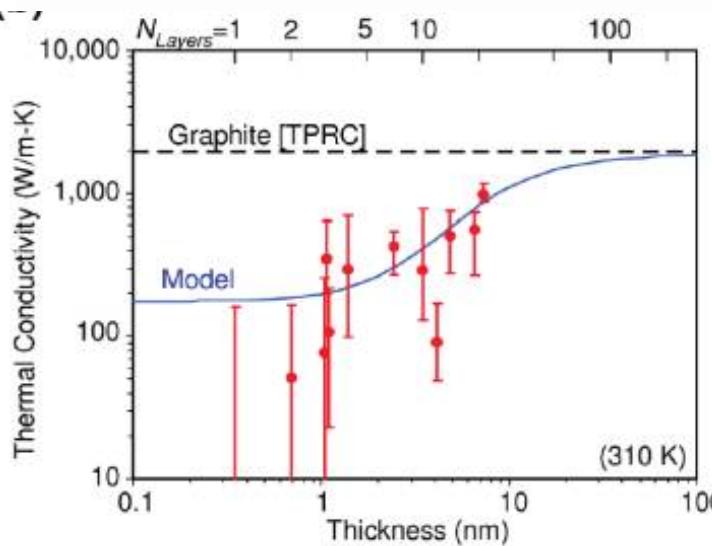
(a)



(b)



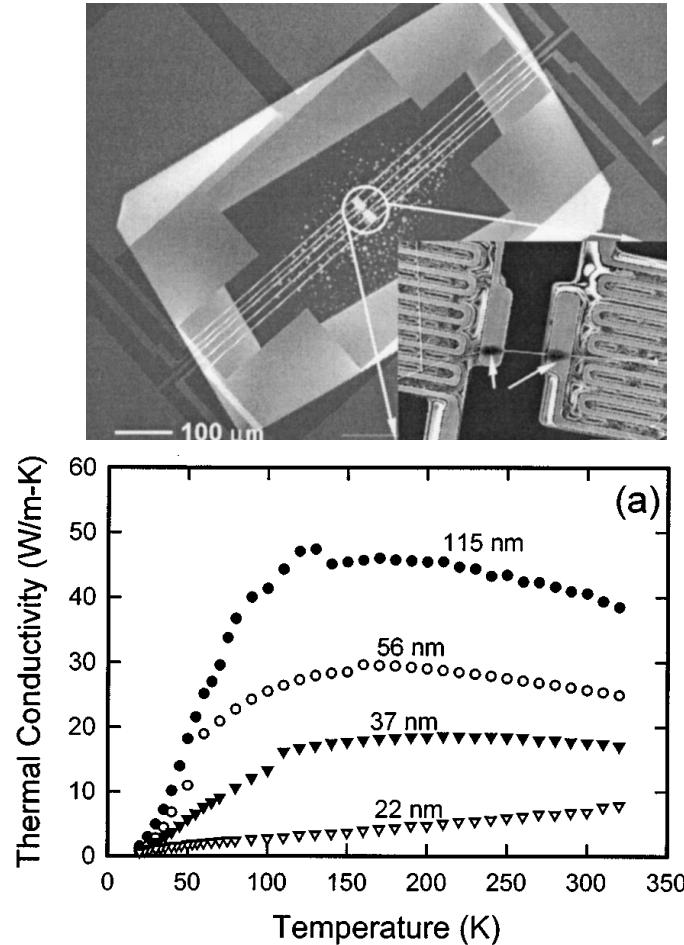
(c)



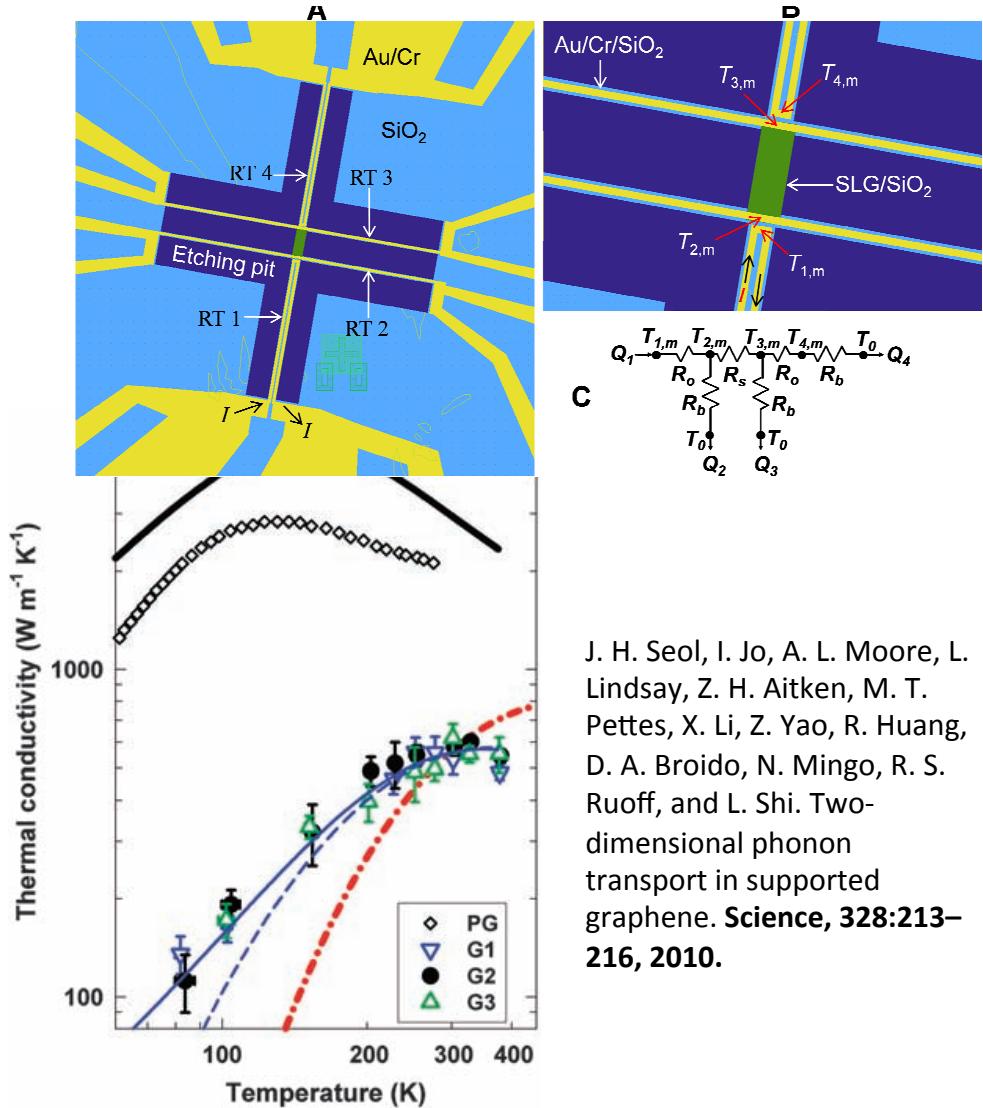
W. Jang, Z. Chen, W. Bao, C. N. Lau, and C. Dames.  
Thickness-dependent thermal conductivity of encased  
graphene and ultrathin graphene. **Nano Letters**,  
**10**:3909–3913, 2010.

# Steady state measurements of $\kappa$ 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 $\kappa$ of thin films

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

# Steady state vs. transient measurements of $\kappa$

**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 $\kappa$

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

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

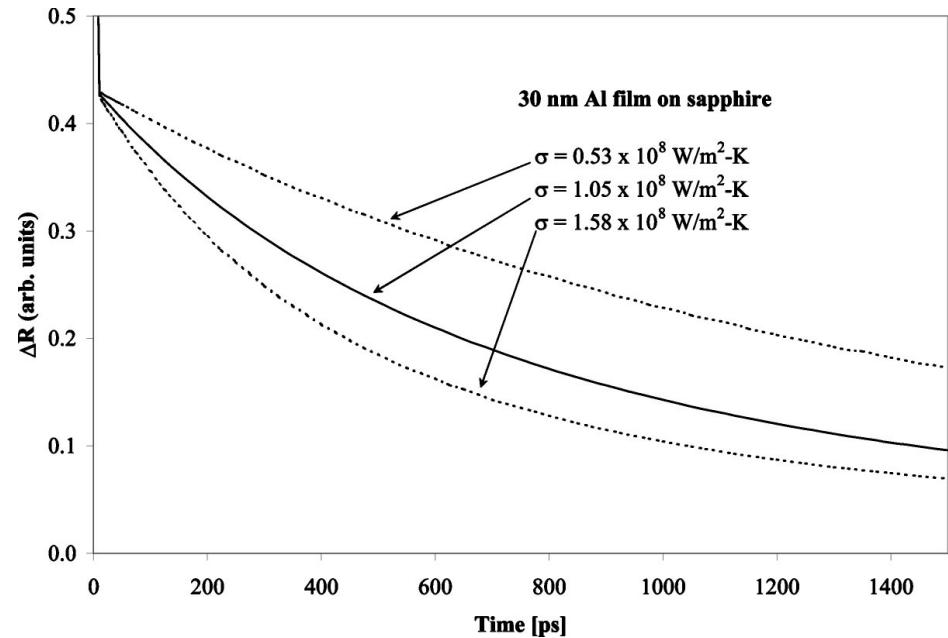


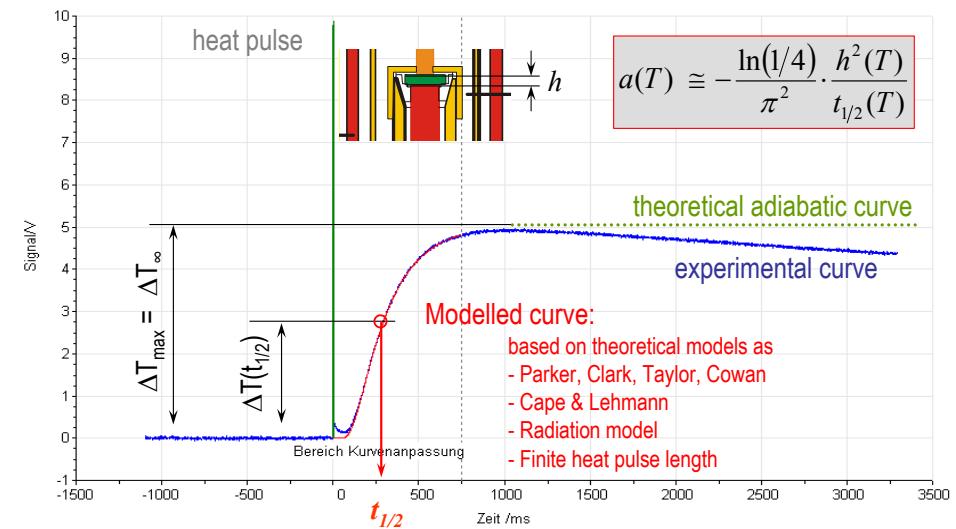
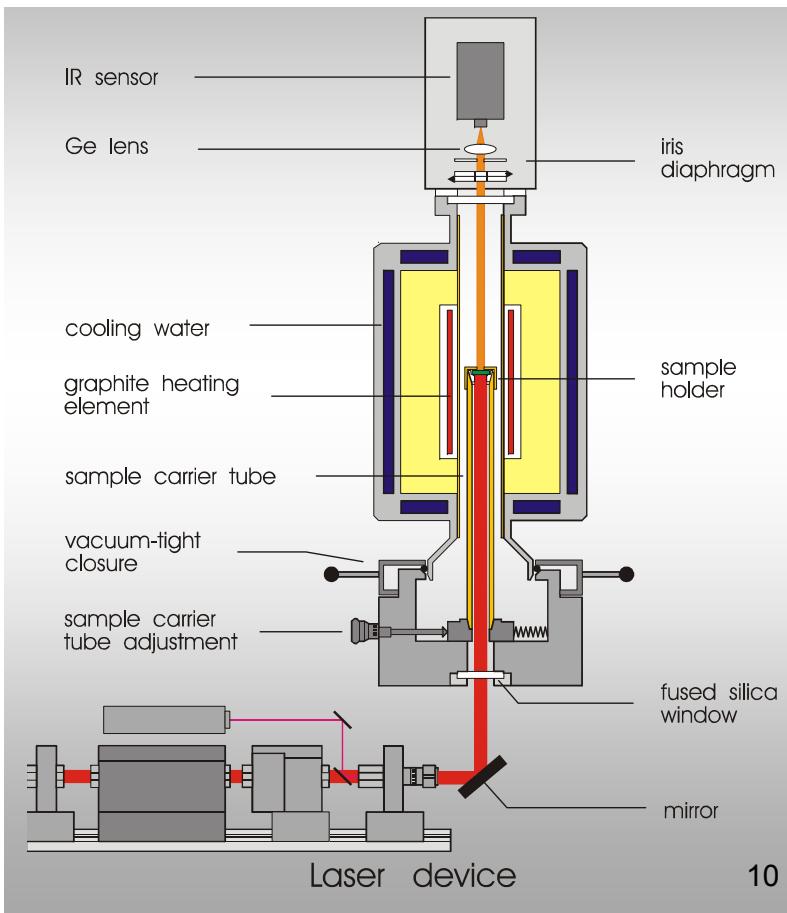
Fig. 2 Modeled thermal response of 30 nm Al film on a sapphire substrate with  $\sigma = 1.05 \times 10^{-8} \text{ W/m}^2\text{-K}$ . The dotted lines are the thermal response for the same film with  $\pm 50\%$  change in  $\sigma$ .

## 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 $\kappa$ in bulk systems

## Laser Flash

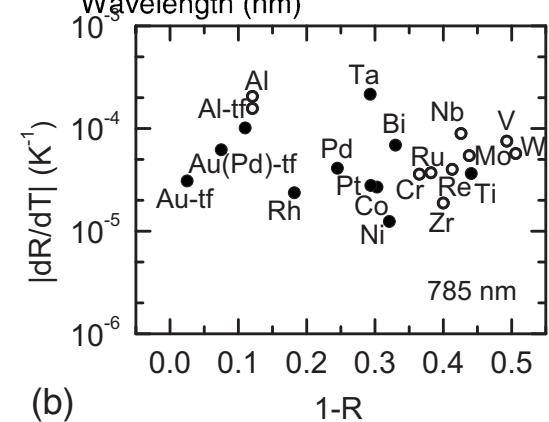
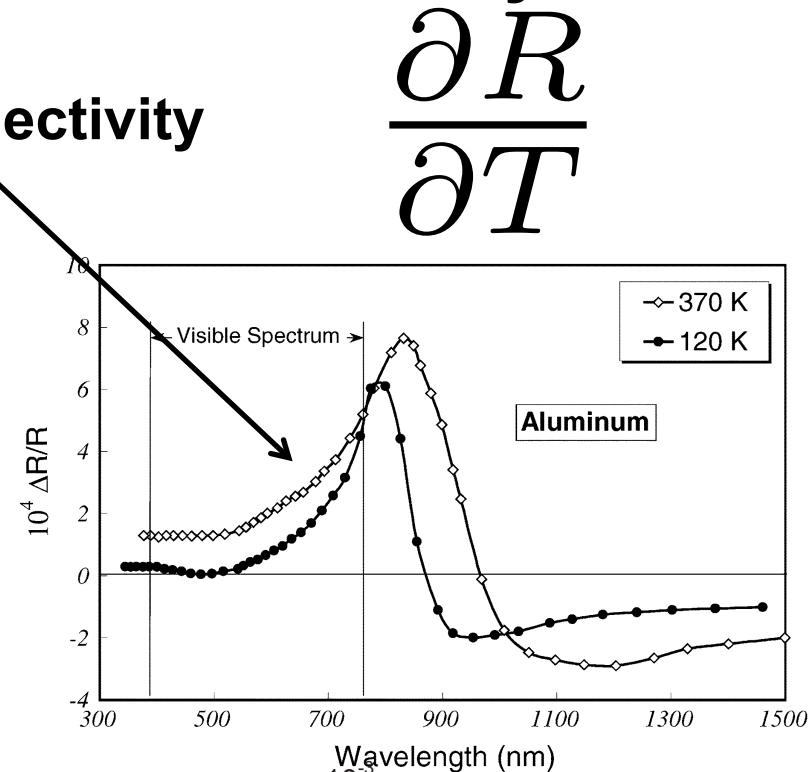
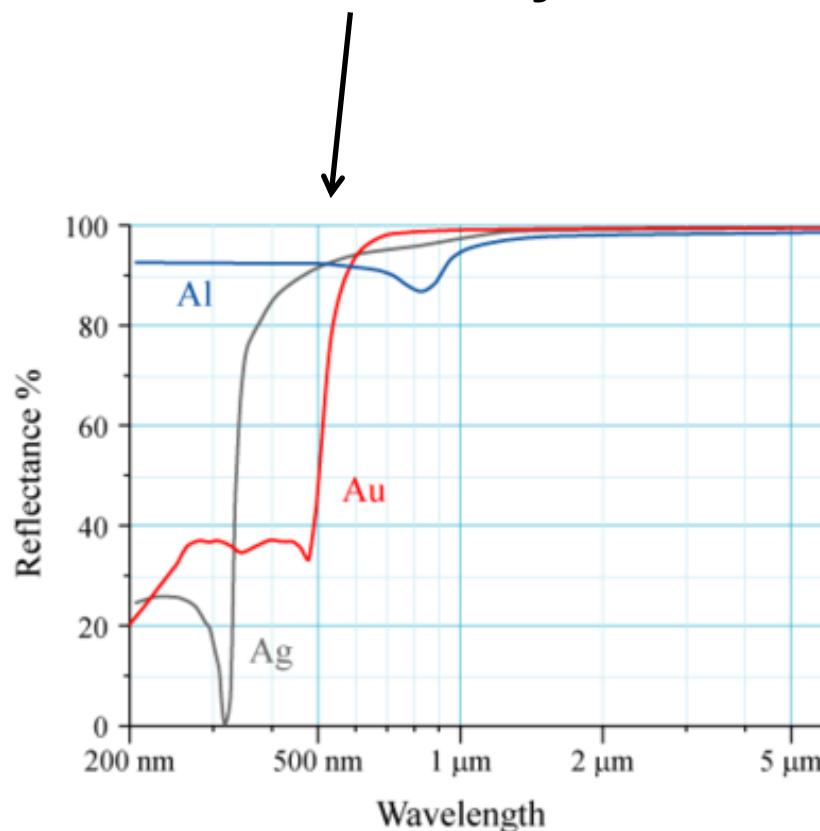


- Very dependent on surface emissivity
- Terrible sensitivity in nanosystems

# Transient measurements of $\kappa$ in nanosystems

## Turn to optics....Thermoreflectivity

### 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.

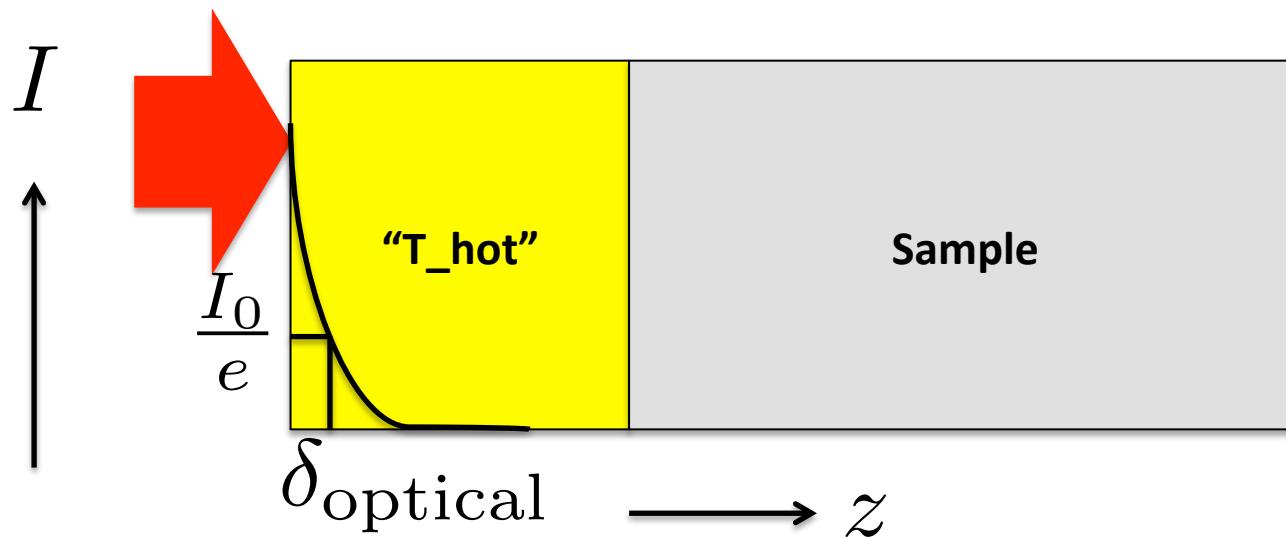
(b)

# Transient measurements of $\kappa$ in nanosystems

## 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”

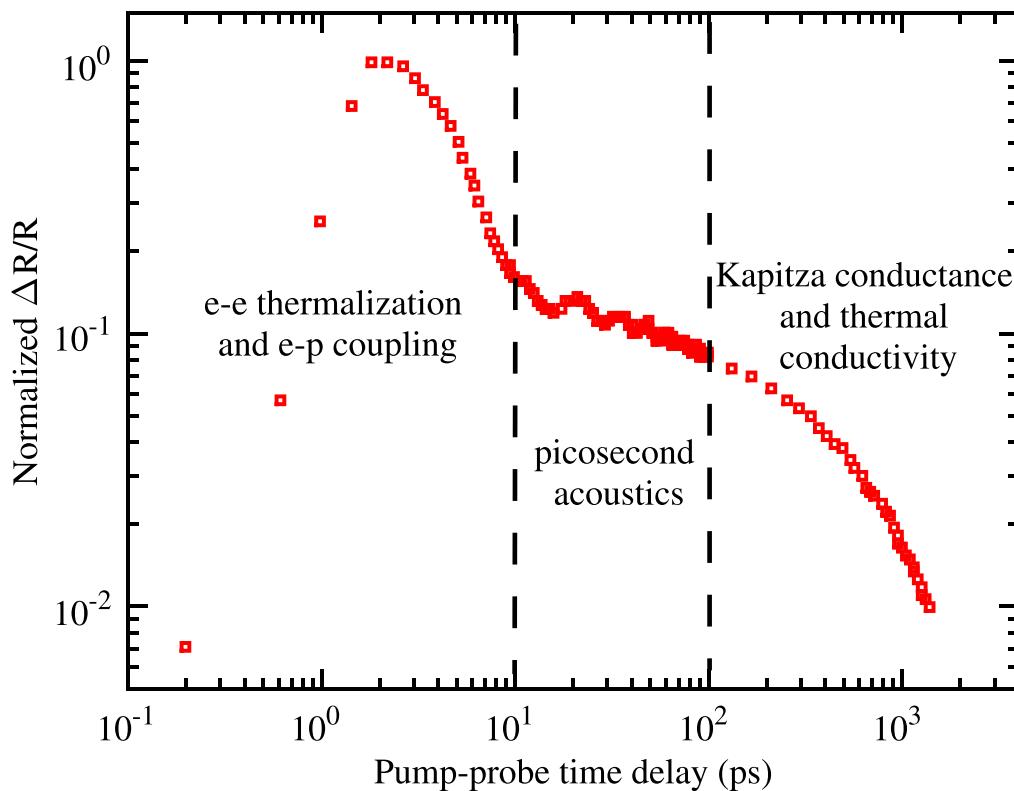
$$\delta_{\text{optical}} = \frac{\lambda}{4\pi k}$$



# Transient measurements of $\kappa$ in nanosystems

## 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. Szwejkowski, 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 $\kappa$ in nanosystems

**Temporal decay of single laser pulse can be used to measure  $\kappa$  of thin films**

**Pump-probe: nanosecond pump**

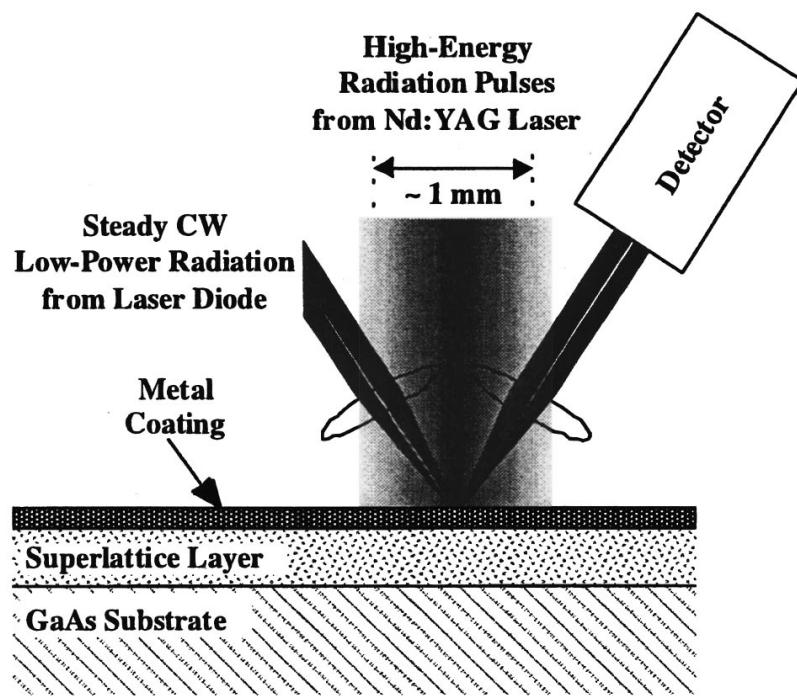


FIG. 1. The thermoreflectance method for measuring the vertical thermal resistance of  $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$  superlattice layers.

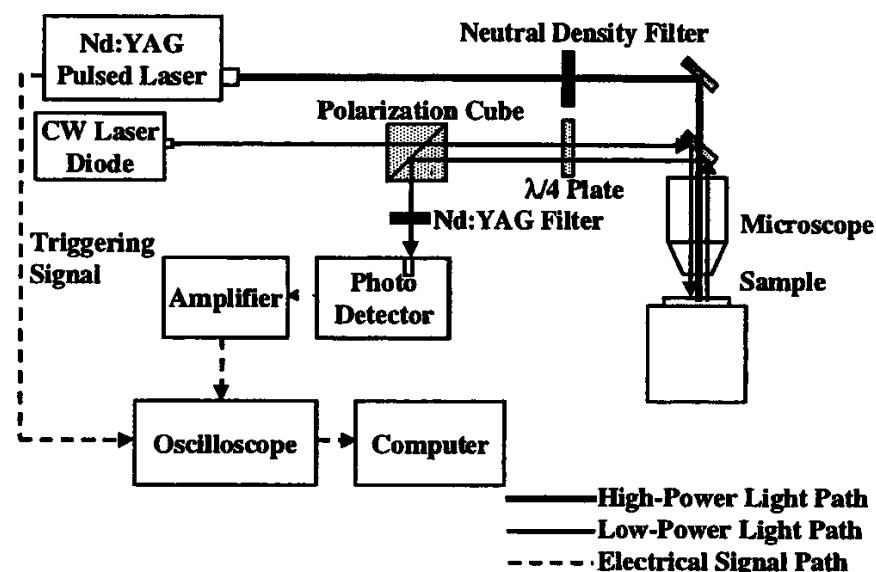
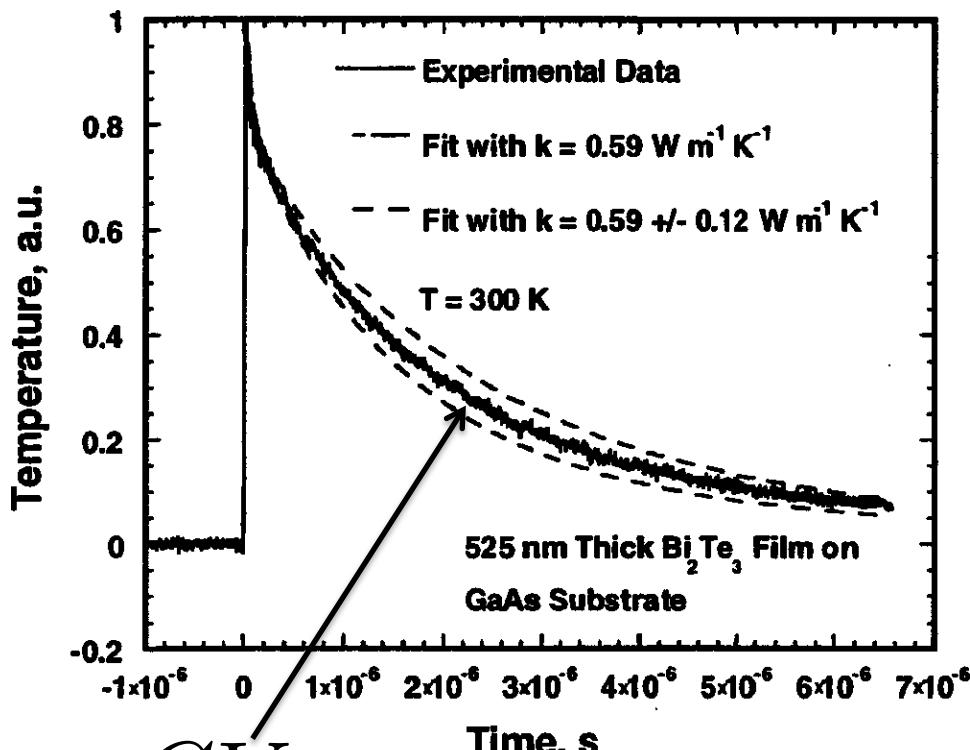


FIG. 2. Diagram describing the paths for radiation and electrical signals in the experimental setup.

# Transient measurements of $\kappa$ in nanosystems

**Temporal decay of single laser pulse can be used to measure  $\kappa$  of thin films**

**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

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

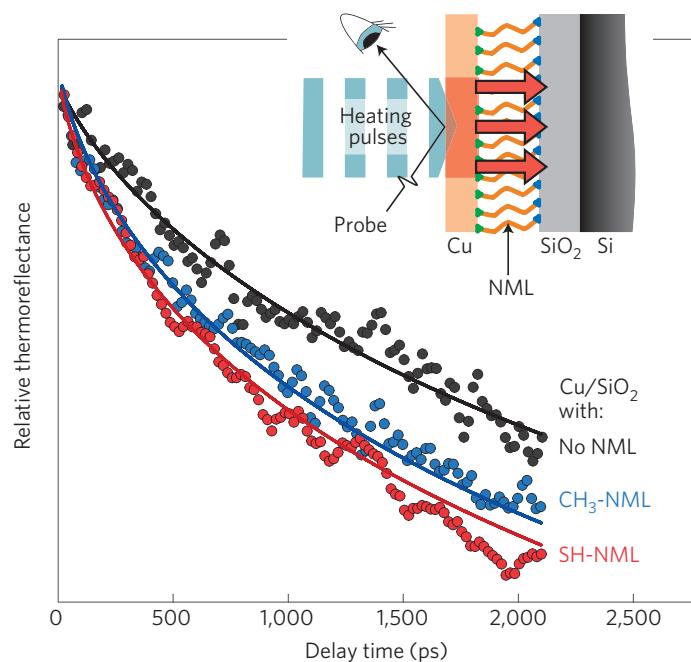
$$\delta = \sqrt{\frac{\tau \kappa}{C}}$$

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

# Transient measurements of $\kappa$ in nanosystems

## Temporal decay of single laser pulse can be used to measure $\kappa$ 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. Kebblinski, and G. Ramanath. Bonding-induced thermal conductance enhancement at inorganic heterointerfaces using nanomolecular monolayers. **Nature Materials**, 12:118–122, 2013.

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  - $3\omega$
  - Thermoreflectance-based techniques
    - FDTR
    - TDTR

# Steady state vs. transient vs. periodic measurements of $\kappa$

**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  
 $q(t)$  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

# Modulated measurements

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

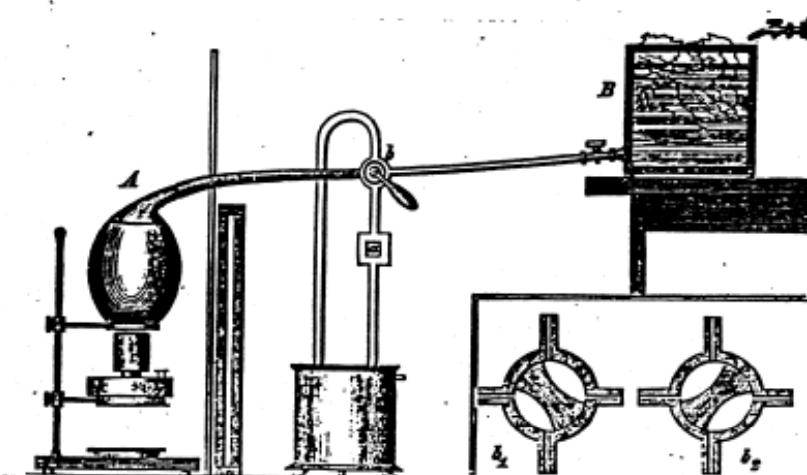
## Ångström method

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  $\Gamma$  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

# Frequency-domain measurements of $\kappa$ in microsystems

## Modified Ångström method

JOURNAL OF APPLIED PHYSICS

VOLUME 95, NUMBER 4

15 FEBRUARY 2004

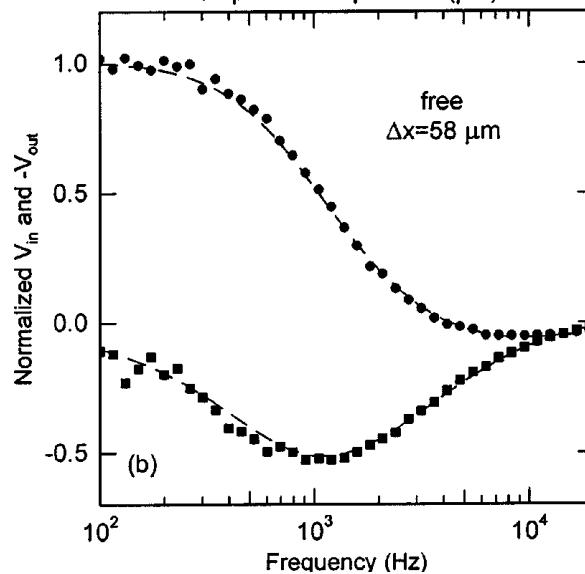
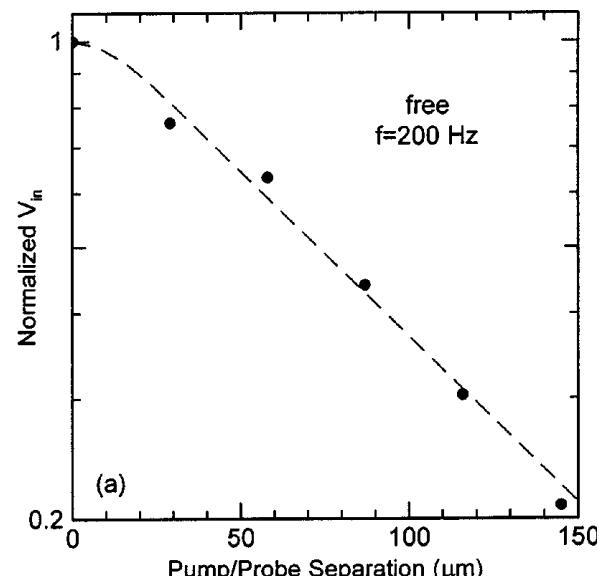
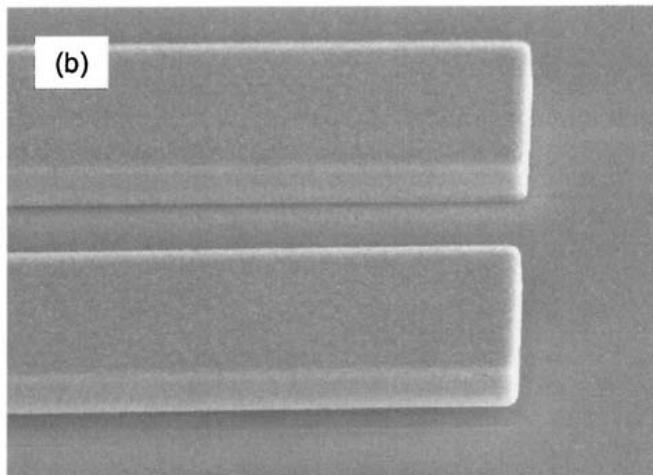
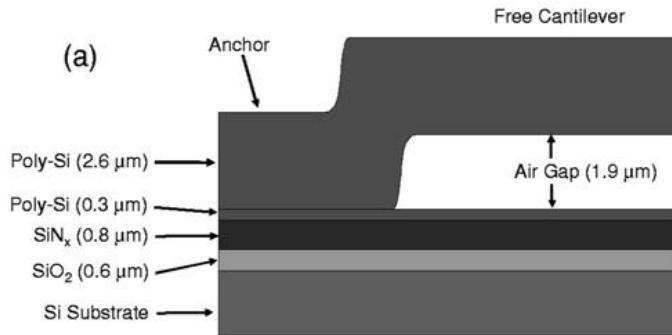
### Thermal contact conductance of adhered microcantilevers

Scott T. Huxtable<sup>a)</sup> 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<sup>b)</sup>

Department of Mechanical and Industrial Engineering, University of Illinois, Urbana, Illinois 61801



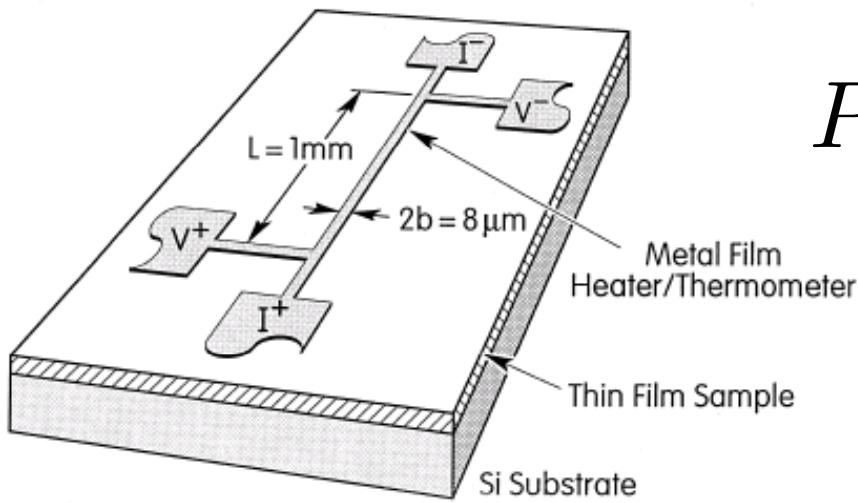
# Frequency-domain measurements of $\kappa$ in thin films

## 3 $\omega$ technique

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

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

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



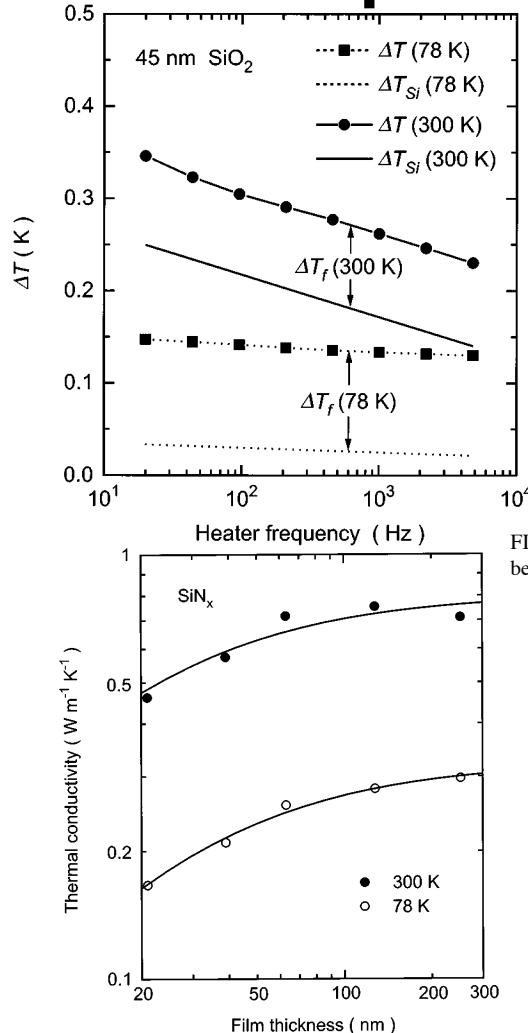
$$\Delta R \propto \exp [i2\omega t]$$

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

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

# Frequency-domain measurements of $\kappa$ in thin films

## 3 $\omega$ 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.

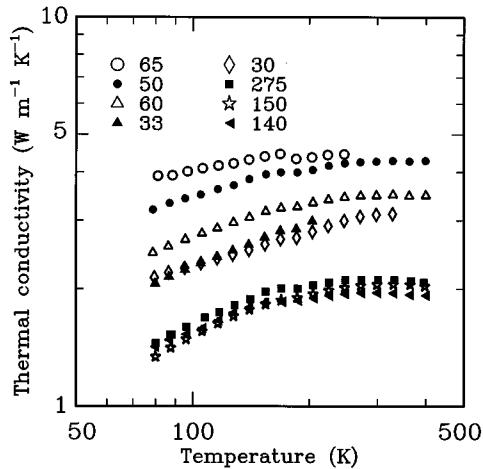
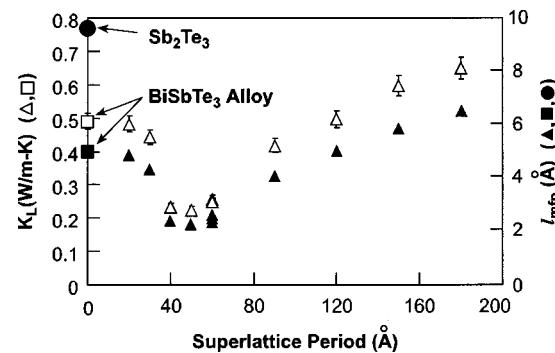
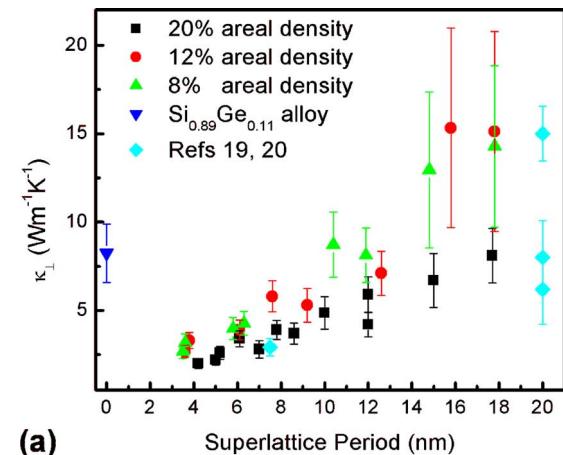
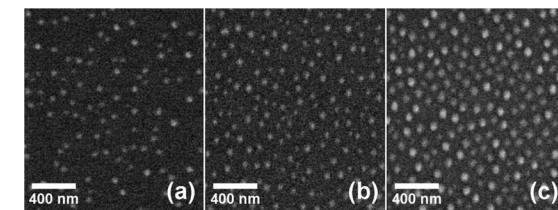


FIG. 2. Thermal conductivity of Si-Ge superlattices. Each symbol is labelled by the superlattice period  $L$  measured in Å.



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

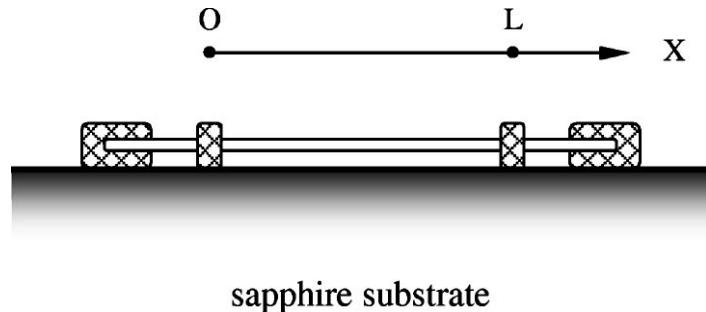
S. M. Lee, D. G. Cahill, and R. Venkatasubramanian. Thermal conductivity of Si-Ge superlattices. *Applied Physics Letters*, 70(22):2957–2959, 1997.



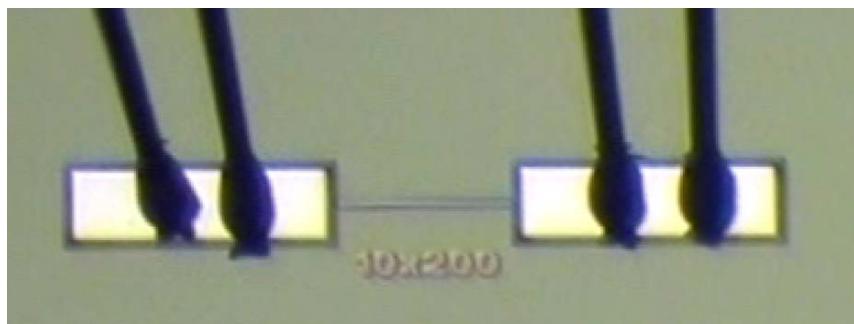
M. L. Lee and R. Venkatasubramanian. Effect of nanodot areal density and period on thermal conductivity in SiGe/Si nanodot superlattices. *Applied Physics Letters*, 92:053112, 2008.

# Frequency-domain measurements of $\kappa$ in thin films

## $3\omega$ 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  $3\omega$  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.

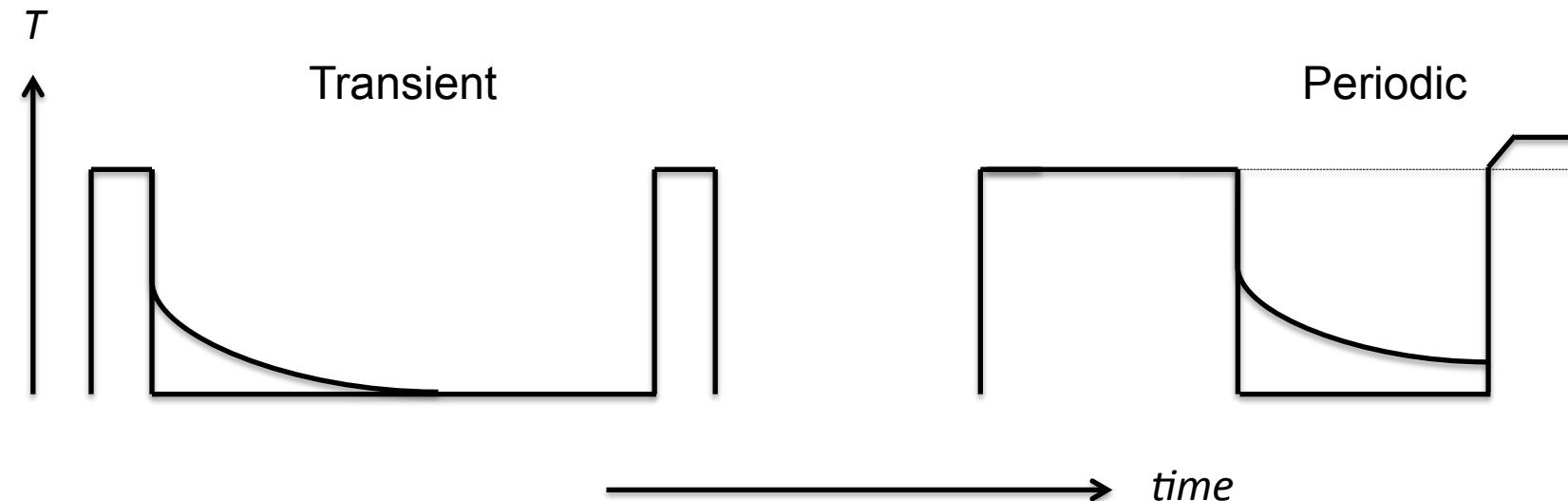
## What about optical periodic heating techniques?

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

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!!!!**

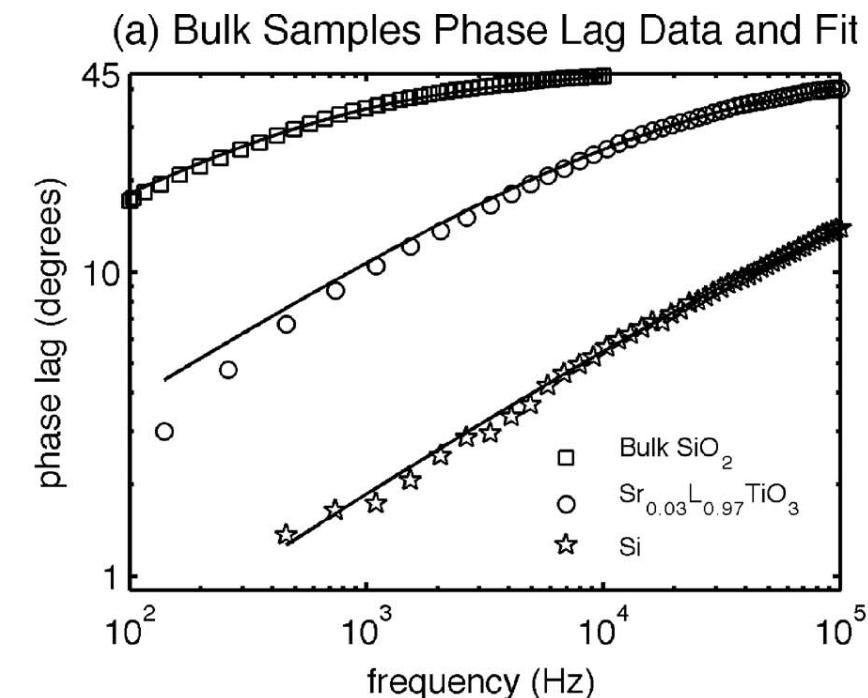
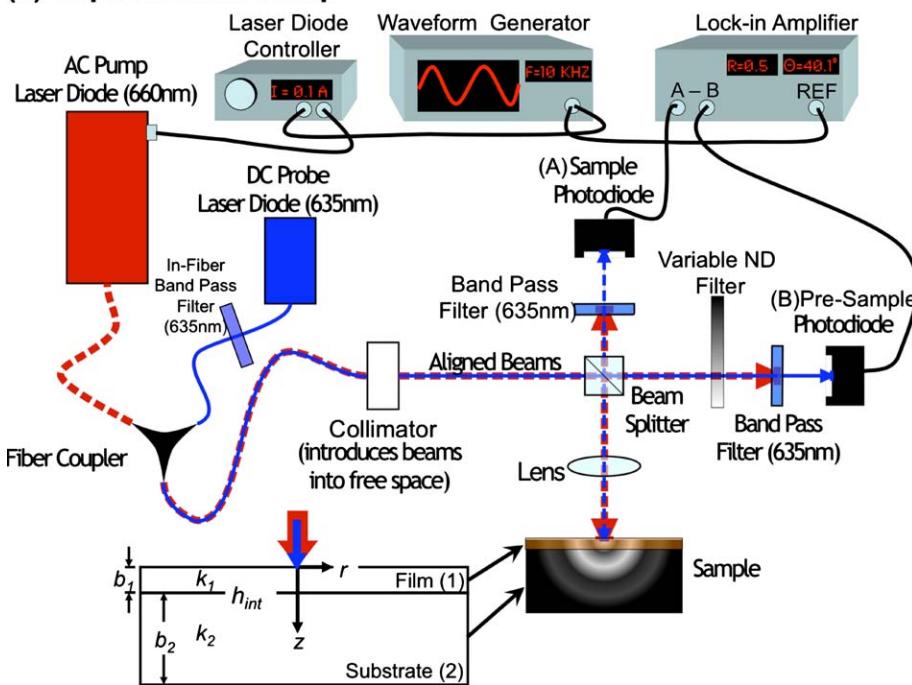
**NEED HEATING EVENT TO BE “FELT” BY NEXT HEATING EVENT**



# Frequency-domain measurements of $\kappa$ in thin films

## Continuous wave frequency domain thermoreflectance (FDTR)

### (a) Experimental Setup

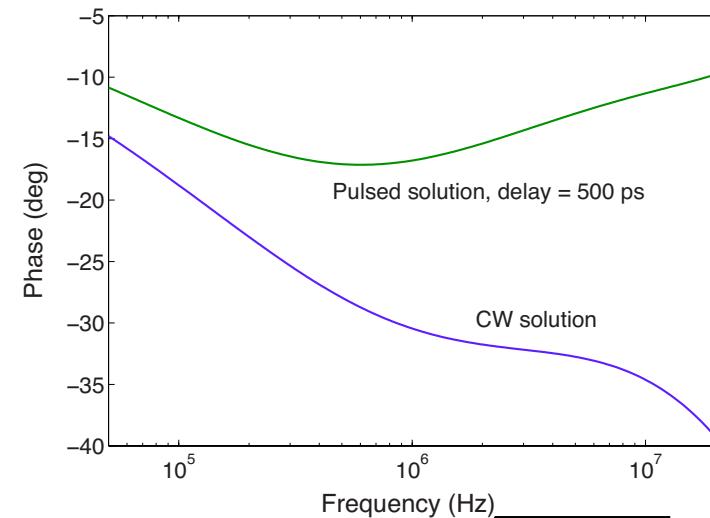
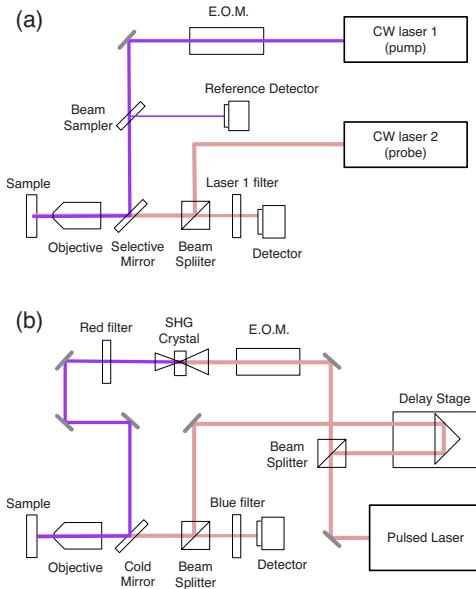


J. A. Malen, K. Baheti, T. Tong, Y. Zhao, J. A. Hudgings, and A. Majumdar. Optical measurement of thermal conductivity using fiber aligned frequency domain thermoreflectance. **Journal of Heat Transfer**, 133(8):081601, 2011.

# Frequency-domain measurements of $\kappa$ in thin films

## Continuous wave vs. pulsed frequency domain thermoreflectance (FDTR)

Different responses, and can resolve different thermal properties



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

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

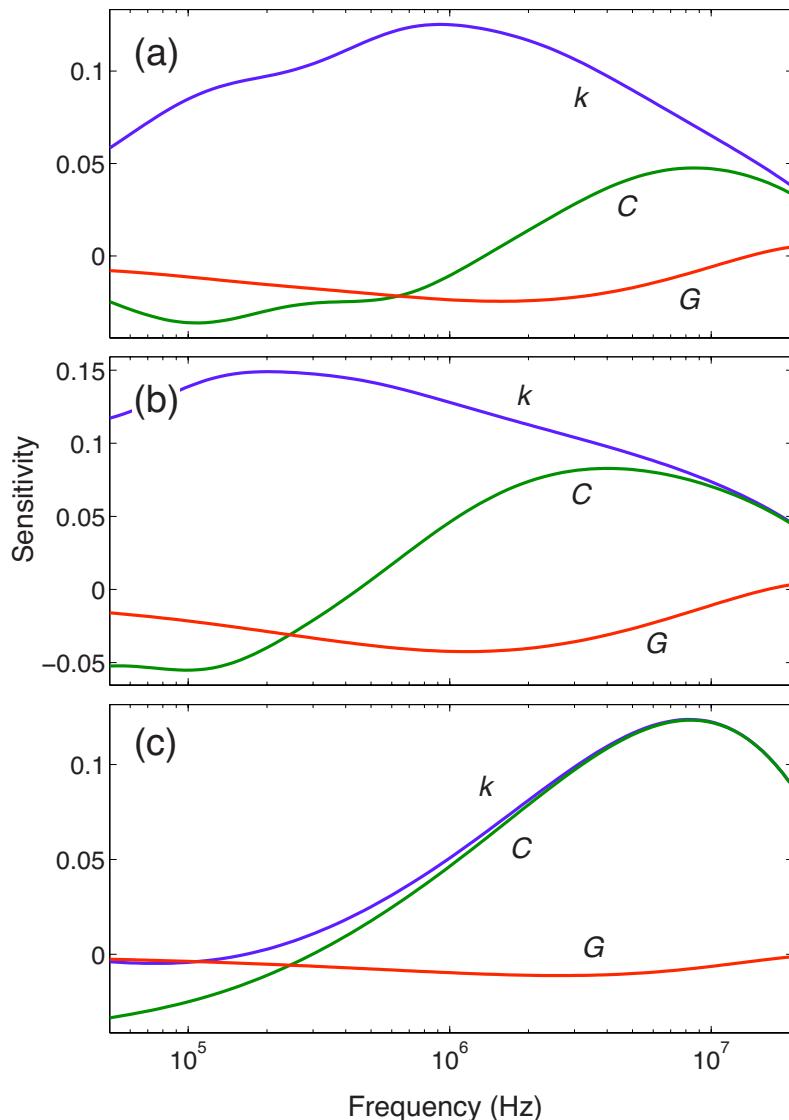
A. J. Schmidt, R. Cheaito, and M. Chiesa. A frequency-domain thermoreflectance method for the characterization of thermal properties. *Review of Scientific Instruments*, 80:094901, 2009.

### Advantage

Optical techniques can operate at higher modulation frequencies  
(smaller  $\delta$ , better resolution to “nano”)

# Frequency-domain measurements of $\kappa$ 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  $\kappa$ ?**

Pyrex ( $\text{SiO}_2$ )

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 $\kappa$ 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 thermoreflectance

Aaron J. Schmidt,<sup>1,2,a)</sup> Ramez Cheaito,<sup>2</sup> and Matteo Chiesa<sup>2</sup>

<sup>1</sup>Department of Mechanical Engineering, Massachusetts Institute of Technology Cambridge, Massachusetts, USA

<sup>2</sup>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)

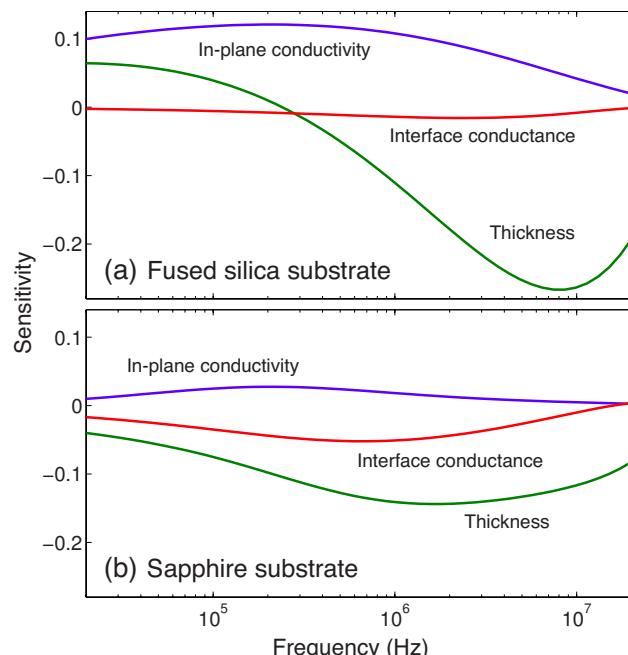


FIG. 5. (Color online) (a) The sensitivity parameter, Eq. (5), for in-plane thermal conductivity, metal-substrate boundary conductance, and metal thickness, for a sample consisting of an 80 nm film of Au deposited on a fused silica substrate. (b) The same sensitivities, calculated for a sapphire substrate.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

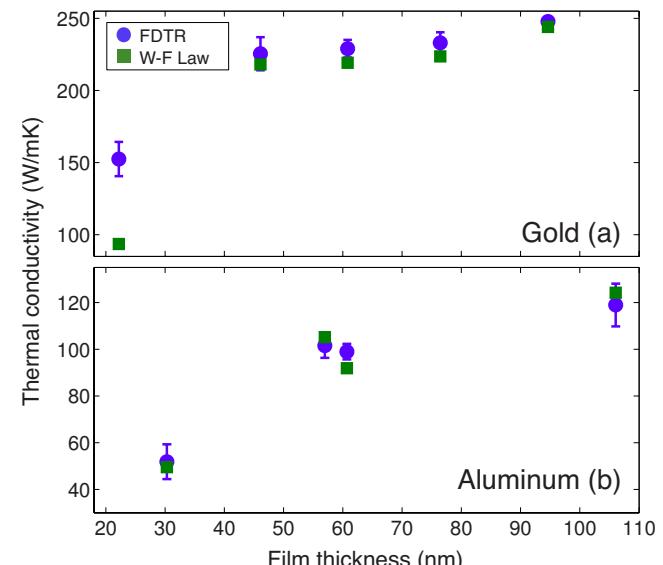
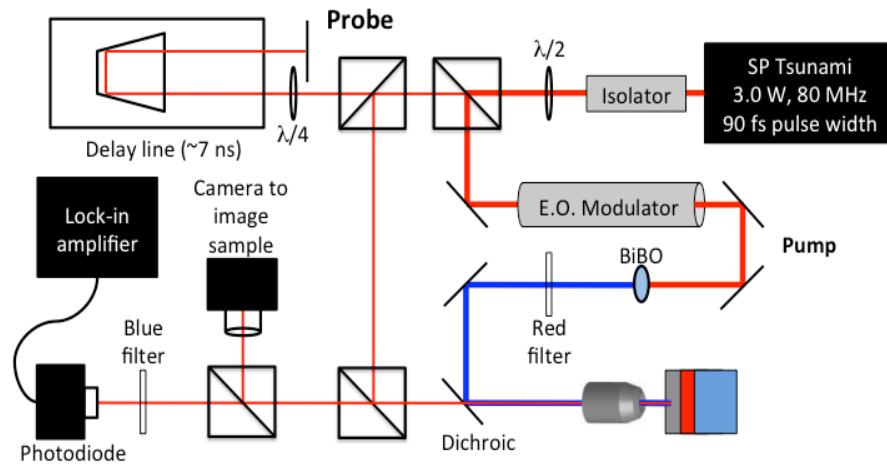


FIG. 4. (Color online) Thermal conductivity data obtained for (a) Au and (b) Al films on fused silica substrates. Circles are values obtained with the FDTR method, while the squares are values computed from electrical conductivity measurements using the WF law.

# Time domain thermoreflectance (TDTR)

Can we achieve transient and periodic thermometry in nanosystems?

Time domain thermoreflectance



Probe TDTR



Probe TDTR (moderate Duty cycle)



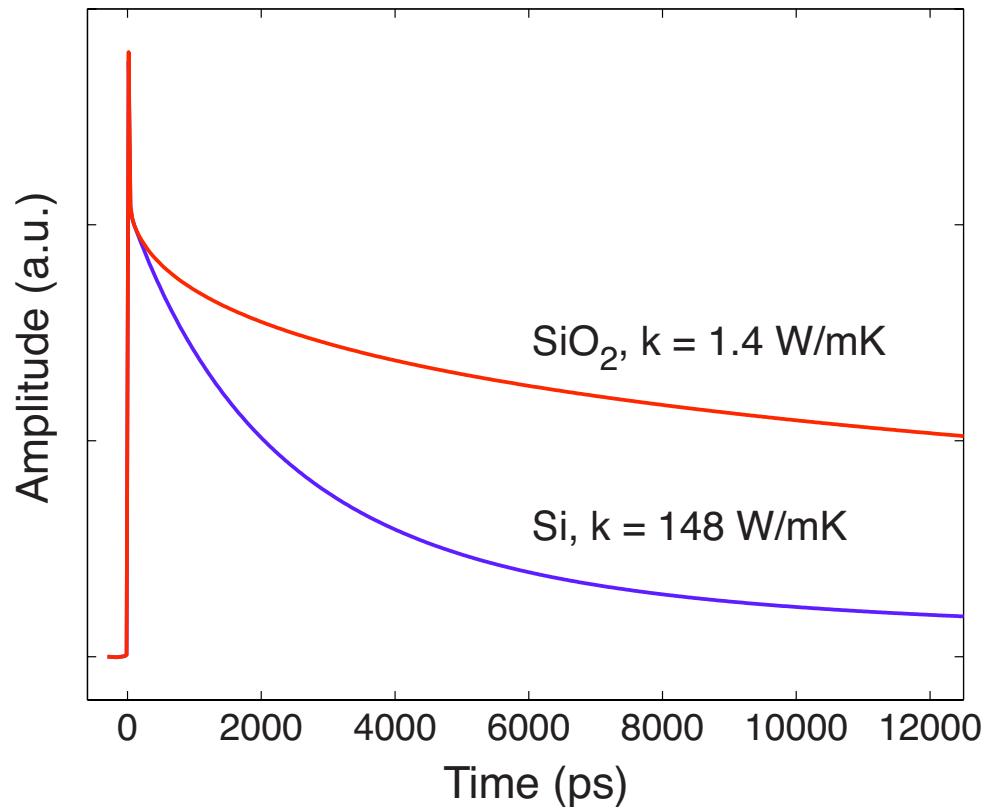
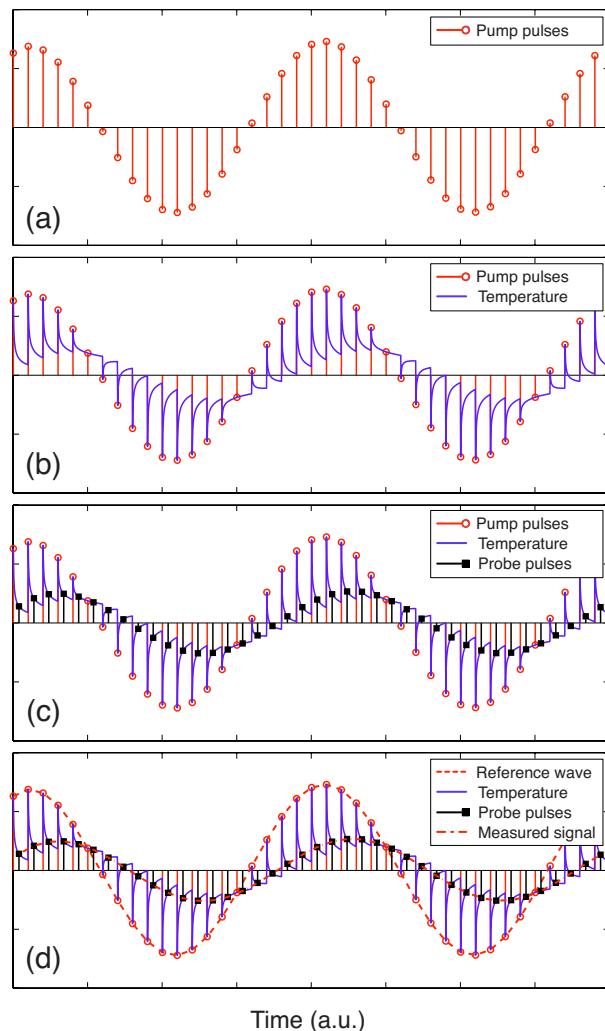
Probe TTR



Pump TTR (low Duty cycle)

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)

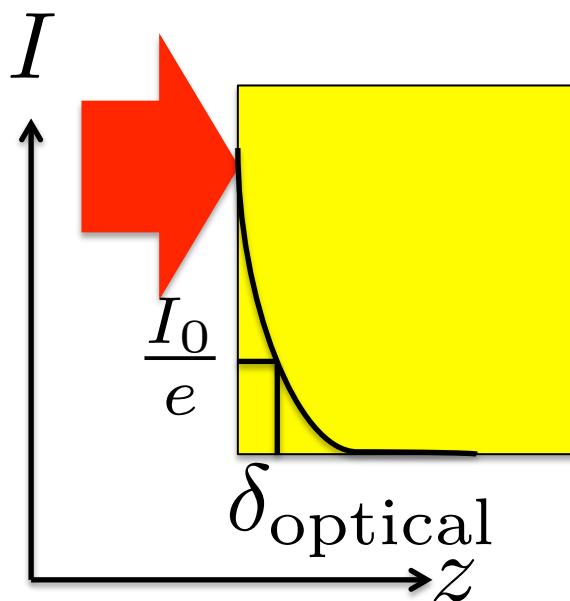


A. J. Schmidt, X. Chen, and G. Chen. Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance. **Review of Scientific Instruments**, 79:114902, 2008.

# Time domain thermoreflectance (TDTR)

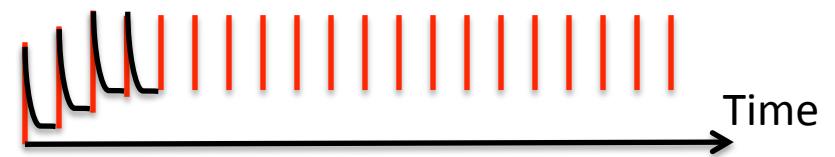
## Spatial regimes in TDTR

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

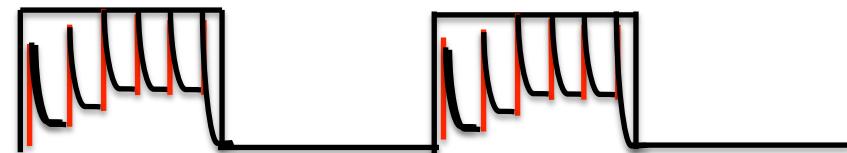


$$\delta_{\text{optical}} = \frac{\lambda}{4\pi k}$$

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



(accumulates due to 80 MHz rep. rate)



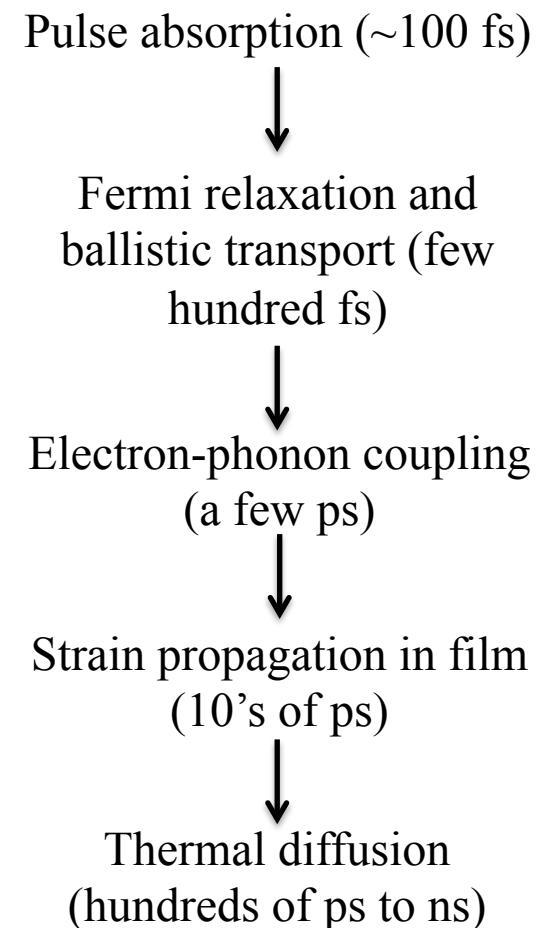
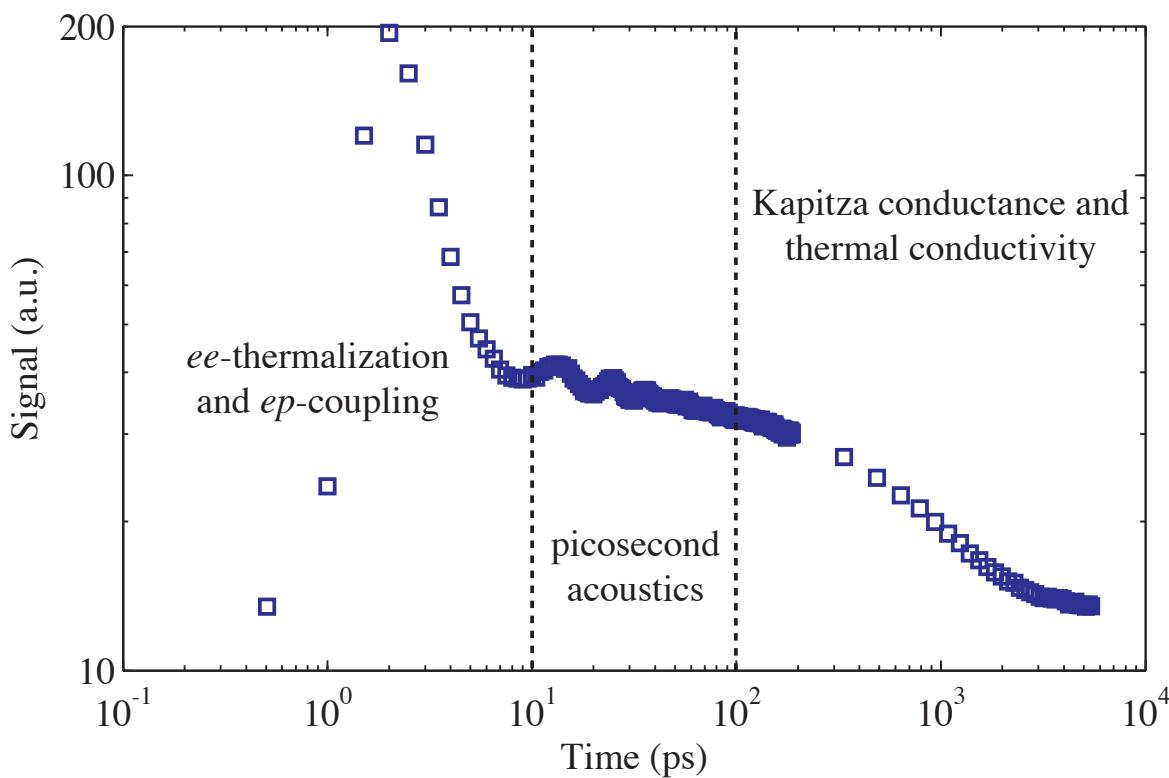
Accumulation leads to **MODULATED** heating event

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

# Time domain thermoreflectance (TDTR)

## Temporal regimes in TDTR

**FANTASTIC temporal resolution (limited by pulse width)**

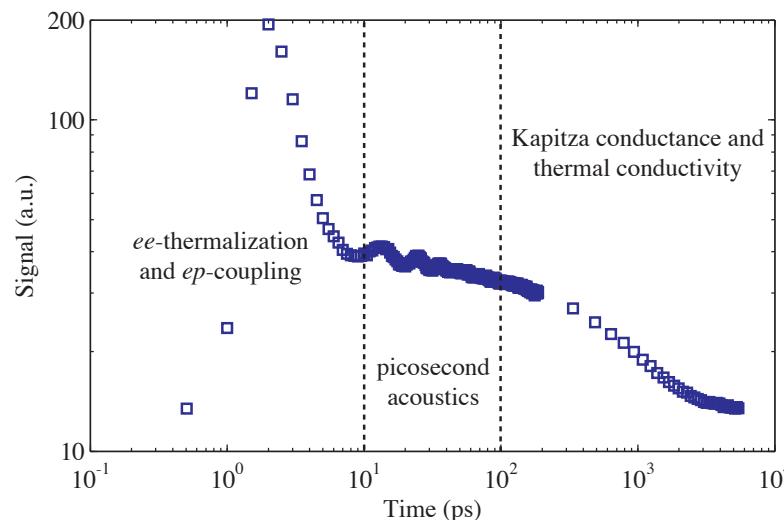


**Now let's look at a few specific examples....**

# Time domain thermoreflectance (TDTR)

## 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 SiO<sub>2</sub> 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 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

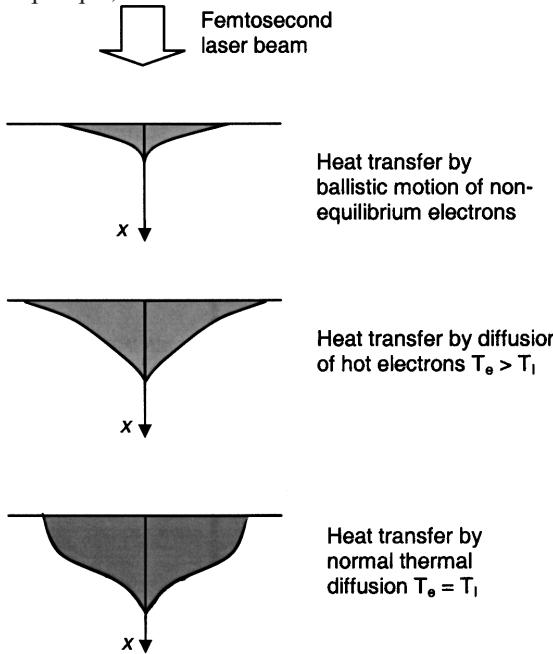
Patrick E. Hopkins

e-mail: [pehopki@sandia.gov](mailto: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

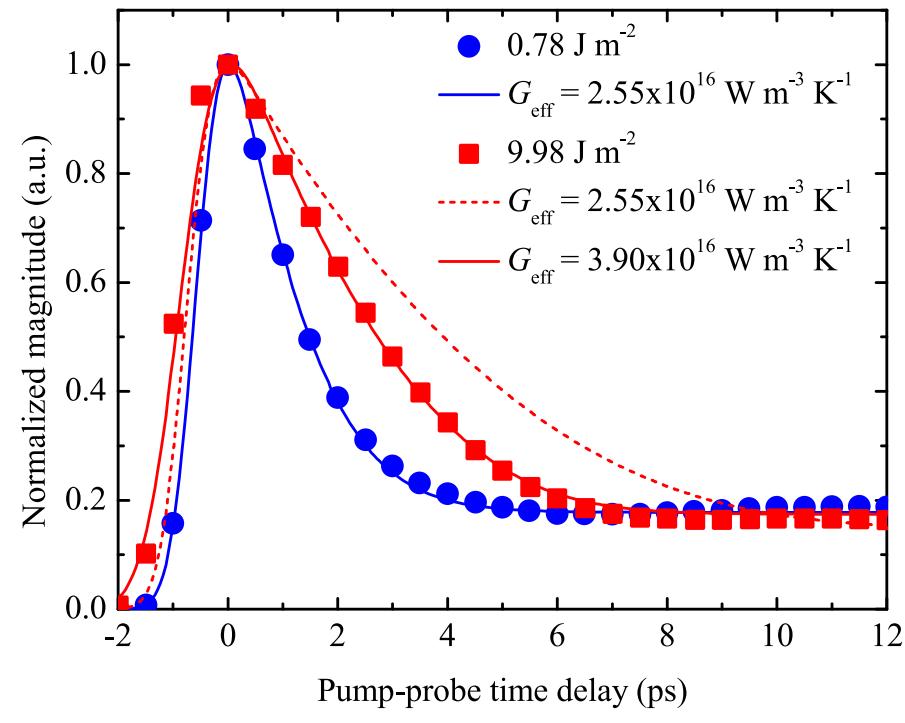
Patrick E. Hopkins,<sup>1,a)</sup> John C. Duda,<sup>1</sup> Bryan Kaehr,<sup>2,3</sup> Xiao Wang Zhou,<sup>4</sup> C.-Y. Peter Yang,<sup>4</sup> and Reese E. Jones<sup>4</sup>

<sup>1</sup>Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA

<sup>2</sup>Advanced Materials Laboratory, Sandia National Laboratories, Albuquerque, New Mexico 87106, USA

<sup>3</sup>Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA

<sup>4</sup>Sandia National Laboratories, Livermore, California 94550, USA



# Time domain thermoreflectance (TDTR)

## Electron-phonon scattering at interfaces



JOURNAL OF APPLIED PHYSICS 117, 105105 (2015)

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

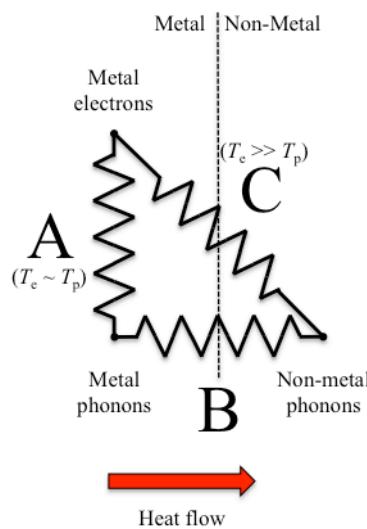
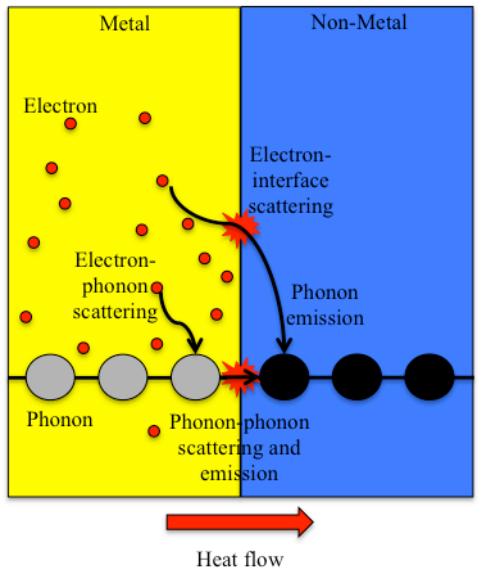
Ashutosh Giri,<sup>1</sup> John T. Gaskins,<sup>1</sup> Brian F. Donovan,<sup>1</sup> Chester Szwajkowski,<sup>1</sup>

Ronald J. Warzoha,<sup>2</sup> Mark A. Rodriguez,<sup>3</sup> Jon Ihlefeld,<sup>3</sup> and Patrick E. Hopkins<sup>1,a)</sup>

<sup>1</sup>Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA

<sup>2</sup>Department of Mechanical Engineering, United States Naval Academy, Annapolis, Maryland 21401, USA

<sup>3</sup>Sandia National Laboratories, Albuquerque, New Mexico 87123, USA

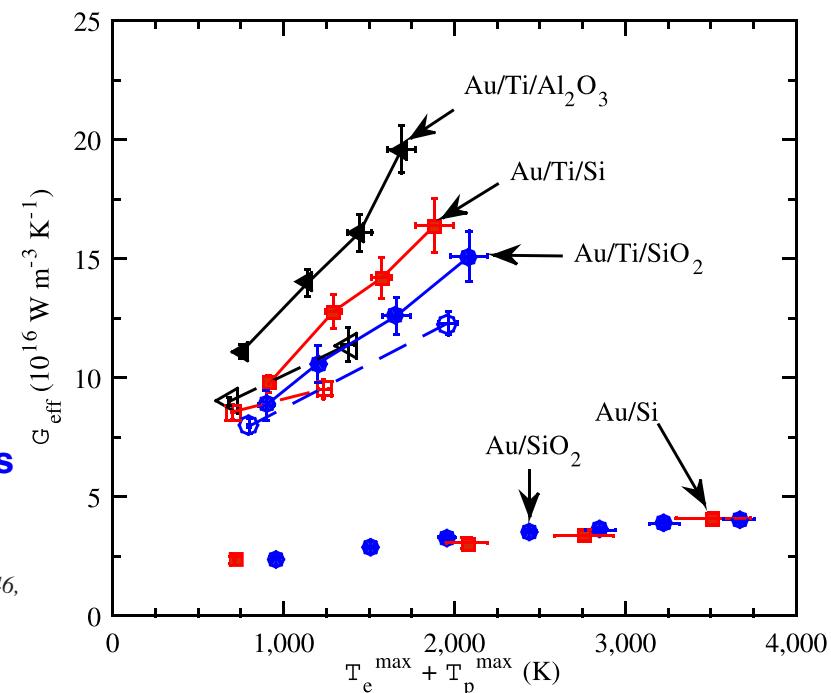


JOURNAL OF APPLIED PHYSICS 105, 023710 (2009)

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

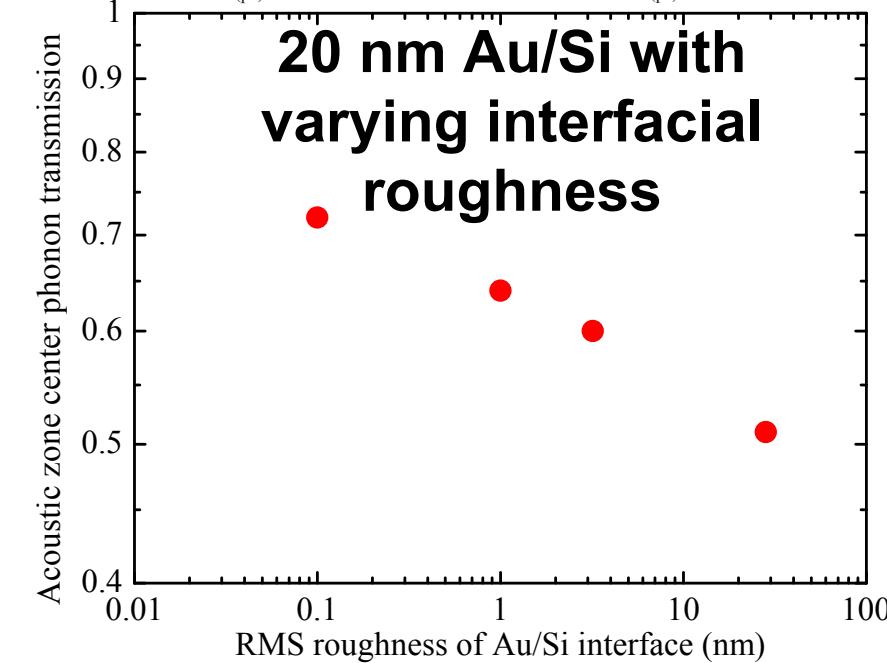
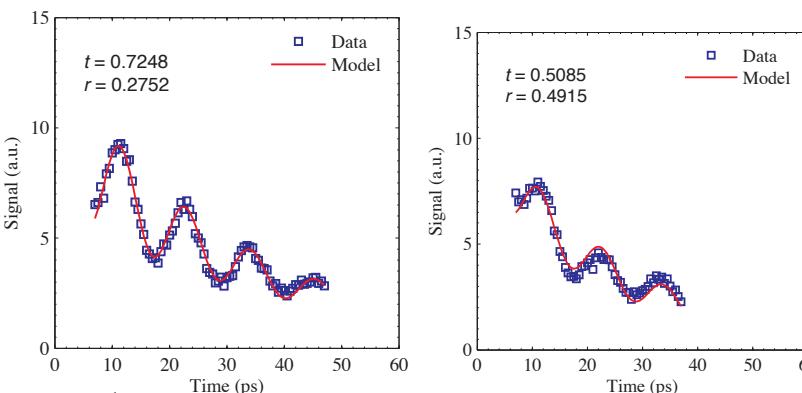
Patrick E. Hopkins,<sup>a)</sup> Jared L. Kassebaum, and Pamela M. Norris

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,<sup>1,a)</sup> C.-Y. P. Yang,<sup>2</sup> B. M. Foley,<sup>1</sup> R. Cheaito,<sup>1</sup> D. L. Medlin,<sup>2</sup> R. E. Jones,<sup>2</sup> and P. E. Hopkins<sup>1,b)</sup>

<sup>1</sup>Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA

<sup>2</sup>Sandia National Laboratories, Livermore, California 94550, USA

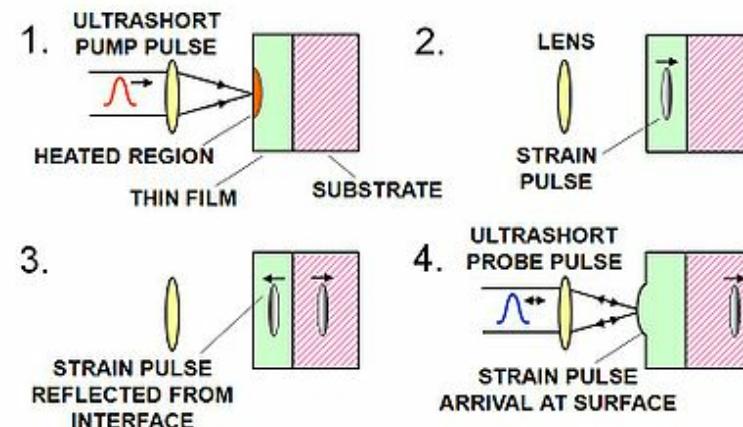


Image from Wikipedia: [http://en.wikipedia.org/wiki/Picosecond\\_ultrasonics](http://en.wikipedia.org/wiki/Picosecond_ultrasonics)

## 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,<sup>1</sup> John C. Duda,<sup>1,2</sup> Thomas E. Beechem,<sup>2</sup> Khalid Hattar,<sup>2</sup> Jon F. Ihlefeld,<sup>2</sup> Douglas L. Medlin,<sup>3</sup> Mark A. Rodriguez,<sup>2</sup> Michael J. Campion,<sup>2,4</sup> Edward S. Piekos,<sup>2</sup> and Patrick E. Hopkins<sup>1,\*</sup>

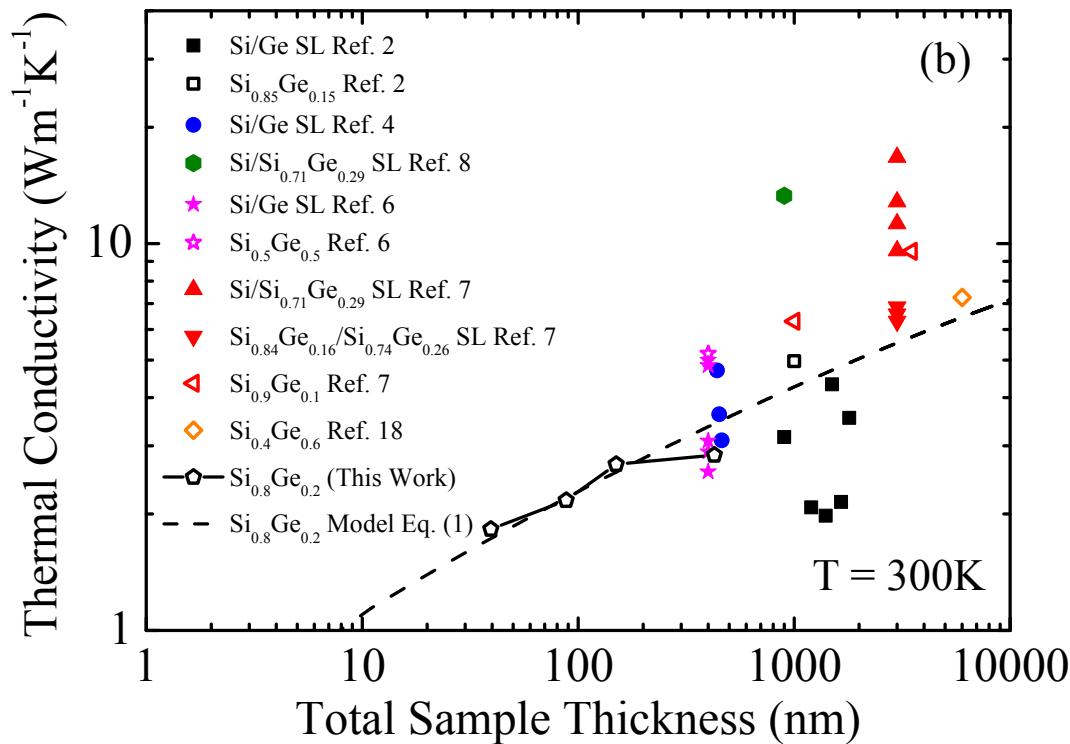
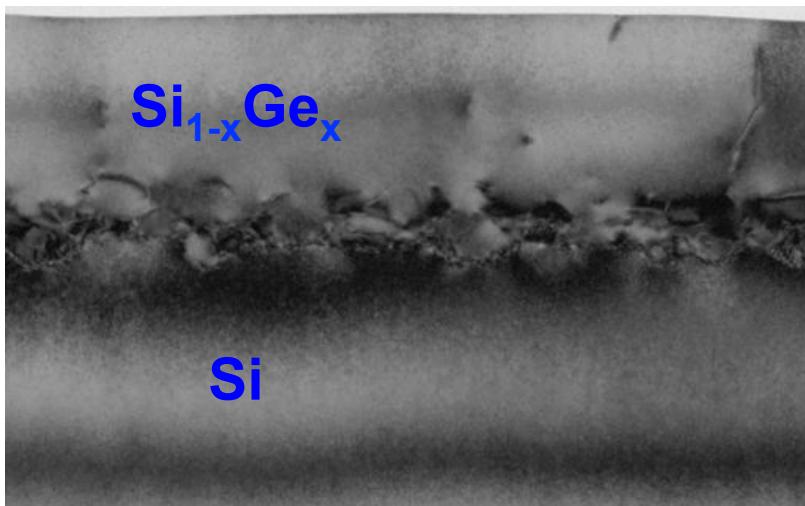
<sup>1</sup>Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA

<sup>2</sup>Sandia National Laboratories, Albuquerque, New Mexico 87123, USA

<sup>3</sup>Sandia National Laboratories, Livermore, California 94550, USA

<sup>4</sup>Massachusetts Institute of Technology, Department of Material Science and Engineering, Cambridge, Massachusetts 02139, USA

(Received 1 June 2012; published 8 November 2012)

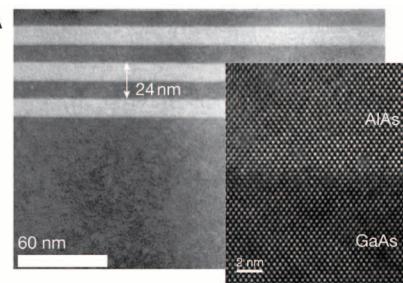


# Time domain thermoreflectance (TDTR)

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

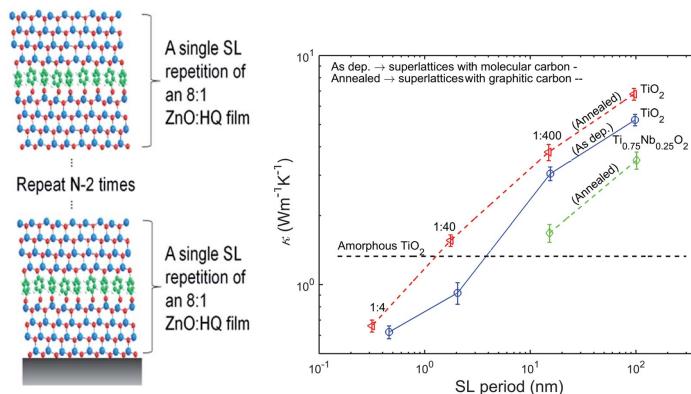
# Coherent Phonon Heat Conduction in Superlattices

Maria N. Luckyanova,<sup>1\*</sup> Jivtesh Garg,<sup>1\*</sup> Keivan Esfarjani,<sup>1</sup> Adam Jandl,<sup>2</sup> Mayank T. Bulsara,<sup>2</sup> Aaron J. Schmidt,<sup>3</sup> Austin J. Minnich,<sup>4</sup> Shuo Chen,<sup>5</sup> Mildred S. Dresselhaus,<sup>6,7</sup> Zhifeng Ren,<sup>8</sup> Eugene A. Fitzgerald,<sup>2</sup> Gang Chen<sup>1†</sup>



## Ultra-low thermal conductivity in $\text{TiO}_2:\text{C}$ superlattices

Janne-Petteri Niemelä,<sup>a</sup> Ashutosh Giri,<sup>b</sup> Patrick E. Hopkins<sup>b</sup> and Maarit Karppinen<sup>\*a</sup>



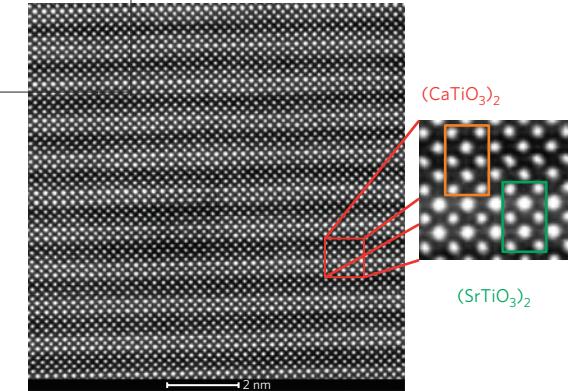
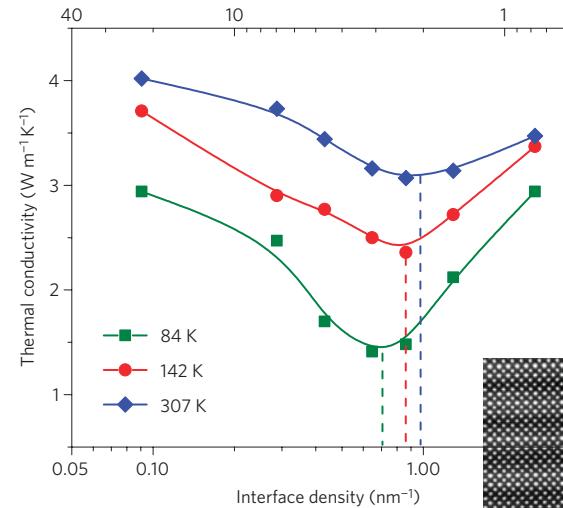
## LETTERS

PUBLISHED ONLINE: 8 DECEMBER 2013 | DOI:10.1038/NMAT382

nature  
materials

# Crossover from incoherent to coherent phonon scattering in epitaxial oxide superlattices

Jayakanth Ravichandran<sup>1,2††</sup>, Ajay K. Yadav<sup>2,3‡</sup>, Rameez Cheaito<sup>4‡</sup>, Pim B. Rossen<sup>3</sup>, Arsen Soukiasian<sup>5</sup>, S. J. Suresha<sup>2</sup>, John C. Duda<sup>4</sup>, Brian M. Foley<sup>4</sup>, Che-Hui Lee<sup>5</sup>, Ye Zhu<sup>6</sup>, Arthur W. Lichtenberger<sup>7</sup>, Joel E. Moore<sup>2,8</sup>, David A. Muller<sup>6,9</sup>, Darrell G. Schlom<sup>5,9</sup>, Patrick E. Hopkins<sup>4</sup>, Arun Majumdar<sup>10</sup>, Ramamoorthy Ramesh<sup>1,2,3,8,11\*</sup> and Mark A. Zurbuchen<sup>12,13,14\*</sup>  
 Period thickness (nm)



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

# Time domain thermoreflectance (TDTR)

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

Thin Solid Films 520 (2012) 6109–6117

APPLIED PHYSICS LETTERS 105, 082907 (2014)



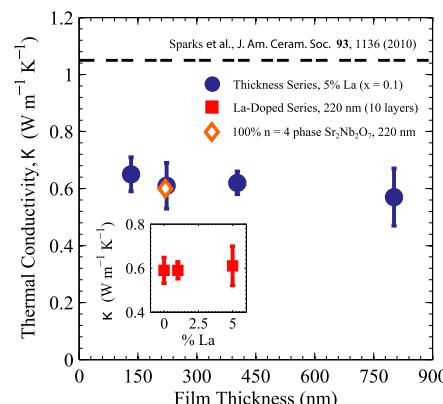
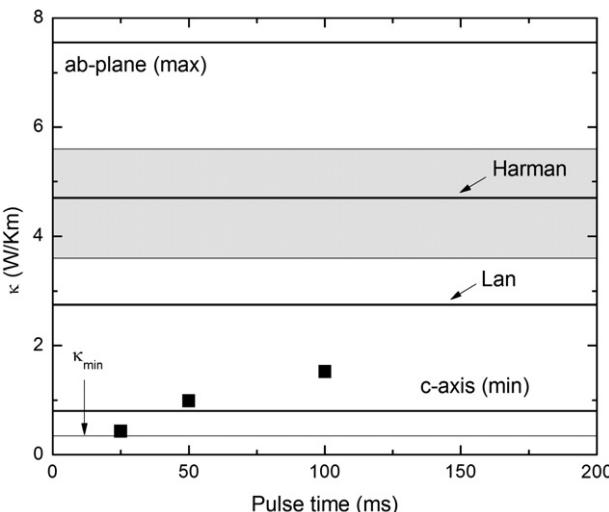
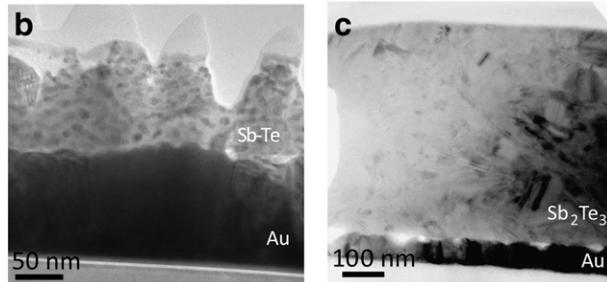
Electrodeposition and characterization of nano-crystalline antimony telluride thin films

J.L. Lensch-Falk<sup>a</sup>, D. Banga<sup>a</sup>, P.E. Hopkins<sup>b,c</sup>, D.B. Robinson<sup>a</sup>, V. Stavila<sup>a</sup>, P.A. Sharma<sup>b</sup>, D.L. Medlin<sup>a,\*</sup>

<sup>a</sup> Sandia National Laboratories, Livermore, CA 94550, USA

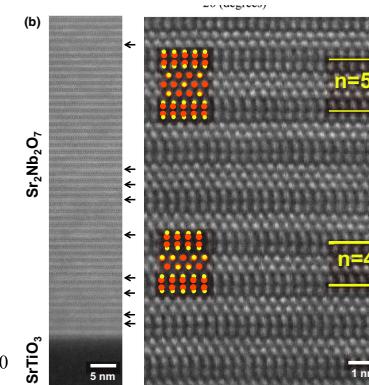
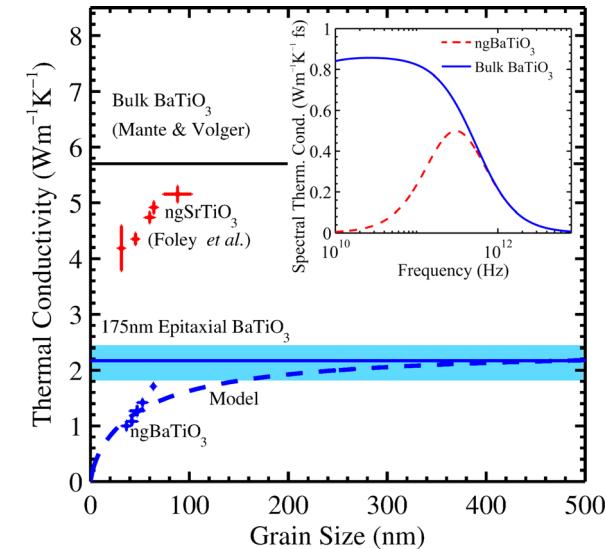
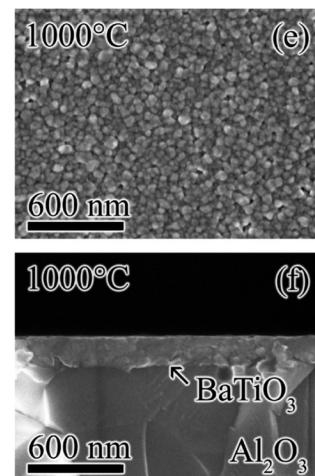
<sup>b</sup> Sandia National Laboratories, Albuquerque, NM 87185, USA

<sup>c</sup> 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,<sup>1</sup> Brian M. Foley,<sup>2</sup> Jon F. Ihlefeld,<sup>3,a)</sup> Jon-Paul Maria,<sup>4</sup> and Patrick E. Hopkins<sup>2,b)</sup>

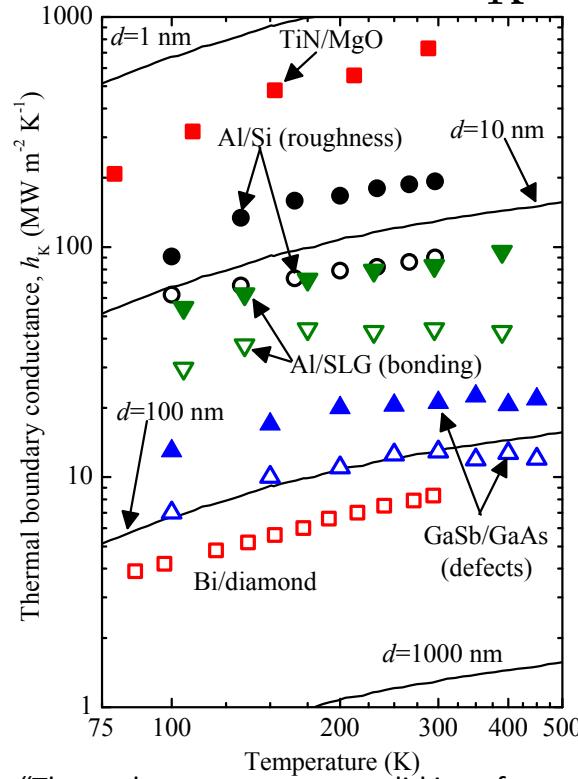


Journal of the American Ceramic Society, 98:624–628, 2015

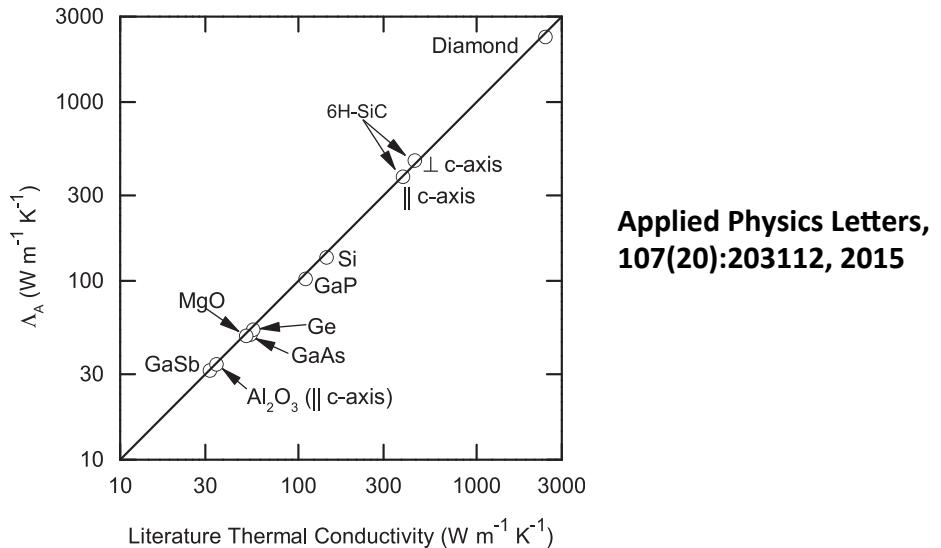
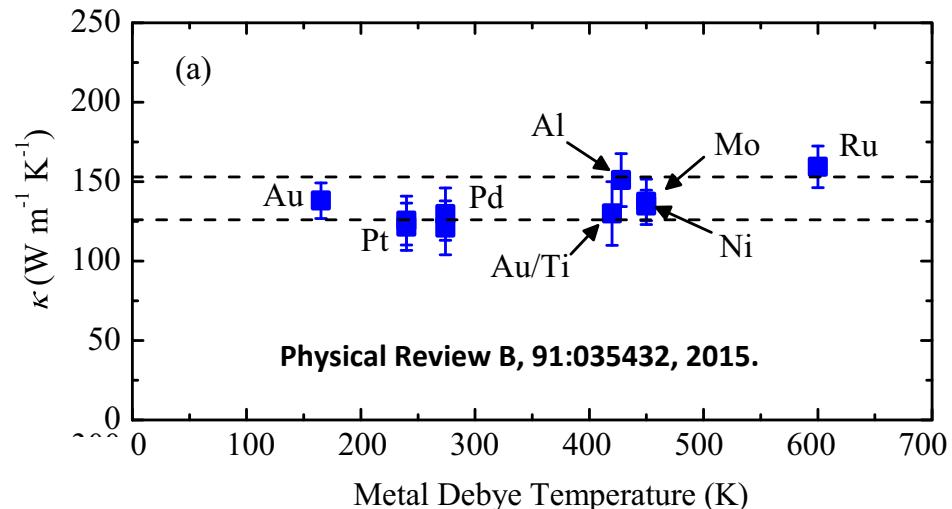
# Time domain thermoreflectance (TDTR)

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

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

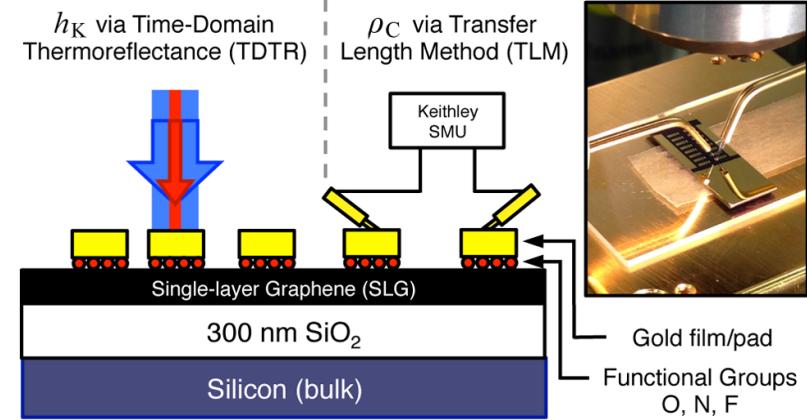
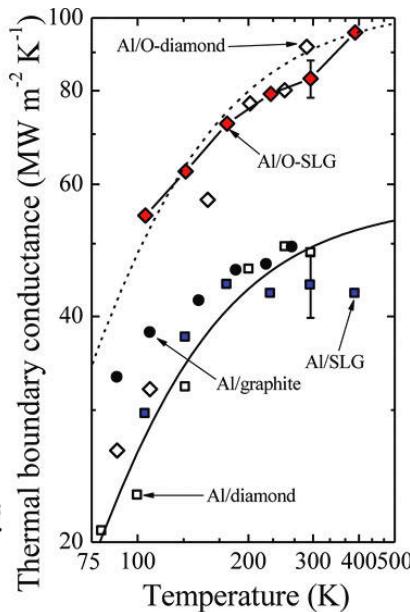
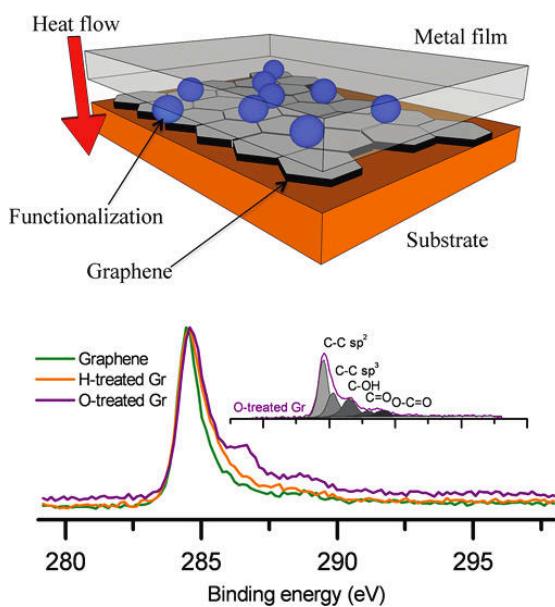


Hopkins, "Thermal transport across solid interfaces with nanoscale imperfections: Effects of roughness, disorder, dislocations and bonding on thermal boundary conductance," *ISRN Mechanical Engineering* 2013, 682586 (2013)



# Time domain thermoreflectance (TDTR)

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



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.

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.

# 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!

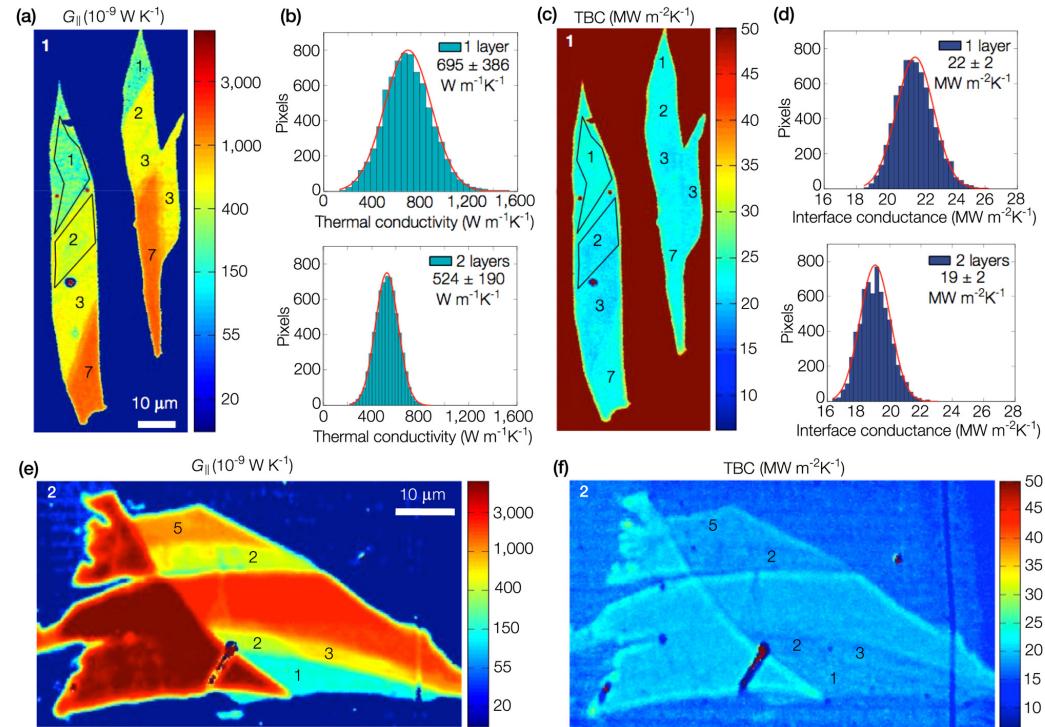
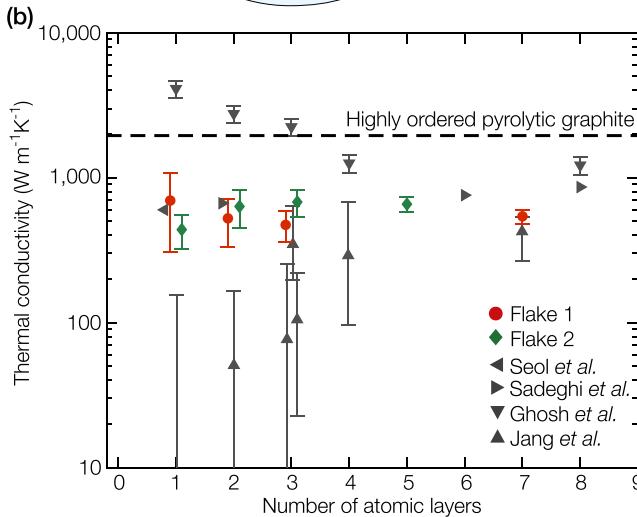
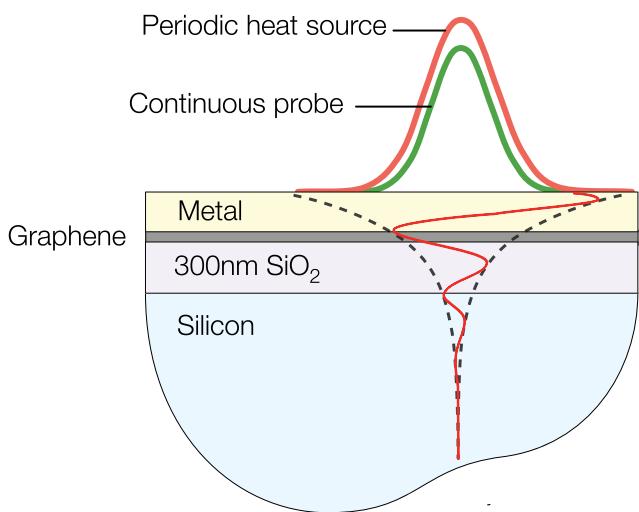


FIG. 8. Thermal conductance maps of the two samples. (a)  $G_{\parallel}$  map of flake 1. (b) Histograms of the thermal conductivity for single-layer graphene and bi-layer graphene analyzed from the polygons in (a). (c) TBC map of flake 1. (d) Histograms of TBC of single-layer graphene and bi-layer graphene, analyzed from the polygons in (c). (e)  $G_{\parallel}$  map of flake 2. (f) TBC map of flake 2. The upper limit of the color bars for (c) is set at  $50 \text{ MW m}^{-2} \text{ K}^{-1}$  to highlight the graphene flake, although the measured value of TBC for Au/Ti/SiO<sub>2</sub> for this sample was closer to  $100 \text{ MW m}^{-2} \text{ K}^{-1}$ . 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 thermoreflectance (TDTR)

## Highly porous/non-conformal films/liquids

### Minimum thermal conductivity considerations in aerogel thin films

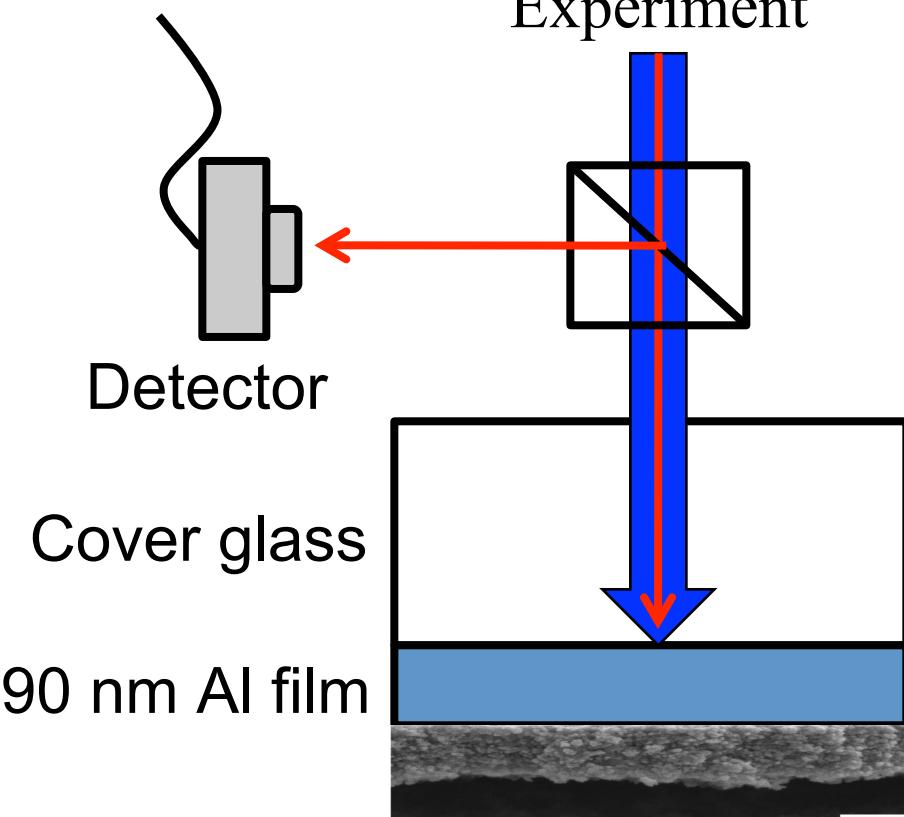
Patrick E. Hopkins,<sup>1,a)</sup> Bryan Kaehr,<sup>2,3</sup> Edward S. Piekos,<sup>2</sup> Darren Dunphy,<sup>3</sup> and C. Jeffrey Brinker<sup>2,3</sup>

<sup>1</sup>*Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904, USA*

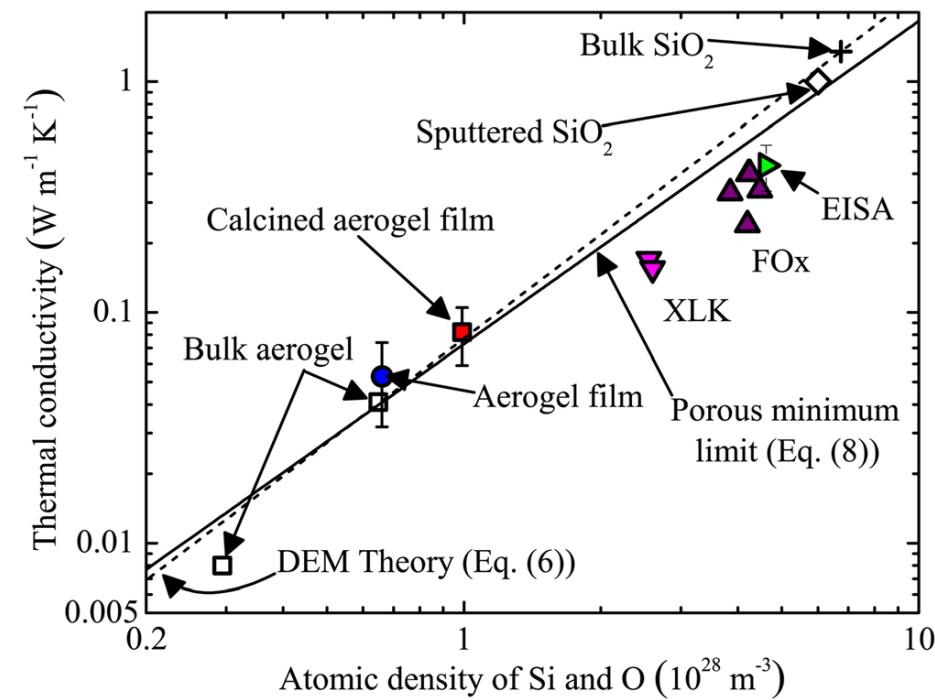
<sup>2</sup>*Sandia National Laboratories, Albuquerque, New Mexico 87123, USA*

<sup>3</sup>*Department of Chemical and Nuclear Engineering, University of New Mexico, Albuquerque, New Mexico 87106, USA*

### Experiment

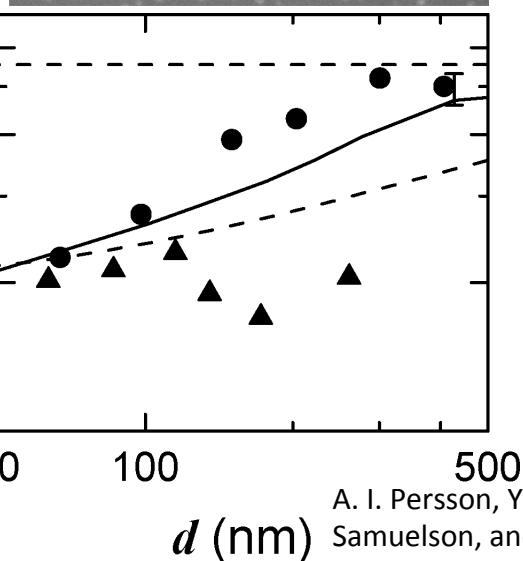
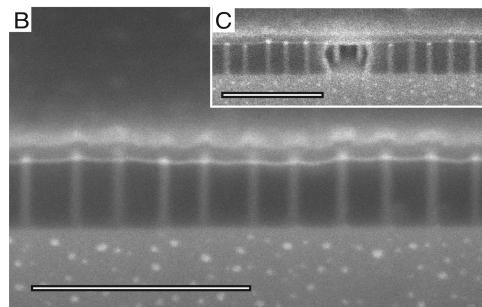
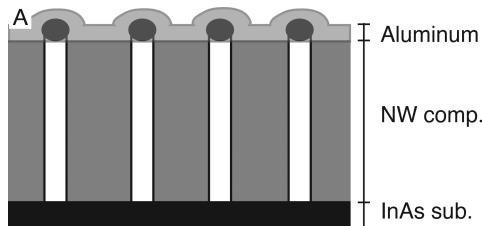


Porous or non-conformal films

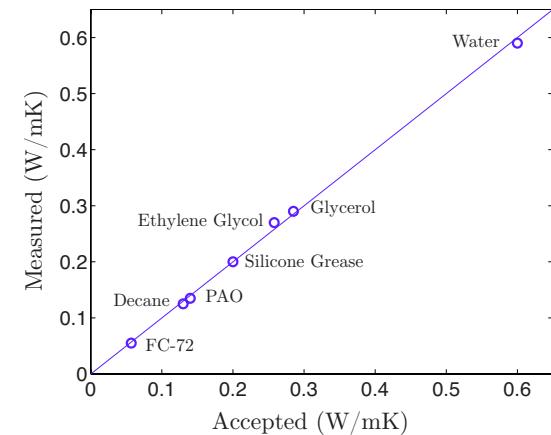
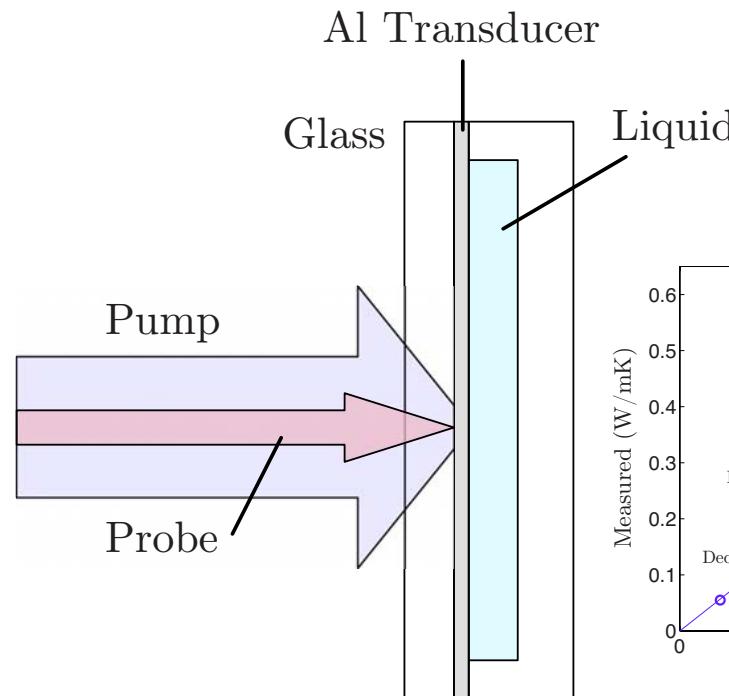


# Time domain thermoreflectance (TDTR)

## 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

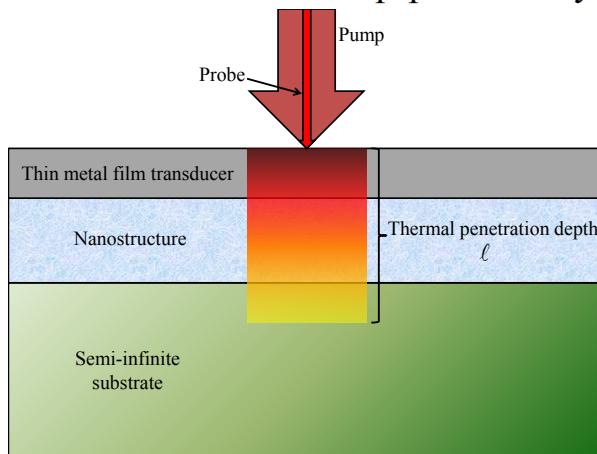
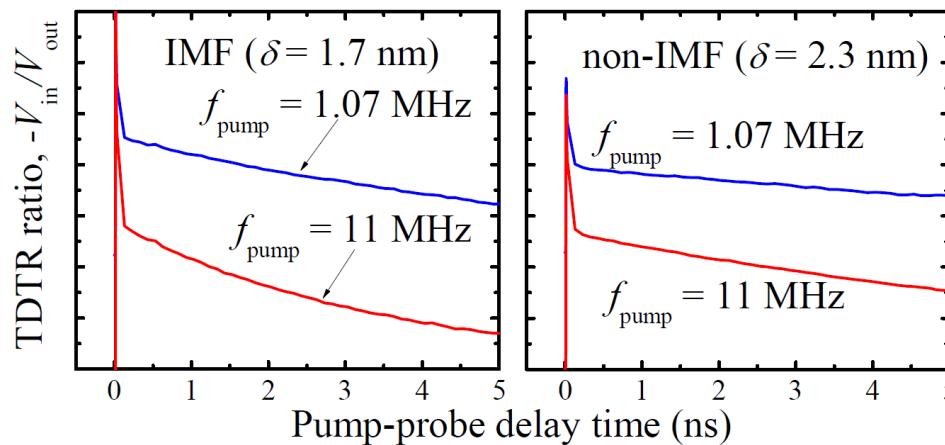
APPLIED PHYSICS LETTERS 98, 161913 (2011)  
Effect of dislocation density on thermal boundary conductance across  
GaSb/GaAs interfaces

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$h_K - \text{Al/GaSb } (f = 11 \text{ MHz})$

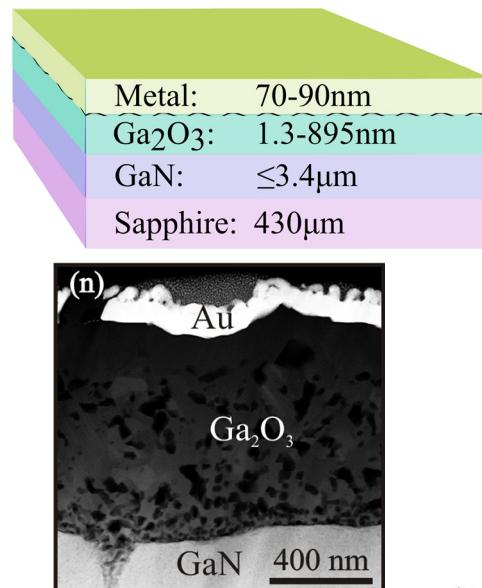
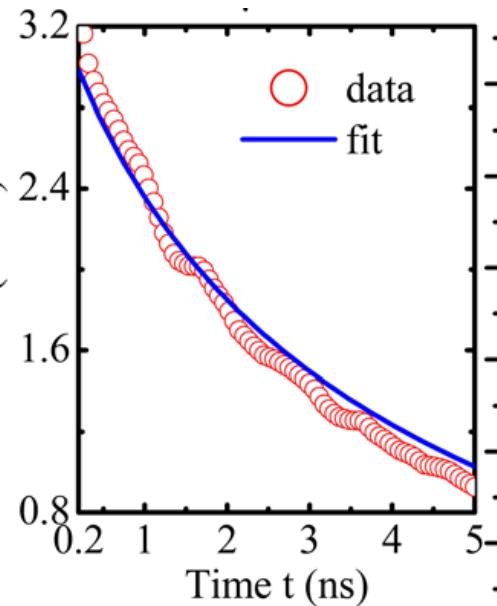
$$\delta_{\text{thermal}} = \sqrt{\frac{\kappa}{\pi f C}} = \sqrt{\frac{2\kappa}{\omega C}}$$

$h_K - \text{GaSb/GaAs } (f = 1.07 \text{ MHz})$

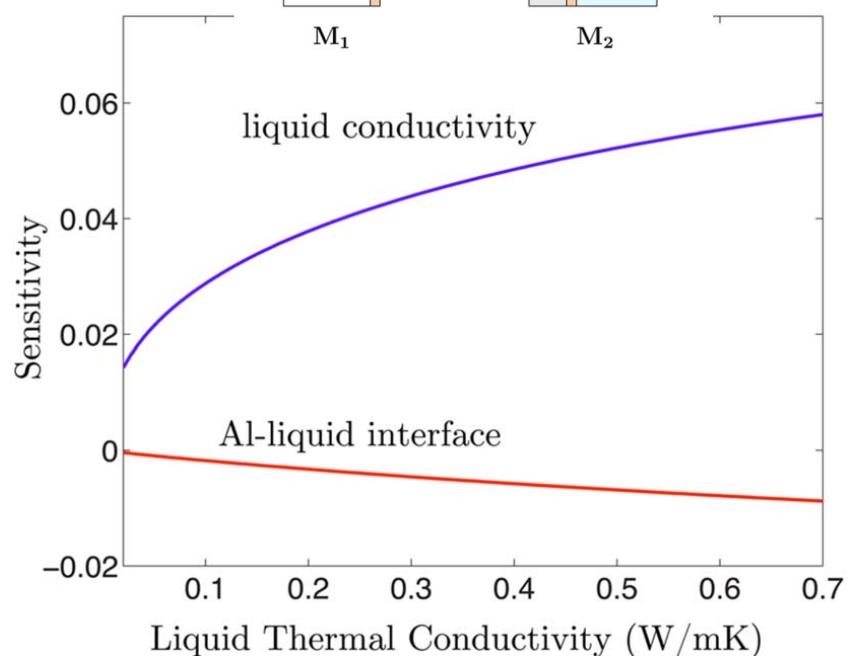
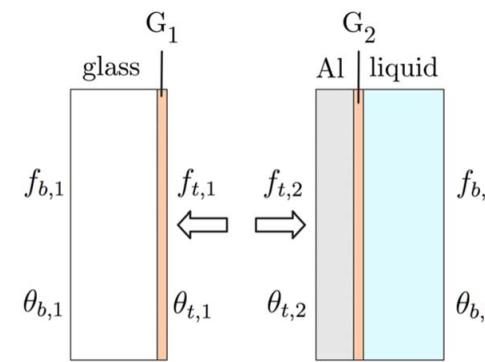
**Increase how “deep” you probe in TDTR by decreasing the frequency**

# Limitations – TDTR (and other thermoreflectance techniques)

## Rough surfaces (~>40 nm RMS-“ish”)



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



C. J. Szwejkowski, 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.

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”)**
  - $3\omega$
  - **Thermoreflectance-based techniques**
    - TDTR
    - FDTR
    - **Limitations and potentials....**

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