The Abdus Salam
International Centre
for Theoretical Physics

2371-10

Advanced Workshop on Energy Transport in Low-Dimensional Systems: Achievements and Mysteries

15 - 24 October 2012

Ultrafast Characterization of Heat Transport and Thermoelectric Energy Exchange at Interfaces (Ballistic and Diffusive Transport of Heat/Energy

Ali SHAKOURI

Birck Nanotechnology Center, Purdue University
West Lafayette
U.S.A.

PURDUE UNIVERSITY Discovery Park

Ballistic and Diffusive Transport of Heat/Energy

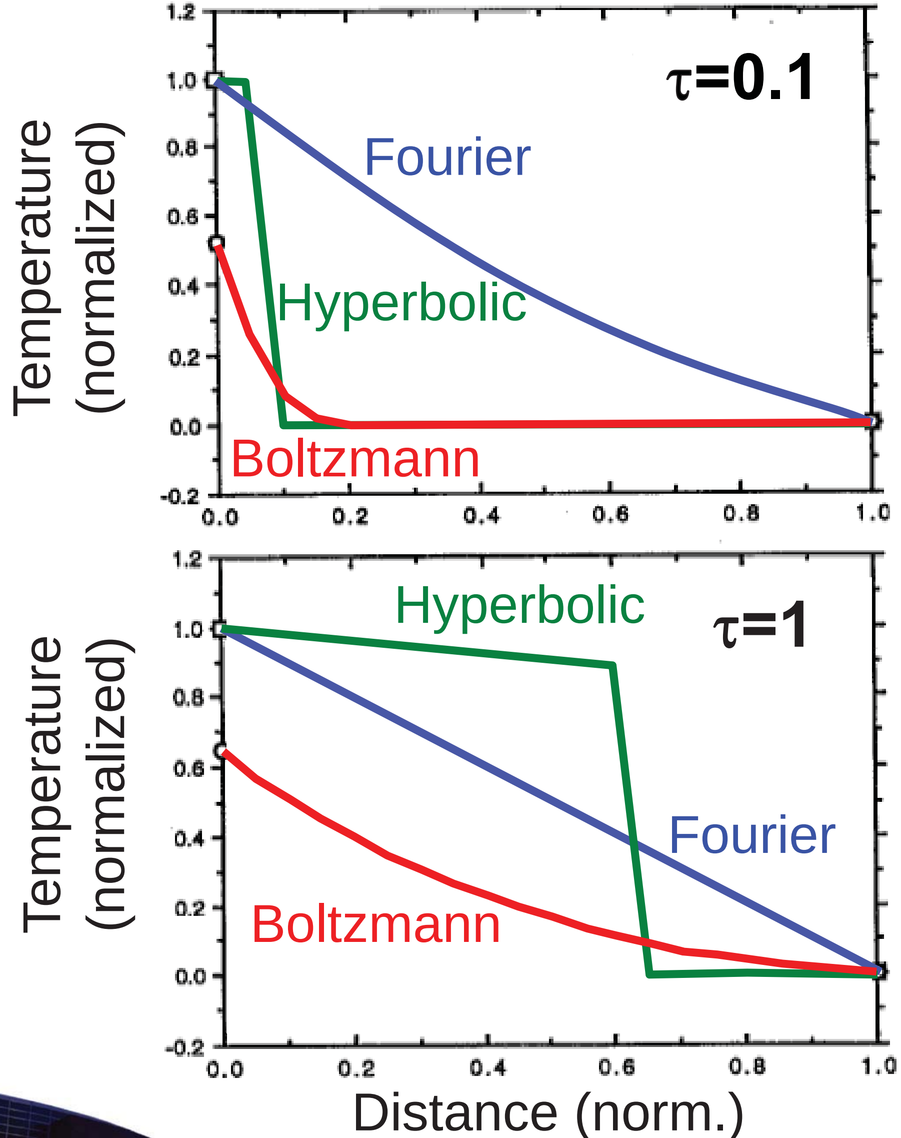
Ali Shakouri
Birck Nanotechnology Center; Purdue University
shakouri@purdue.edu

Acknowledgement: **DOE/EFRC (CEEM Center), DARPA, ONR, AFOSR, NSF, SRC-IFC, CEA, Intel, Wyle Lab**

Advanced Workshop on Energy Transport in Low Dimensional Systems
17 October 2012

QUANTUM ELECTRONICS

Diffusive or Ballistic Propagation of Heat



L=0.1μm
Diamond

Fourier

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \qquad \alpha = \kappa / C$$

Hyperbolic Heat (Cattaneo)

$$\tau_R \frac{\partial^2 T}{\partial t^2} + \frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2}$$

Boltzmann (Equation Phonon Radiative Transfer)

$$\frac{\partial N_q}{\partial t} + \mathbf{v} \cdot \nabla N_q = \left(\frac{\partial N_q}{\partial t} \right)_{scat}$$

Phonon Number: $N_q(x,t)$

A. A. Joshi and A. Majumdar; J. Appl. Phys. 74, p. 31 (1993)

Analogy with Drift-Diffusion

PURDUE UNIVERSITY Discovery Park

$$J = \sigma E - e D \nabla n \quad ; \quad \sigma = e n \mu$$

μ: mobility; D: diffusion coefficient

Einstein Relation: $D/\mu = k_B T/e$

Luttinger's Gravitation Field Analogy

Ψ(x,t): Fictitious mechanical field

↔ T(x,t): Temperature

Luttinger J M 1964 Phys. Rev. 135 A1505
Luttinger J M 1964 Phys. Rev. 136 A1481
Shastry B S 2009 Rep, Prog, Phys 72, 016501

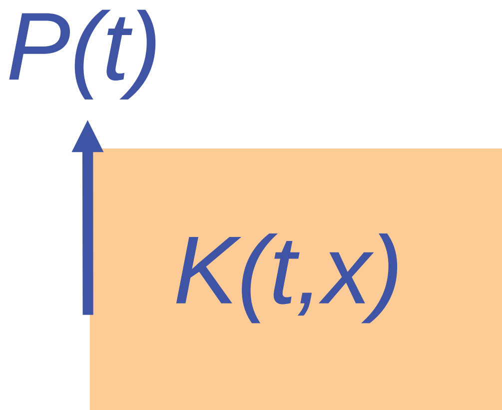
QUANTUM ELECTRONICS

Shastry: energy density propagation

N_2: The Green's function of the total <u>energy density propagation</u> K(t,x) in a solid material when there is delta-function excitation P(t)

$$N_2(\omega,q) = \frac{K(\omega,q)}{P(\omega)}$$

$$N_2(\omega,q) = \frac{\imath + \tau_q \omega}{\omega - \imath \tau_q \omega^2 + \imath D_Q q^2}$$

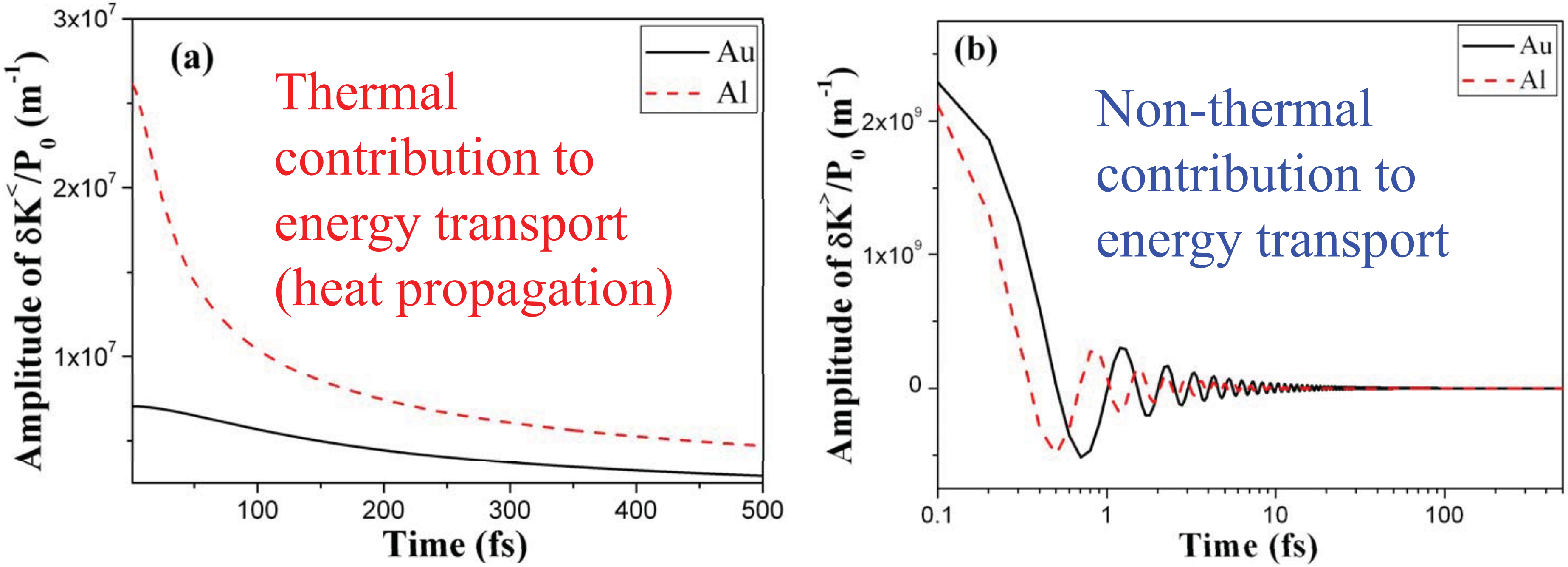


- τ_q total relaxation time of energy carriers (function of wavevector q)
- D_Q heat diffusion constant

<u>B. S. Shastry, Rep, Prog, Phys **72**, 016501, (2009)</u>

Top surface energy density response: Thermal and Non-thermal contributions

PURDUE UNIVERSITY Discovery Park



Damped oscillation of period θ

$$\theta = 2\sqrt{3}\,\frac{a}{v_F} \Rightarrow \;(1\,fs)$$

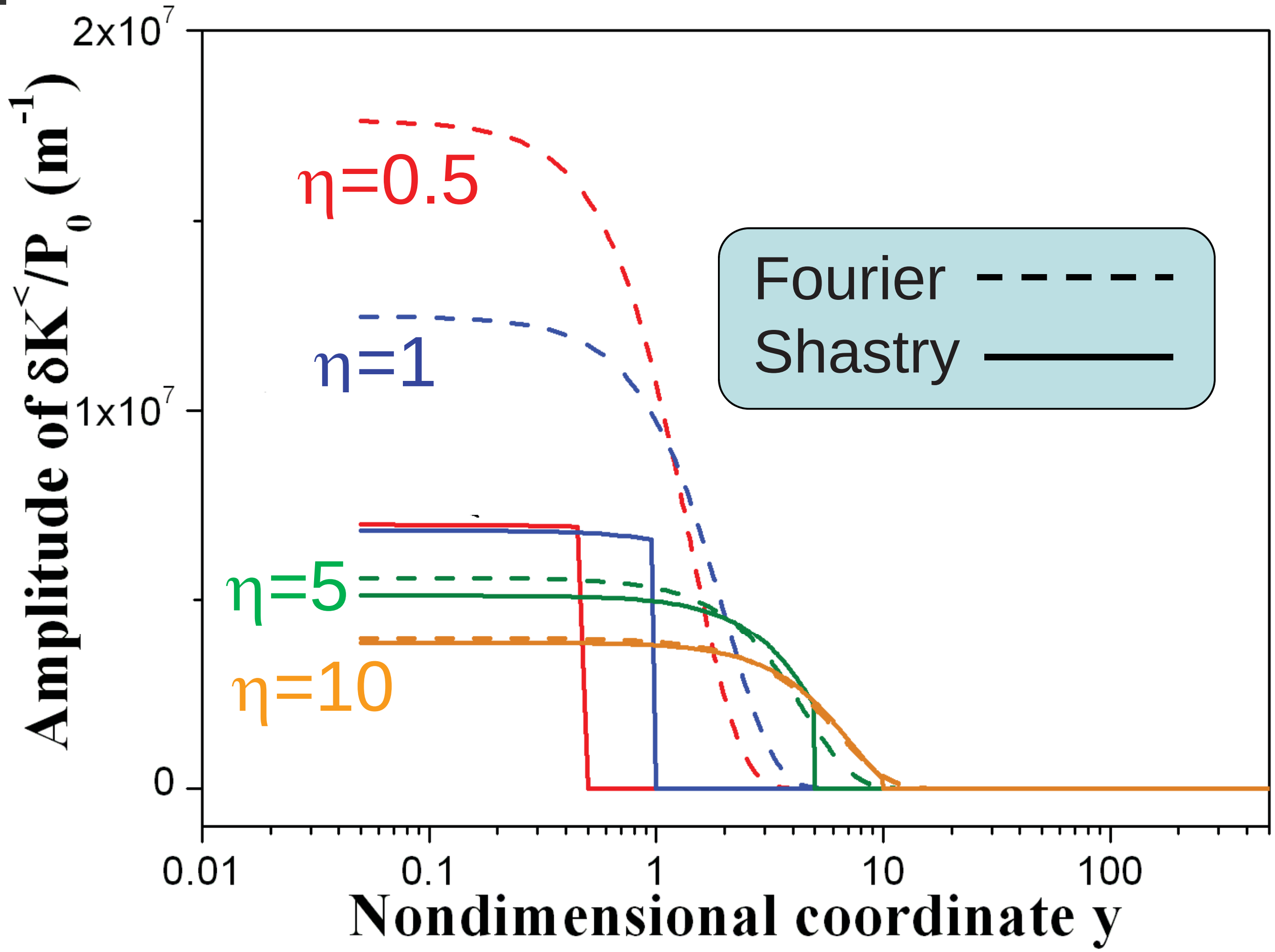
a: lattice constant.
v_F: Fermi velocity.

Oscillations in the total energy density at the top free surface of the metal:
- *due to the Bragg reflection of ballistic electrons from the Brillouin Zone boundaries.*

Y. Ezzahri and A. Shakouri; Phys. Rev. B, 79, 184303, (2009)

QUANTUM ELECTRONICS

Thermal contribution to energy transport (Temperature distribution in space)



Non Dimensional Variables

$\eta = \mathbf{t} / (\tau_q = \text{const.})$

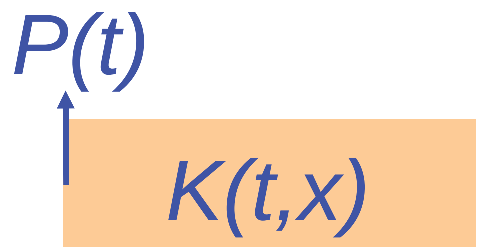
$\mathbf{y} = \mathbf{x} / \mathrm{sqrt}(D_Q \tau_q)$

Shastry predicts <u>temperature wavefronts</u> similar to Cattaneo (ballistic heat equation) but the momentum-dependence of relaxation time can affect the shape.

Y. Ezzahri and A. Shakouri; Journal of Heat Transfer, 2011

Energy density propagation in TE materials

Total <u>energy density propagation</u> K(t,x) in a thermoelectric material when there is delta-function excitation P(t)



- Heat Diffusion D_Q
- Charge Diffusion D_C

- ξ coupling factor between charge and energy density

$$\xi = \frac{Z^* T}{1 + Z^* T}$$

- Z^* high frequency limit of figure of merit

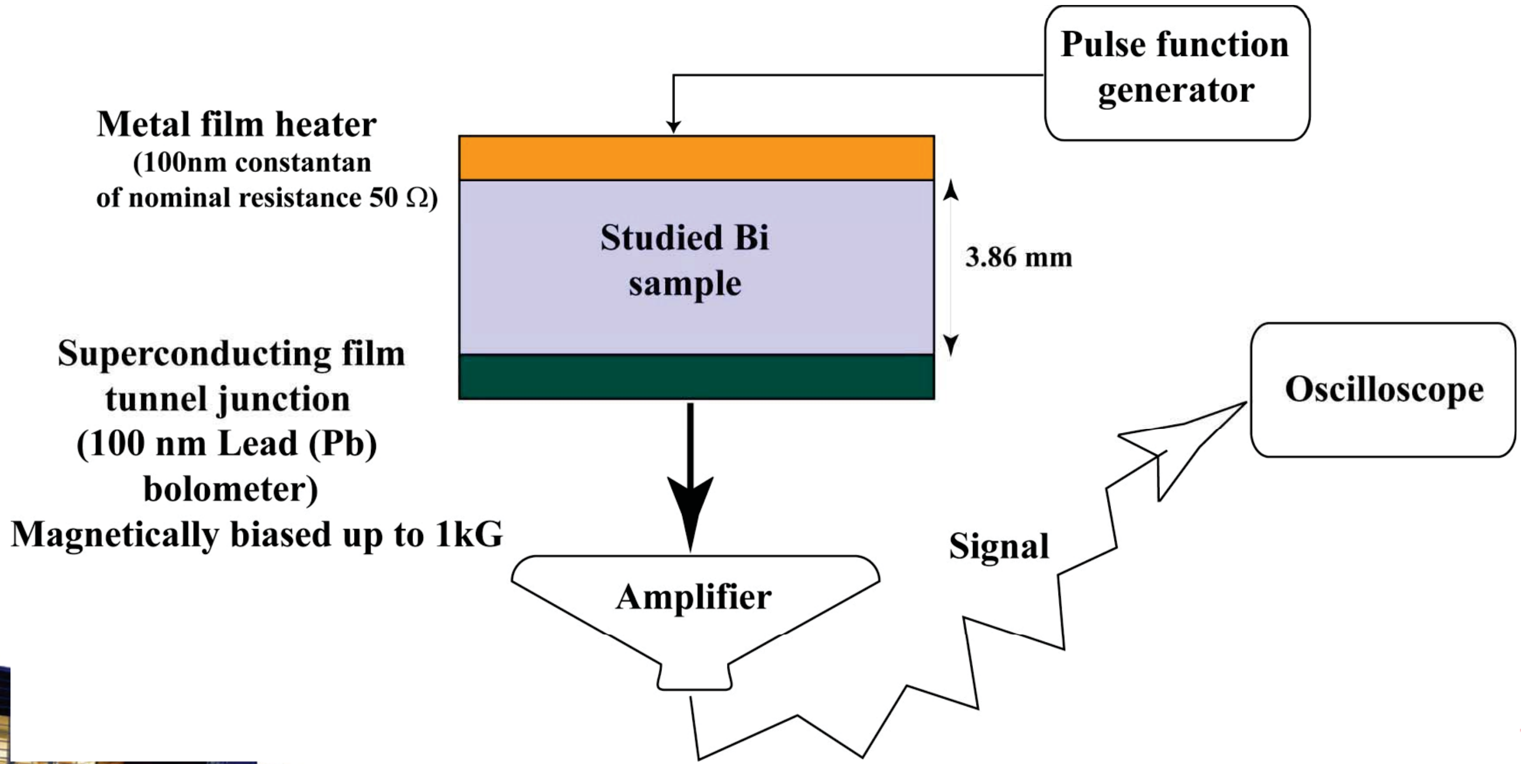
B. S. Shastry, Rep, Prog, Phys ***72****, 016501, (2009)*

Observation of second sound in bismuth

V. Narayanamurti and R. C. Dynes, Phys. Rev. Lett, 28, 1461, (1972).

Younes Ezzahri

Experiment principle



Results

Propagation along the C3 axis

PURDUE UNIVERSITY Discovery Park

Younes Ezzahri

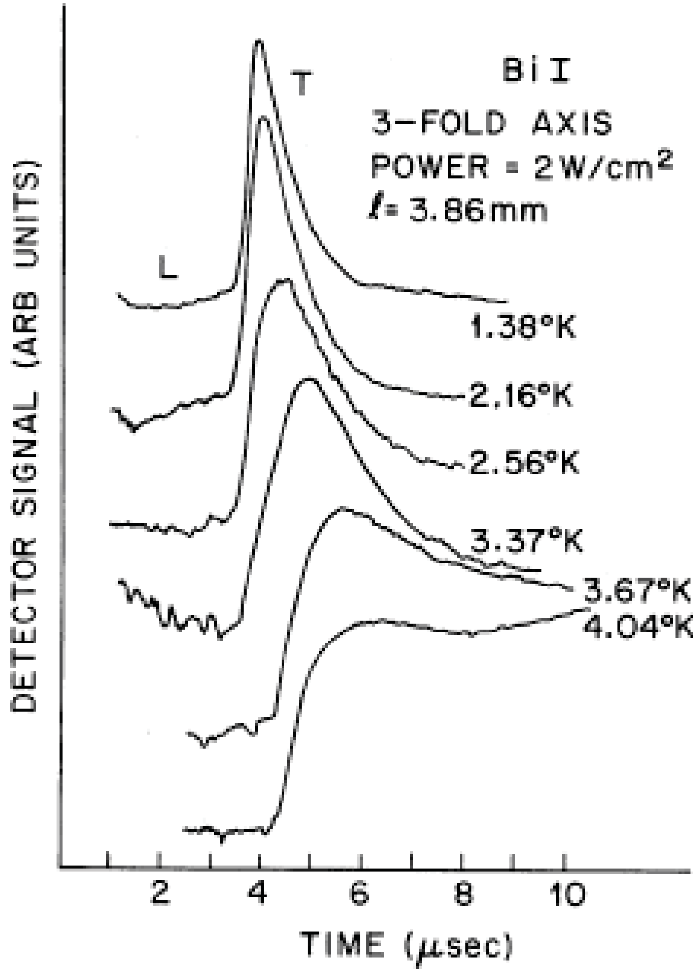


FIG. 1. Heat pulses in Bi along the C_3 axis.

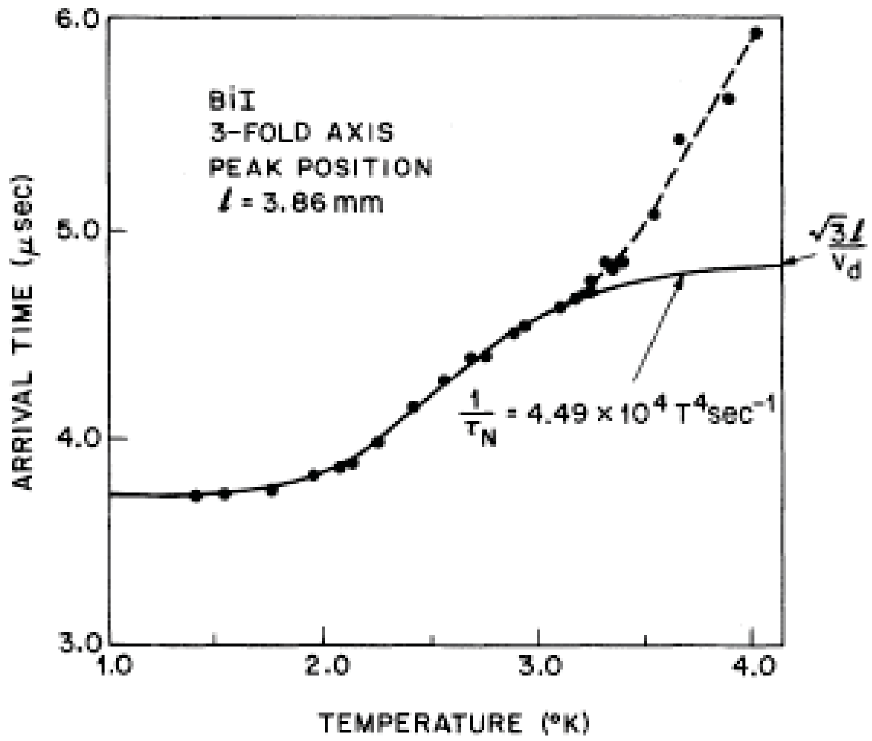


FIG. 2. Arrival time of the peak position as a function of temperature from the data of Fig. 1. Solid line, calculated from the dispersion relation for second sound (see text).

- Observation of a ballistic transverse phonon propagation.
- Transition to second sound.

QUANTUM ELECTRONICS

Narayanamurti, and Dynes, PRL 28, 1461 (1972)

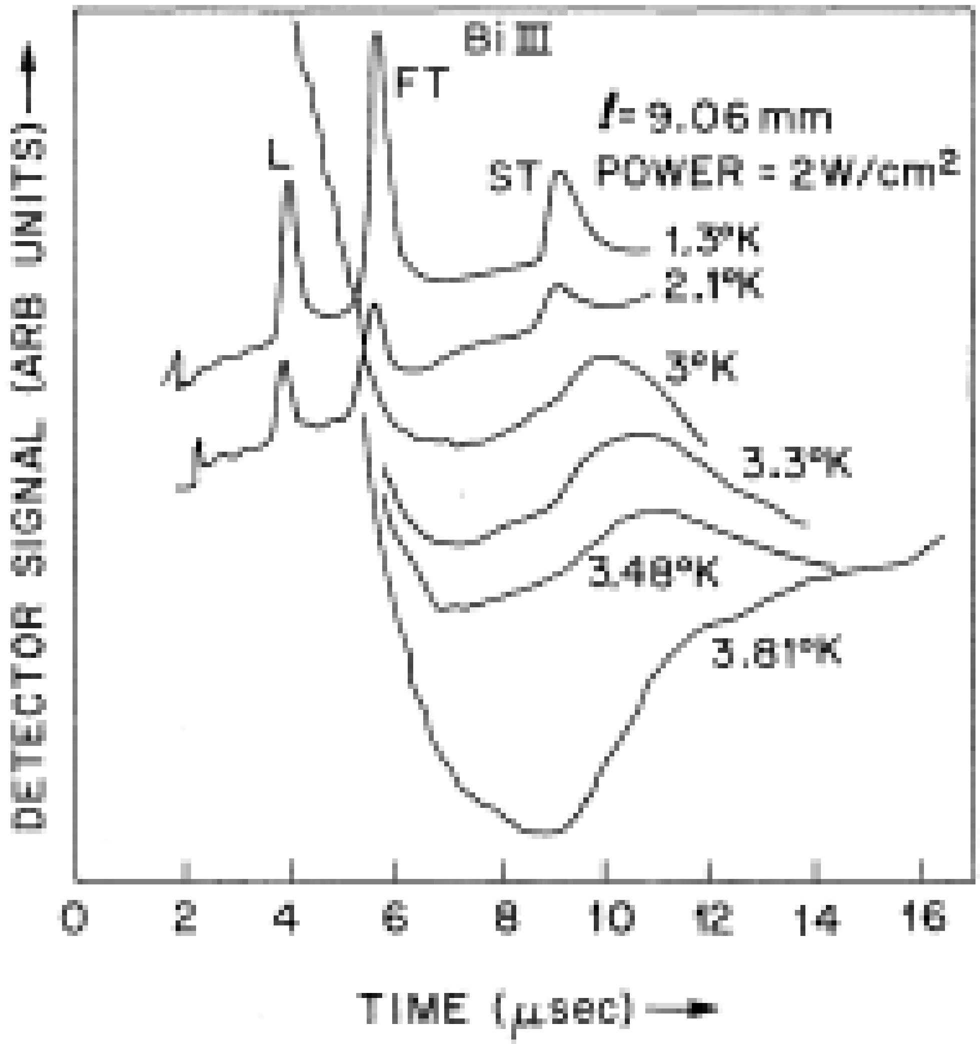


FIG. 3. Heat pulses in Bi along the twofold axis.

Second sound -heat pulse experiments

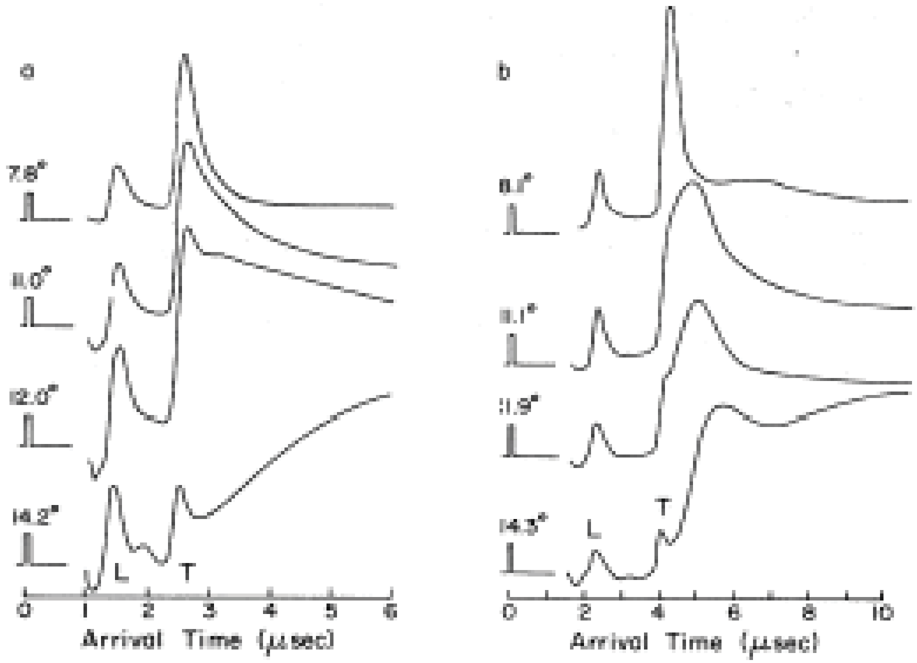


FIG. 1. Heat pulses in NaF in the ⟨100⟩ direction at temperatures in the vicinity of the thermal conductivity maximum. (a) Singly grown NaF, separation between heater and bolometer $l = 7.8$ mm. (b) Triply grown NaF, $l = 12.7$ mm. Crystal faces approximately 12 mm × 12 mm. The letters L and T mark the longitudinal

McNelly, et al, Phys. Rev. Lett. 24, 100 (1970).

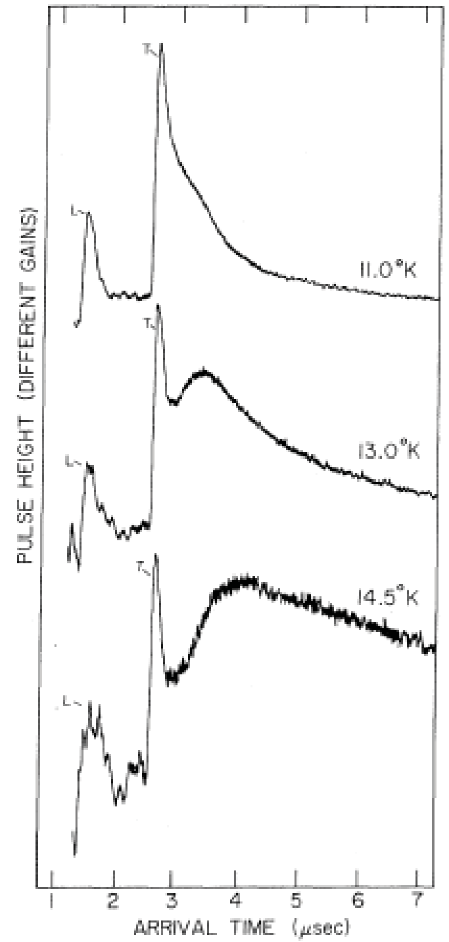


FIG. 4. Heat pulses in a pure NaF sample ($l = 7.9$ mm) in the ⟨100⟩ direction for several different temperatures. L and T mark the peaks of the longitudinal and transverse ballistic pulses, respectively. Note the appearance of a third distinct pulse (second sound).

H. Maris, Brown Univ.

Jackson and Walker, Phys. Rev. B, 1428 (1971)

Different macroscopic equations describing heat wave propagation

- **Cattaneo-Vernotte type: Telegraph equation**

$$\begin{cases} C_V \dfrac{\partial T}{\partial t} + div(Q) = 0 \\ \tau \dfrac{\partial Q}{\partial t} + Q = -\beta \nabla T \end{cases} \Rightarrow \frac{\partial^2 T}{\partial t^2} + \frac{1}{\tau}\frac{\partial T}{\partial t} = \frac{\beta}{\tau C_V}\nabla^2 T$$

- **Jeffreys type**

$$\begin{cases} C_V \dfrac{\partial T}{\partial t} + div(Q) = 0 \\ \tau \dfrac{\partial Q}{\partial t} + Q = -\beta \nabla T - \tau \beta_1 \dfrac{\partial}{\partial t} \nabla T \Rightarrow \dfrac{\partial^2 T}{\partial t^2} + \dfrac{1}{\tau}\dfrac{\partial T}{\partial t} = \dfrac{\beta}{\tau C_V}\nabla^2 T + \dfrac{\beta_1}{C_V}\nabla^2 \dfrac{\partial T}{\partial t} \\ \beta = \beta_1 + \beta_2 \end{cases}$$

β_1: Effective thermal conductivity.

β_2: Elastic thermal conductivity.

Younes Ezzahri (UCSC, now at Univ. Poitier)

QUANTUM ELECTRONICS

Macroscopic equations of Guyer and Krumhansl

$$\begin{cases} C_V \dfrac{\partial T}{\partial t} + div(Q) = 0 \\ \dfrac{\partial Q}{\partial t} + \dfrac{v^2}{3}\nabla T + \dfrac{1}{\tau_R} Q = \dfrac{\tau_N v^2}{5}\left[\nabla^2 Q + 2\nabla div(Q)\right] \end{cases} \Rightarrow \frac{\partial^2 T}{\partial t^2} + \frac{1}{\tau_R}\frac{\partial T}{\partial t} = \frac{v^2}{3C_V}\nabla^2 T + \frac{3}{5}\tau_N v^2 \nabla^2 \frac{\partial T}{\partial t}$$

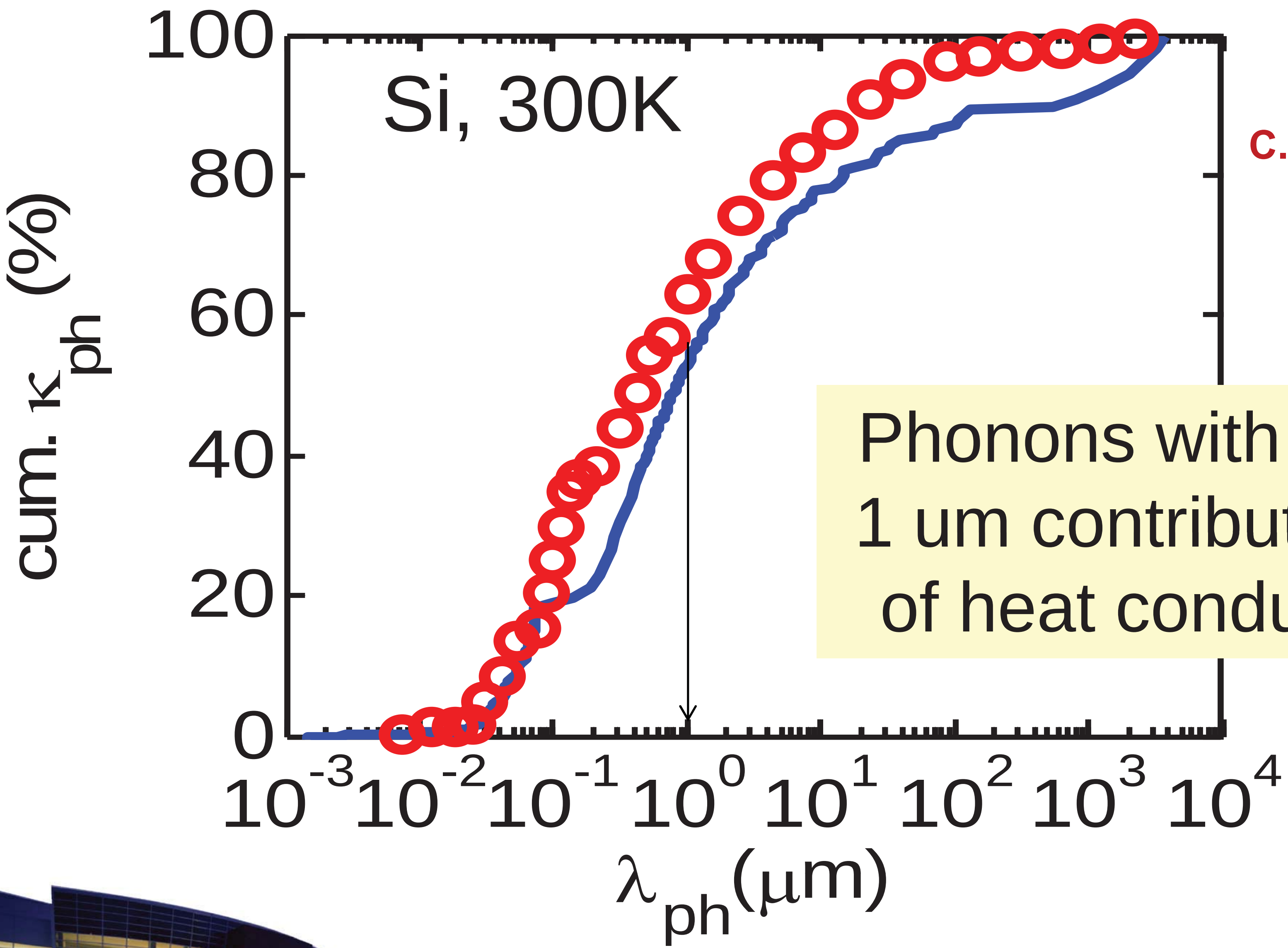
Jeffrey's type equation with and effective thermal diffusivity:

$$\alpha_1 = \frac{\beta_1}{C_V} = \frac{3}{5}\tau_N v^2$$

Viscosity of phonon gas

Younes Ezzahri (UCSC, now at Univ. Poitier)

Cummulative κ_{ph} vs. mfp at 300K



C. Jeong and M. Lundstrom (Purdue)

Phonons with mfp > 1 um contribute 50% of heat conduction.

Thin film silicon thermal conductivity

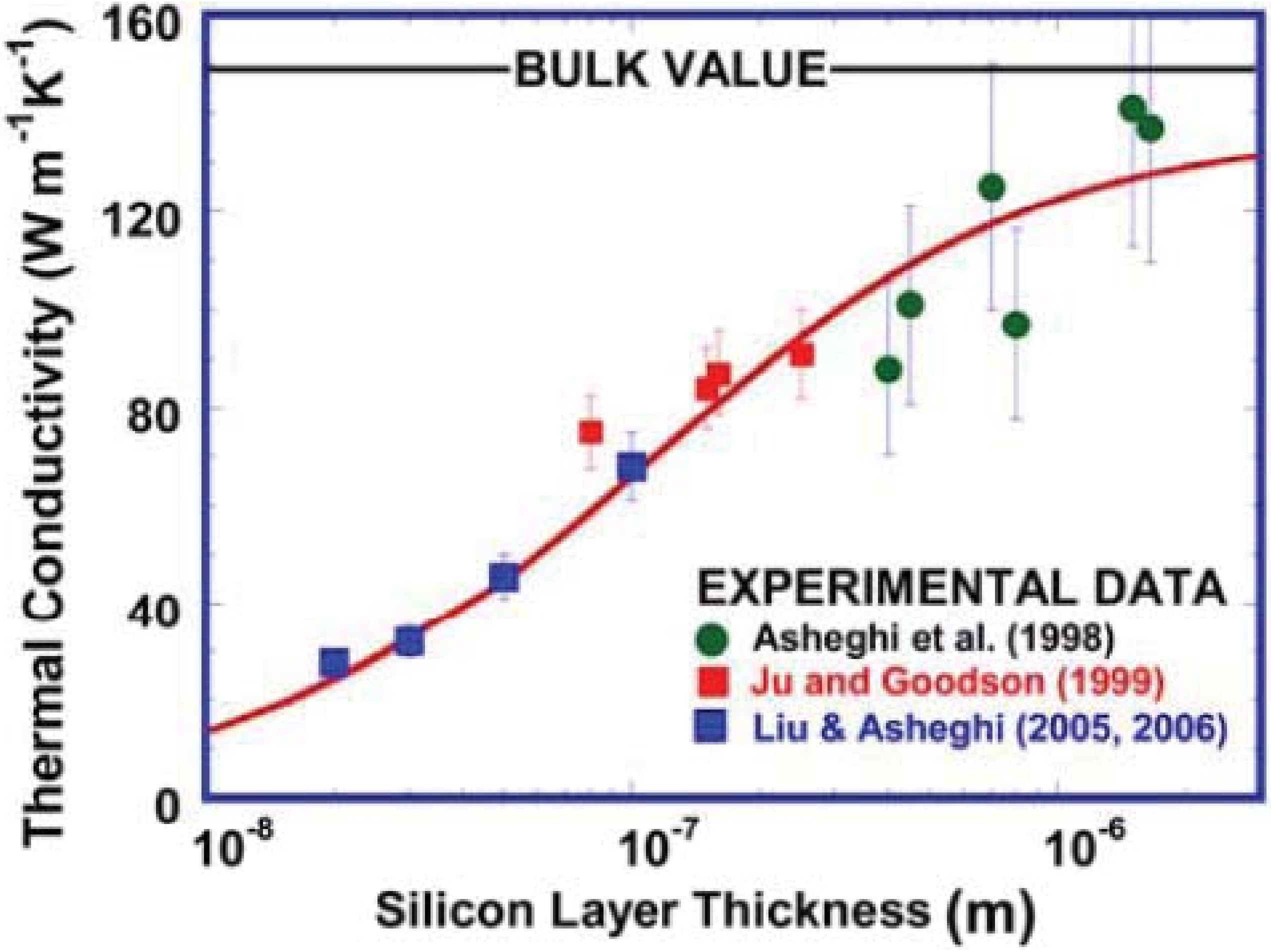
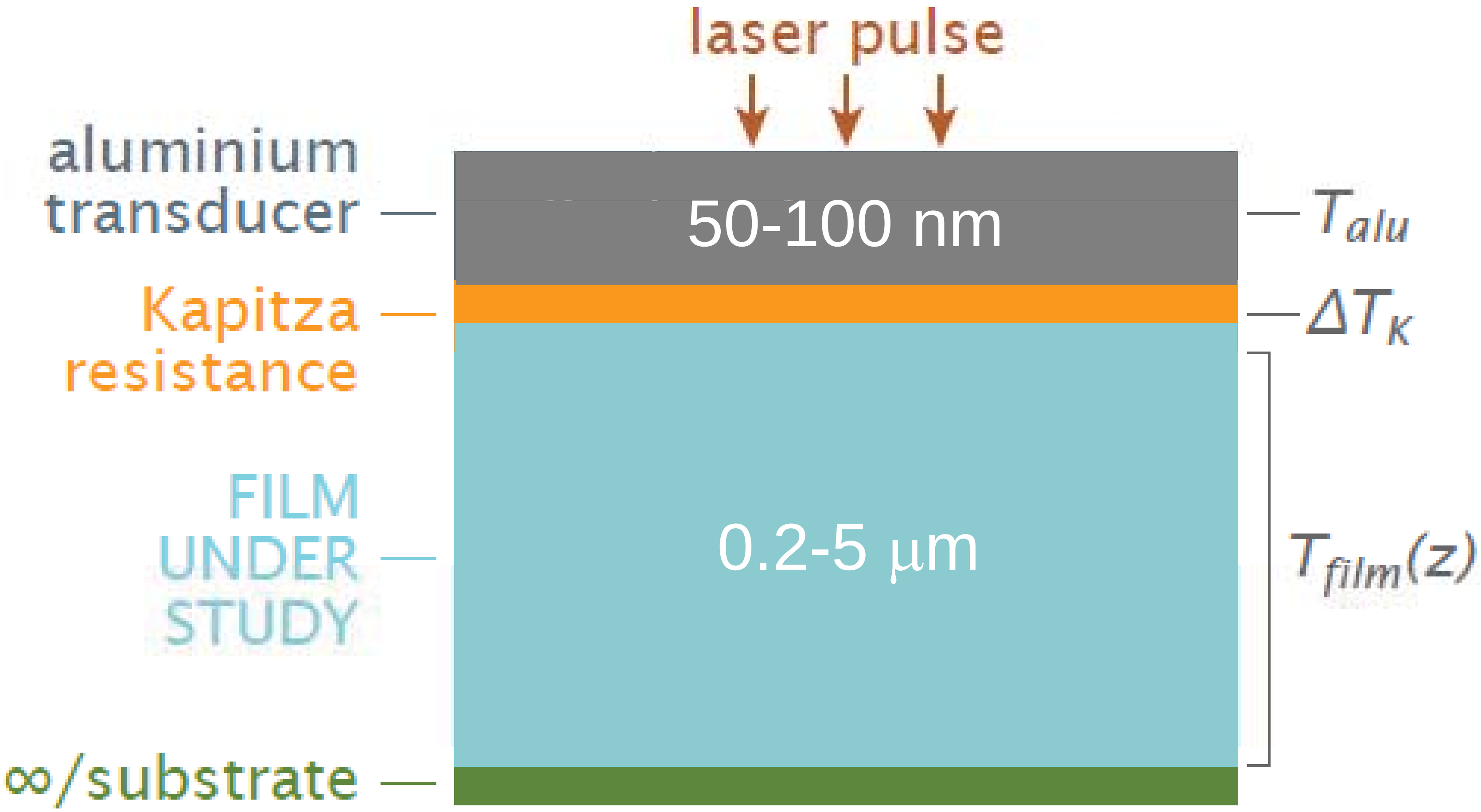


Fig. 1. *Thermal conductivity of silicon thin films versus film thickness at room temperature (courtesy of Prof. M. Asheghi [27], [159]).*

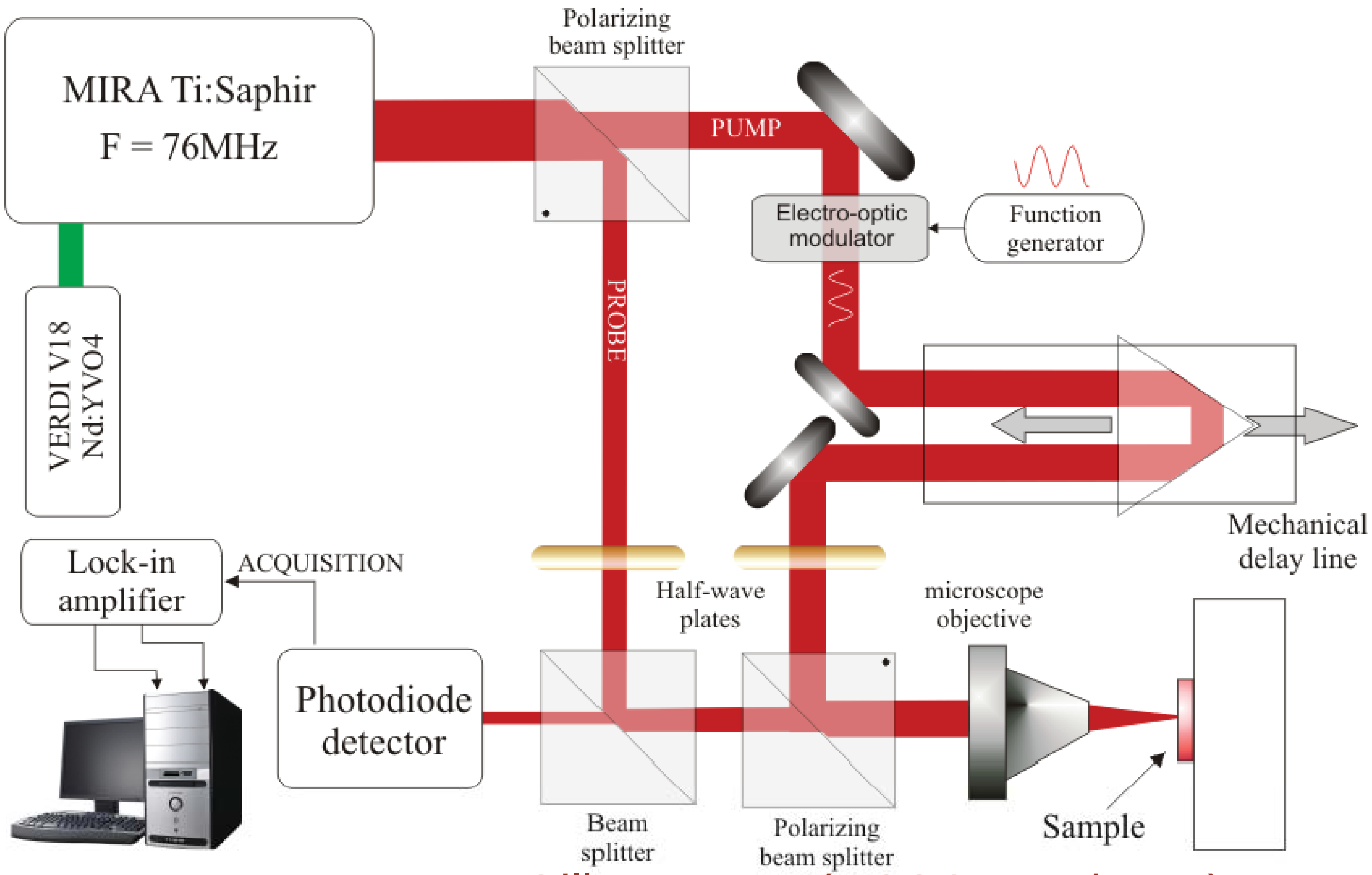
Time Domain Thermoreflectance (TDTR)



David G. Cahill, *Rev. Sci. Instrum.* 75, 5119 (2004).

Time Domain ThermoReflectance

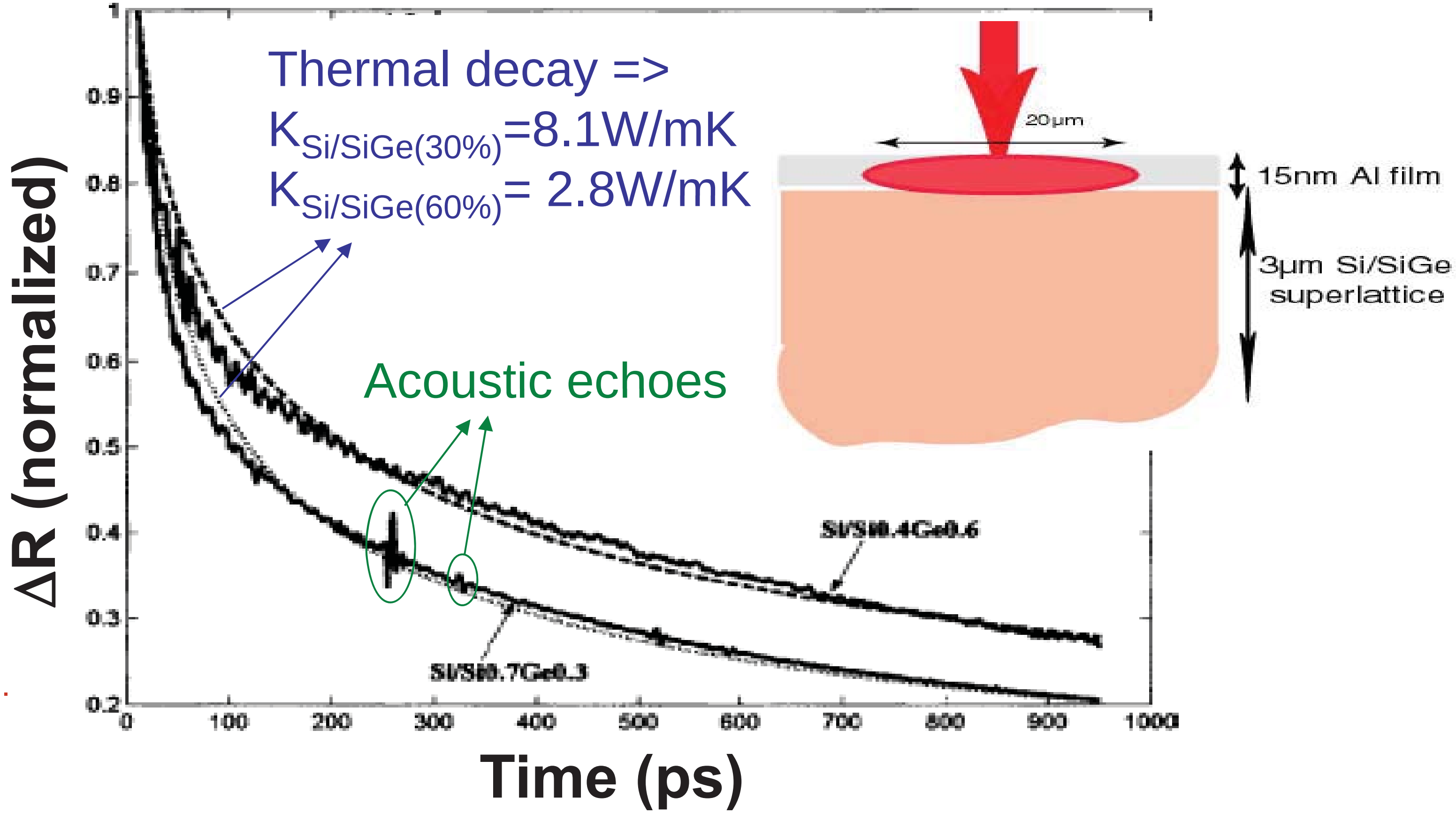
- **Modulated** & **delayed** femtosecond laser pulse used as a **Pump.**
- A **Probe** beam measures **reflectivity variation on the surface**
- The lock-in amplifier gives the **In-phase (V_{in})** and **Out-of-phase (V_{out})** signals.



Gilles Pernot (UCSC, Bordeaux)

Thin Film Thermal Characterization

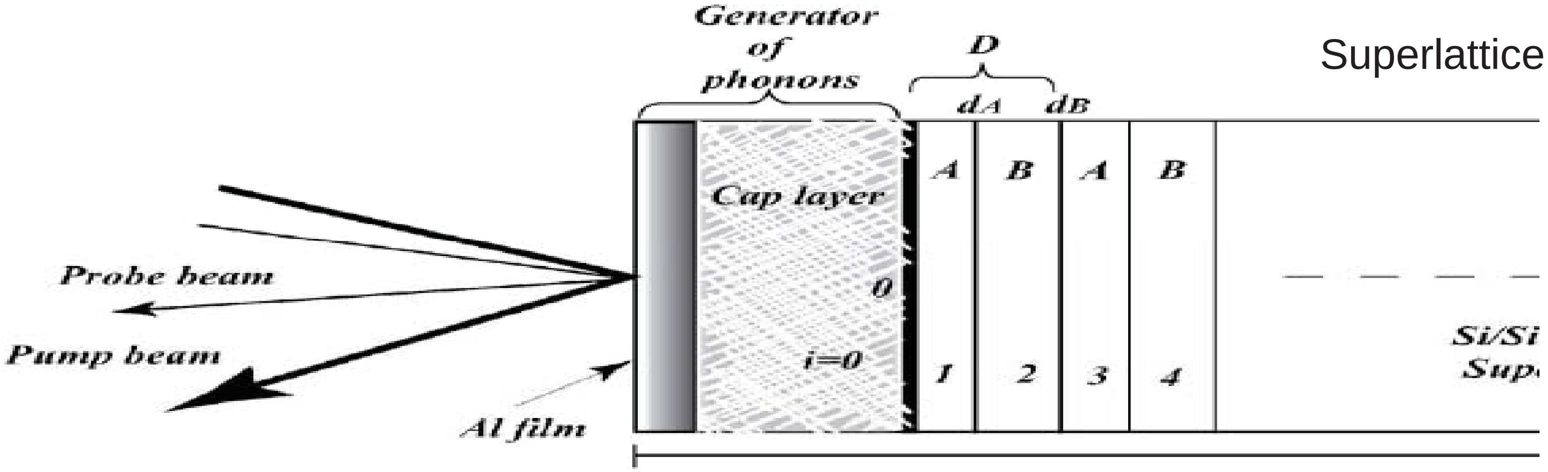
PURDUE UNIVERSITY Discovery Park

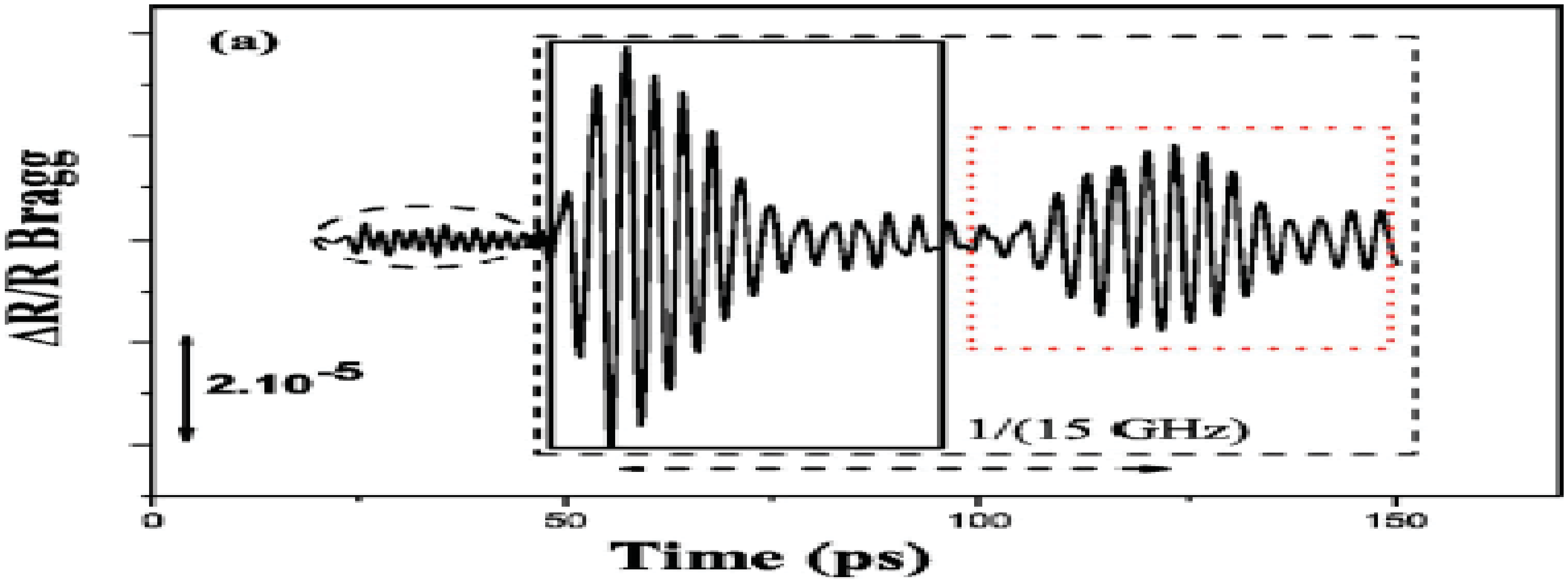


Y. Ezzahri et al. Appl. Phys. Letters 2005

Metal film is heated by laser pulse and it acts both as a heat source and a transducer (creates acoustic waves). It can characterize thermal interface resistances as well as interface quality (acoustic mismatch).

Phonon echoes in SiGe superlattices

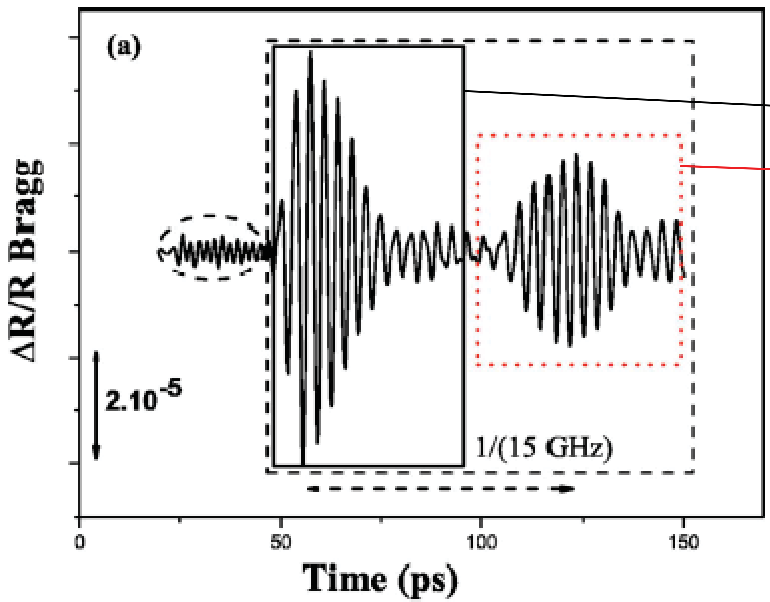


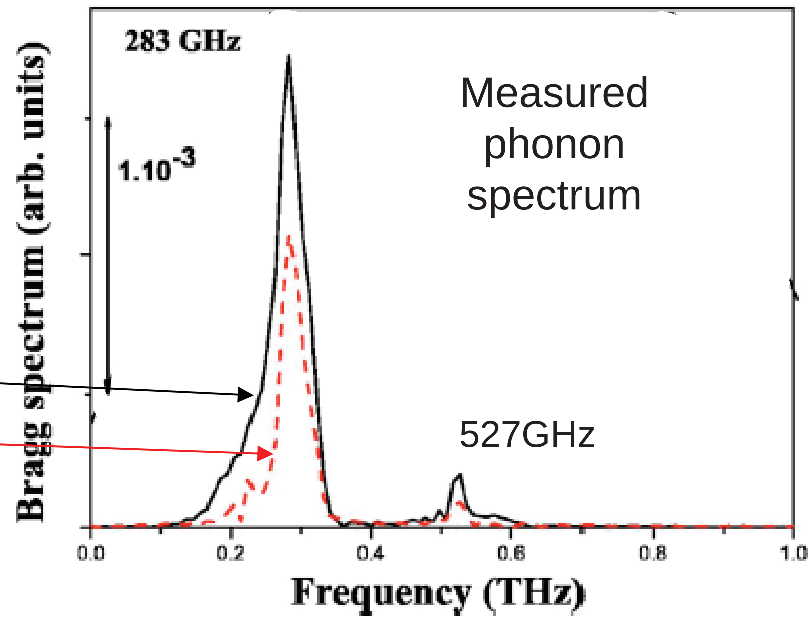


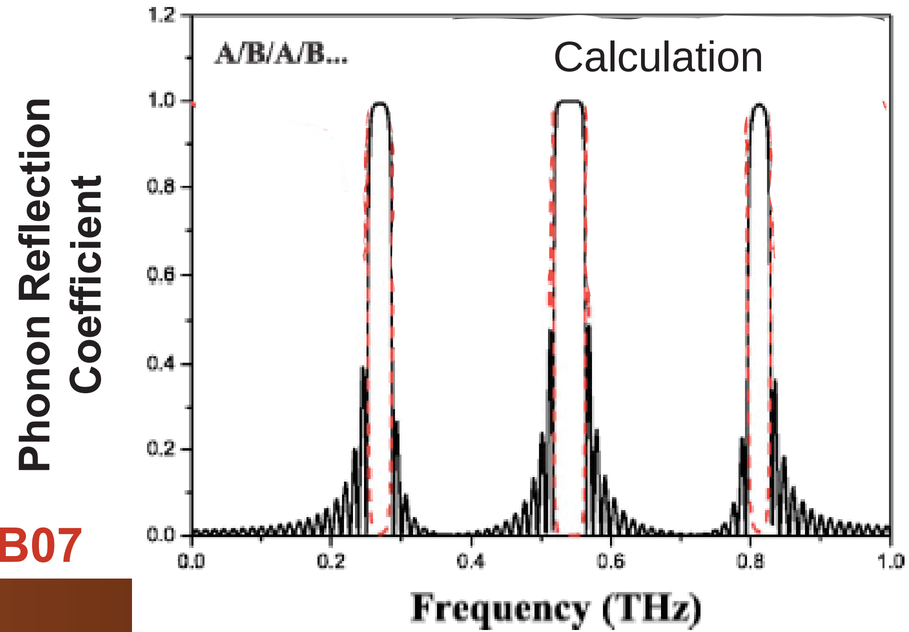
Y. Ezzahri, S. Dilhaire et al. PRB07

QUANTUM ELECTRONICS

Phonon minibands in SiGe superlattices

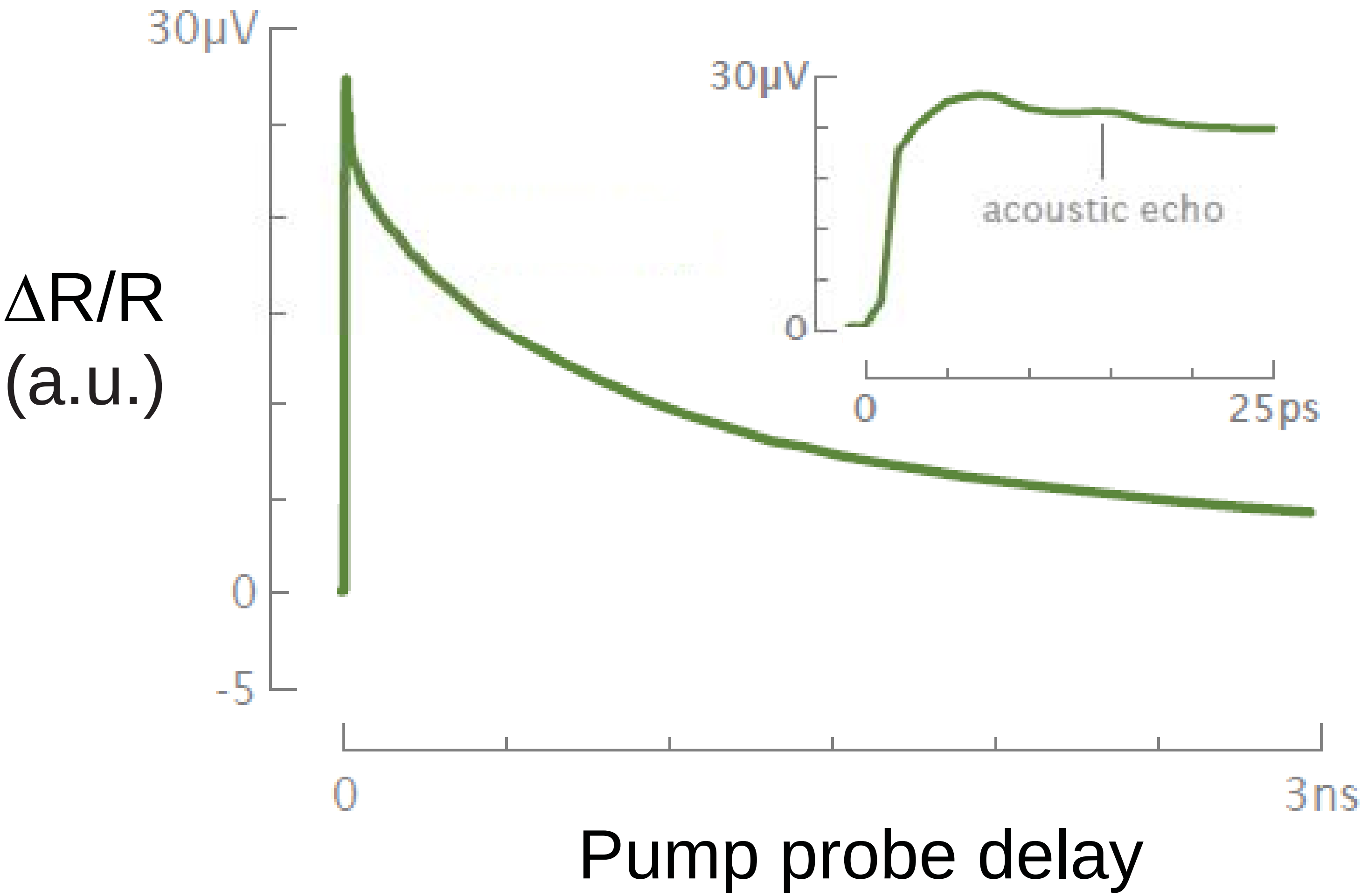






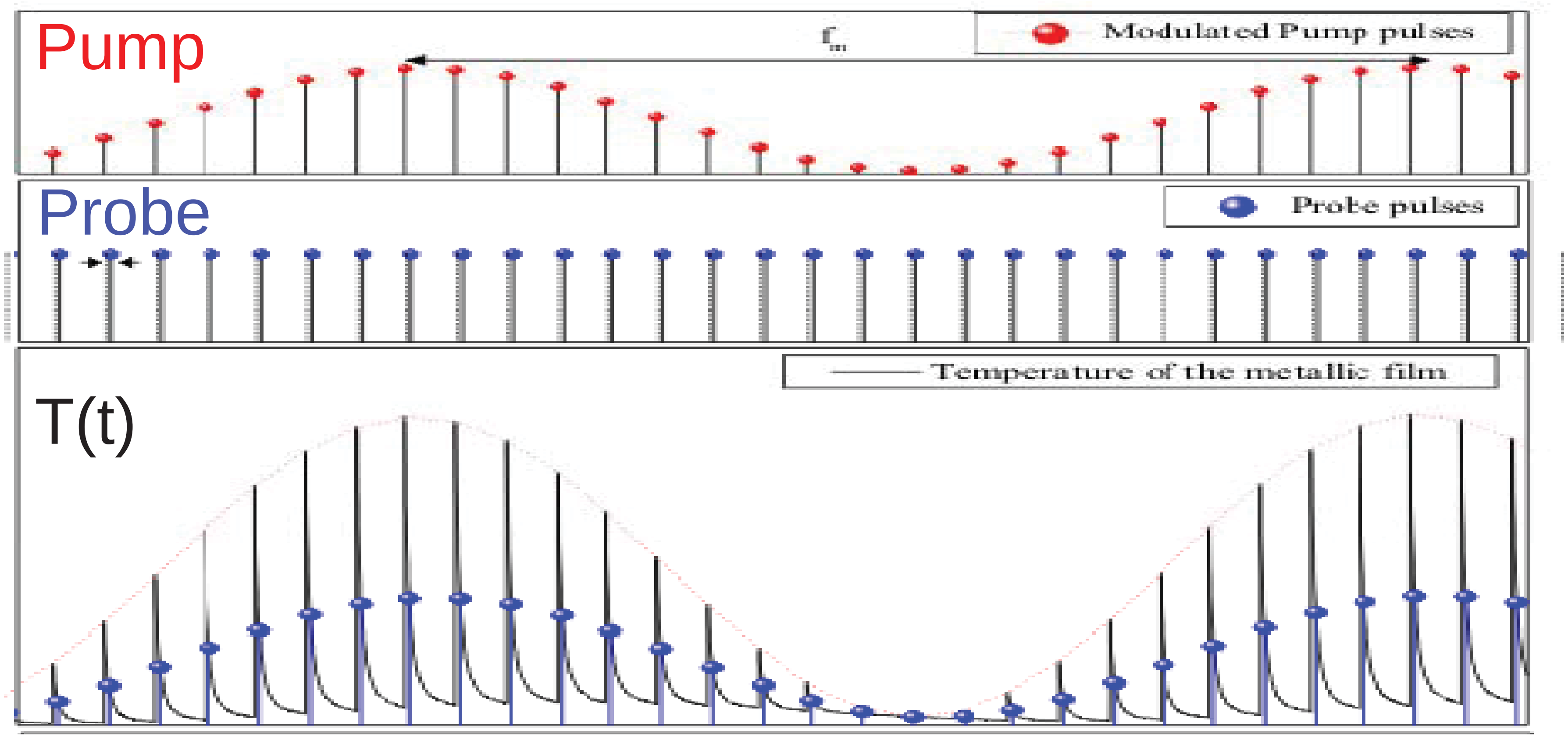
Y. Ezzahri, S. Dilhaire et al. PRB07

Femtosecond laser heat dissipation in thin film



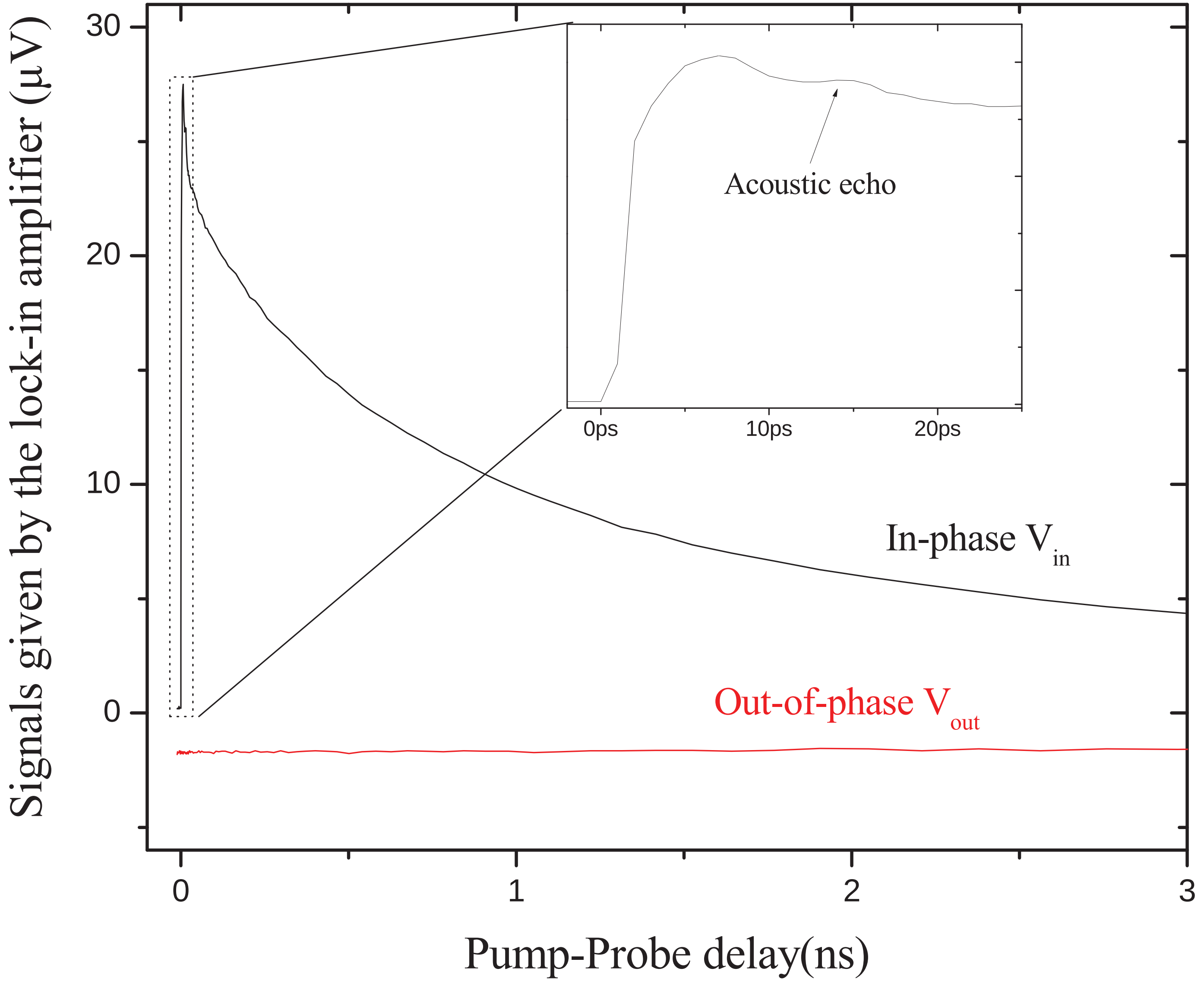
Bjorn Vermeersch, Gilles Pernot, et al. (to be submitted)

Optical sampling by TDTR



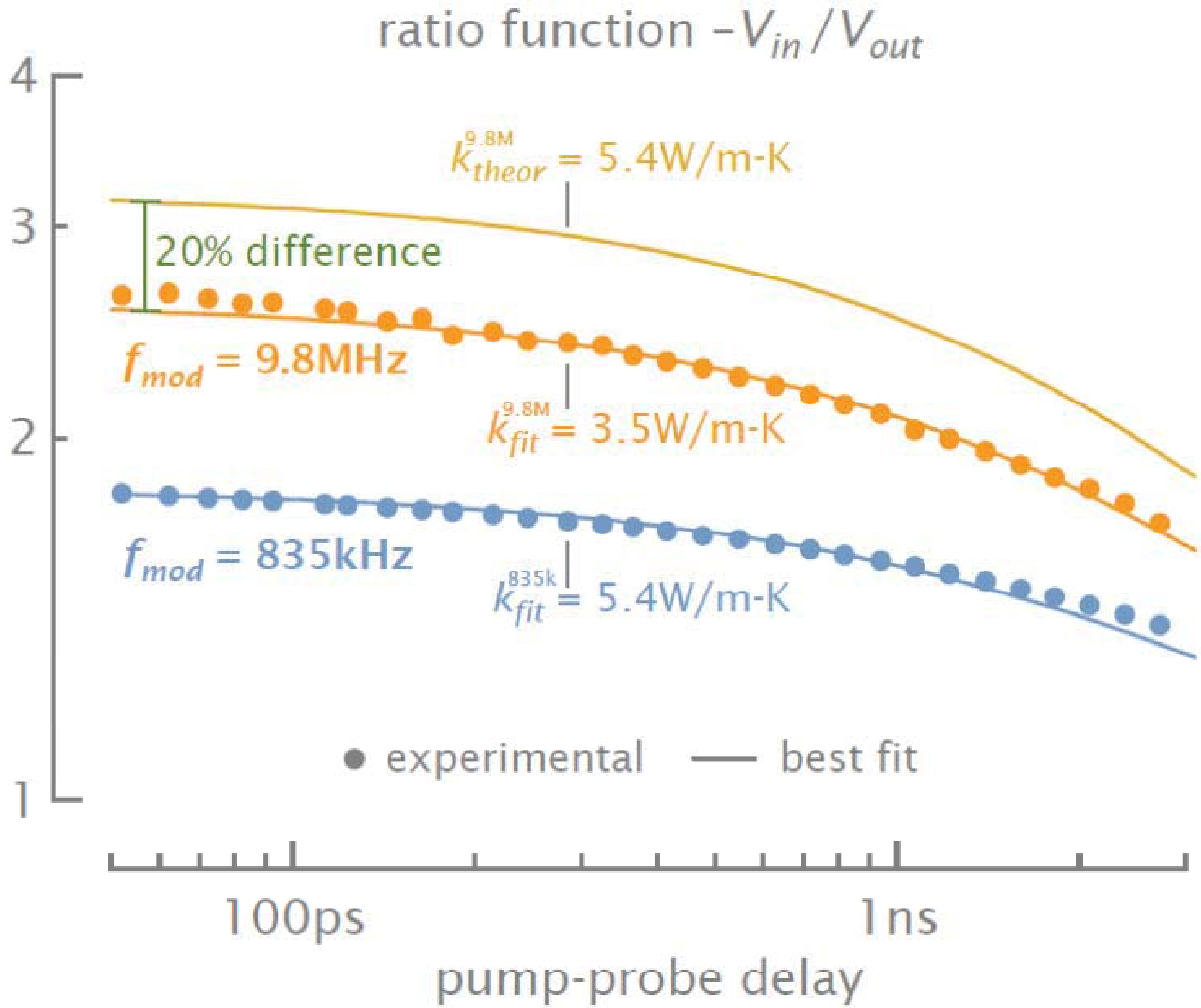
Gilles Pernot (UCSC/Bordeaux)

Optical sampling by TDTR

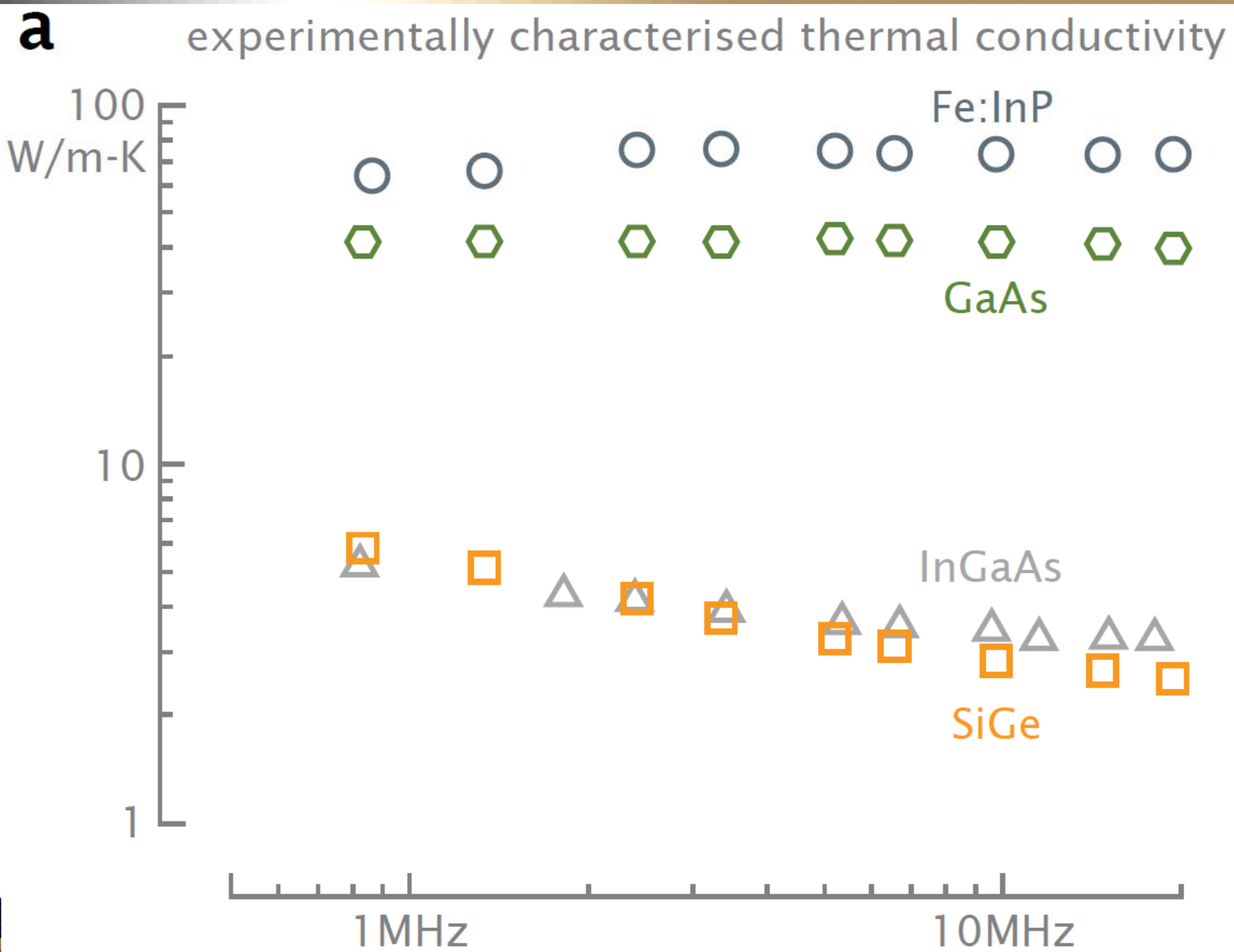


David G. Cahill, *Rev. Sci. Instruments* 75, 5119 (2004)

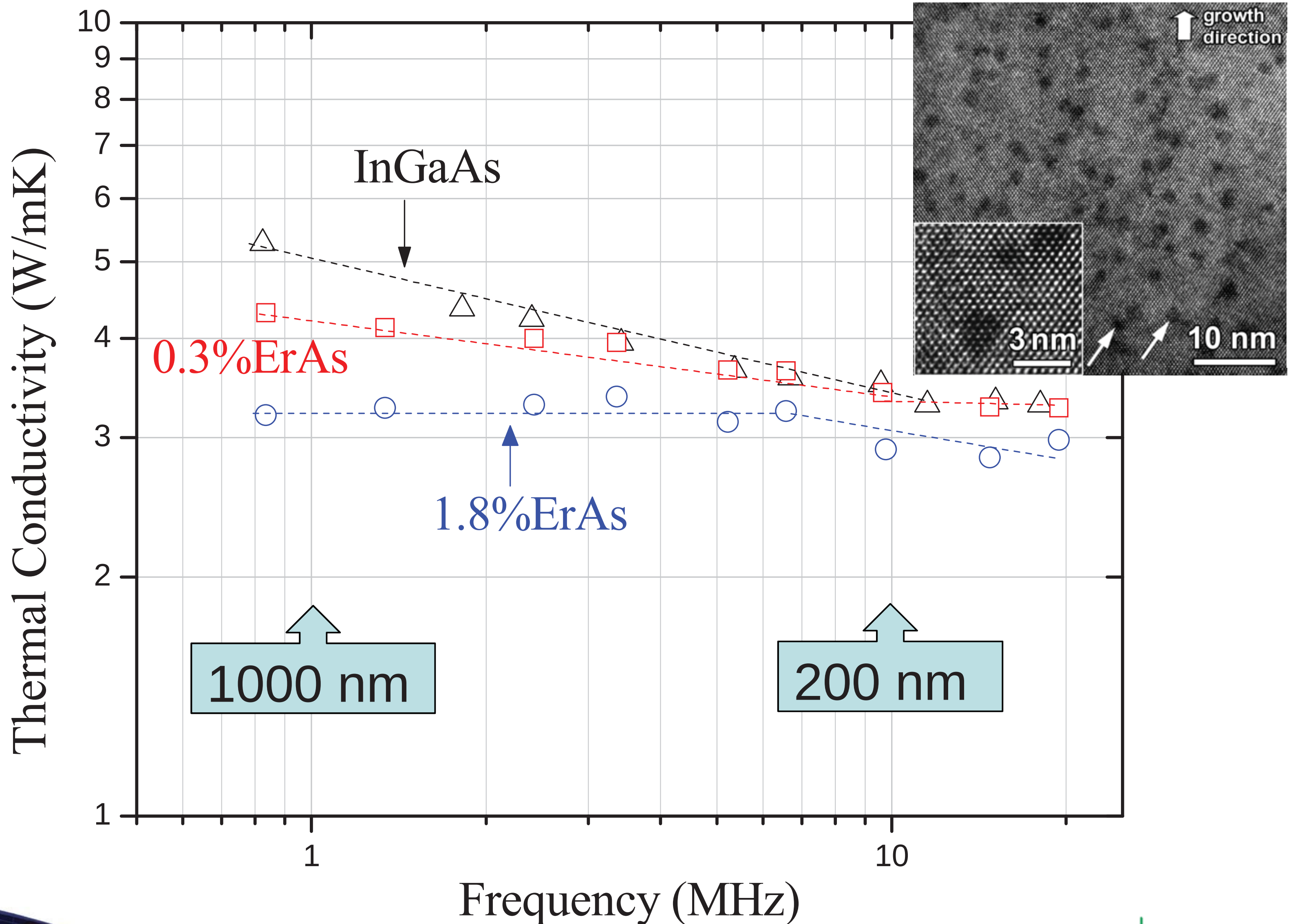
Heat Decay at different modulation frequencies



Effective thermal conductivity at different modulation frequencies

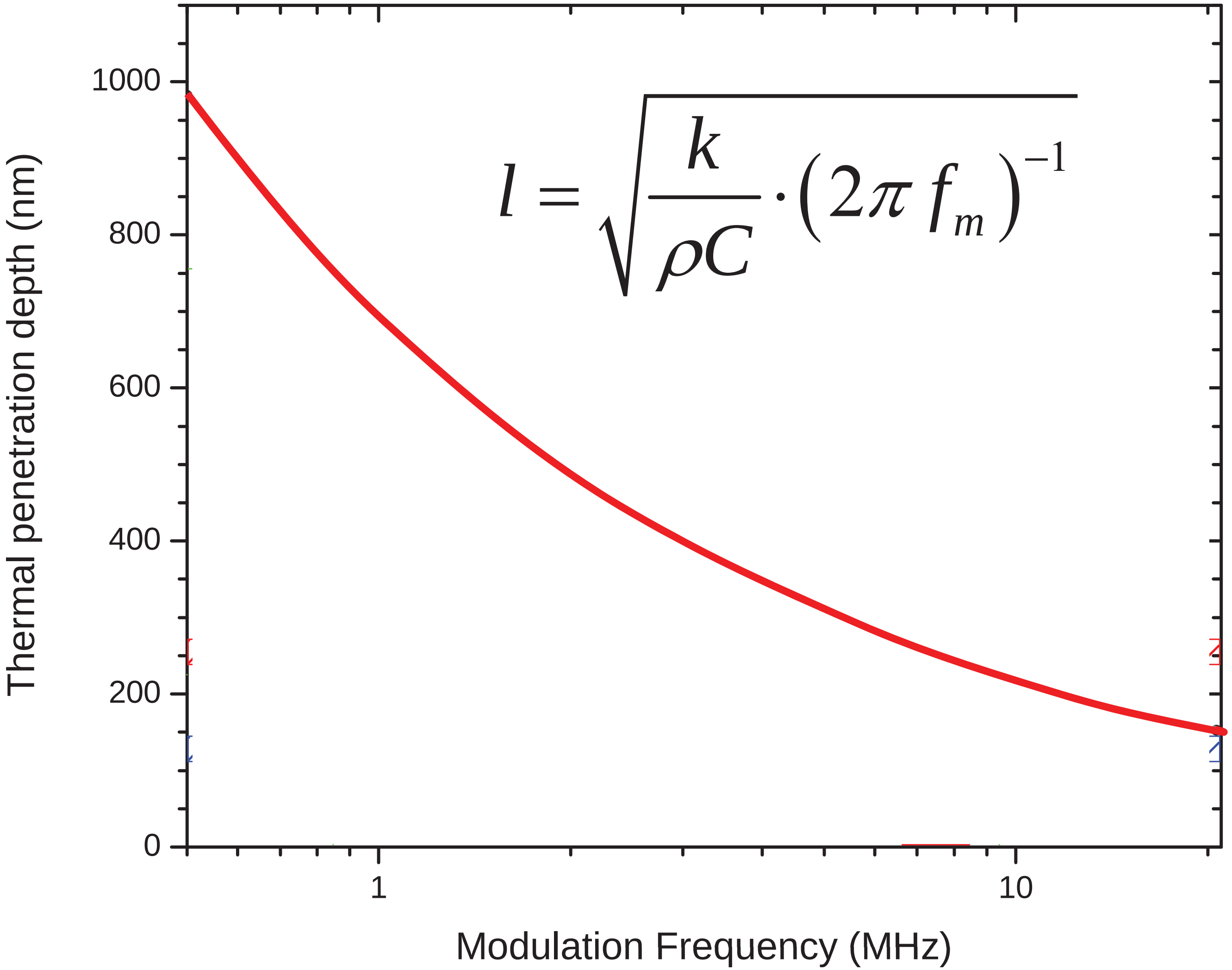


FREQUENCY-DEPENDENT THERMAL CONDUCTIVITY



Gilles Pernot, H. Lu, P. Burke et al., MRS 2012

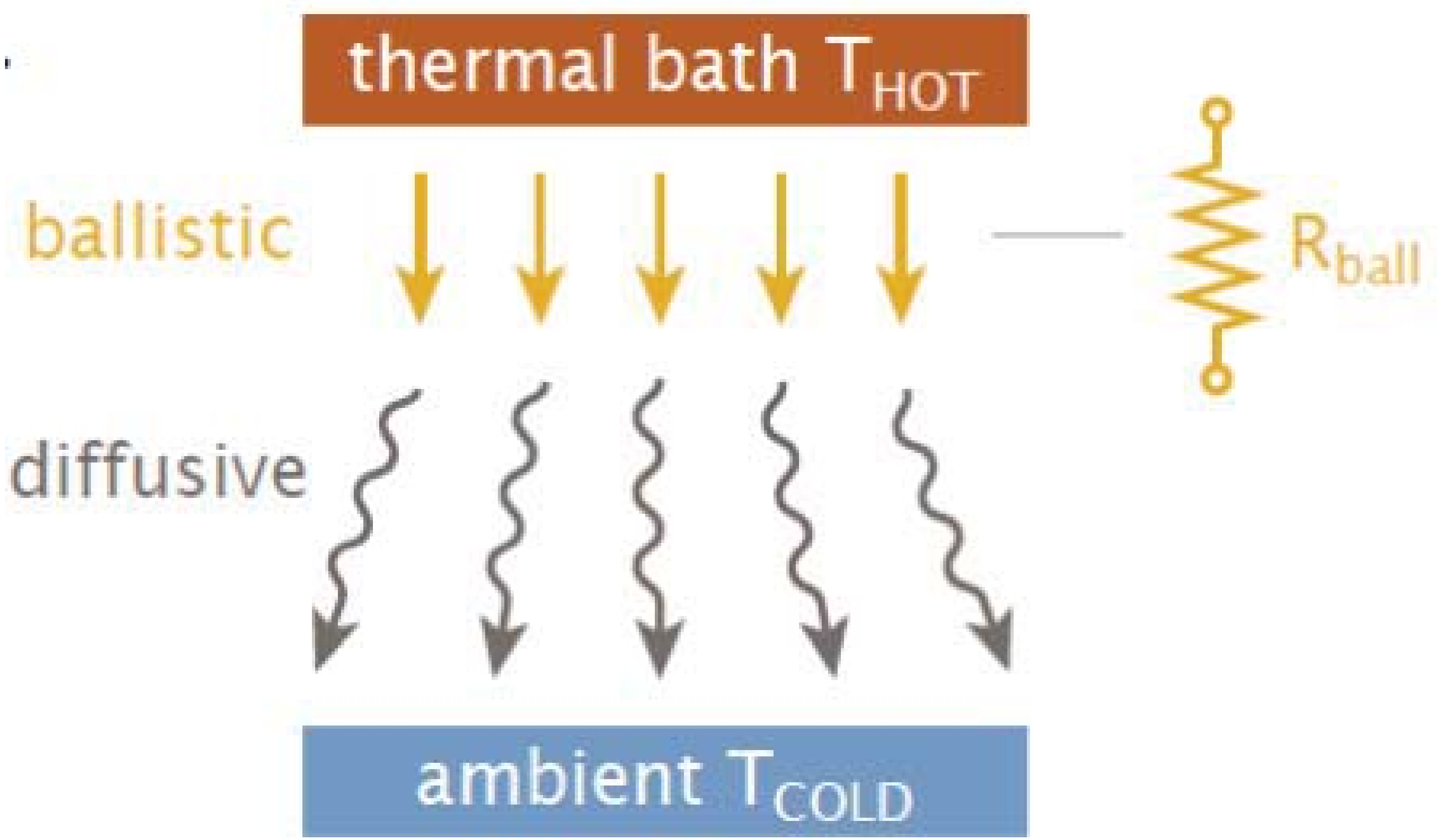
Thermal penetration length



$$l = \sqrt{\frac{k}{\rho C} \cdot (2\pi f_m)^{-1}}$$

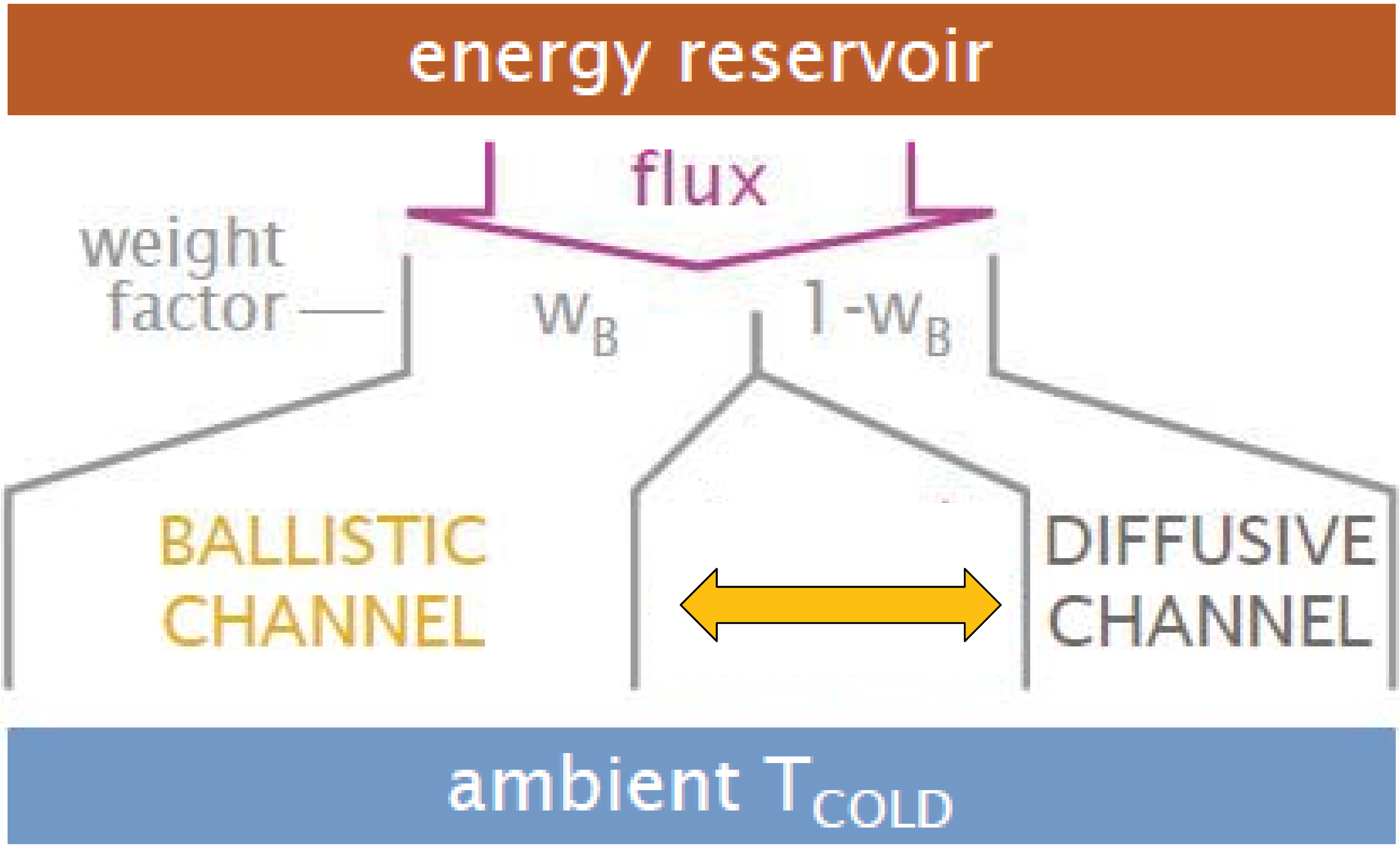
Gilles Pernot (UCSC)

Roll of Ballistic Heat Transport (current theory)



Bjorn Vermeersch, Gilles Pernot, et al. (to be submitted)

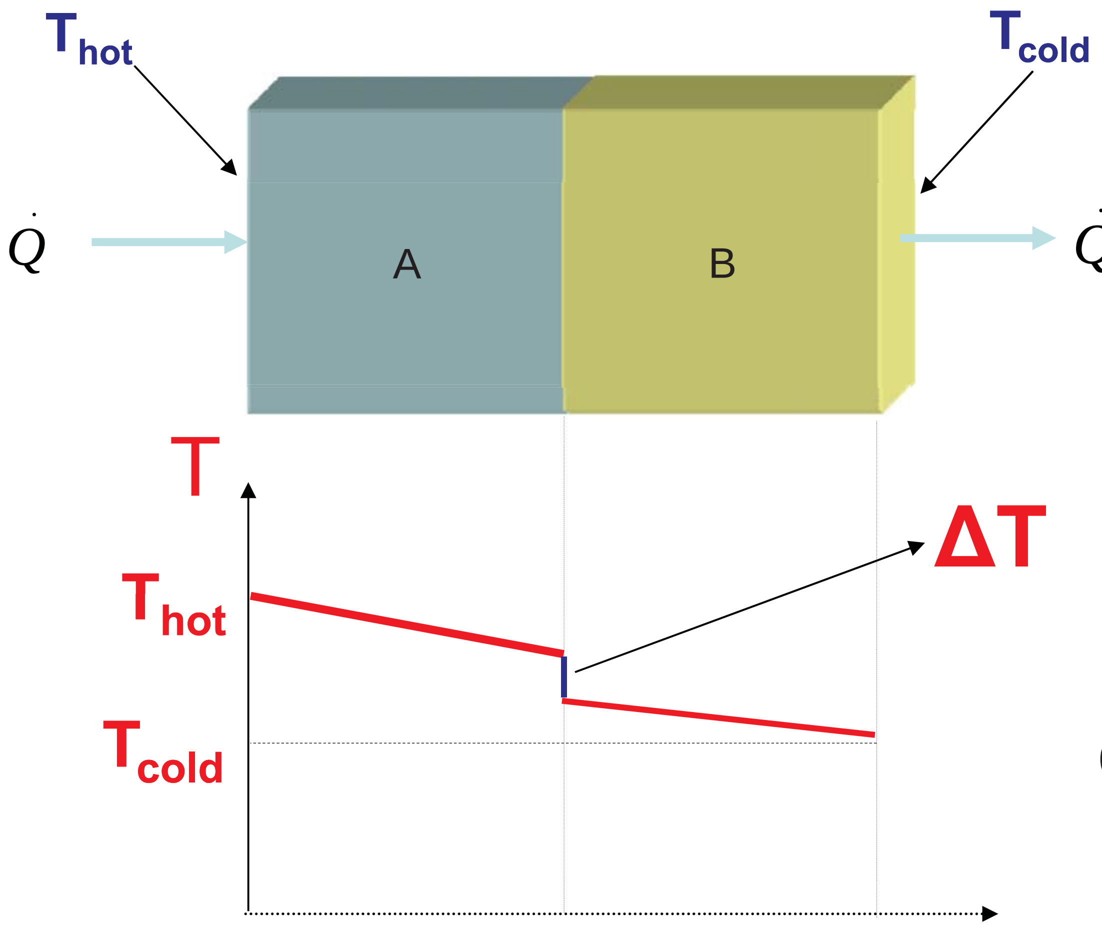
Ballistic/Diffusive Heat Transport



Alexei Maznev et al. PRB 2011

Bjorn Vermeersch, Gilles Pernot, et al. (to be submitted)

Thermal Boundary Conductance

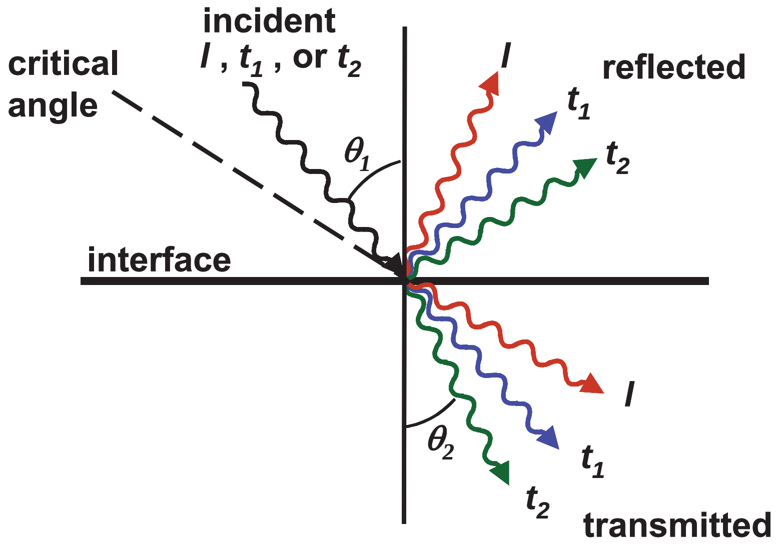


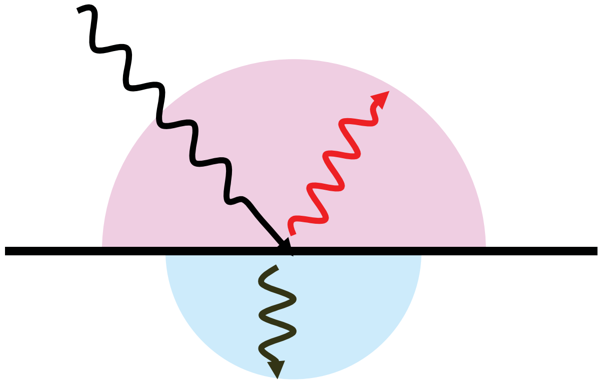
$$h = \frac{\dot{Q}}{A \Delta T}$$

h= Thermal boundary conductance

1/h = Thermal boundary resistance

Transmission Probabilities





Acoustic Mismatch Model (AMM)

Khalatnikov (1952)

$$\alpha_{1\to 2} = \frac{4Z_1 Z_2}{(Z_1 + Z_2)^2}$$

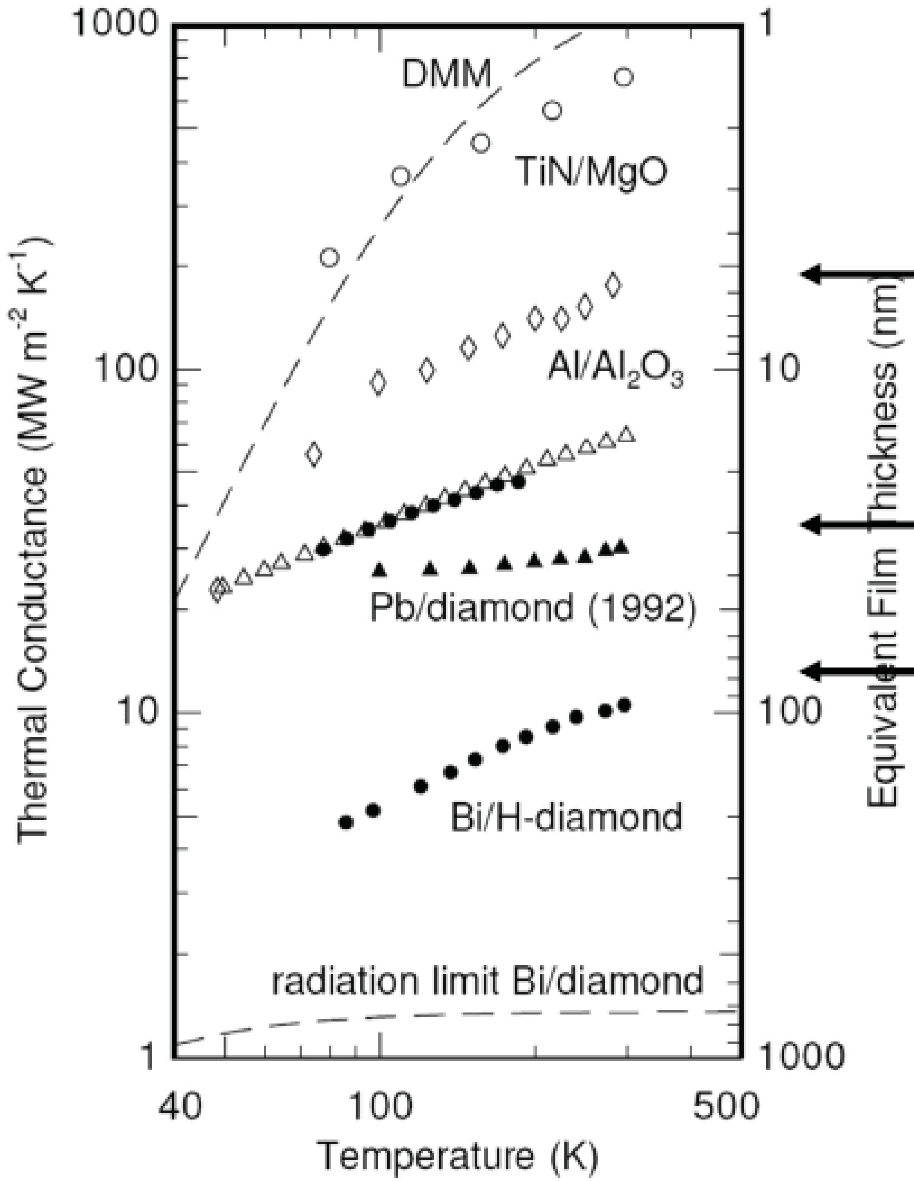
$$Z_i = \rho_i c_i$$

Diffuse Mismatch Model (DMM)

Swartz and Pohl (1989)

$$\alpha_{1\to 2}(\omega) = \frac{[\sum_j N_{2,j}(\omega) c_{2,j}(\omega)]}{[\sum_{i,j} N_{i,j}(\omega) c_{i,j}(\omega)]}$$

Interface thermal conductance



W/Al_2O_3

Au/water

$PMMA/Al_2O_3$

nanotube/alkane

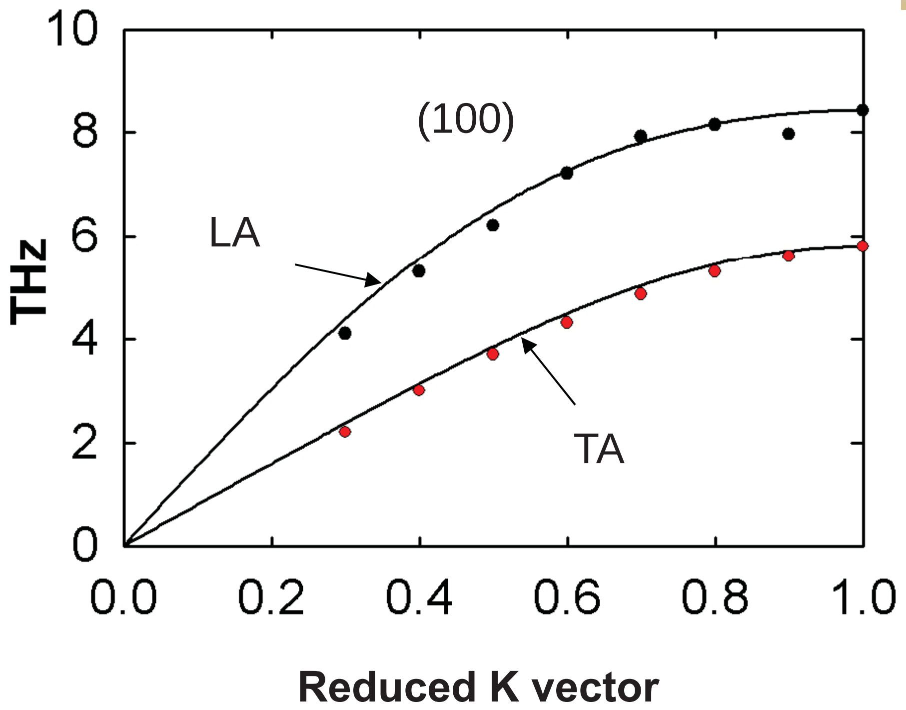
$L = \Lambda/G$

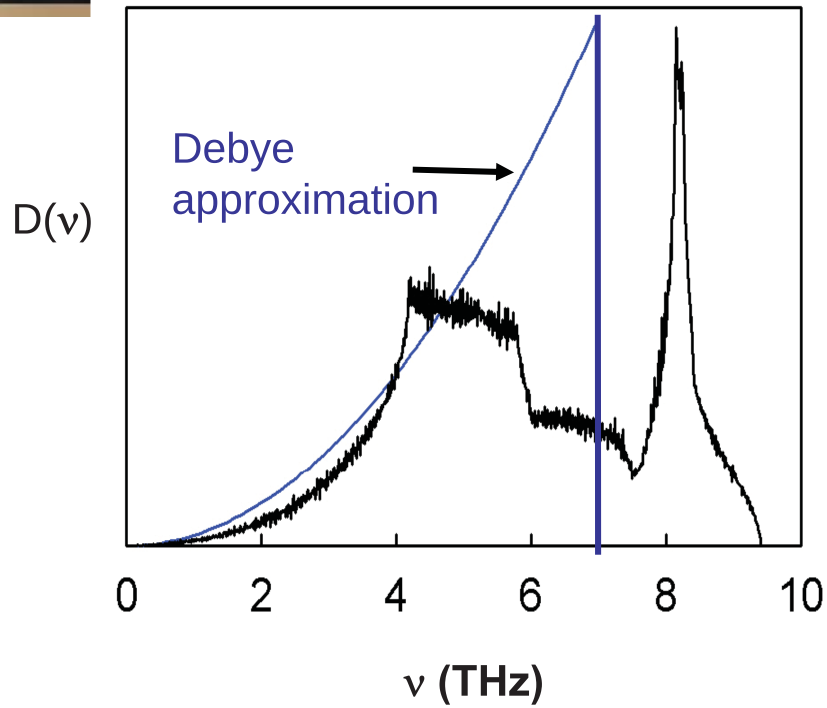
$\Lambda = 1\ W\ m^{-1}\ K^{-1}$

David Cahill (UIUC)

QUANTUM ELECTRONICS

Generalized DMM





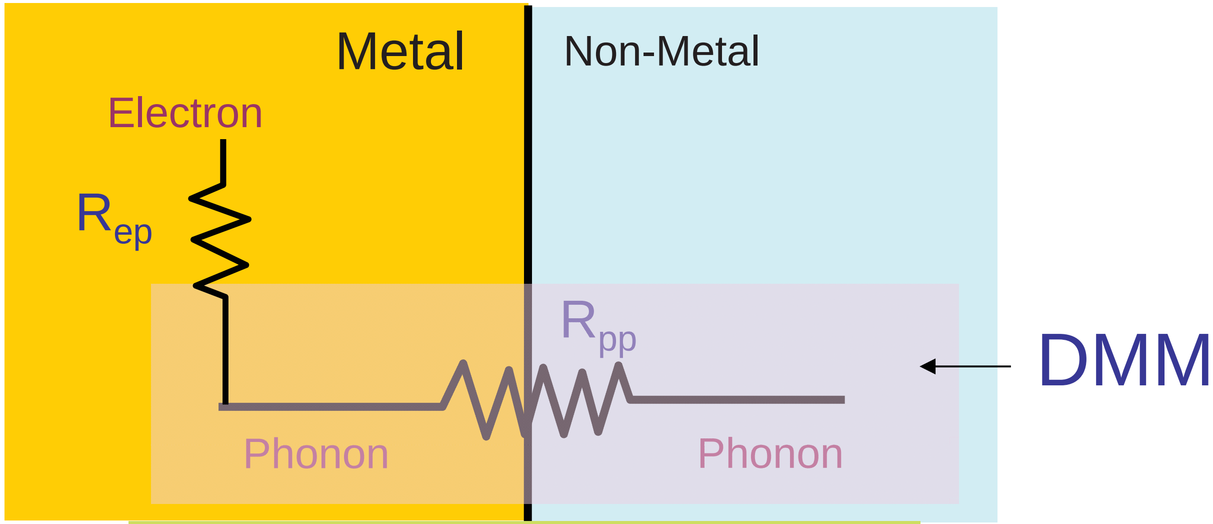
$$\dot{Q}_{1\to 2} = \frac{1}{2}\sum_{j}\int_{0}^{(\pi/2)}\int_{0}^{\omega_{max}} \underbrace{(\hbar\omega)}_{Phonon\ Energy}\ \underbrace{\alpha_{1\to 2}(\theta, j, \omega)}_{Transmissivity}\ \underbrace{N_{1,j}(\omega)}_{Phonon\ Density}\ \underbrace{c_{1,j}(\omega)}_{Phonon\ Speed}\ \sin(\theta)\cos(\theta)\,d\theta\,d\omega$$

$$h = \frac{\dot{Q}_{1\to 2}(T_1) - \dot{Q}_{1\to 2}(T_2)}{A(T_1 - T_2)}$$

DMM based on a real dispersion relation and a real density of states

Majumdar et al. UC Berkeley

Thermal Resistance of Metal-Nonmetal Interfaces



$$h \approx \frac{h_{ep} h_{pp}}{h_{ep} + h_{pp}}; \quad h_{ep} = \sqrt{G k_p}$$

$$R = R_{ep} + R_{pp}$$

Phonon thermal conductivity of metal

$$k_p = \frac{1}{3} C_p v_p \ell_p$$

$$v_p \propto \sqrt{\frac{E}{\rho}}$$

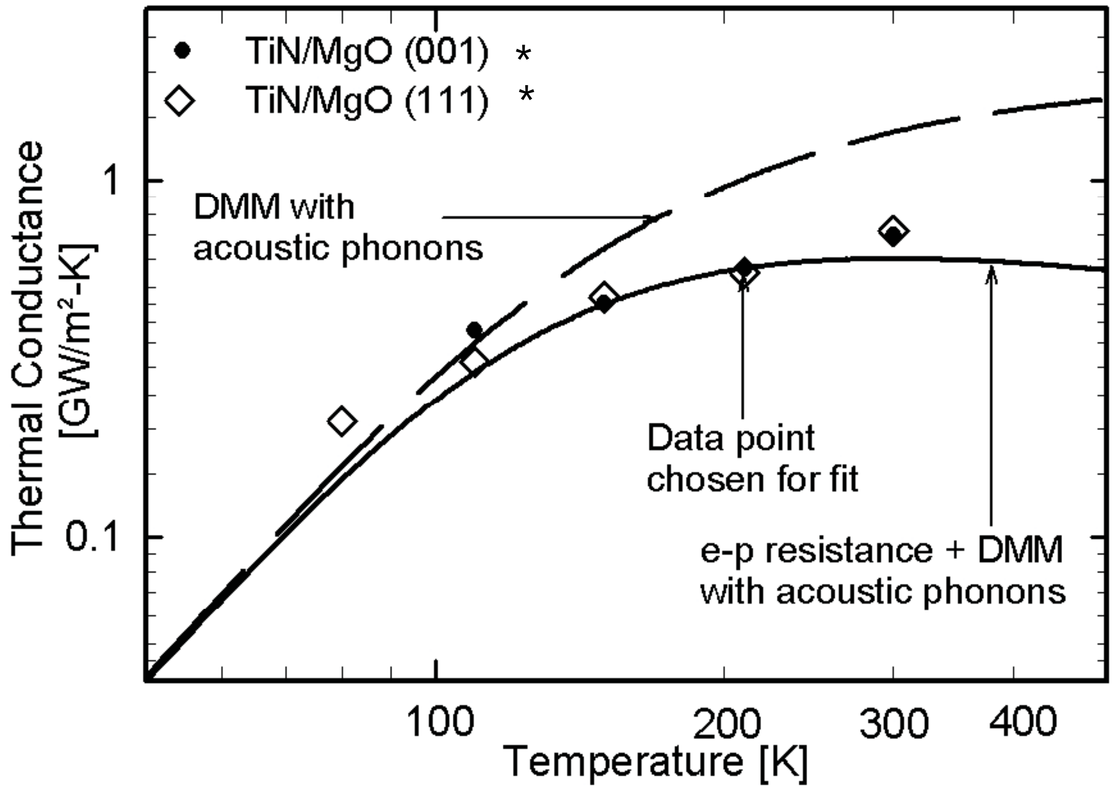
$$h_{ep} \propto \frac{1}{\sqrt{T}}$$

$$G \approx 10^{16} - 10^{17} \frac{W}{m^3 K}; \quad k_p \approx 10 - 20 \frac{W}{mK}$$

$$h_{ep} \approx 0.3 - 1.4 \frac{GW}{m^2 K}$$

Thermal Boundary Resistance of Metal-Nonmetal Interfaces

PURDUE UNIVERSITY Discovery Park



$$h \propto \frac{1}{\sqrt{T}}$$

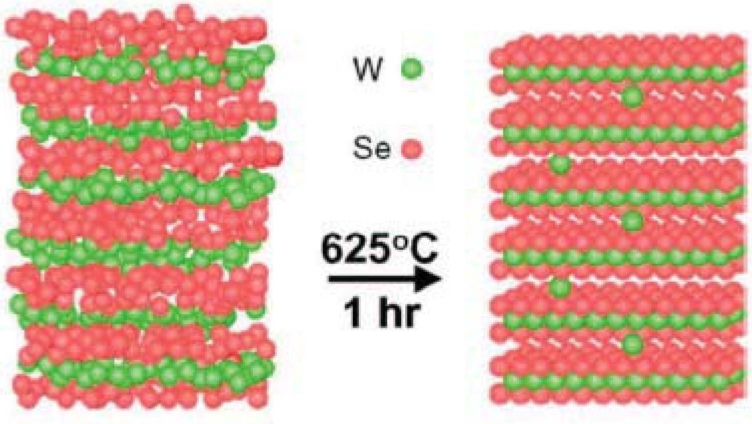
APPLIED PHYSICS LETTERS VOLUME 84, NUMBER 23 7 JUNE 2004

Role of electron–phonon coupling in thermal conductance of metal–nonmetal interfaces

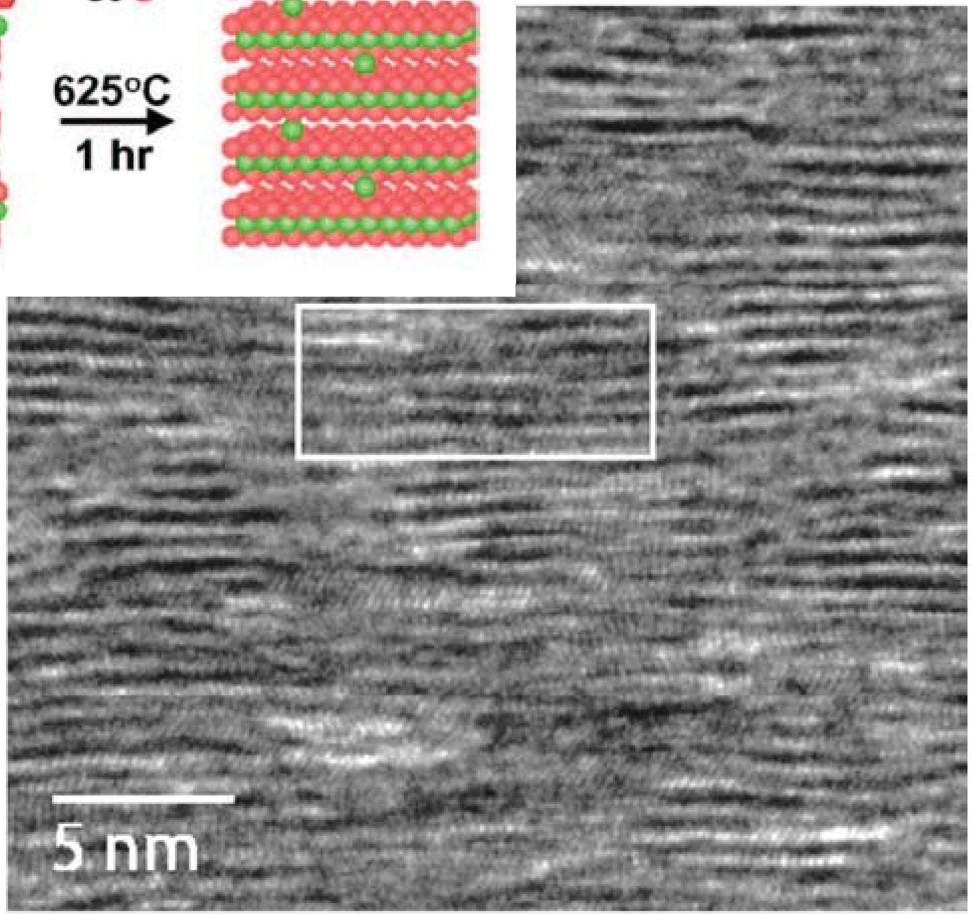
Arun Majumdar[a)]
Department of Mechanical Engineering and Materials Science Division, Lawrence Berkeley National Laboratory, University of California, Berkeley, California 94720

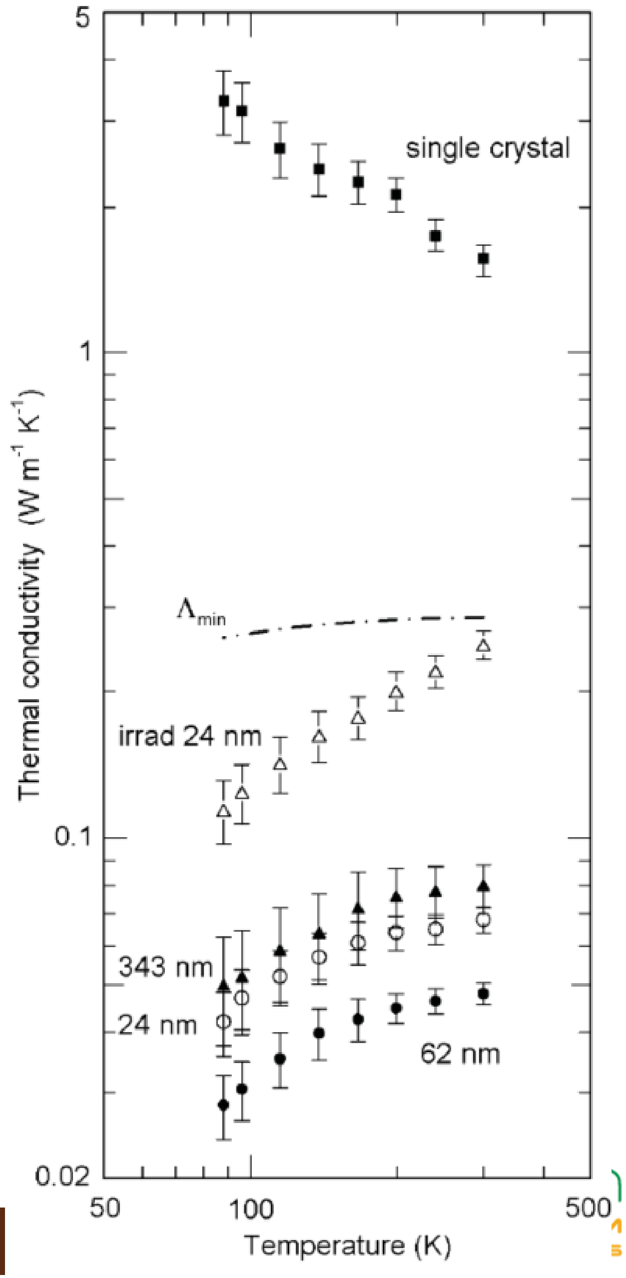
Pramod Reddy
Applied Science and Technology Program, University of California, Berkeley, California 94720

Extremely low thermal conductivity



WSe_2

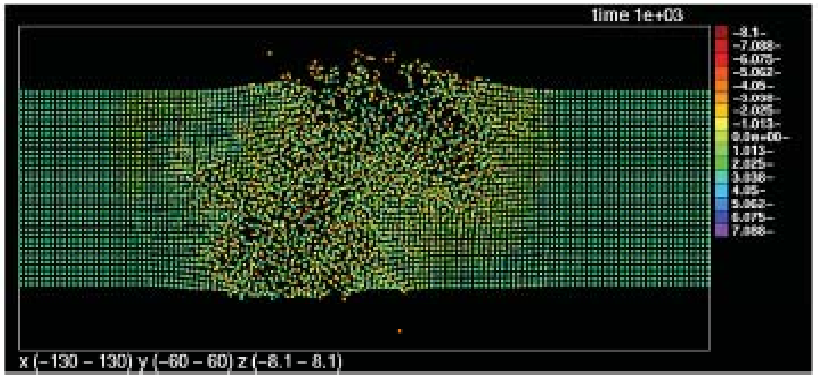




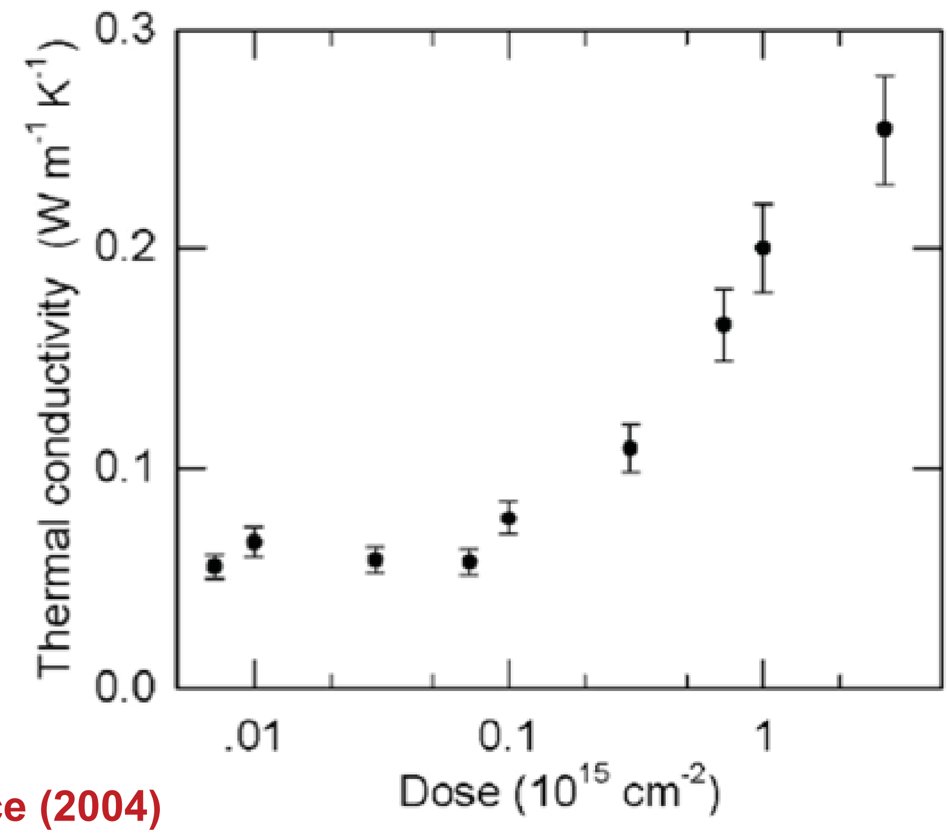
D. Cahill, D. Johnson, et al. Science (2004)

Ion irradiation of WSe_2

MD simulation of 1 MeV Kr impact on Au



- Heavy ion irradiation (1 MeV Kr^+) of 24 nm WSe_2 film.
- Novel behavior: ion damage causes the thermal conductivity to *increase*.



D. Cahill, D. Johnson, et al. Science (2004)

Summary

- Ballistic-diffusive heat transport
  - Cattaneo’s and Shastry’s formalisms
  - Second sound
- Phonon mean free paths
- Time-domain thermoreflectance
  - Picosecond acoustics
  - Frequency-dependent thermal conductivity
- Interface thermal resistance (DMM, AMM, experimental results)

Cahill, Ford, Goodson, Mahan, Majumdar, Maris, Merlin, and Phillpot, "Nanoscale thermal transport," *J. Appl. Phys.* 93, 793 (2003)

David G. Cahill, *Rev. Sci. Instrum.* 75, 5119 (2004)

Y. Ezzahri and A. Shakouri; *Phys. Rev. B,* 79, 184303, (2009)

Acknowledgement

PURDUE UNIVERSITY Discovery Park

Research Professors: Zhixi Bian, Kaz Yazawa

Postdocs/Graduate Students: Kerry Maize, Hiro Onishi, Tela Favaloro, Phil Jackson, Oxana Pantchenko, Amirkoushyar Ziabari, Bjorn Vermeersch, Je-Hyeong Bahk, Yee Rui Koh

Collaborators: John Bowers, Art Gossard (UCSB), Tim Sands, Yue Wu (Purdue), Rajeev Ram (MIT), Venky Narayanamurti (Harvard), Arun Majumdar (Berkeley/ARPA-E), Josh Zide (Delaware), Lon Bell (BSST), Yogi Joshi, Andrei Federov (Georgia Tech), Kevin Pipe (Michigan), Stefan Dilhaire (Bordeaux), Natalio Mingo (CEA), Mike Isaacson, Sriram Shastry, Joel Kubby, Ronnie Lipschutz, Melanie Dupuis, Ben Crow, Steve Kang (UCSC), Bryan Jenkins, Susan Kauzlarich (Davis)

Alumni: Younes Ezzahri (Prof. Univ. Poitier), Daryoosh Vashaee (Prof. Oklahoma State), Zhixi Bian (Adj. Prof. UCSC), Mona Zebarjadi (Prof. Rutgers), Yan Zhang (Tessera), Rajeev Singh (PV Evolutions), James Christofferson (Microsanj), Kazuhiko Fukutani (Canon), Je-Hyoung Park (Samsung) , Javad Shabani (postdoc, Harvard), Xi Wang (InterSil), Helene Michel (CEA), Gilles Pernot (Bordeaux), Ramin Sadeghian (H2scan), Shila Alavi (UCSC ASL), Tammy Humphrey, David Hauser