UVA | SCHOOL of ENGINEERING & APPLIED SCIENCE

Controlling heat transfer through interfacial coupling of photons, electrons and phonons

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- H. Olson (Laser Thermal)
- J. Gaskins (Laser Thermal)
- K. Aryana (NASA)
- R. Cheaito (Google)
- Md. S. B. Hoque (UVA)
- W. Hutchins (UVA)
- T. Pfeifer (ONRL/UVA)

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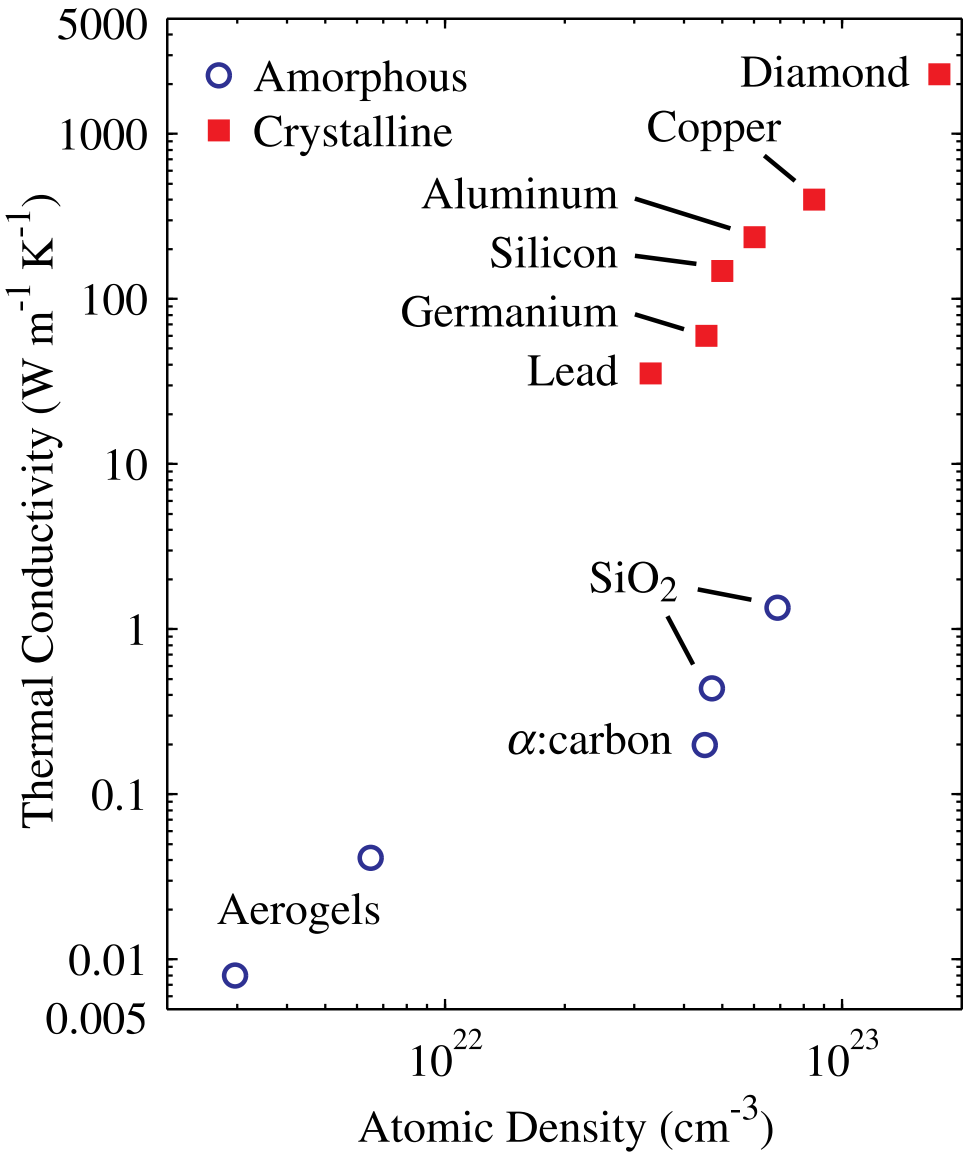
- R. Ramesh (Rice)
- J. Ravichandran (USC)
- C. Nuckolls (Columbia)
- P. Kim (Harvard)
- A. Khan (U. South Carolina)
- J.P. Maria (Penn State)
- S. King (Intel)
- S. Walton (NRL)
- O. Prezhdo (USC)
- E. Runnerstrom (ARO)
- J. Caldwell (Vanderbilt)
- S. Pantelides (Vanderbilt)

E. Hoglund (ORNL) OAK RIDGE National Laboratory J. Hachtel (ORNL)

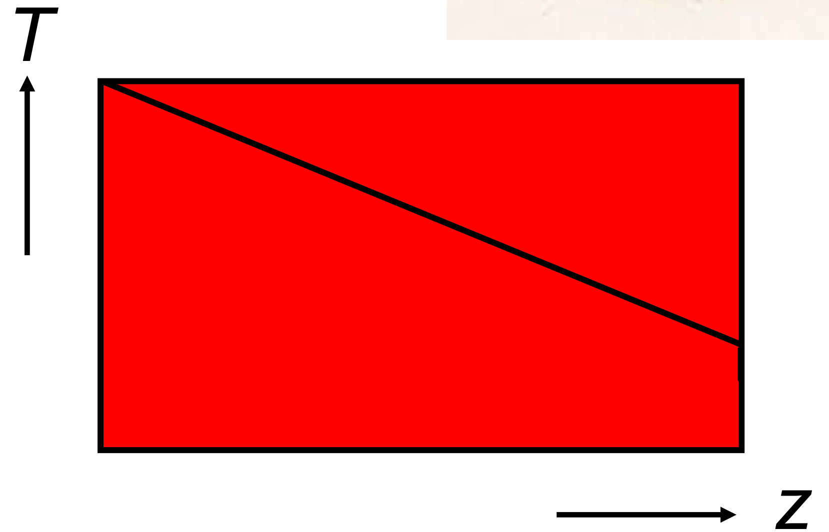
Thermal conductivity of materials – Macroscopic



PRL **110**, 015902 (2013)

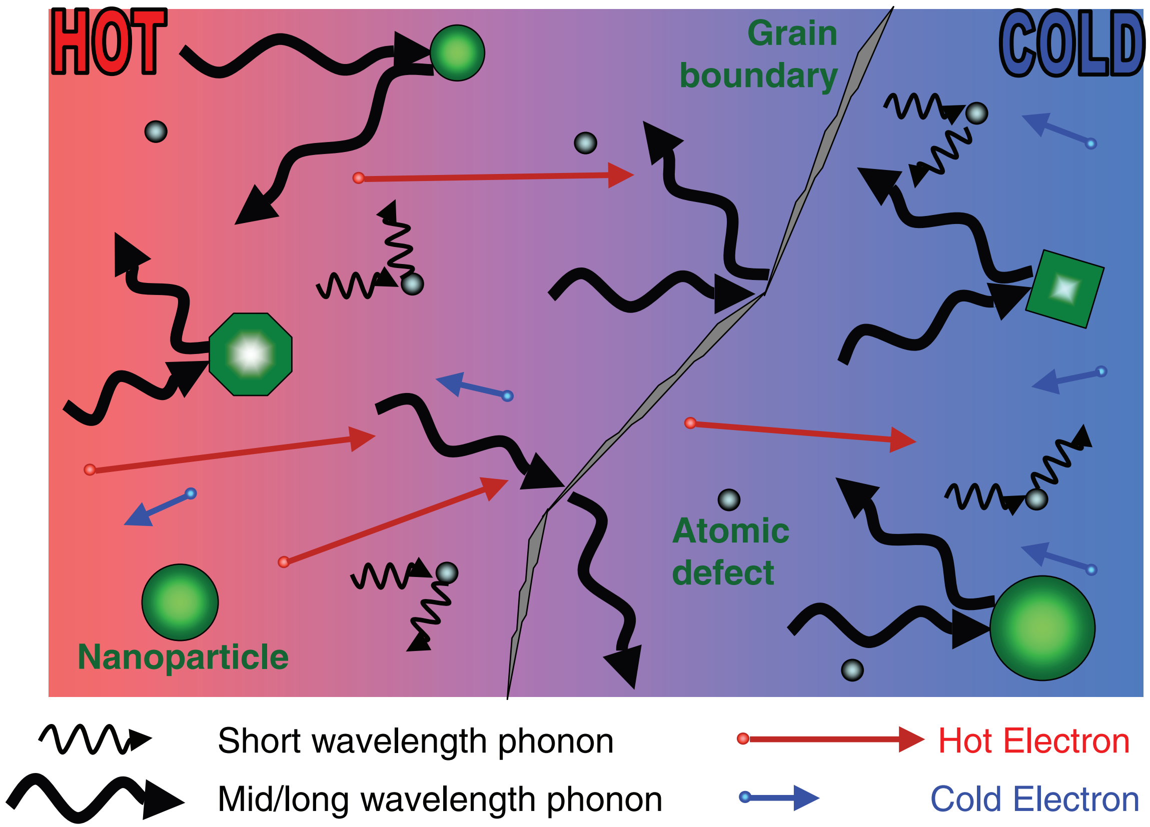
The Fourier Law

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



A nanoscopic view with Kinetic Theory

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



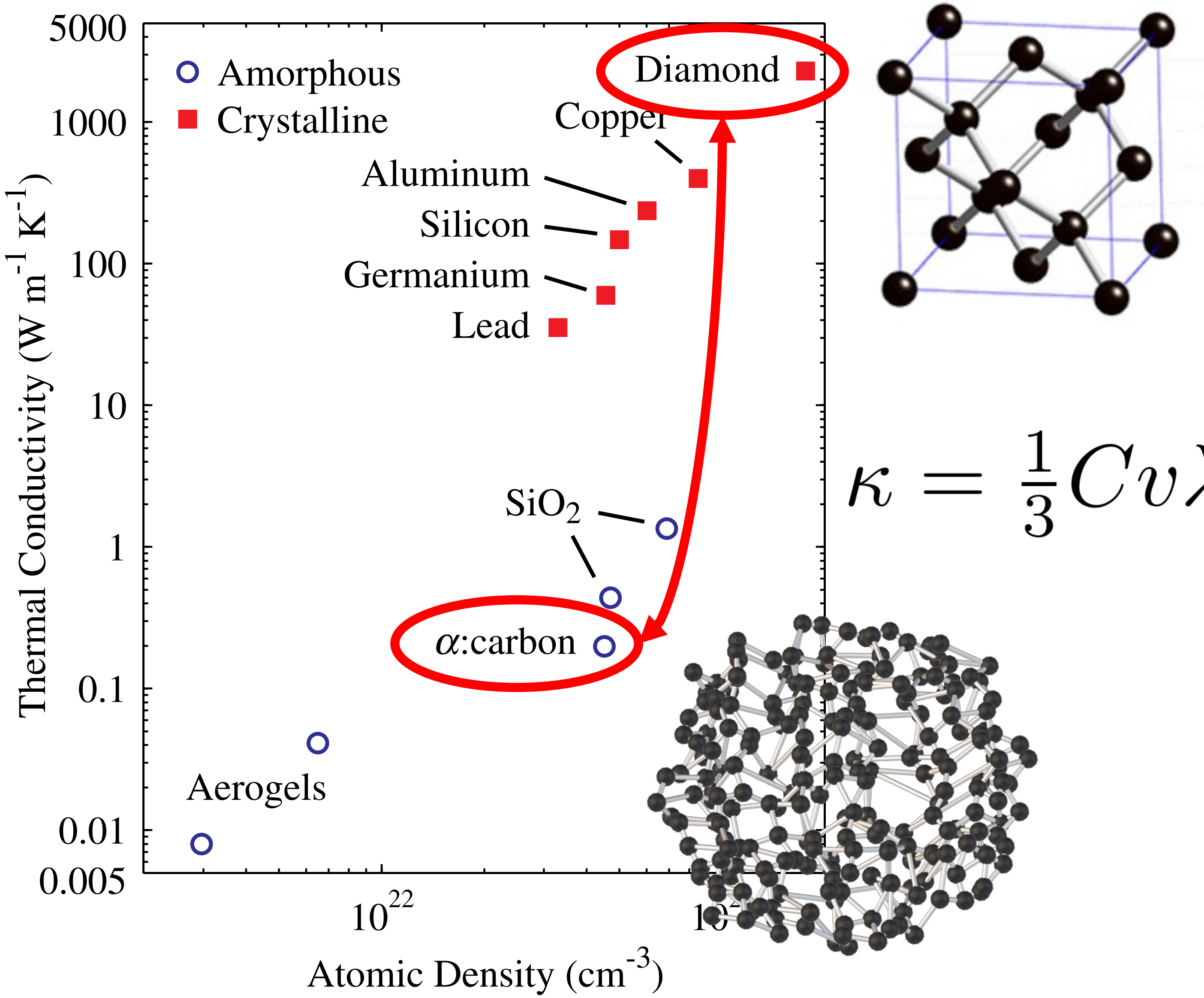
C: Heat capacity
"How much energy electrons/phonons store"

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

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

Adv. Mat. **22**, 3970

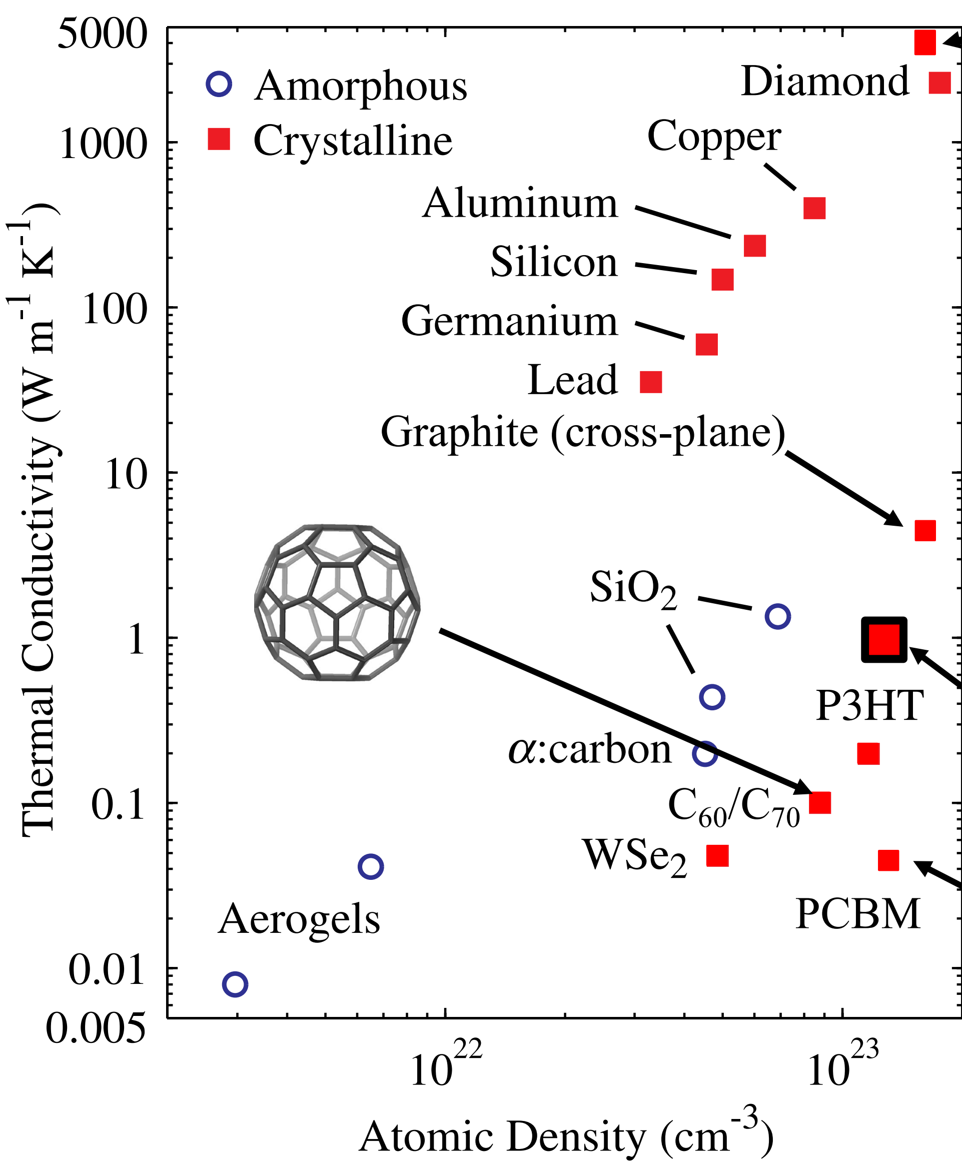
Thermal conductivity of materials – Nanoscopic



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

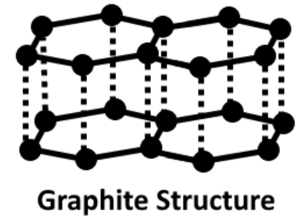
PRL **110**, 015902 (2013)

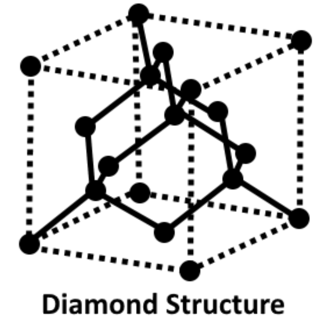
Engineering the thermal conductivity of materials: Nano HX

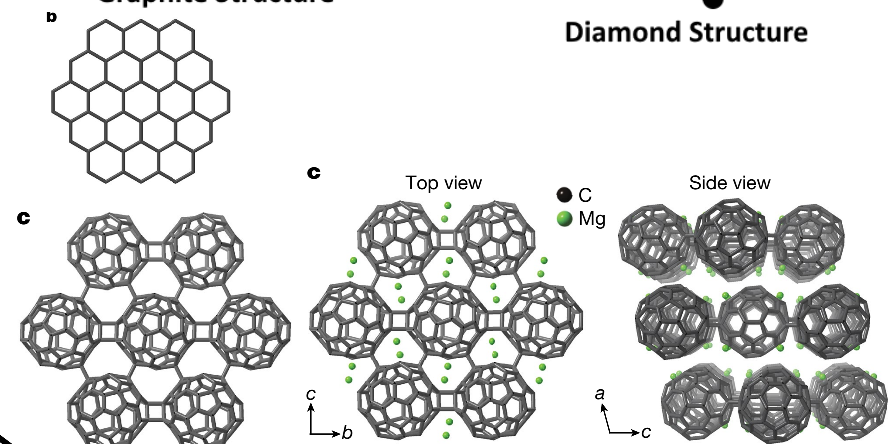


PRL **110**, 015902 (2013)

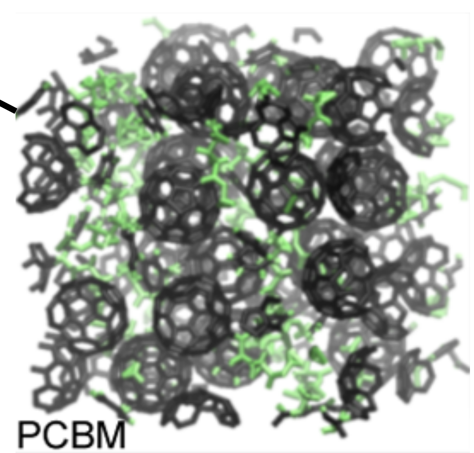
Graphene (in-plane)



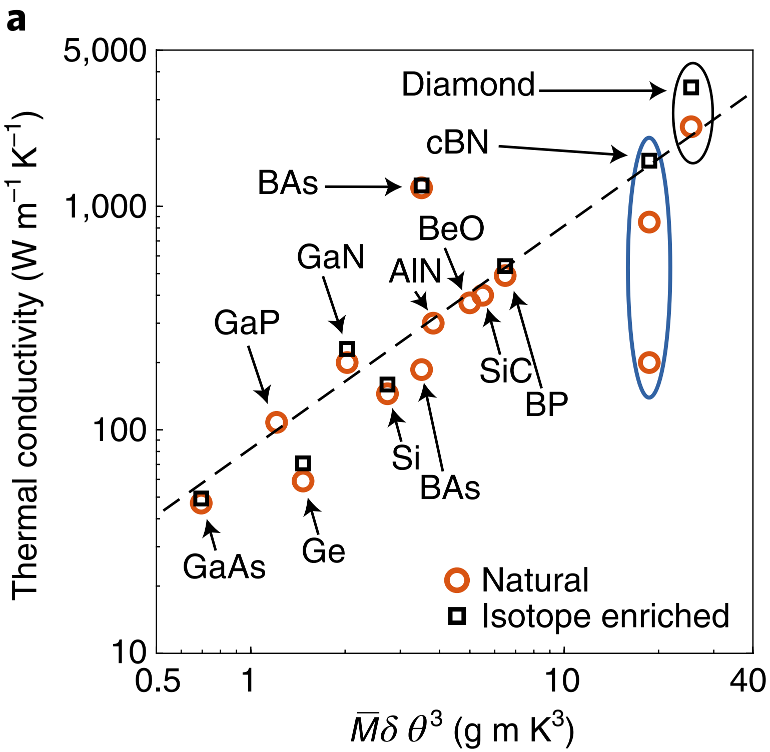




Graphullerene *Nature* 613, 71 (2023)

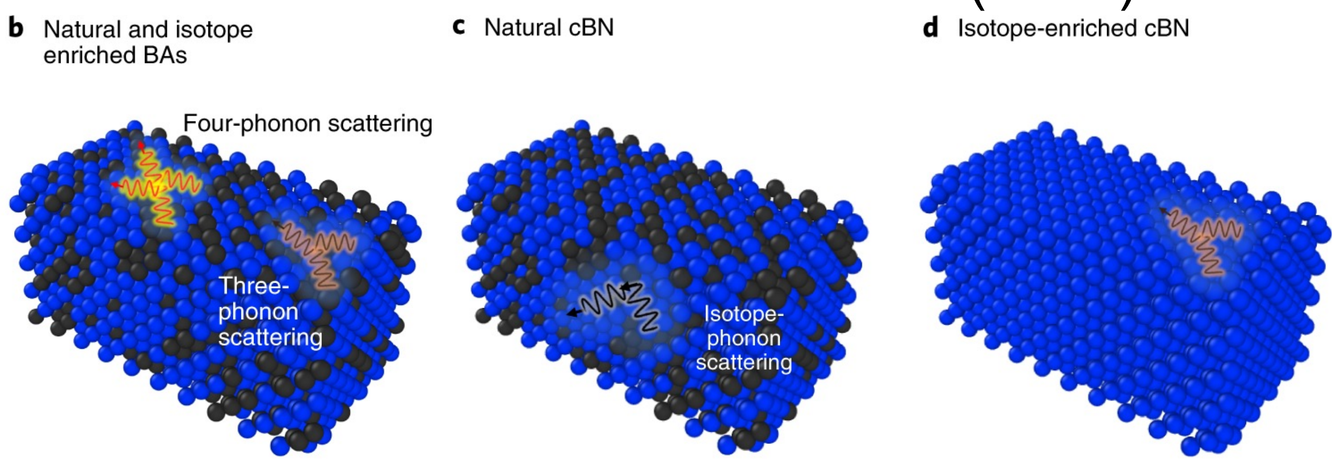


Thermal conductivity of materials: "Engineering defects"



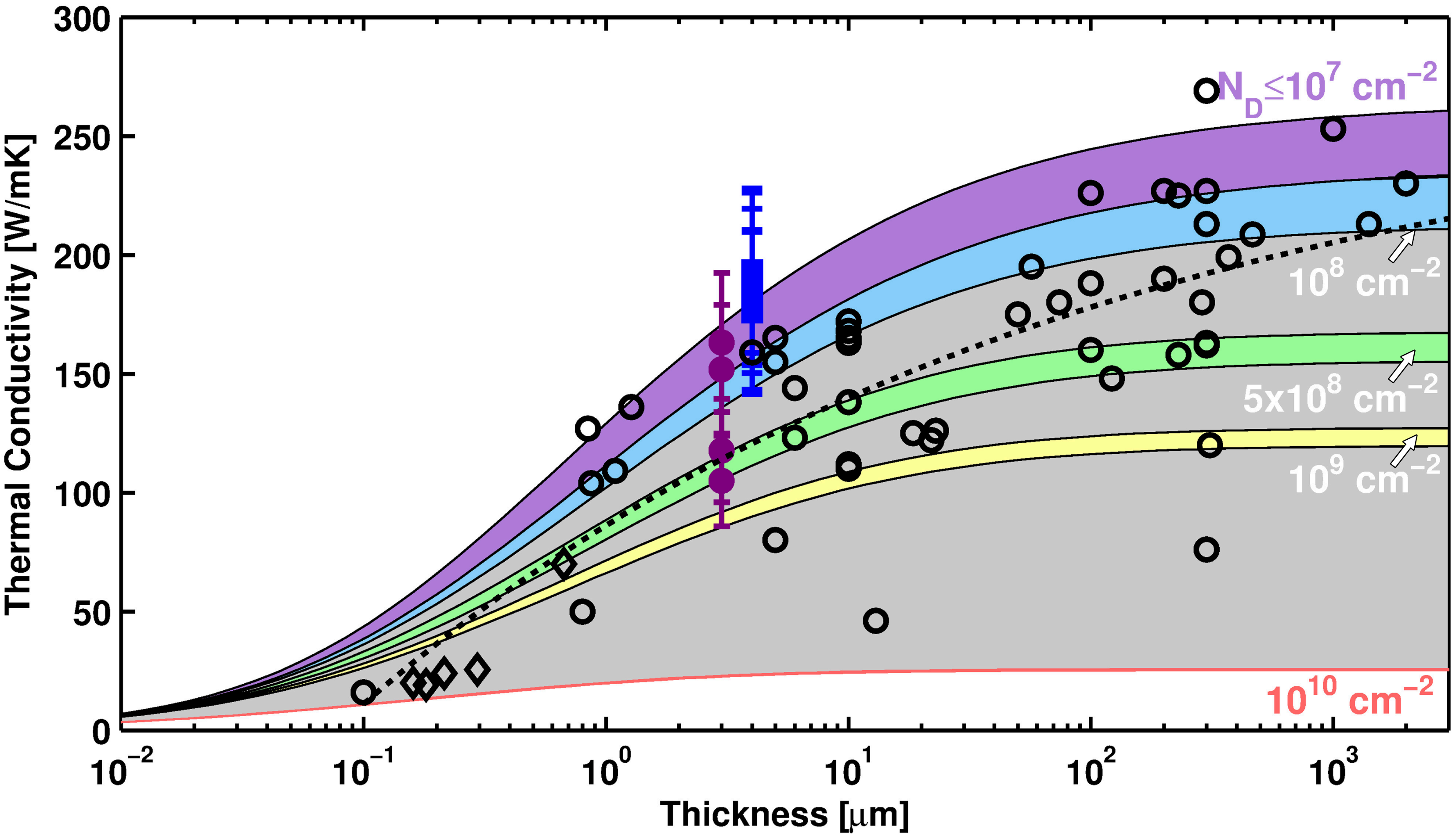
Giri and Hopkins
Nature Materials **19**, 482 (2020)

- Isotope enrichment reduces phonon-defect scattering
- Acoustic phonon bunching and large A-O bandgap reduces phonon-phonon scattering (originally discovered by Lindsay *et al. Phys. Rev. Lett.* **111**, 025901 (2013)

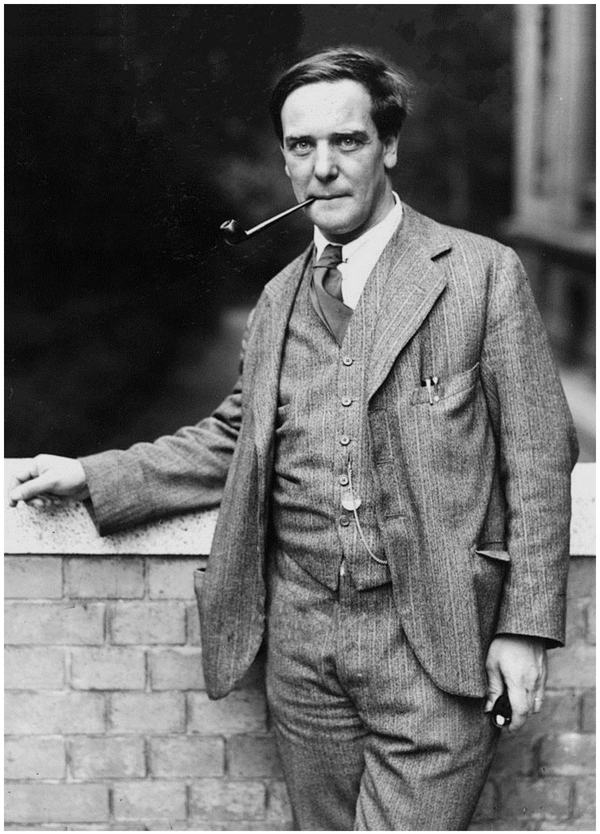


Thermal conductivity of materials: Defects and size effects

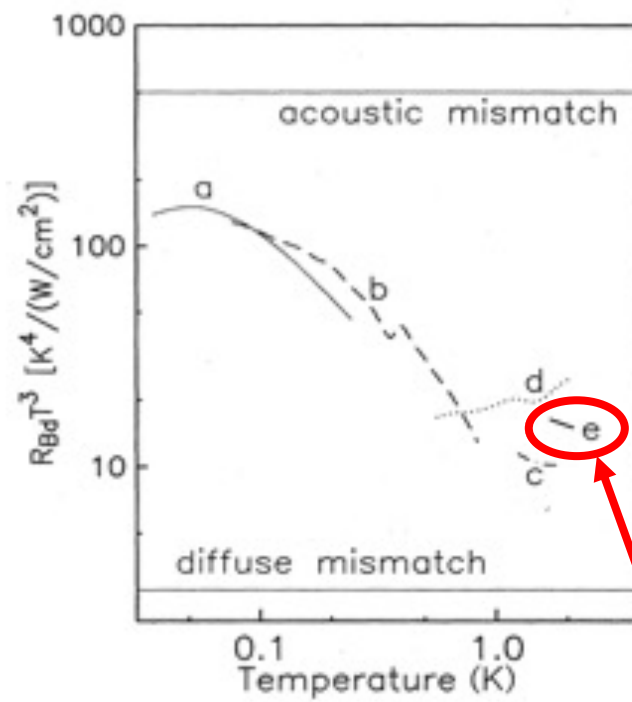
Survey of literature thermal conductivity of GaN
J. Appl. Phys. **120**, 095104



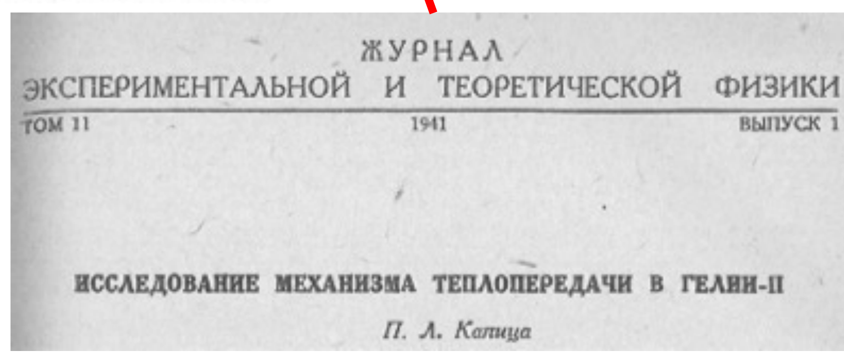
The founding father of the "Kapitza Resistance"



Pyotr Kapitsa



Plot from Fig. 1 in Swartz and Pohl, "Thermal boundary resistance," *Rev. Mod. Phys.* **61**, 605 (1989)



ЖУРНАЛ
ЭКСПЕРИМЕНТАЛЬНОЙ И ТЕОРЕТИЧЕСКОЙ ФИЗИКИ

ТОМ 11 1941 ВЫПУСК 1

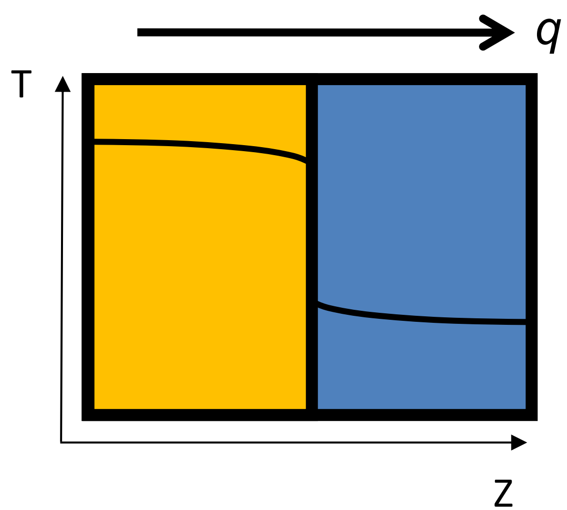
ИССЛЕДОВАНИЕ МЕХАНИЗМА ТЕПЛОПЕРЕДАЧИ В ГЕЛИИ-II

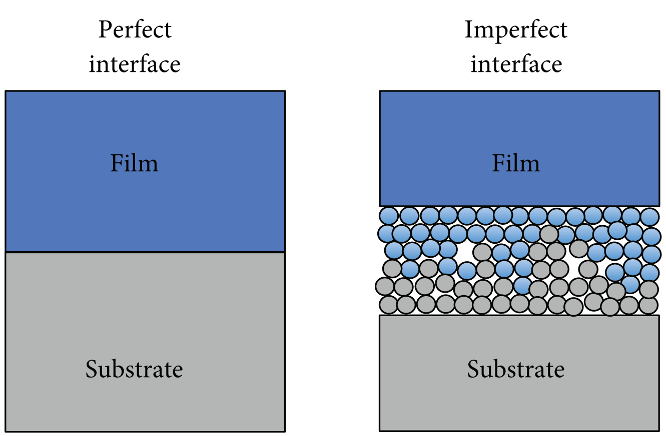
П. Л. Капица

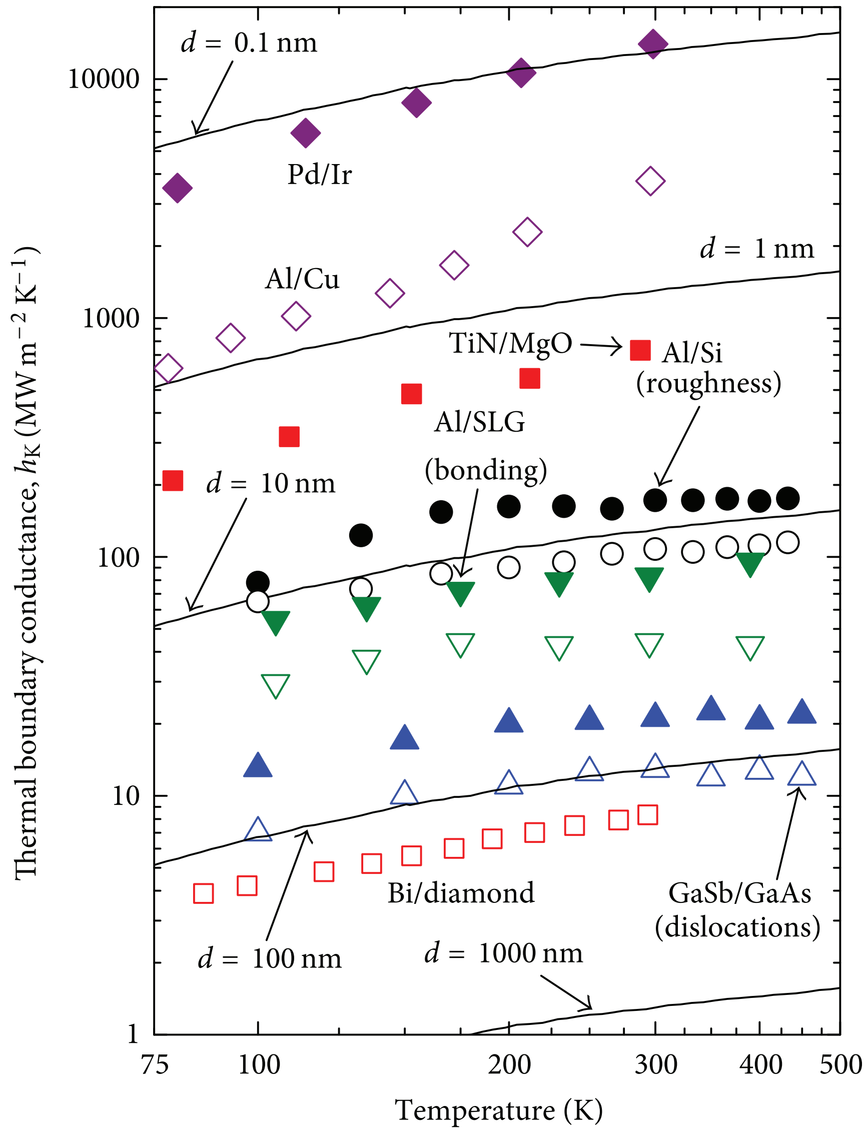
P. L. Kapitza. The study of heat transfer in helium II. Zhurnal eksperimentalnoi i teoreticheskoi fiziki, 11:1–31, 1941.

Thermal boundary conductance – nanoscale resistances

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

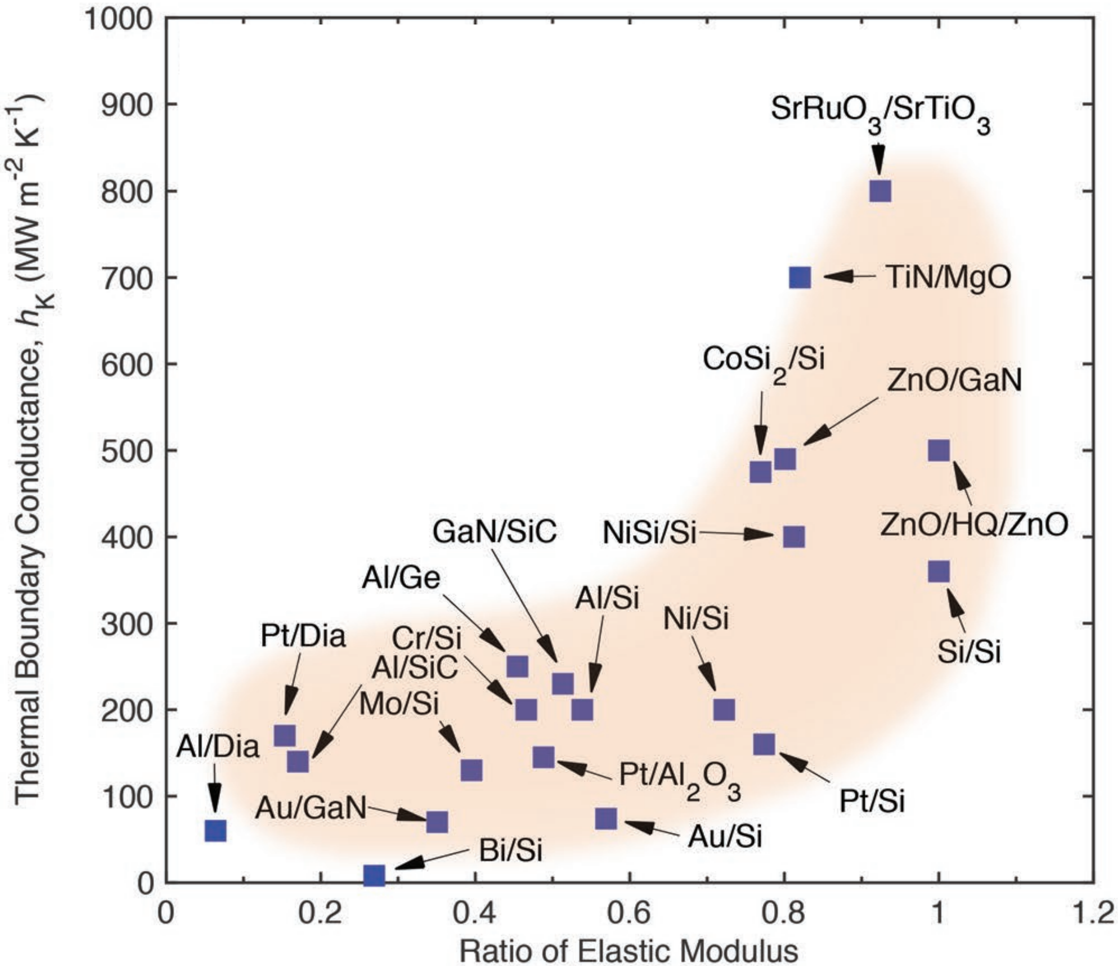






ISRN Mech. Eng. **2013**, 682586

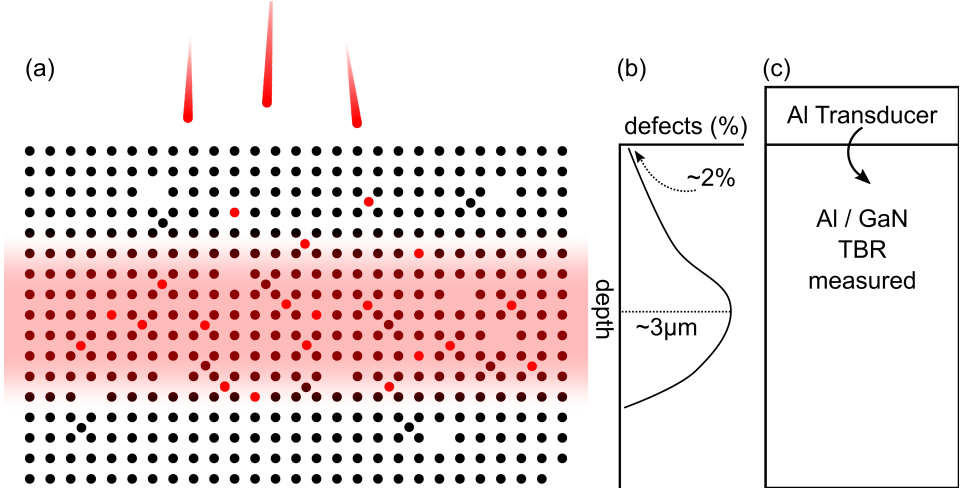
Phonon dominated TBC: metal/nonmetal interfaces

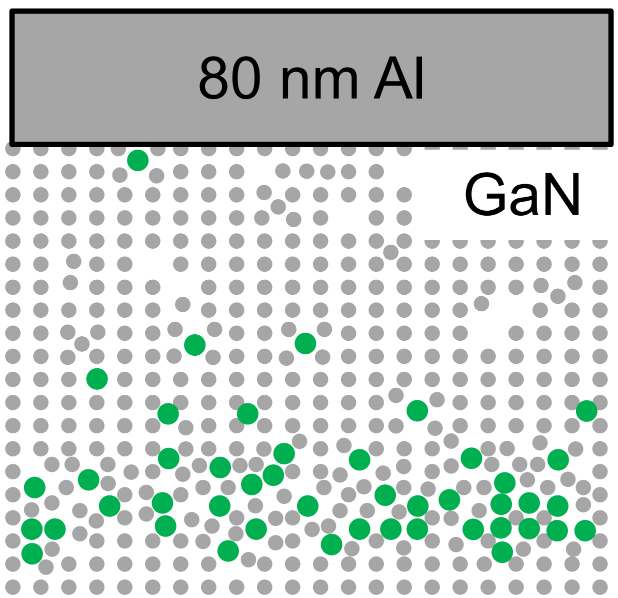


<u>Recent Review</u>

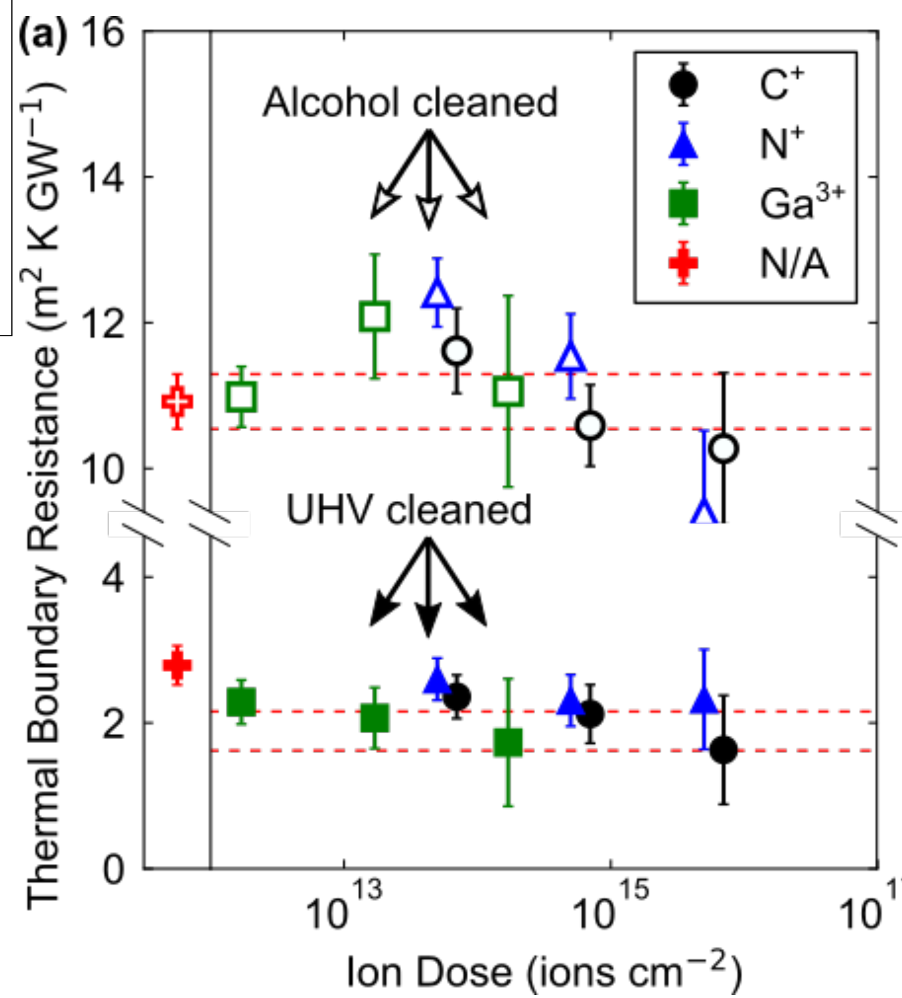
Advanced Functional Materials **30**, 1903857 (2020)

Phonon dominated TBC: "Engineering defects"



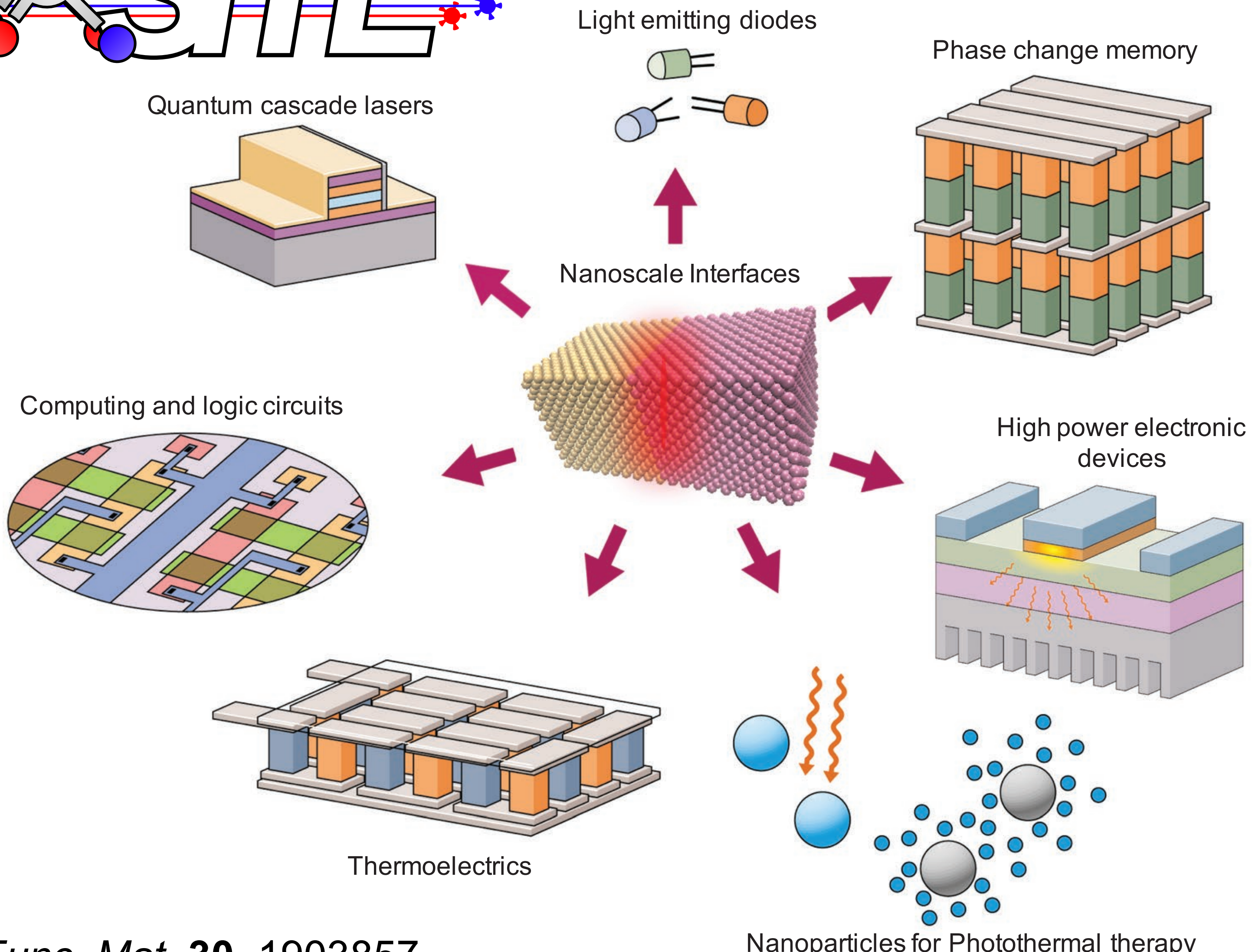


400 keV He+ implants: end of range dep



Pfeifer *et al. Under review*

Thermal properties of nanomaterials and interfaces



Adv. Func. Mat. **30**, 1903857

Outline

Theme: Coupling of carriers across interfaces lead to unique energy states and modes of heat transfer

- How do we measure the thermal conductivity of materials and the thermal boundary resistance at their interfaces?

- Thermal conductivity of superlattices
  - Minimum thermal conductivity and creating “crystals of interfaces”

- Interfacial heat transfer control of the IR properties of solids
  - Near field radiative interfacial heat transfer with plasmon-polaritons and phonon-polaritons

- Transient temperature changes during plasma-surface interactions
  - “Plasma cooling”

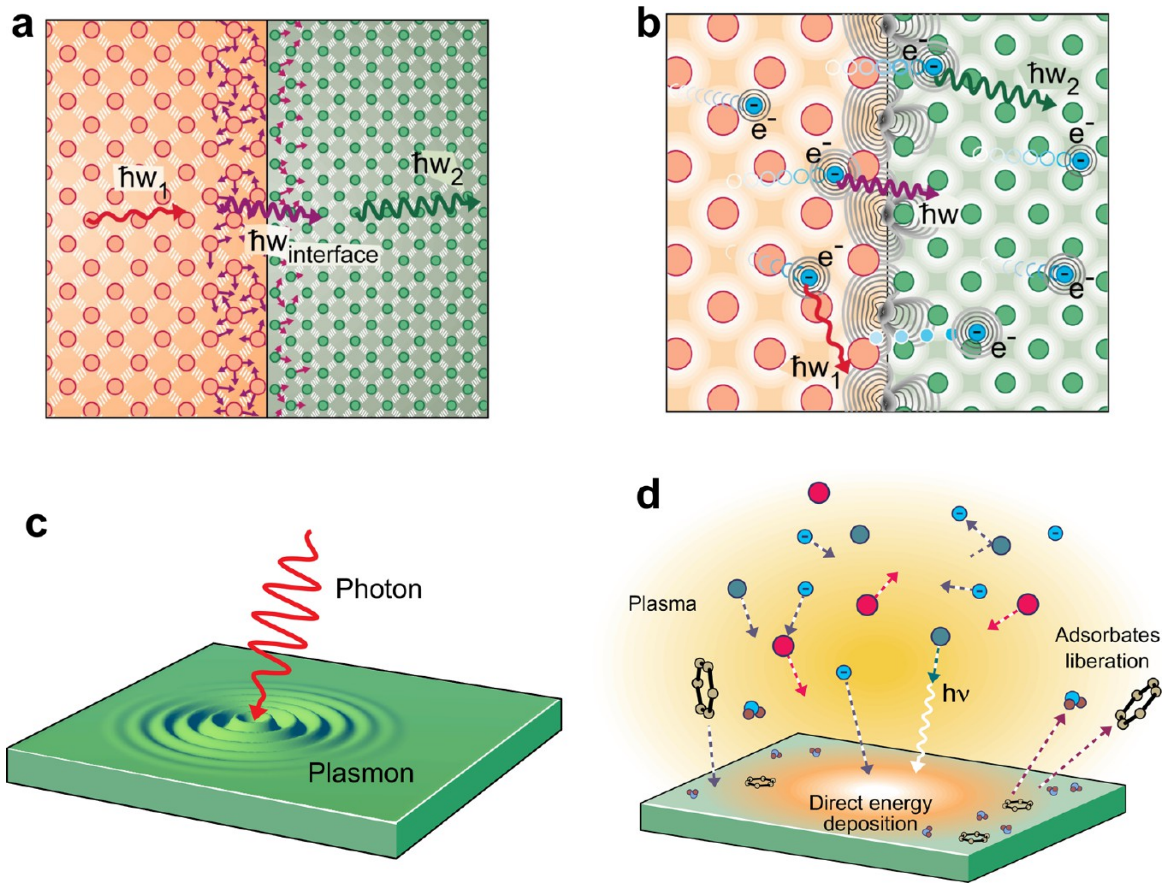
Recent review and perspective: *ACS Nano* 17, 14253 (2023)

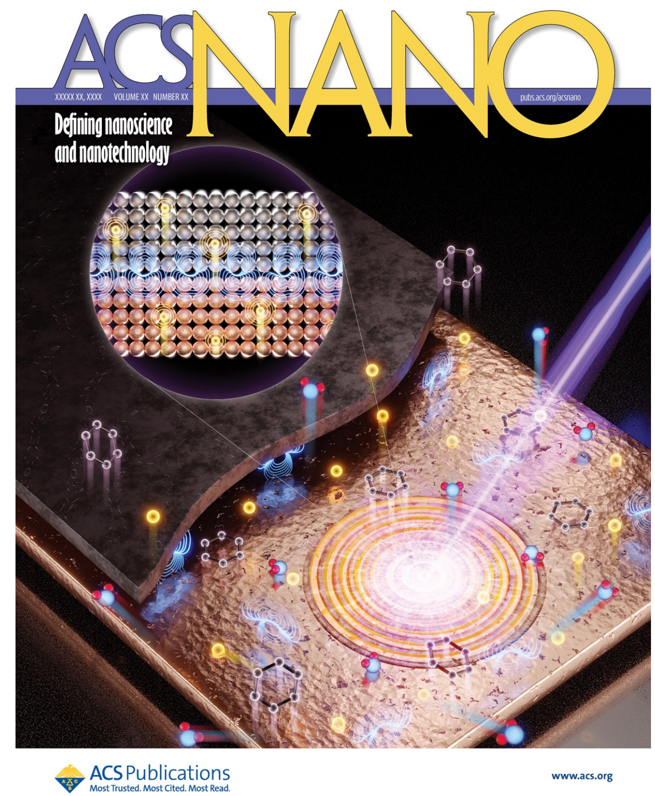
ACS NANO

www.acsnano.org

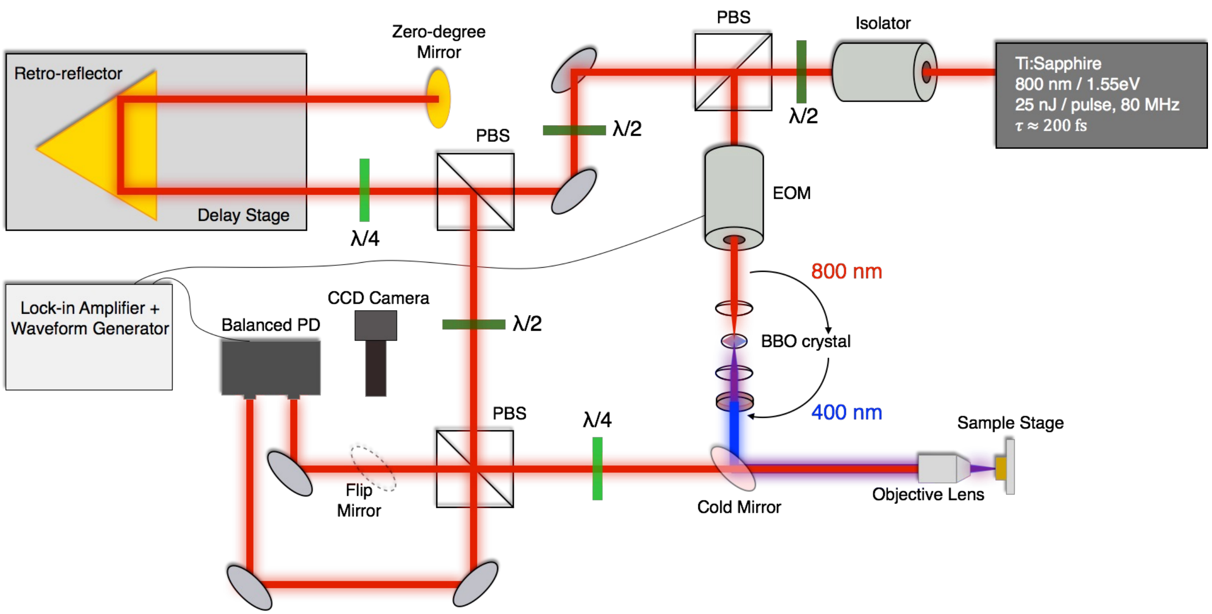
Ultrafast and Nanoscale Energy Transduction Mechanisms and Coupled Thermal Transport across Interfaces

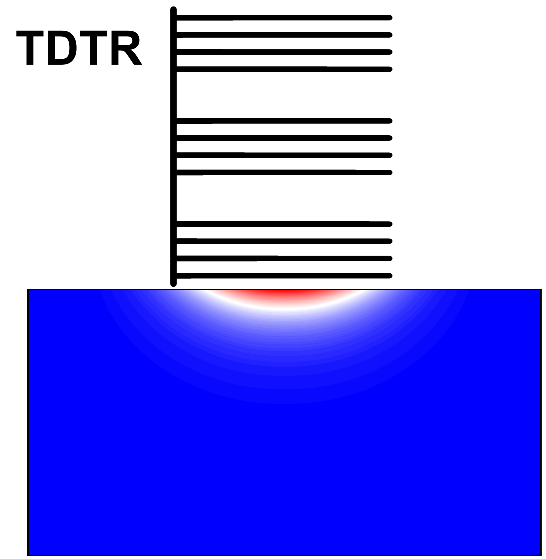
Ashutosh Giri,* Scott G. Walton, John Tomko, Niraj Bhatt, Michael J. Johnson, David R. Boris, Guanyu Lu, Joshua D. Caldwell, Oleg V. Prezhdo, and Patrick E. Hopkins*

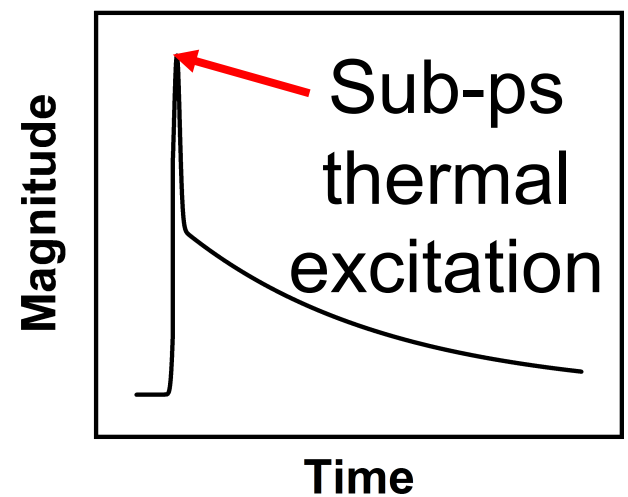




How do we measure the thermal properties? TDTR



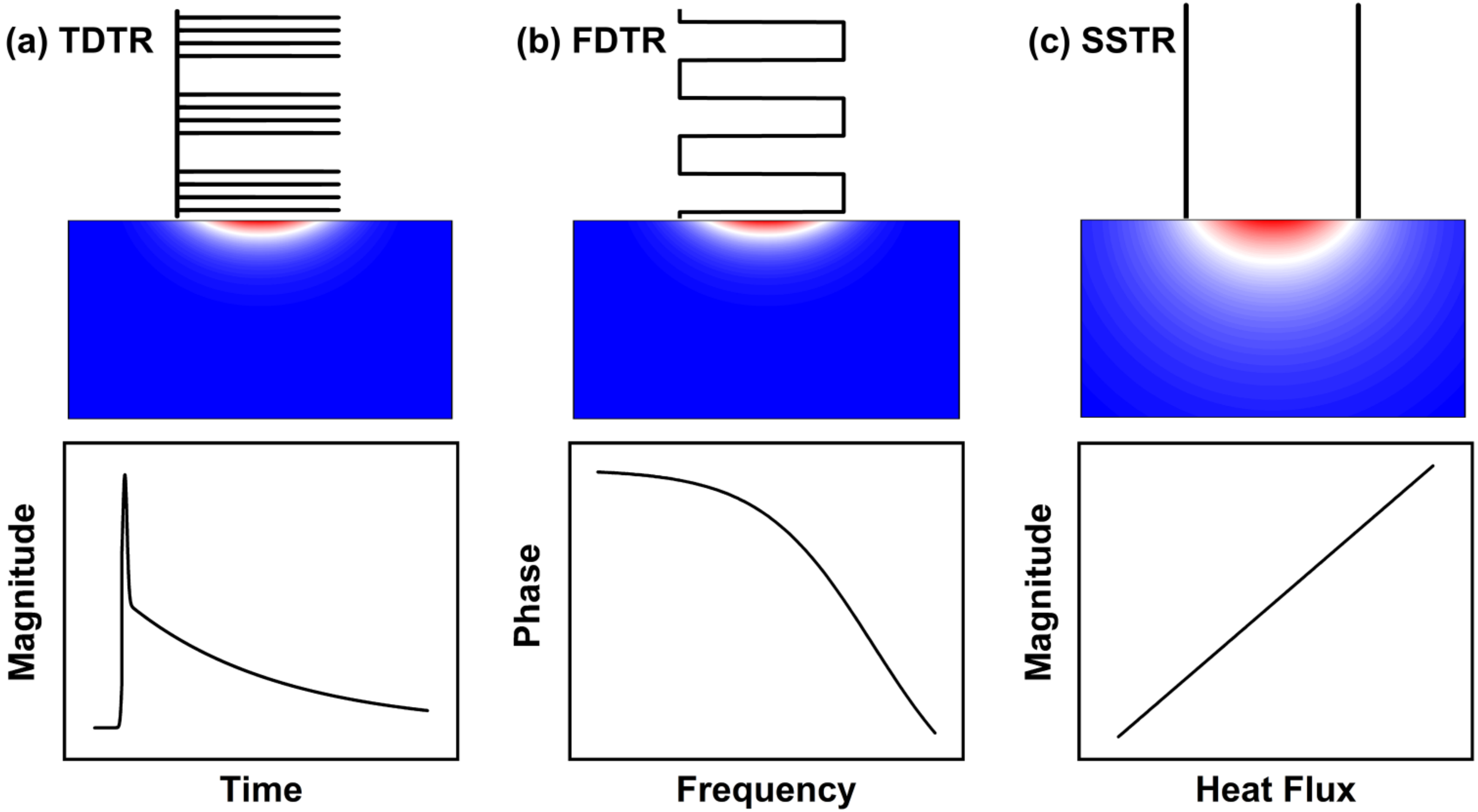




TDTR Reviews and Analyses

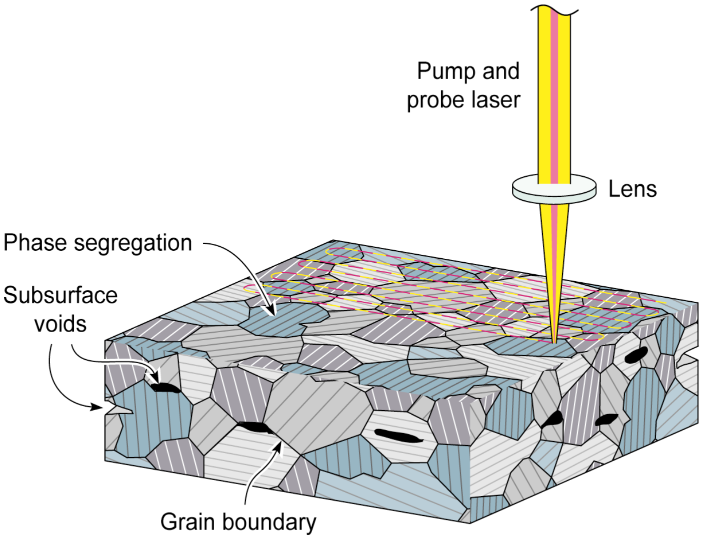
Rev. Sci. Instr. **75**, 5119;
Rev. Sci. Instr. **79**, 114902
J. Heat Trans. **132**, 081302;
Ann. Rev. Heat Trans. **16**, 159

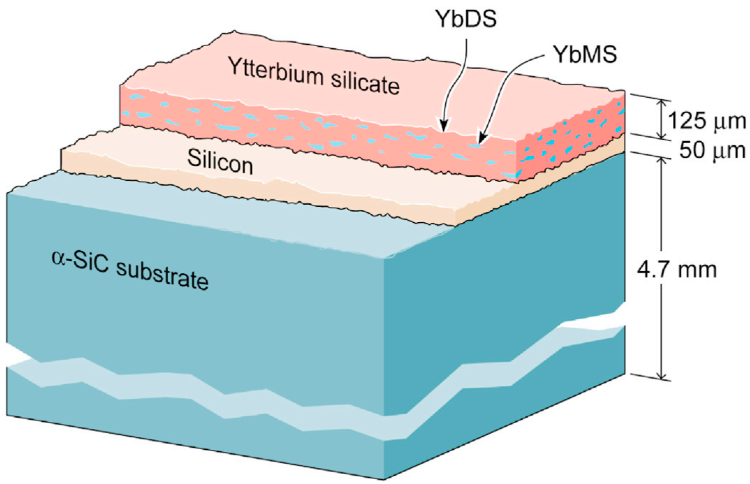
The “flavors” of thermoreflectance

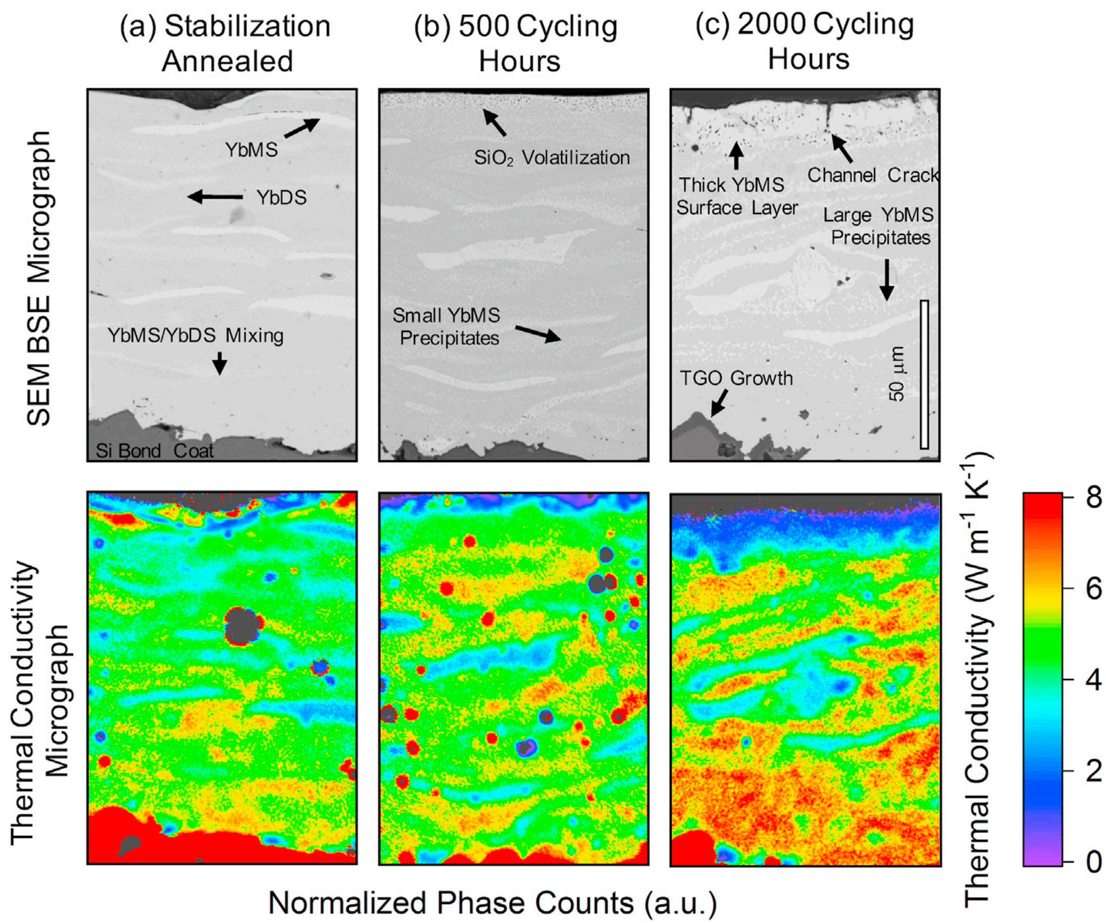


J. Appl. Phys. **126**, 150901

Local thermal conductivity measurements and spatial maps

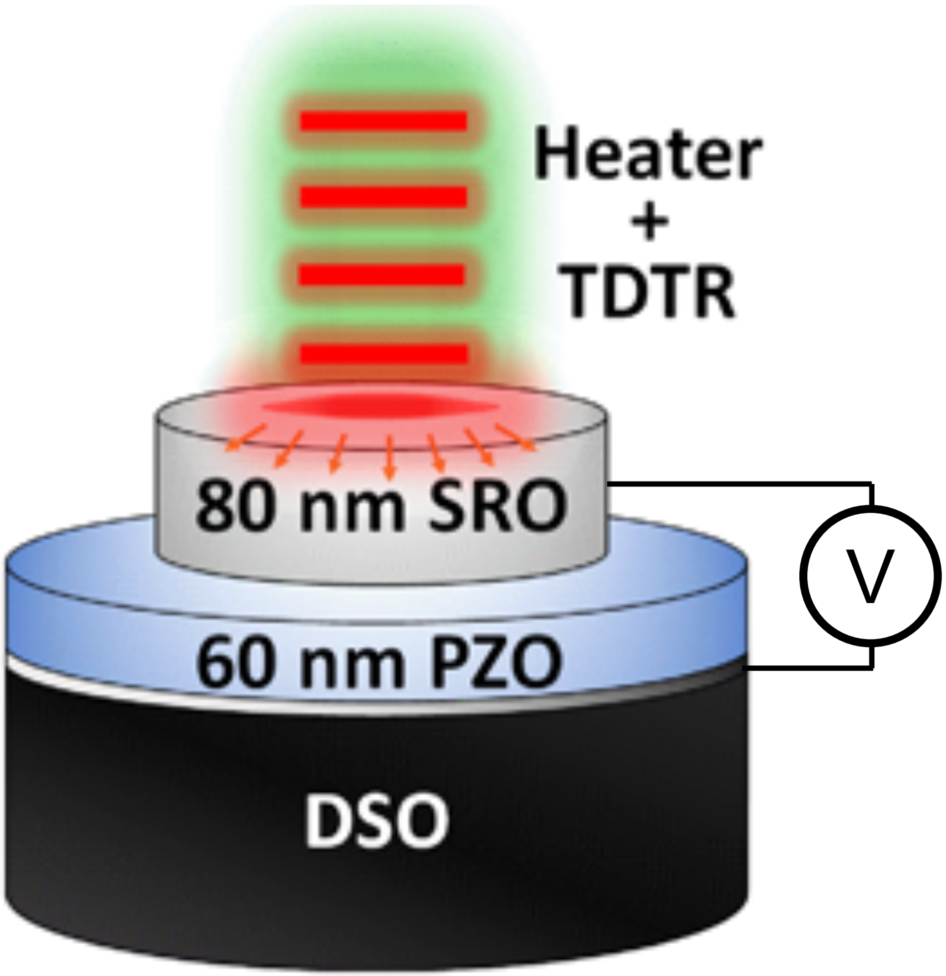


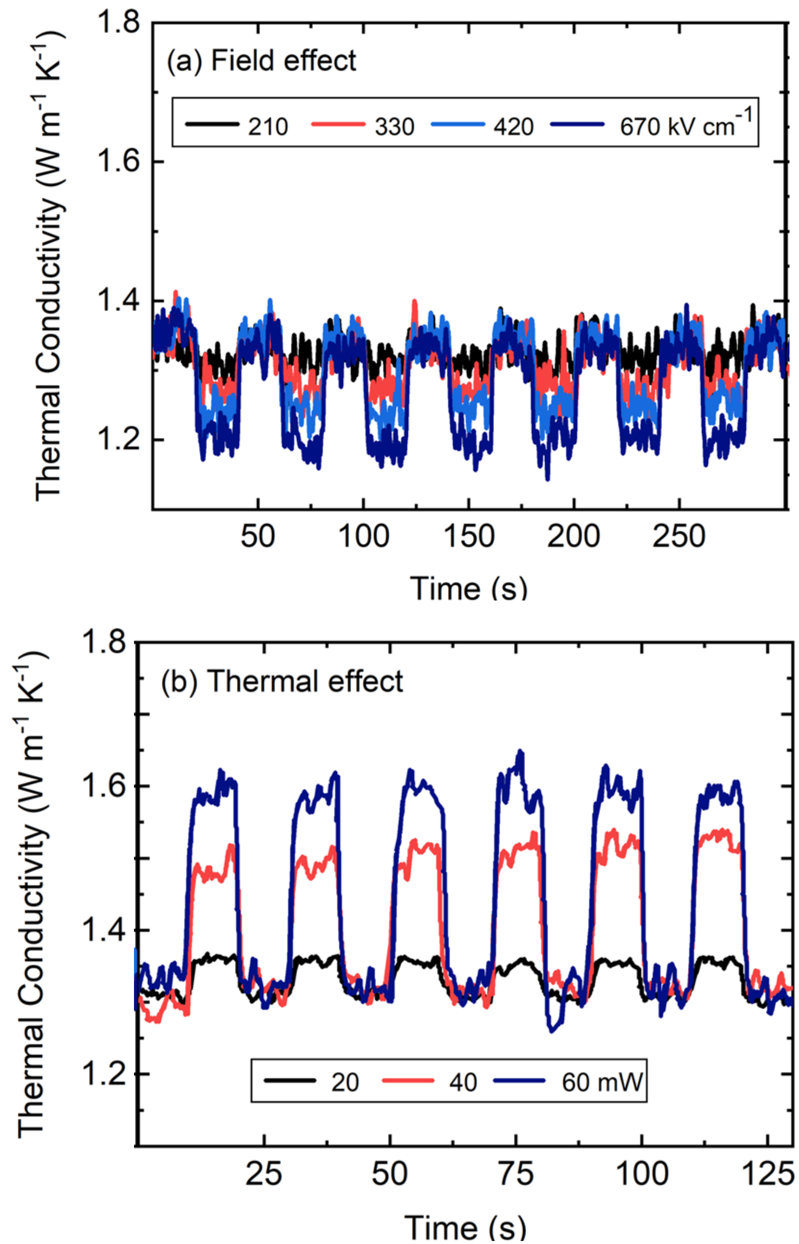




Olson *et al.*, *Materials Today Phys.* **17**, 100304 (2021)

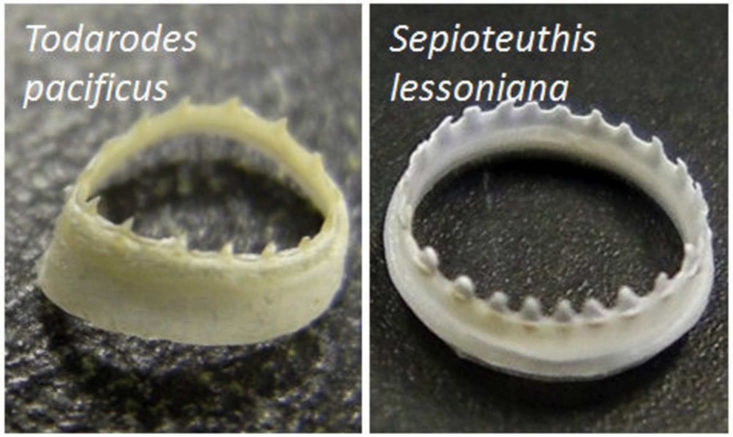
Actively switching thermal conductivity w/ FE/AFE domains

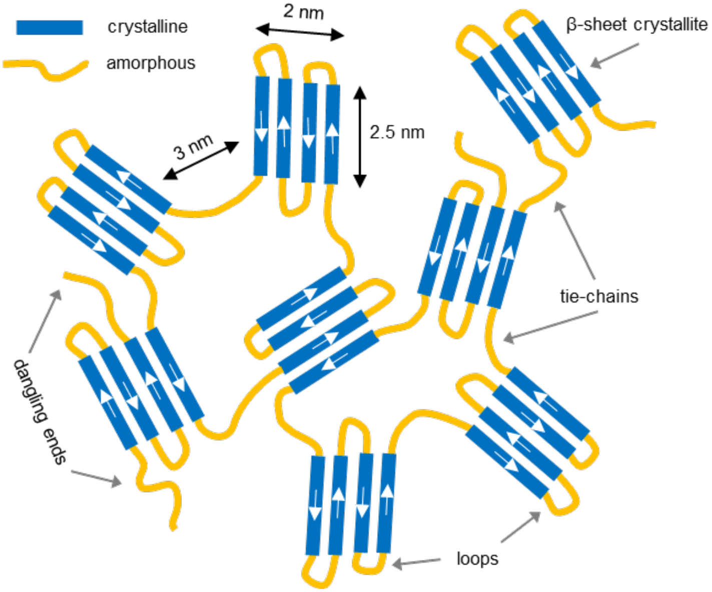


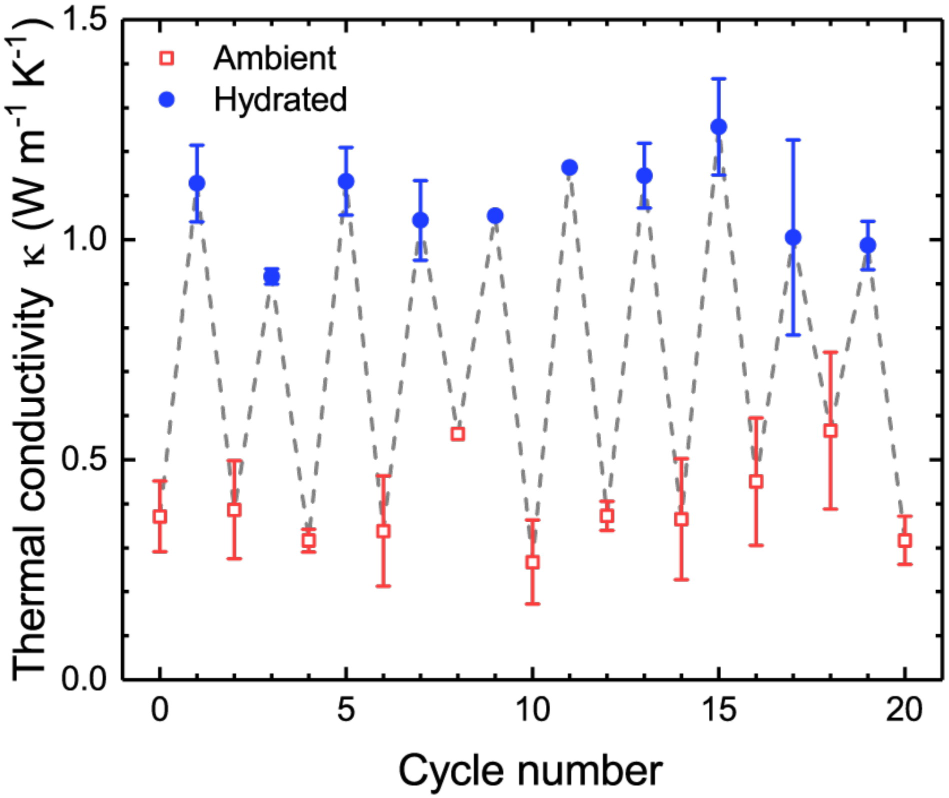


Aryana *et al.*, *Nat. Comm.* **13**, 1573 (2022)

Actively switching thermal conductivity in bio materials

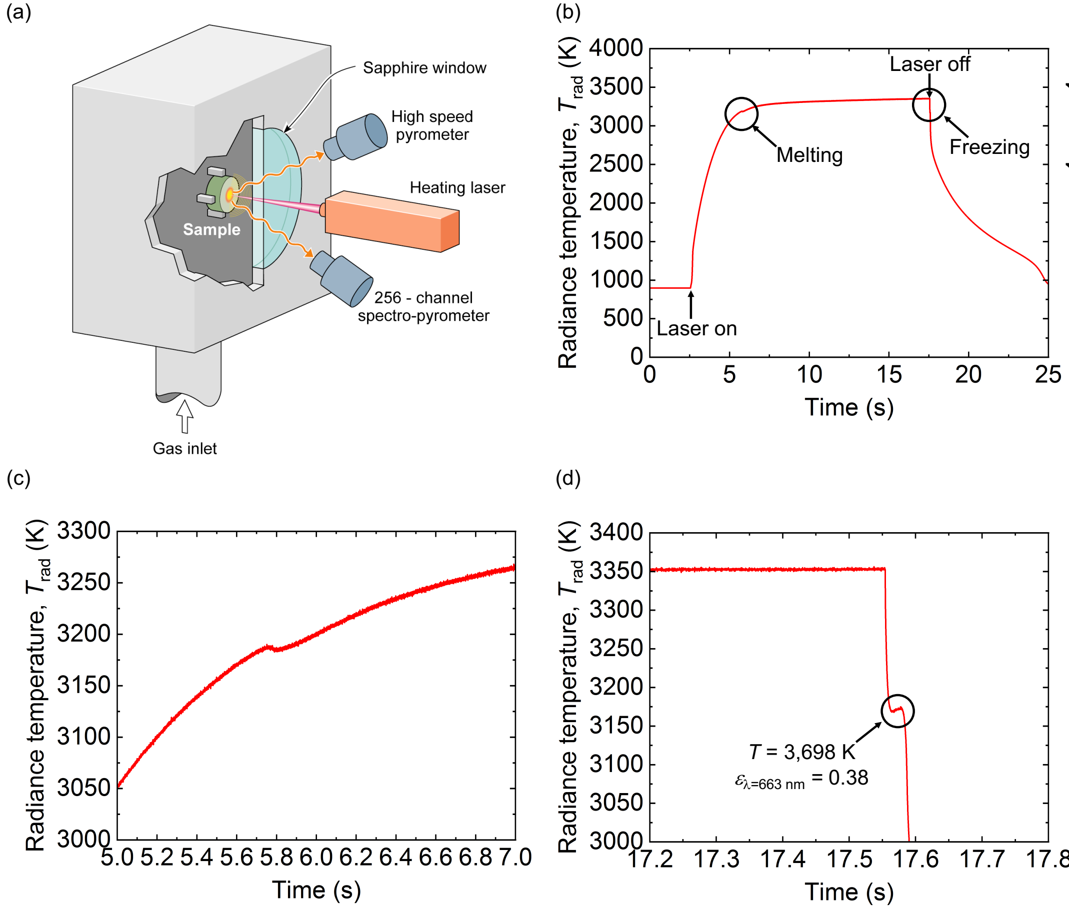


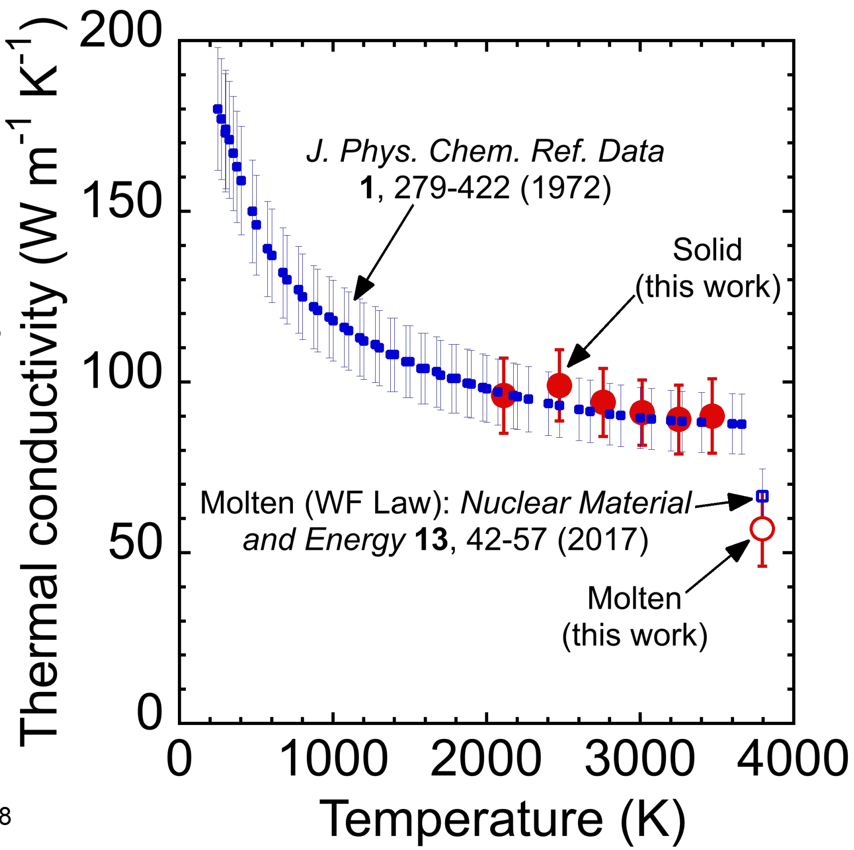




Tomko *et al.*, *Nat. Nano.* **13**, 959 (2018)

Thermometry at ultrahigh temperatures

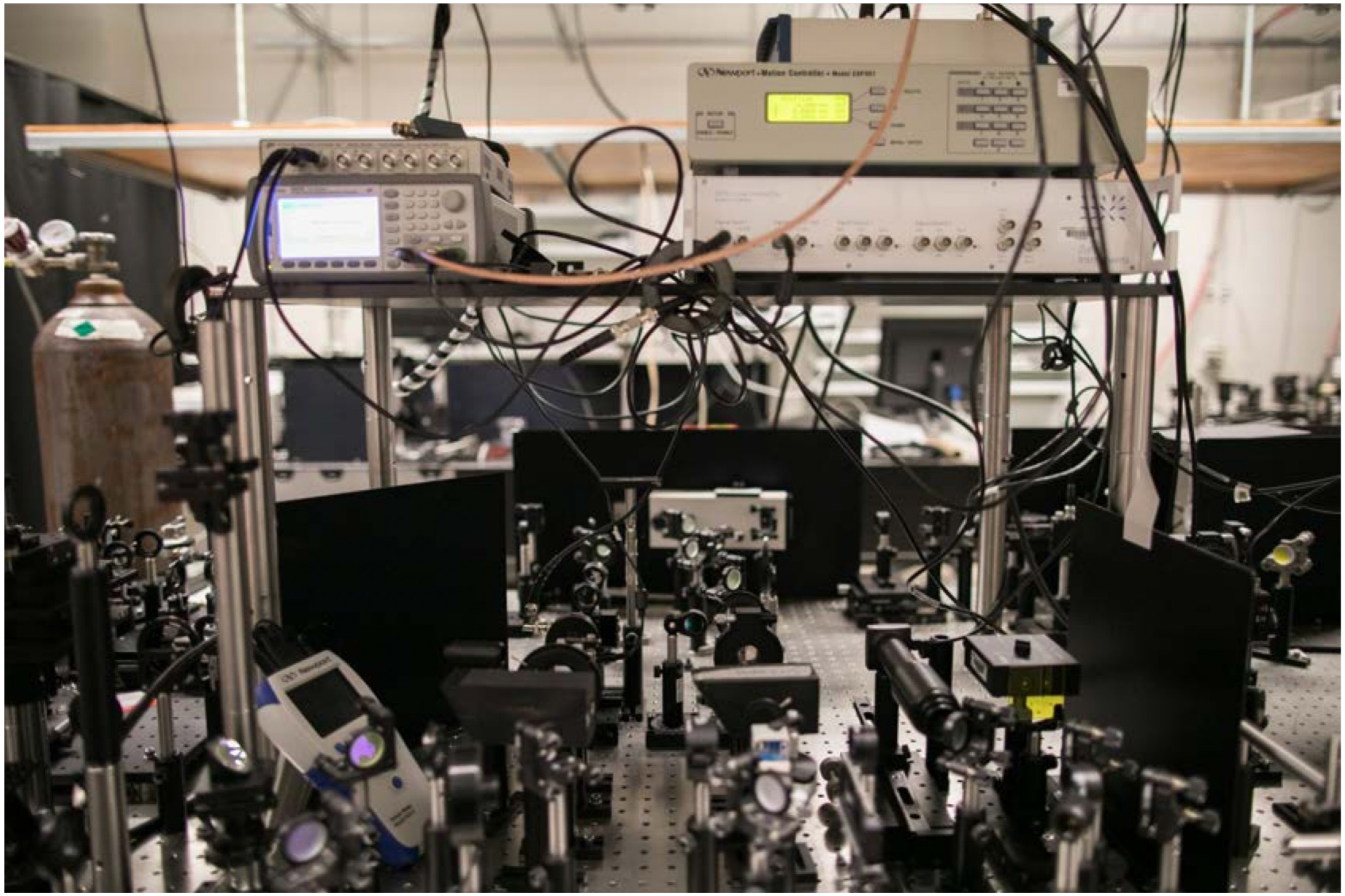




arXiv: 2309.01062

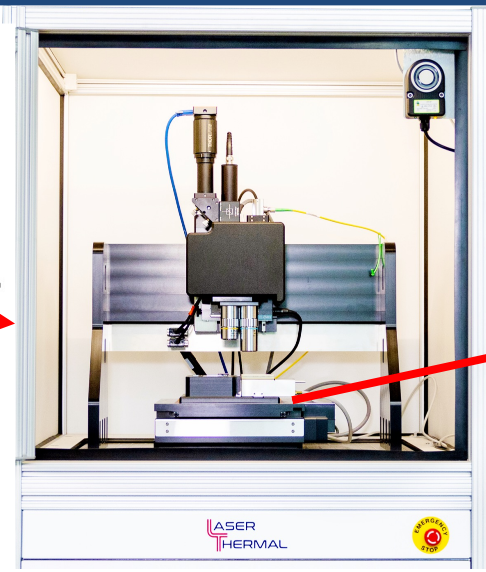
So why don’t we all have thermoreflectance systems????

The typical thermoreflectance set up



A LOT of optics, upkeep and expertise for analysis

SSTR-F: Commercialized for turn-key thermal conductivity microscope for bulk materials, thin films and interfaces



LASER THERMAL

Disclosure: Hopkins co-Founder

Outline

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  - **Minimum thermal conductivity and creating "crystals of interfaces"**

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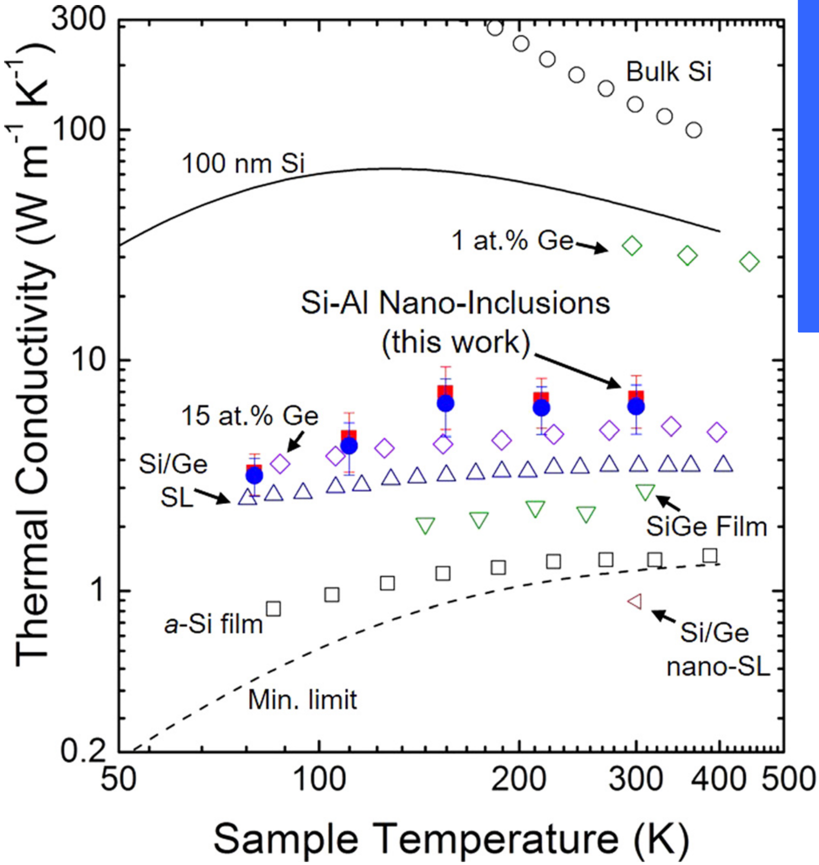
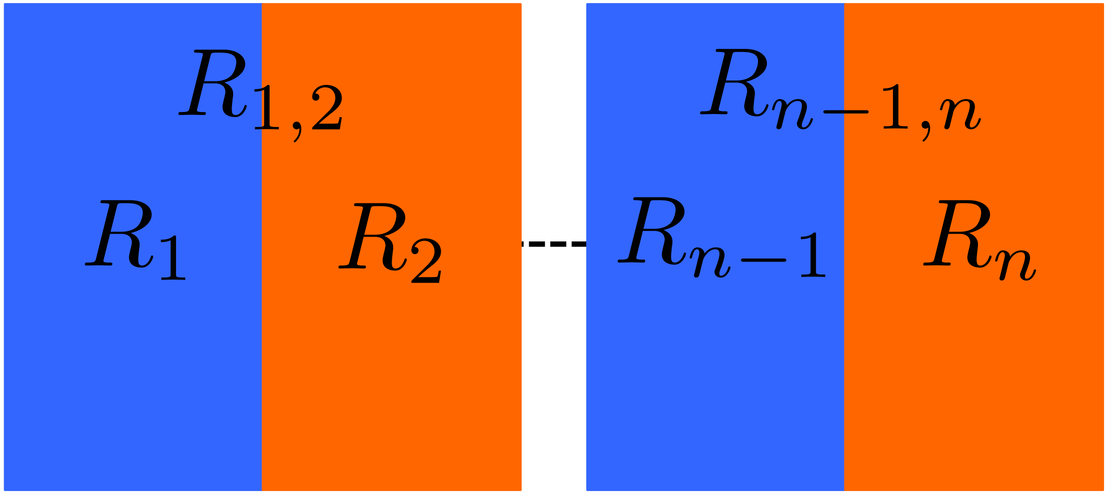
Phonon thermal conductivity in superlattices: incoherent



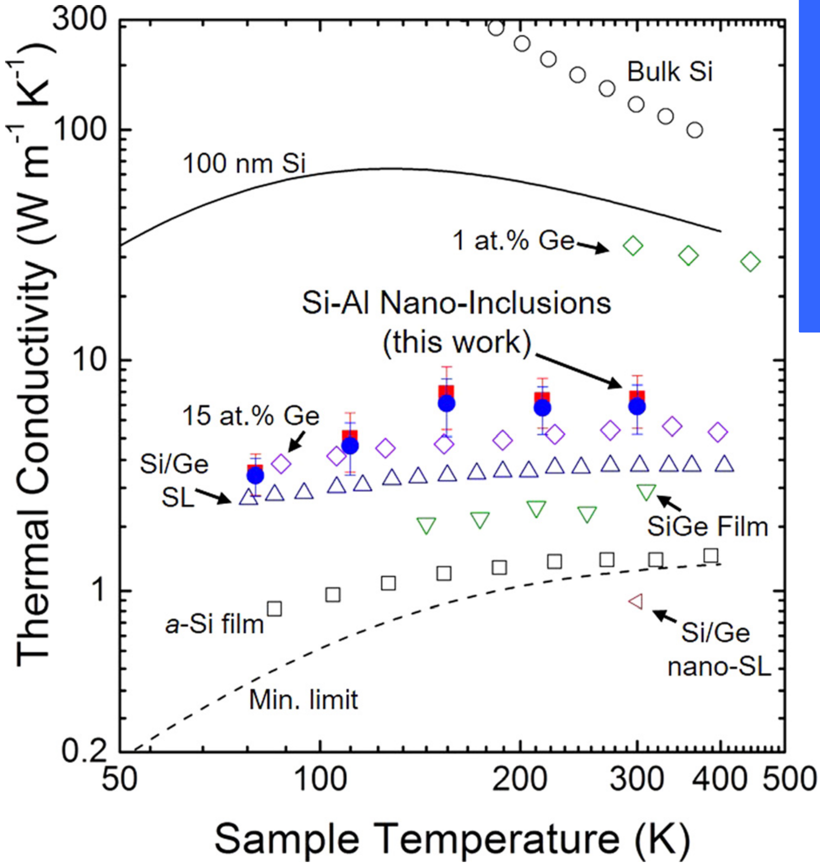
APL **112**, 213103



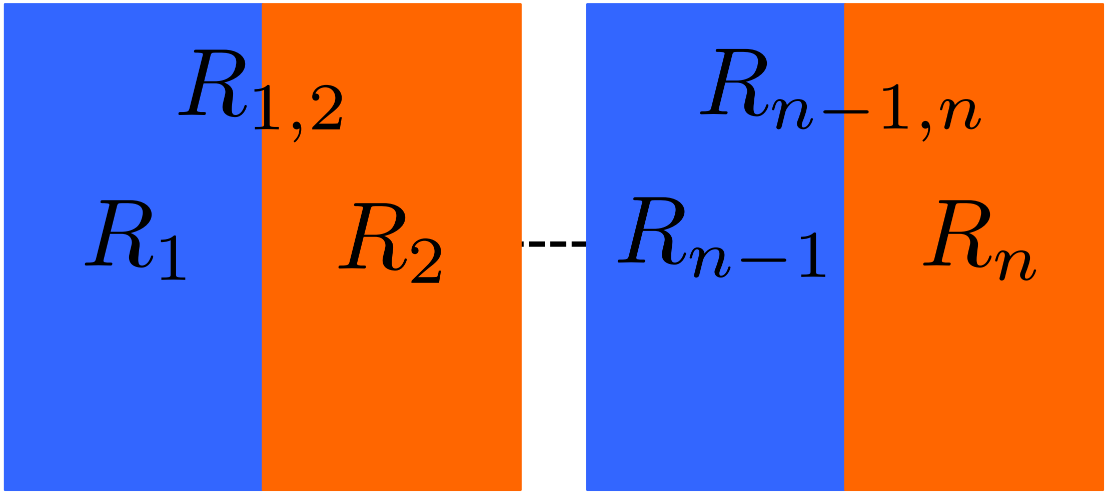
$$1/\kappa \propto 1/h = \sum_n R_j$$

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

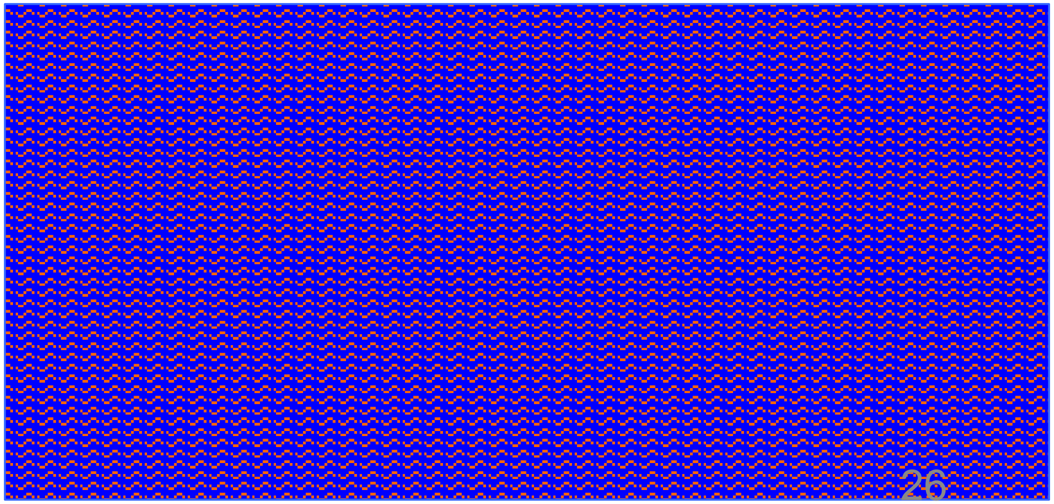
Phonon thermal conductivity in superlattices: coherent



APL **112**, 213103



What if layers are "linked"?
– coherent transport

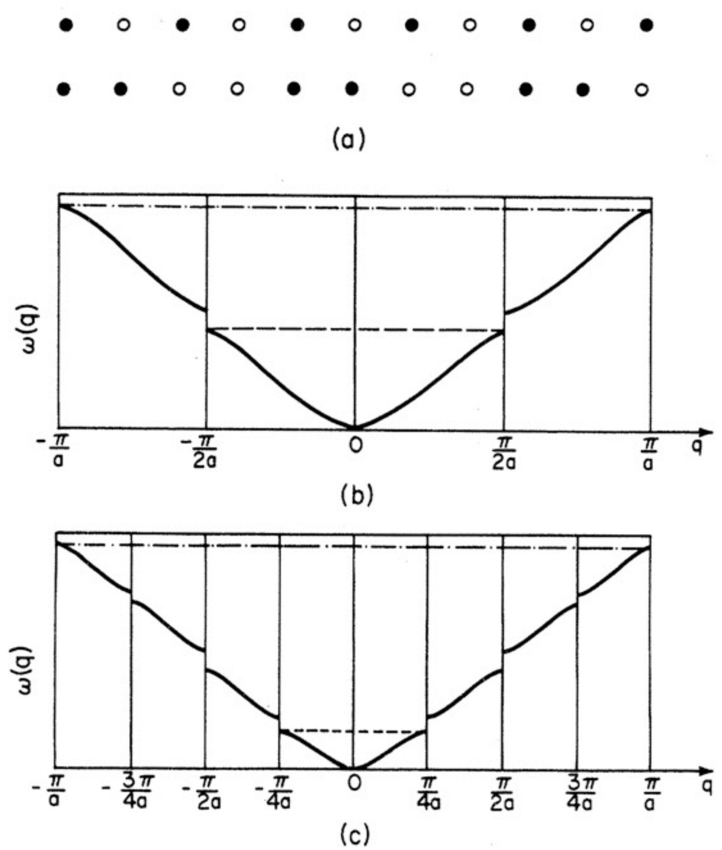


The minimum thermal conductivity of superlattices

PHYSICAL REVIEW B VOLUME 25, NUMBER 6 15 MARCH 1982

Thermal conductivity of superlattices

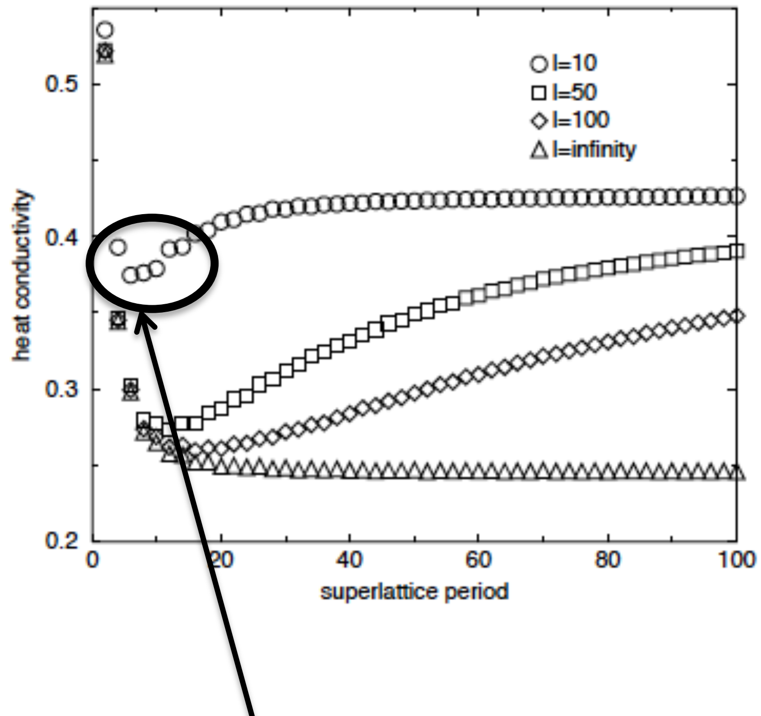
Shang Yuan Ren* and John D. Dow
Department of Physics and Coordinated Science Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801
(Received 21 September 1981)



VOLUME 84, NUMBER 5 PHYSICAL REVIEW LETTERS 31 JANUARY 2000

Minimum Thermal Conductivity of Superlattices

M. V. Simkin and G. D. Mahan
Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996-1200 and Solid State Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, Tennessee 37831
(Received 23 July 1999)

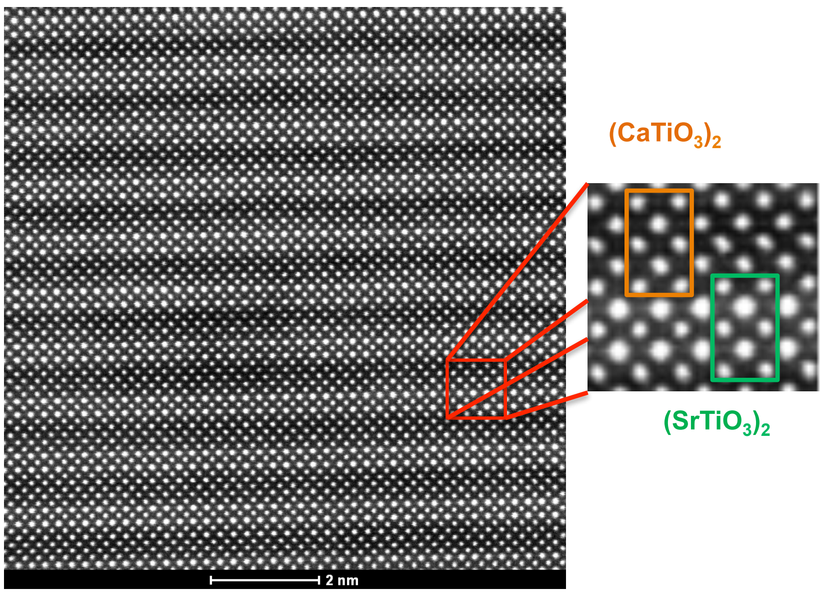


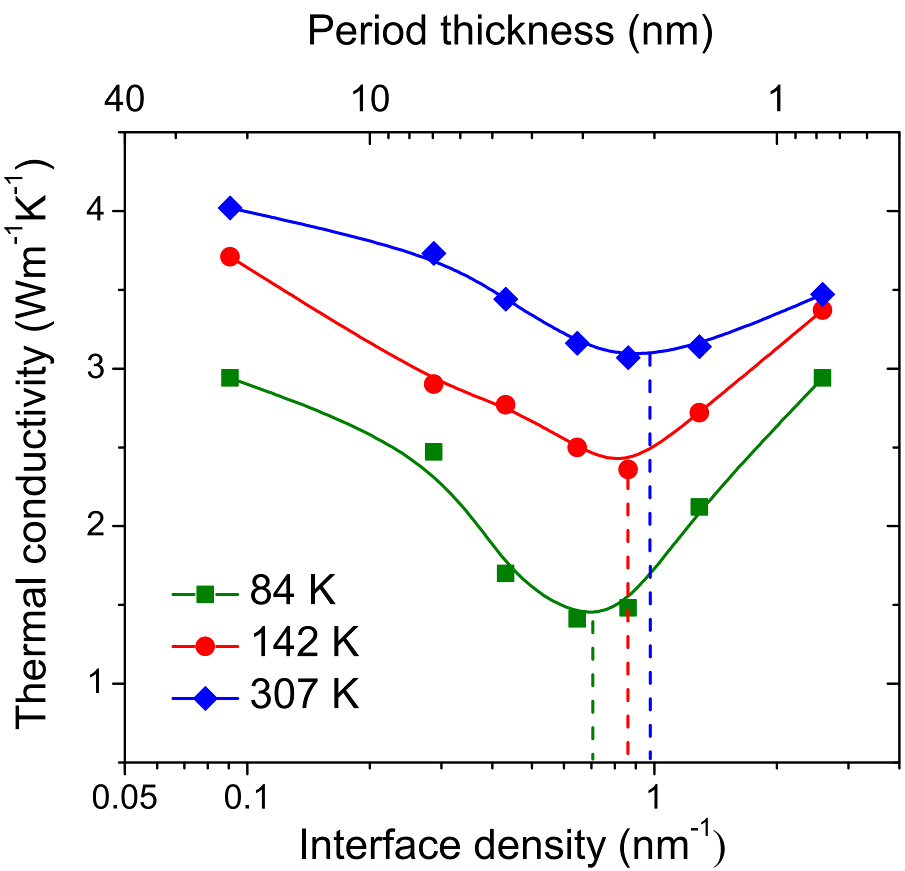
Mini-band formation leads to a minimum in the superlattice thermal conductivity

Interfacial periodicity can lead to “mini-band” formation

Experimental evidence of minimum thermal conductivity

- More pronounced minimum at low T, thermal conductivity measurements show trends of mini-band formation



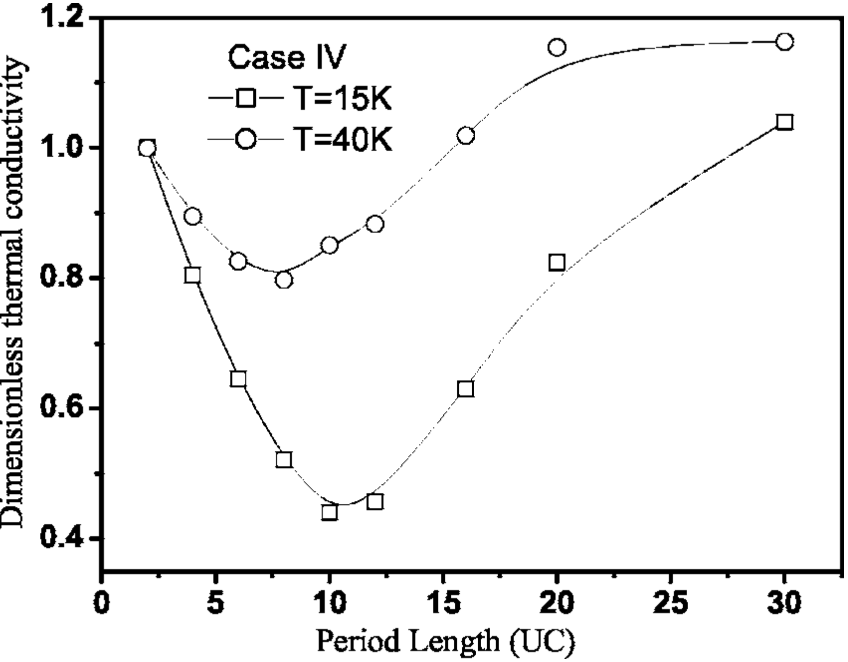


SL design to manipulate coherent phonon transport

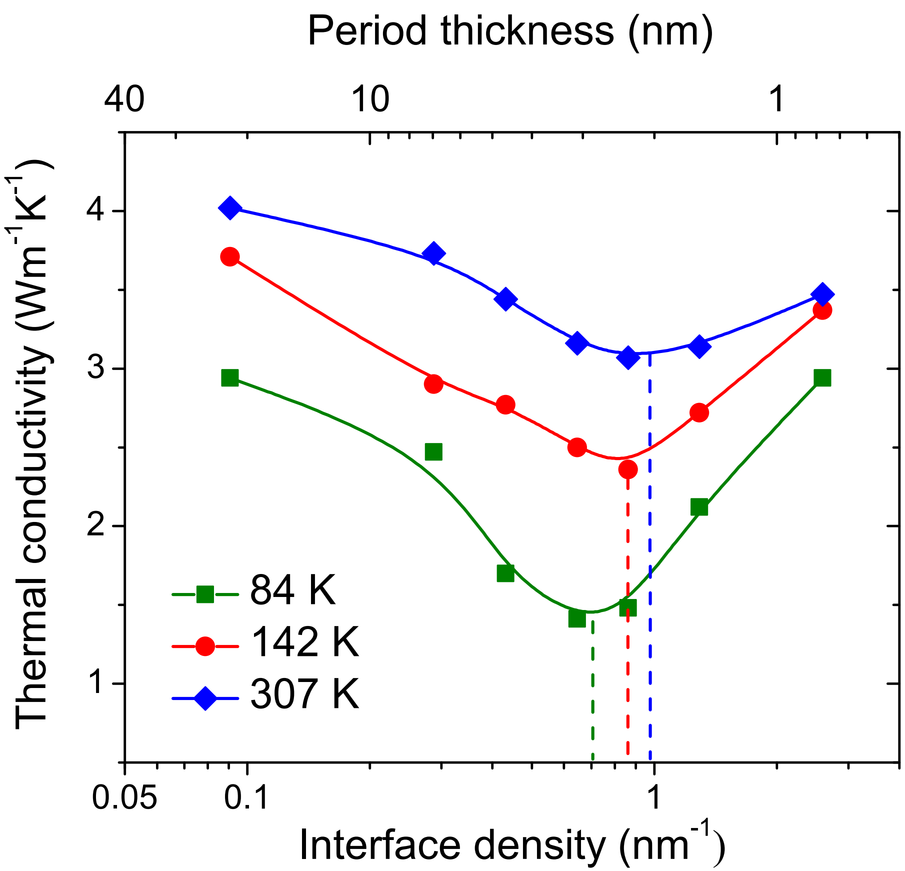
Ravichandran *et al.* *Nature Materials* **13**, 168 (2013)

Experimental evidence of minimum thermal conductivity

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



Phys. Rev. B **72**, 174302



SL design to manipulate coherent phonon transport

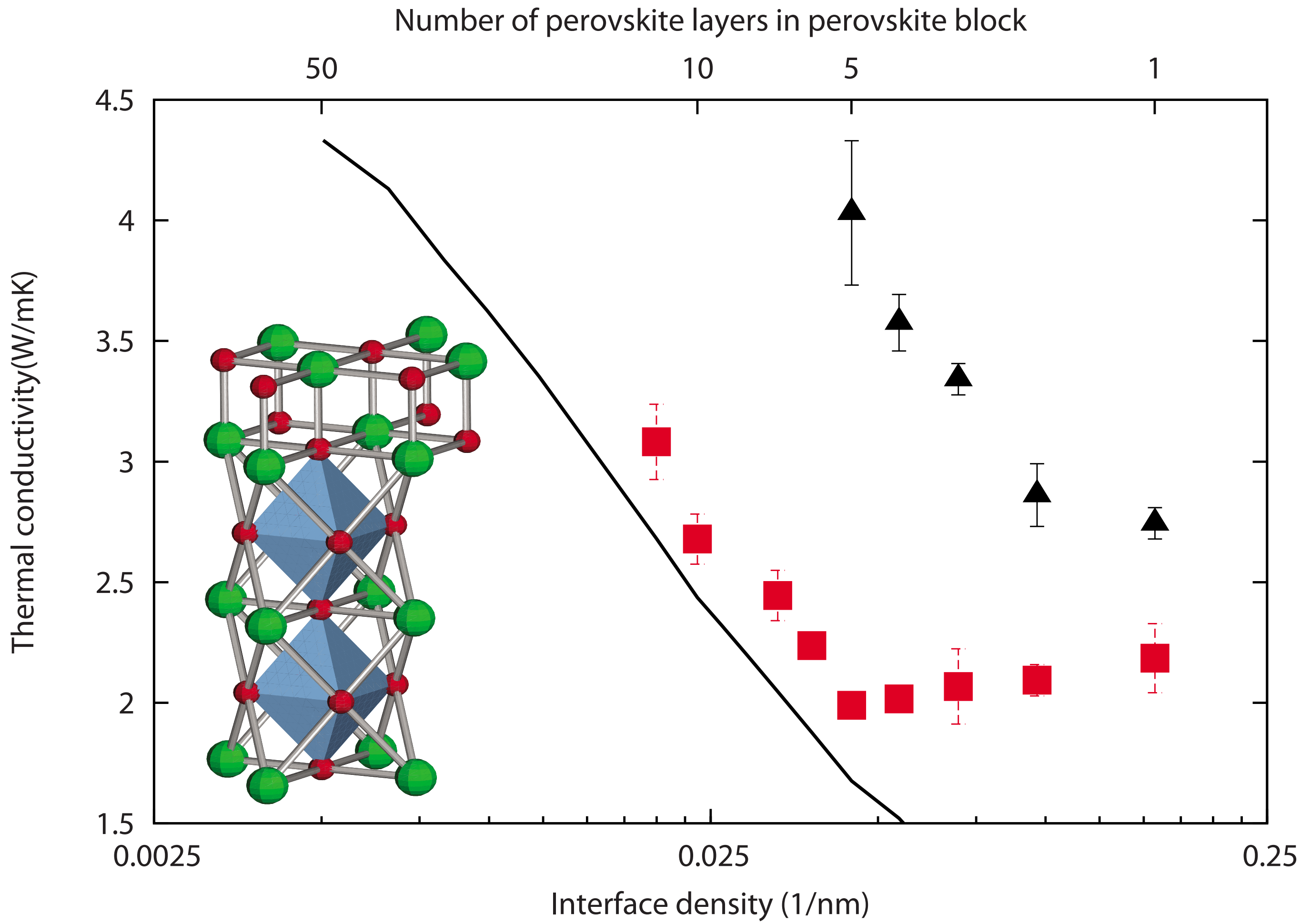
Ravichandran *et al. Nature Materials* **13**, 168 (2013)

”Naturally occurring superlattices”: Layered phase crystals

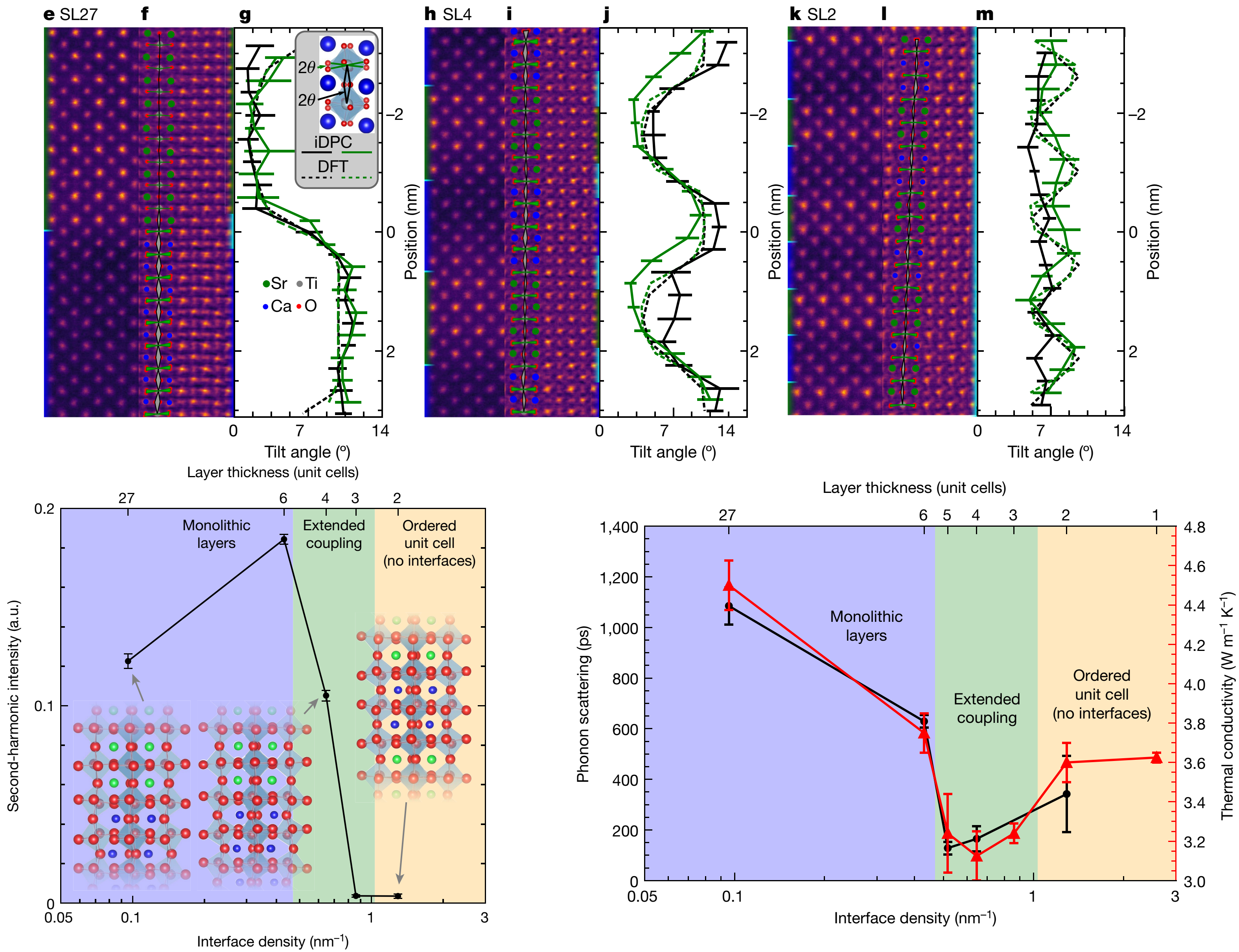
APPLIED PHYSICS LETTERS **95**, 161906 (2009)

Crossover in thermal transport properties of natural, perovskite-structured superlattices

Aleksandr Chernatynskiy,[1] Robin W. Grimes,[2] Mark A. Zurbuchen,[3] David R. Clarke,[4] and Simon R. Phillpot[1,a)]

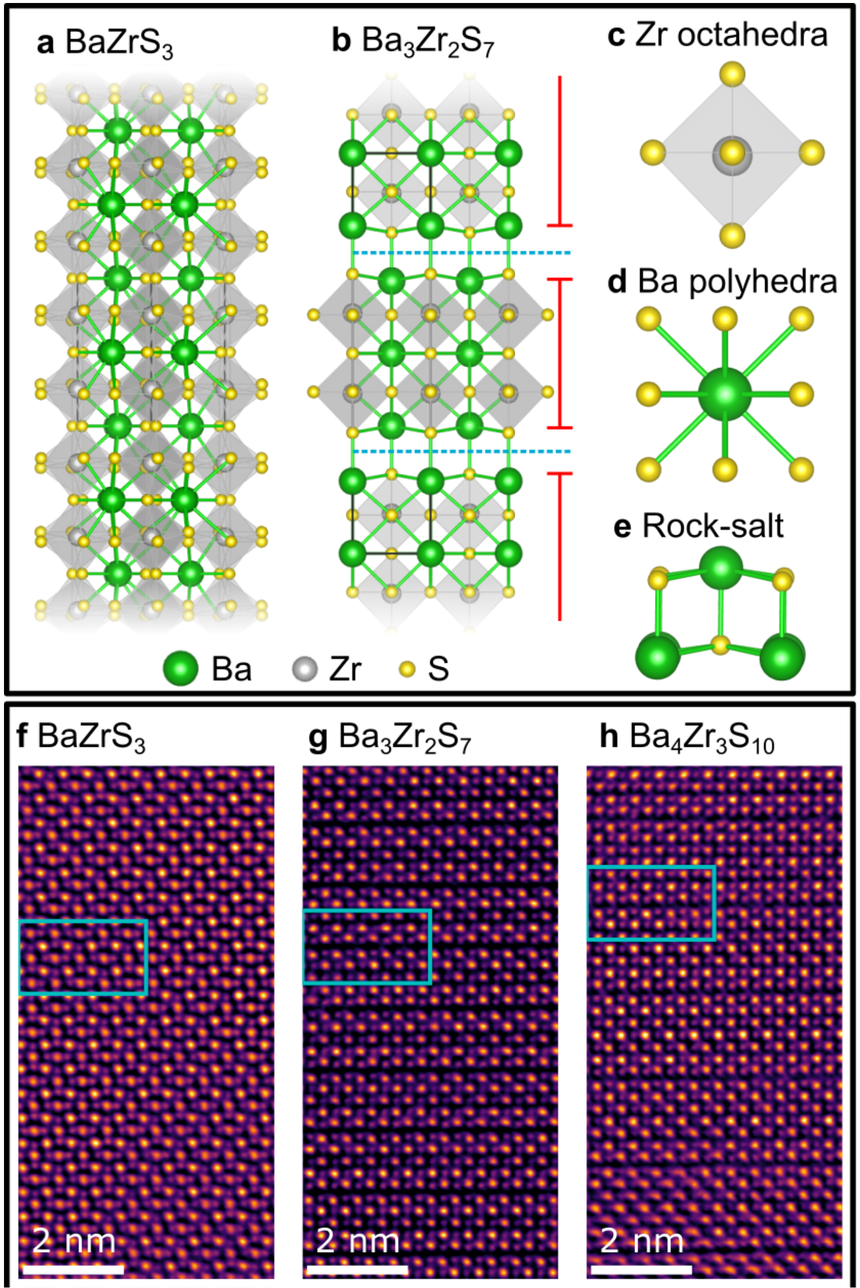


Experimental evidence of phonon coherence



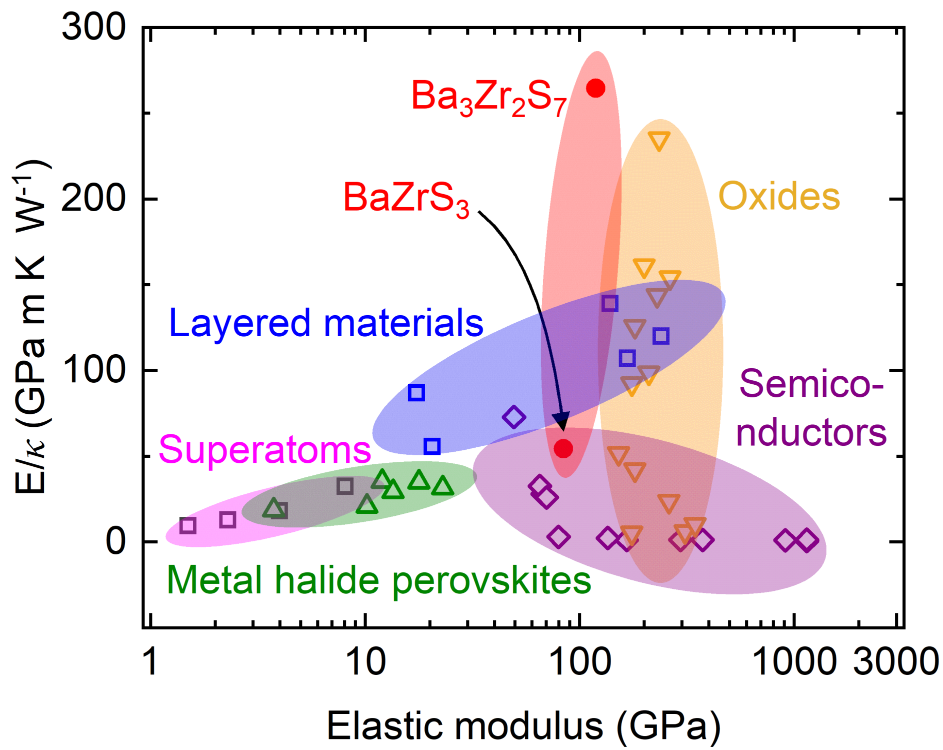
Hoglund *et al. Nature* **601**, 556 (2022)

Barium Zirconium Sulfide:Ruddlesden-Popper chalcogenides



$Ba_3Zr_2S_7$: "stiff phonon glass"

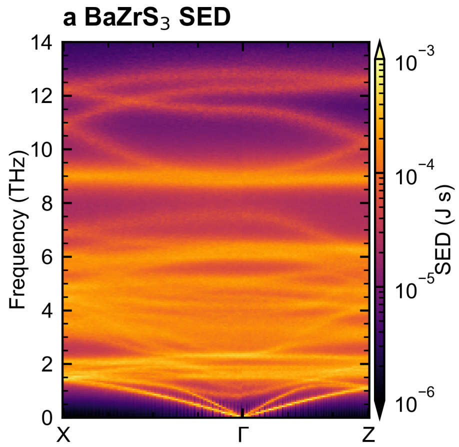
- Large anharmonicity
- Layered structure
- Covalent bonding

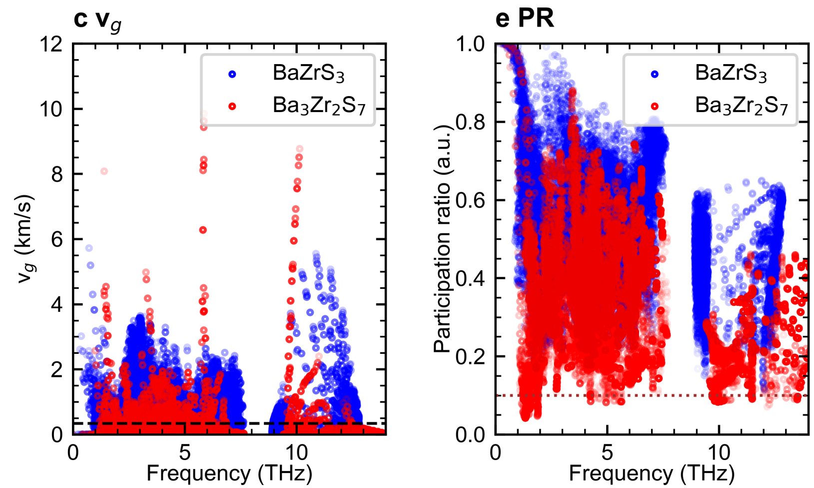


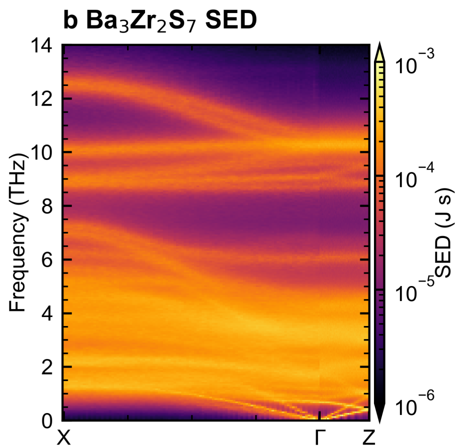
JMR **34**, 3819 (2019)
arXiv: 2312.02534

RP BZS: Scattering, low group velocity, localization

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







$Ba_3Zr_2S_7$: "stiff phonon glass"

- Strong scattering of all modes
- Low group velocity across Brillouin Zone

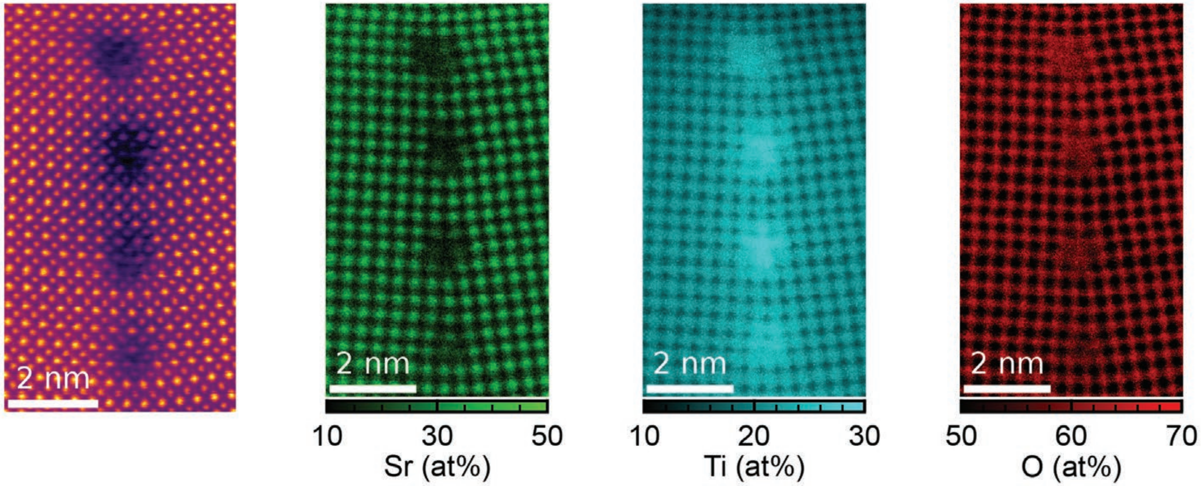
Ruddlesden-Popper chalcogenides push the limit of mechanical stiffness and glass-like thermal conductivity in crystals

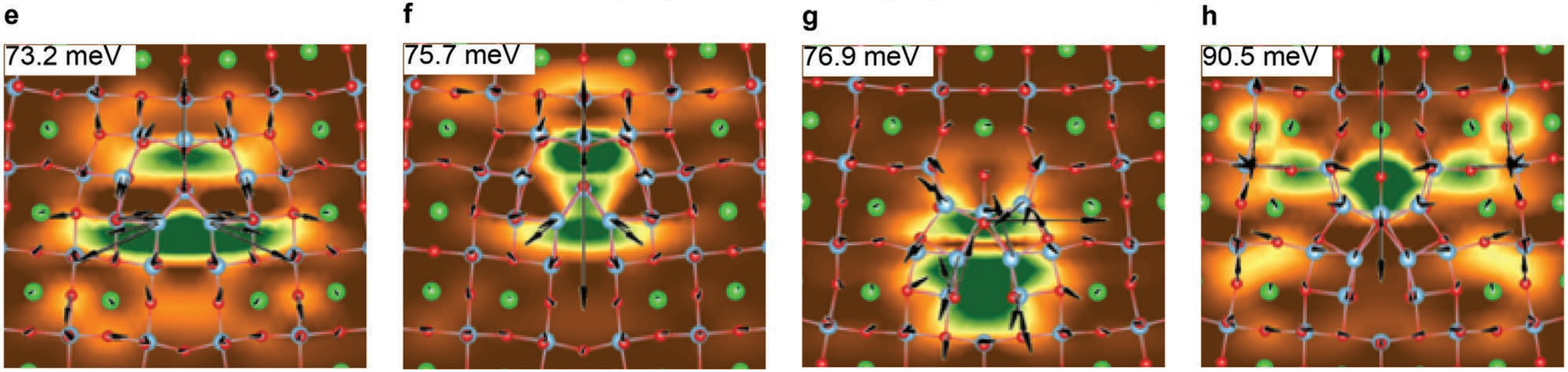
Md Shafkat Bin Hoque, Eric R. Hoglund*, Boyang Zhao*, De-Liang Bao, Hao Zhou, Sandip Thakur, Eric Osei-Agyemang, Khalid Hattar, Ethan A. Scott, Mythili Surendran, John A. Tomko, John T. Gaskins, Kiumars Aryana, Sara Makarem, Ganesh Balasubramanian, Ashutosh Giri, Tianli Feng, Jordan A. Hachtel, Jayakanth Ravichandran, Sokrates T. Pantelides, and Patrick E. Hopkins*

arXiv: 2312.02534

Advanced microscopy (at ORNL) leads to discovery and understanding of emergent phononic properties

Localized vibrational modes at grain boundaries (STO)
Vibrational modes at grain boundaries are different from “bulk”

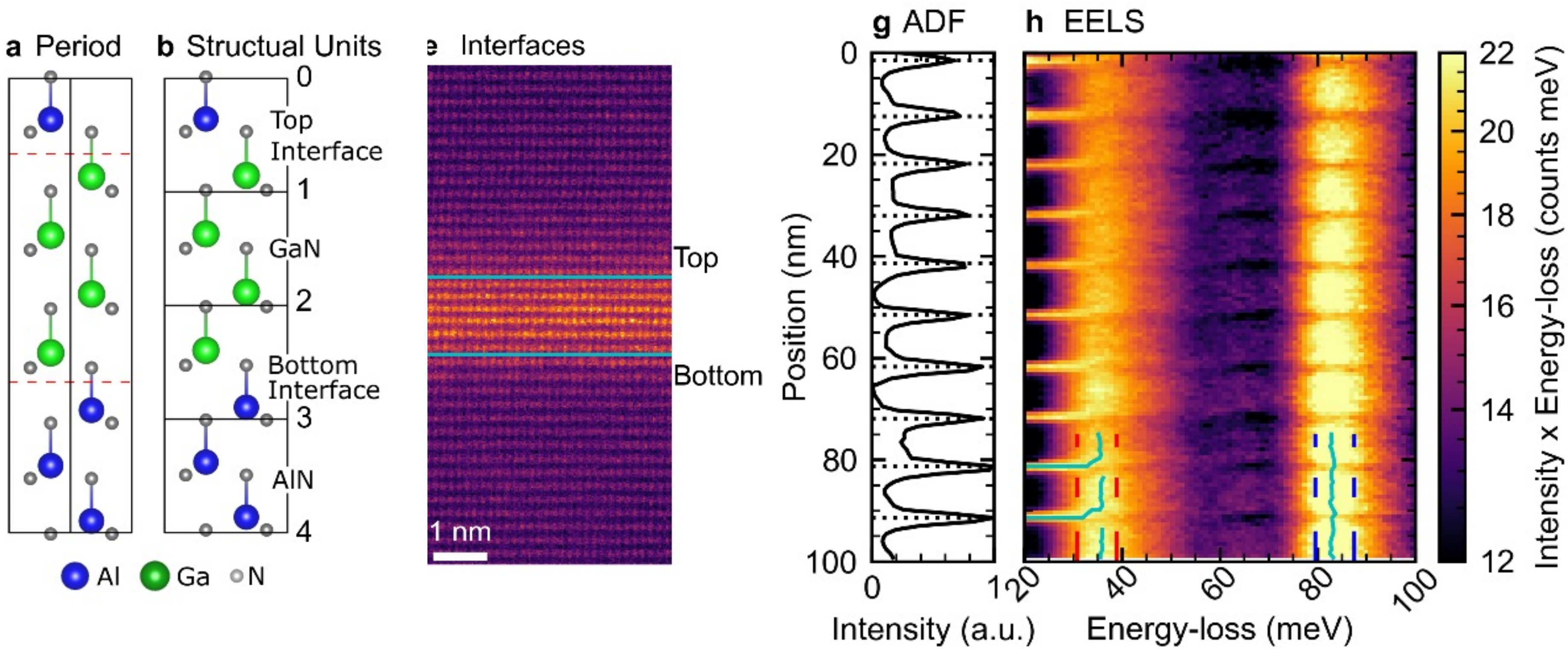




Hoglund *et al. Adv. Mat.* **35**, 2208920

Advanced microscopy (at ORNL) leads to discovery and understanding of emergent phononic properties

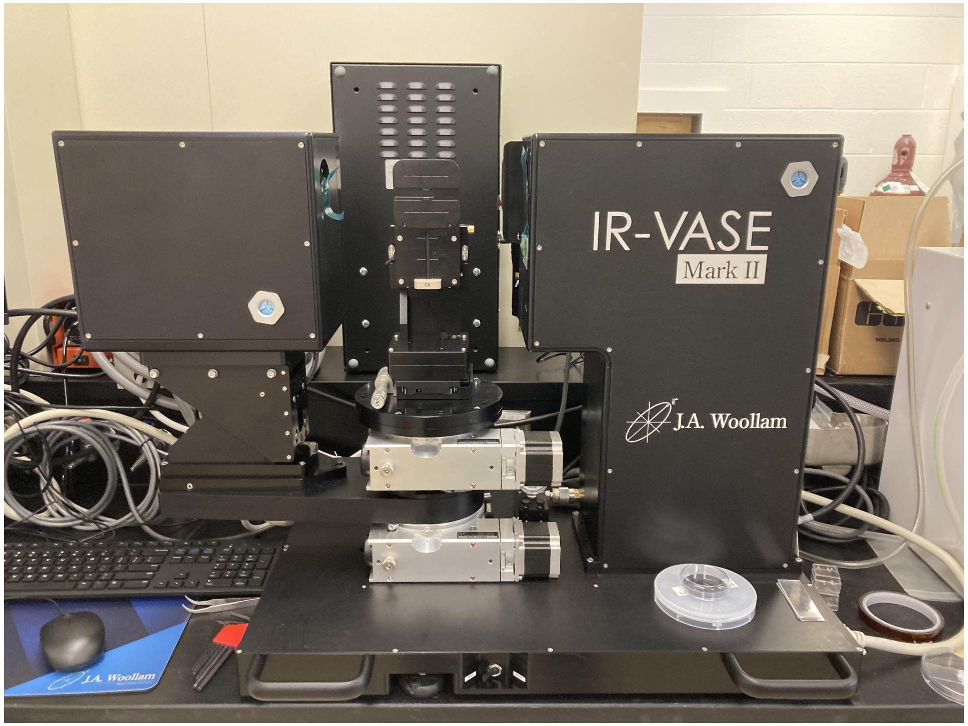
Asymmetry in vibrational modes at polar interfaces
Phononically, AlN/AlGaN is different from AlGaN/AlN

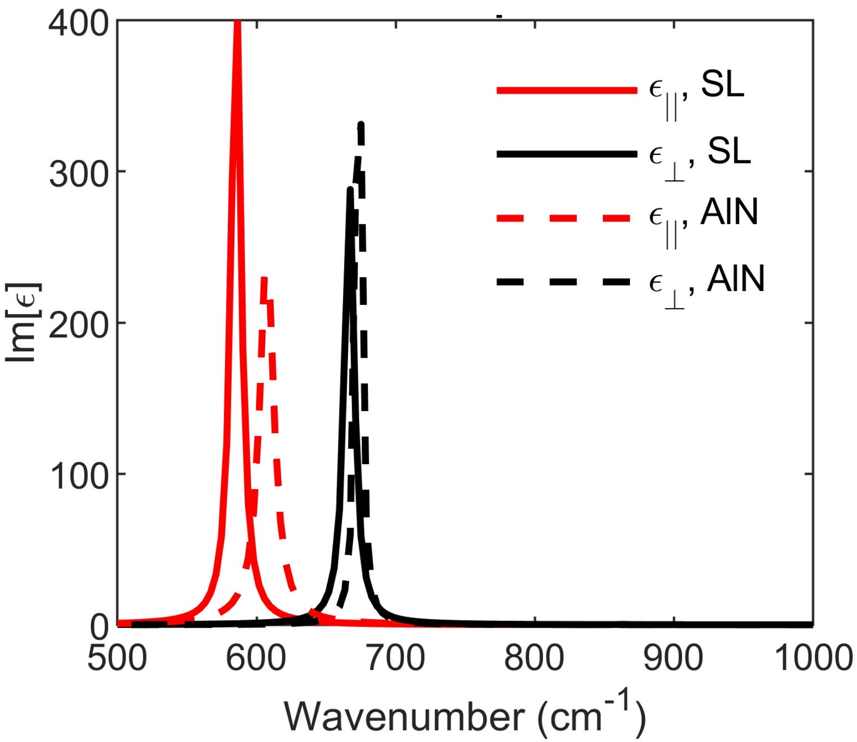


Hoglund *et al. under review*

New techniques we're standing up at UVA to complement microscopy collaborations at ORNL

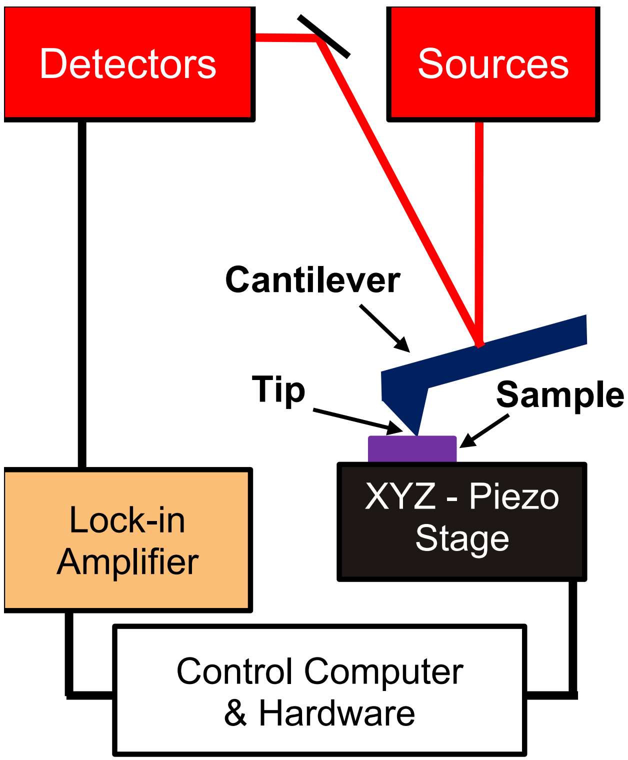
Infrared variable angle spectroscopic ellipsometry



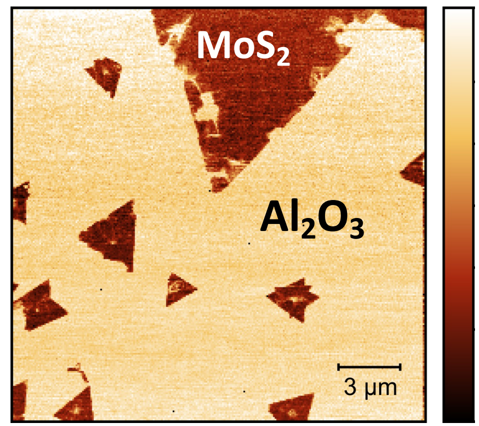


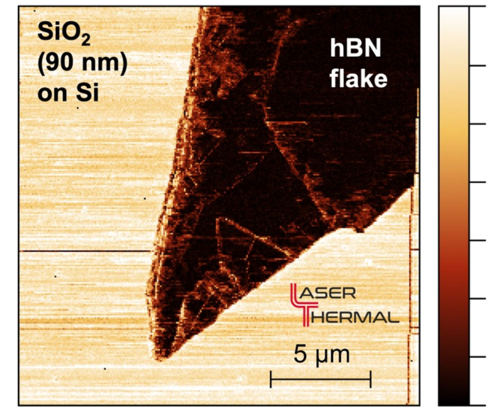
New techniques we're standing up at UVA to complement microscopy collaborations at ORNL

Nanoscale Thermal Microscopy (NTM)
Pump-probe (SSTR) with ~10 nm areal resolution

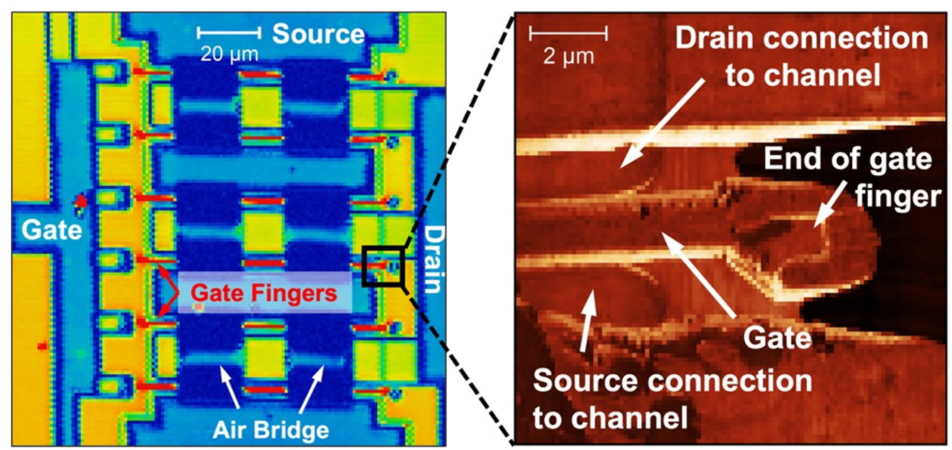


LASER THERMAL





SSTR-F Thermoreflectance Map | **NTM Thermoreflectance Map**



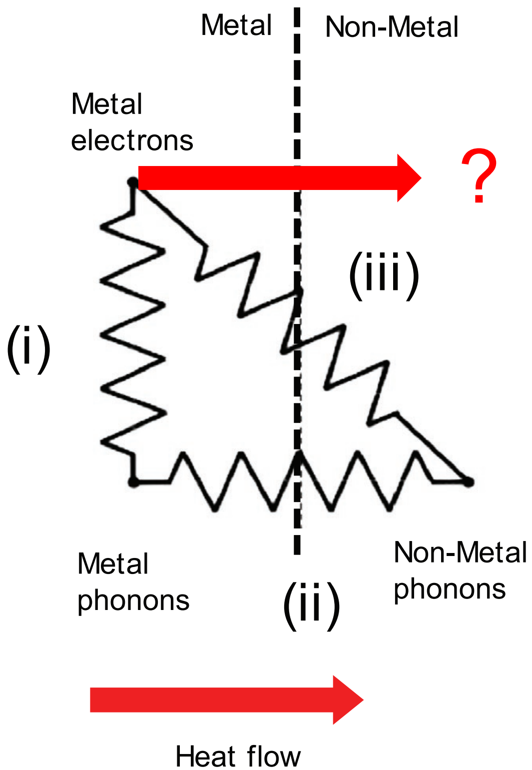
~1 μm pixel resolution Limit of SSTR-F → 110 nm pixel resolution (shown) *and smaller* w/ NTM

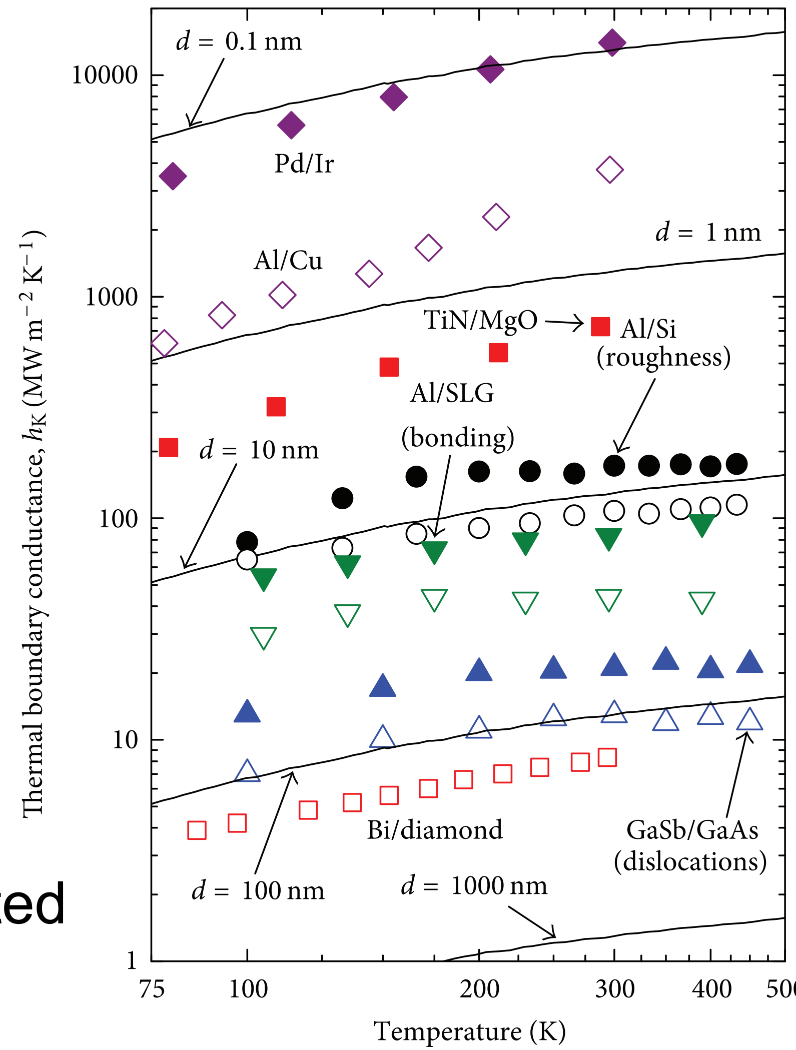
Outline

Theme: Coupling of carriers across interfaces lead to unique energy states and modes of heat transfer

- How do we measure the thermal conductivity of materials and the thermal boundary resistance at their interfaces?
- Thermal conductivity of superlattices
  - Minimum thermal conductivity and creating “crystals of interfaces”
- **Interfacial heat transfer control of the IR properties of solids**
  - **Near field radiative interfacial heat transfer with plasmon-polaritons and phonon-polaritons**
- Transient temperature changes during plasma-surface interactions
  - “Plasma cooling”

Phonon dominated TBC < electron dominated TBC

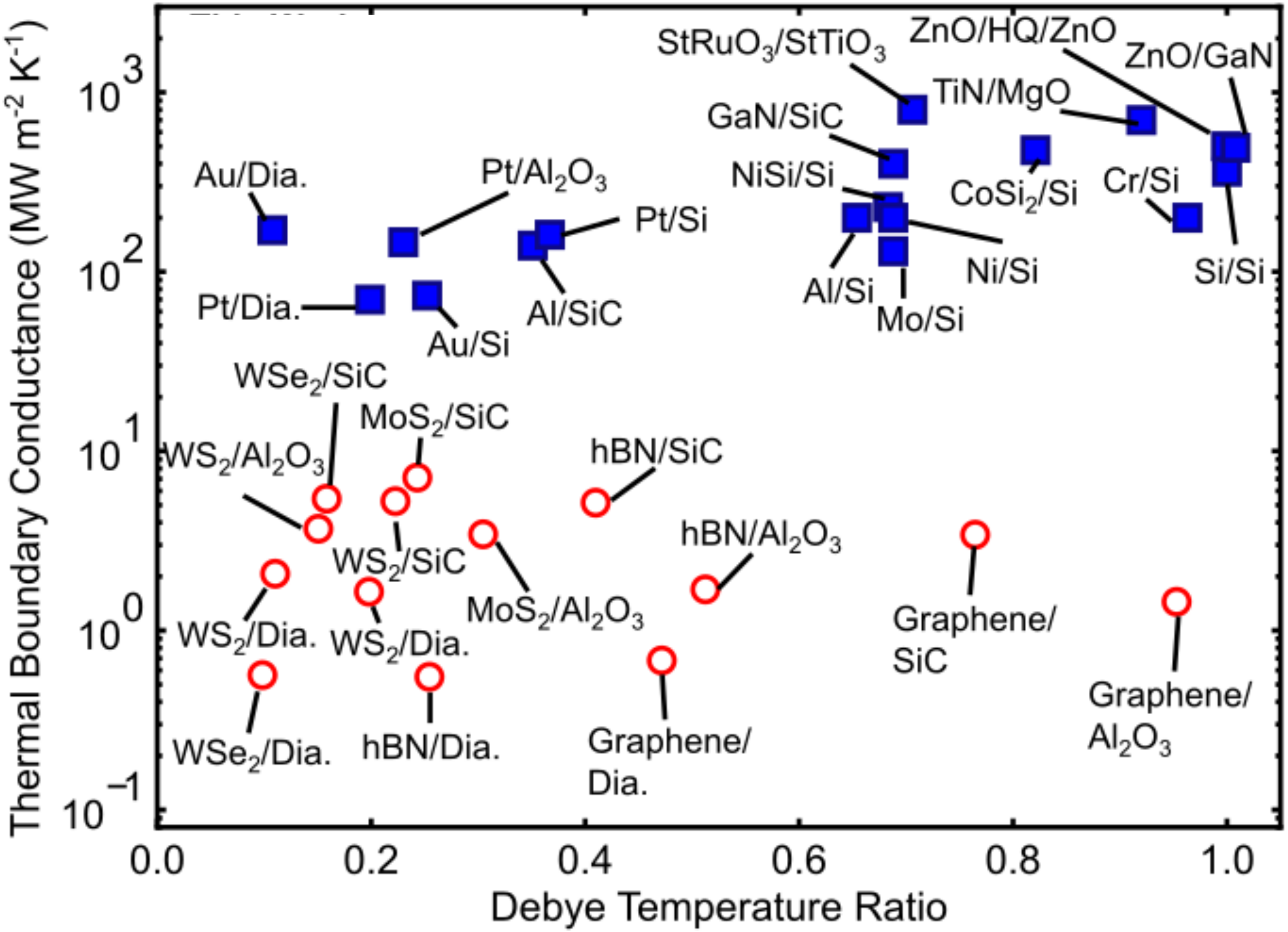




Metal/non-metal = Phonon dominated
Does it have to be?

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

TBCs are even lower at interfaces of 2D materials



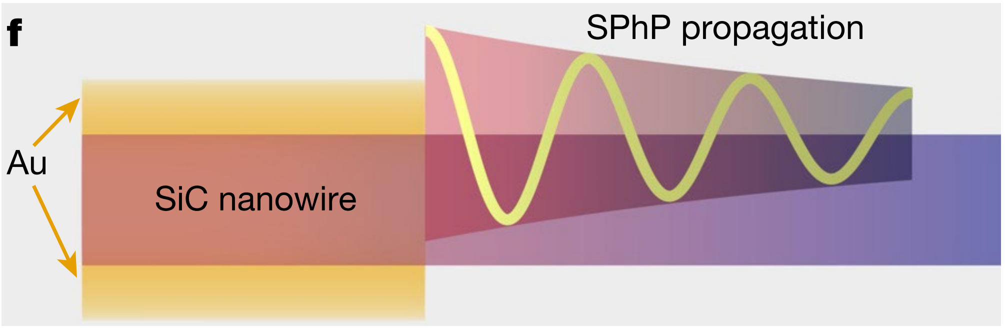
arXiv: 2401.09594

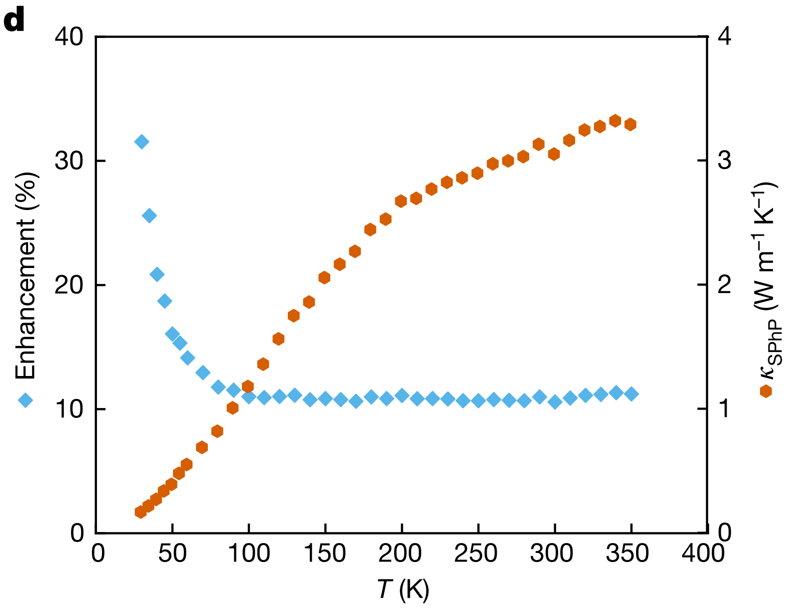
How can we increase TBC across metal/nonmetal interfaces?

Remarkable heat conduction mediated by non-equilibrium phonon polaritons

Zhiliang Pan[1], Guanyu Lu[1], Xun Li[2], James R. McBride[3], Rinkle Juneja[2], Mackey Long[4], Lucas Lindsay[2], Joshua D. Caldwell[1] & Deyu Li[1]✉

Nature | Vol 623 | 9 November 2023 | **307**





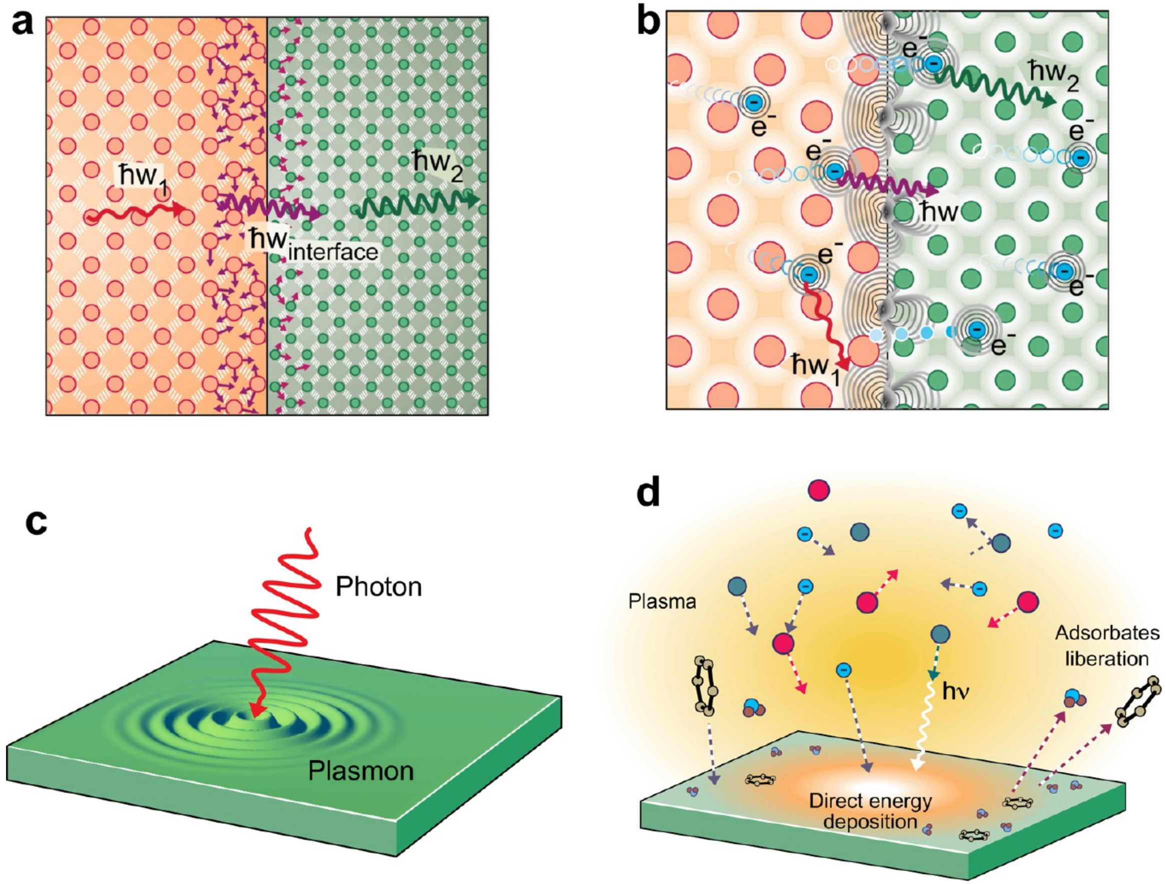
Recent review and perspective: *ACS Nano* 17, 14253 (2023)

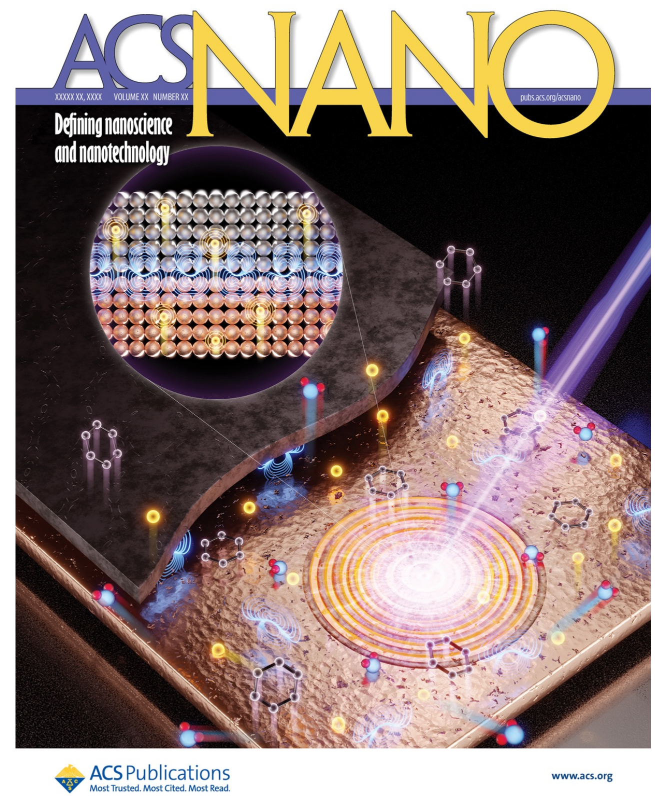
ACS NANO

www.acsnano.org

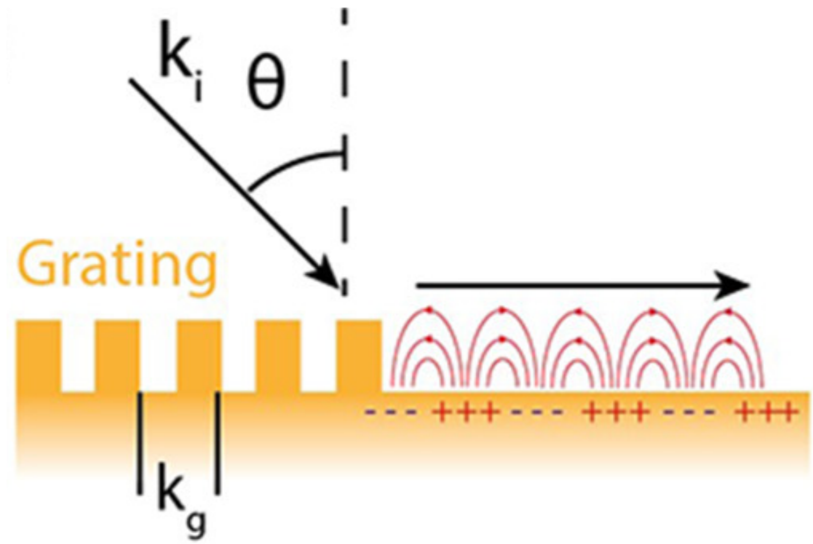
Ultrafast and Nanoscale Energy Transduction Mechanisms and Coupled Thermal Transport across Interfaces

Ashutosh Giri,* Scott G. Walton, John Tomko, Niraj Bhatt, Michael J. Johnson, David R. Boris, Guanyu Lu, Joshua D. Caldwell, Oleg V. Prezhdo, and Patrick E. Hopkins*

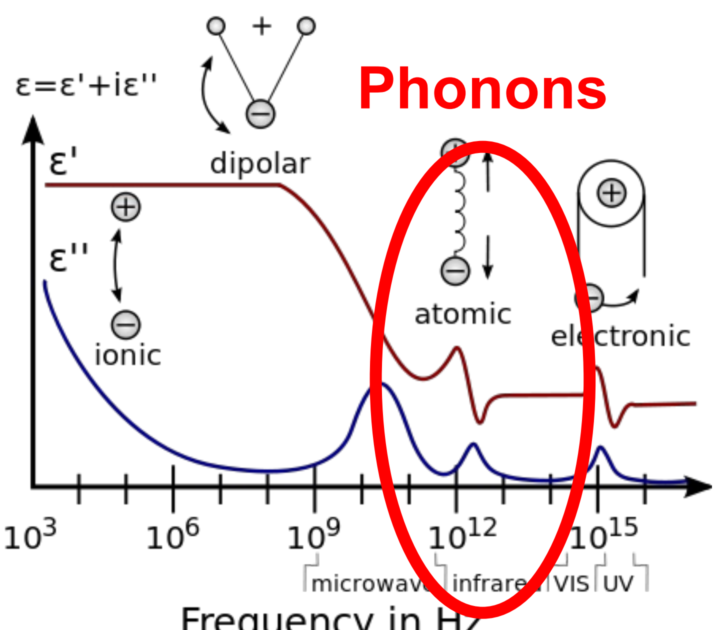




Phonon-polaritons: enhancing heat transfer?



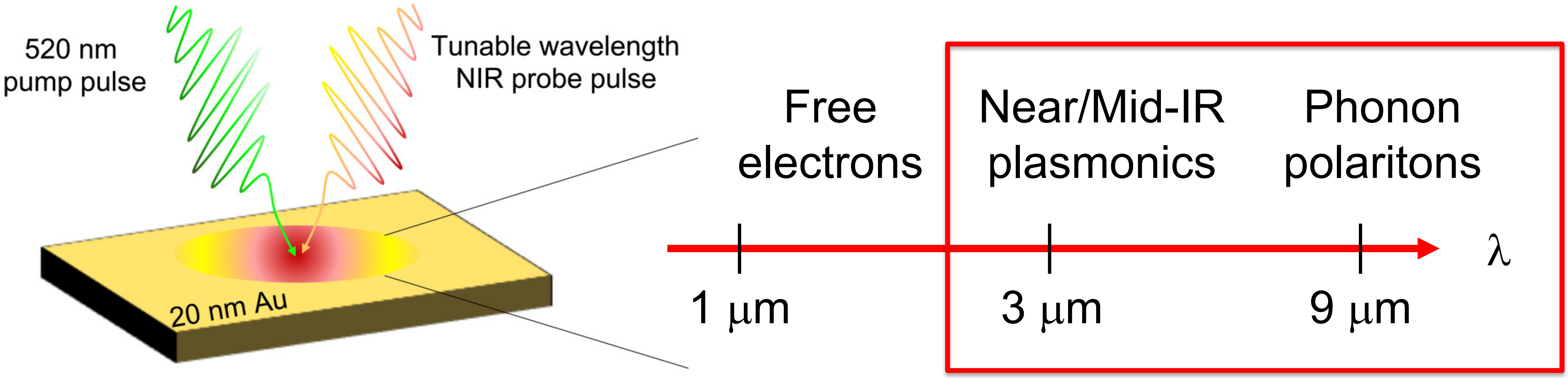
J. Appl. Phys. **125**, 191102 (2019)



- Light couples with electric dipole creating quasiparticle
- At IR wavelengths: Phonon polariton!
- PhP quasiparticles can propagate at ~1% of the speed of light!
- Prior evidence of thermal conductivity enhancement during excitation of PhPs

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

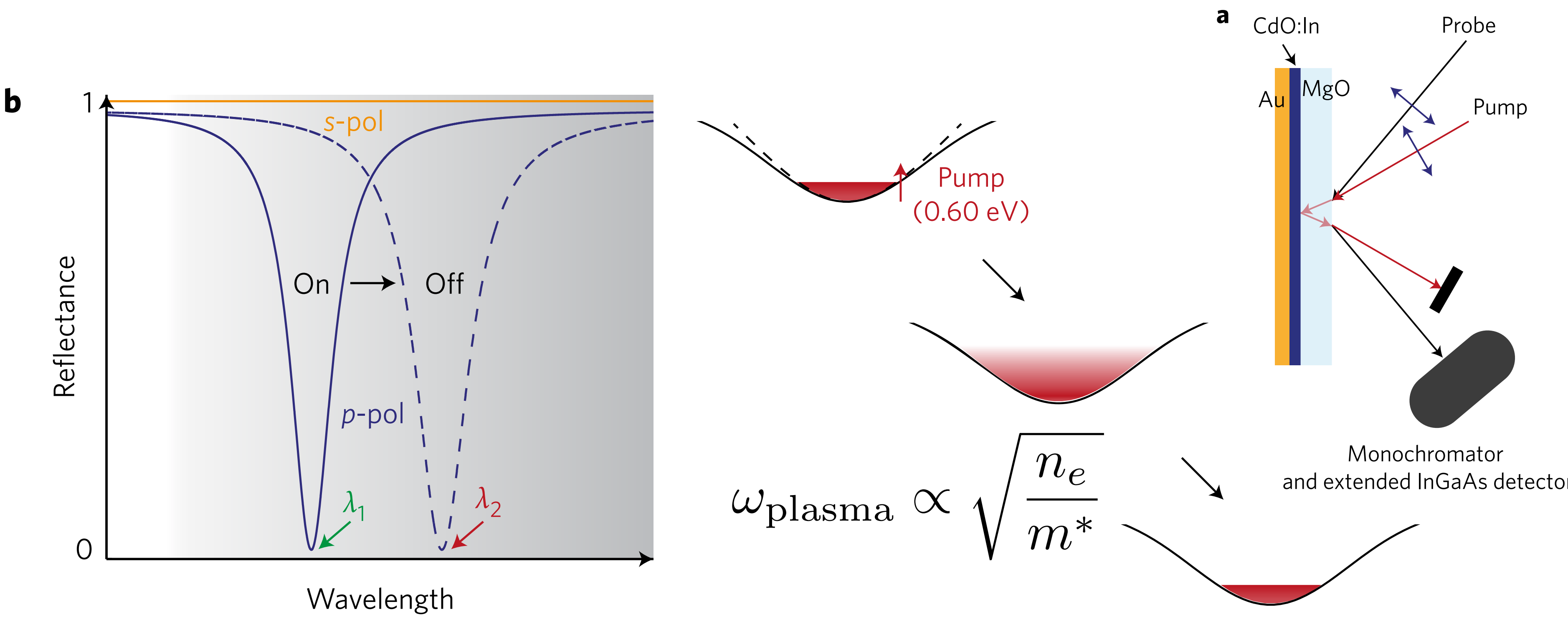
Pumping with heat, probing in IR away from e^- transitions



- **Thermally modulated plasmon-polariton in CdO**
  - Electron TBC controls plasmon response
- **Thermally modulated phonon-polariton in h-BN**
  - Broadband radiative flux from Au heat PhP

Can we use interfacial heat transfer to manipulate the optical properties of materials?

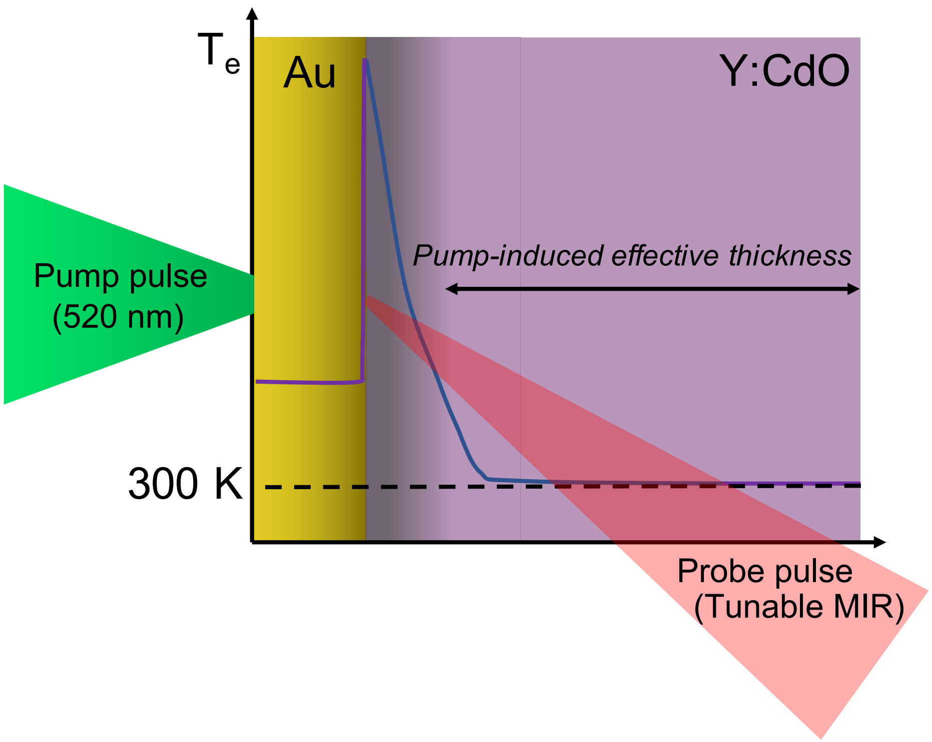
Carrier distributions modulate optical properties of ENZ plasmonic modes in CdO

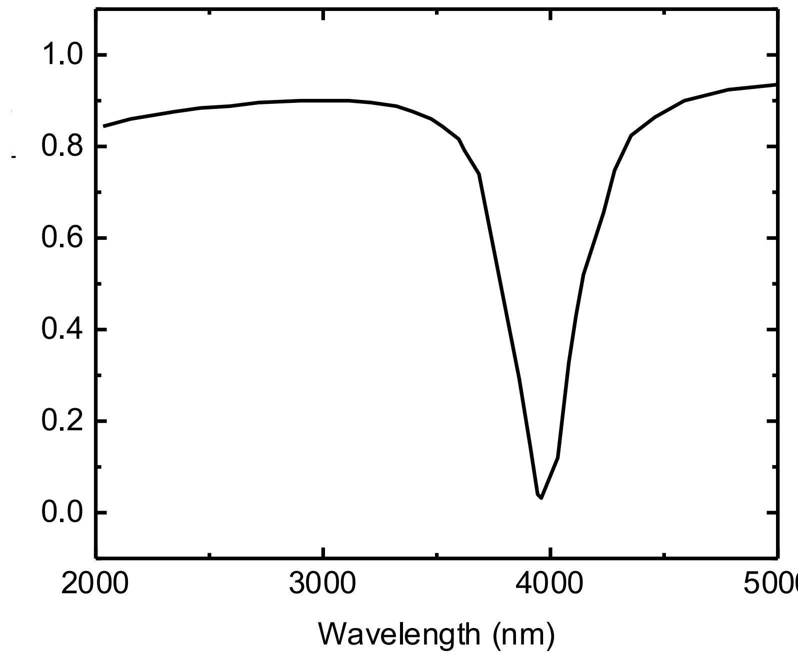


Nature Photonics **11**, 390

Recall earlier example

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



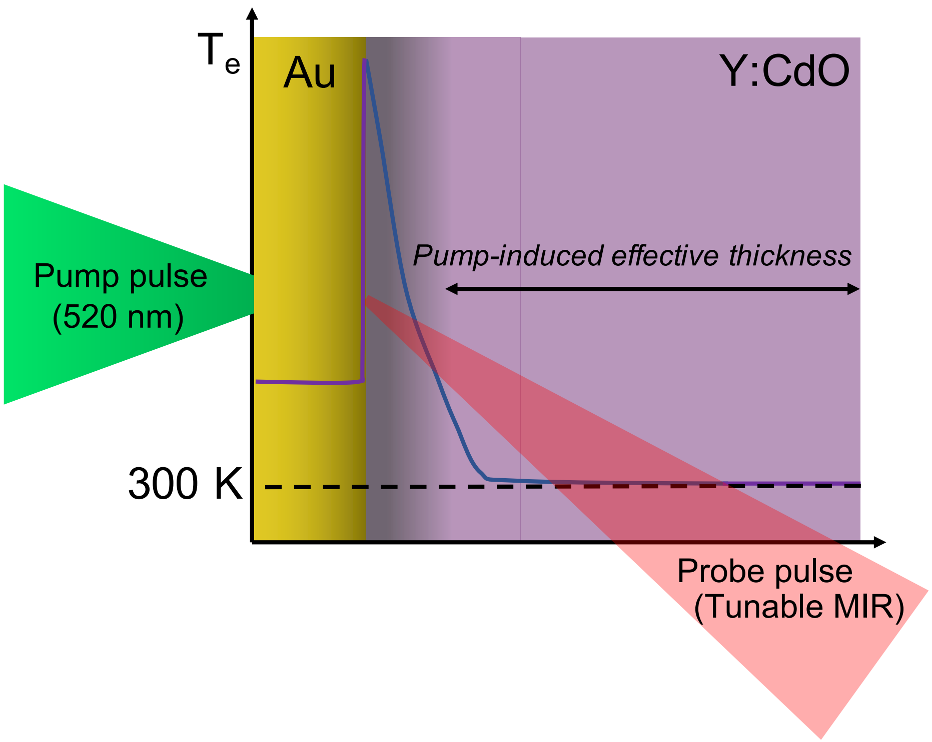


Tomko *et al. Nature Nano.* **16**, 47

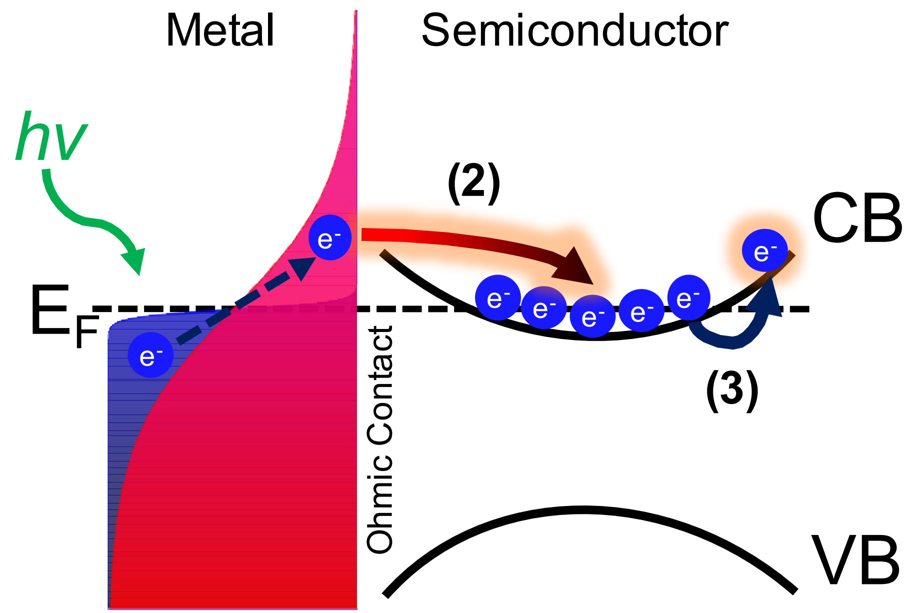
Collaboration: Caldwell (Vanderbilt), Maria (PSU), Prezhdo (USC)

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

- Energy easily transmitted across interface when out of equilibrium with phonons
- Slowly “goes back” across the interface when diffusive
- **This “ballistic thermal injection” is different than charge injection**



Ballistic thermal injection
(Energy transfer)



Tomko *et al. Nature Nano.* **16**, 47

Collaboration: Caldwell (Vanderbilt), Maria (PSU), Prezhdo (USC)

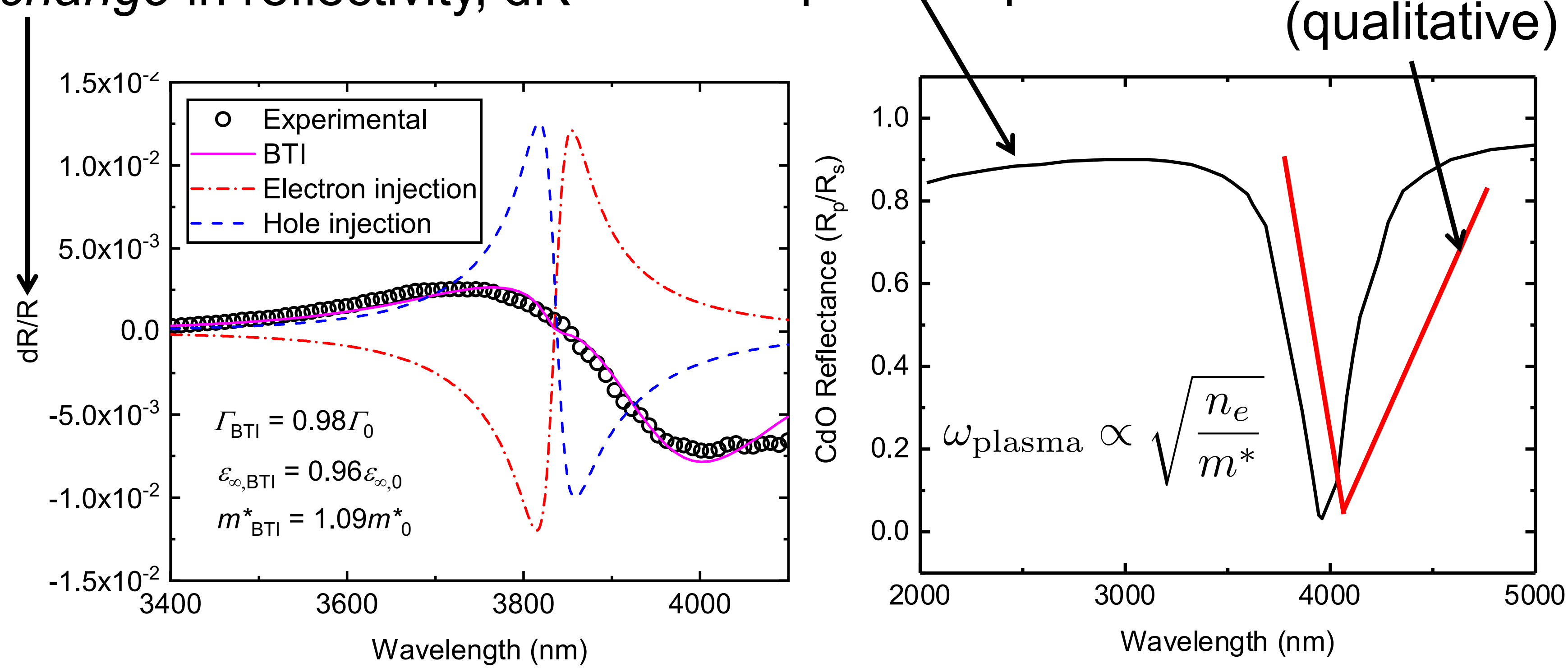
Pump electrons in Au, probe plasmon in CdO

Asymmetric red shift in ENZ plasmon mode due to BTI

Note we are measuring *change* in reflectivity, dR

Au/CdO/sapphire absorption response

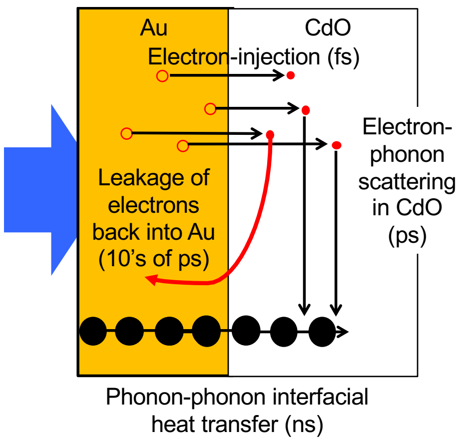
Thermo-modulated response from indirect heating form Au film (qualitative)



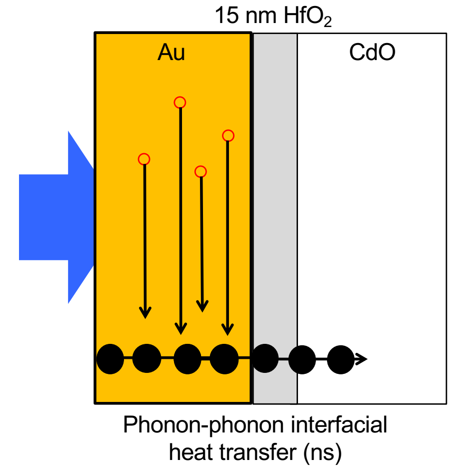
Tomko *et* al. *Nature Nano.* **16**, 47

Collaboration: Caldwell (Vanderbilt), Maria (PSU), Prezhdo (USC)

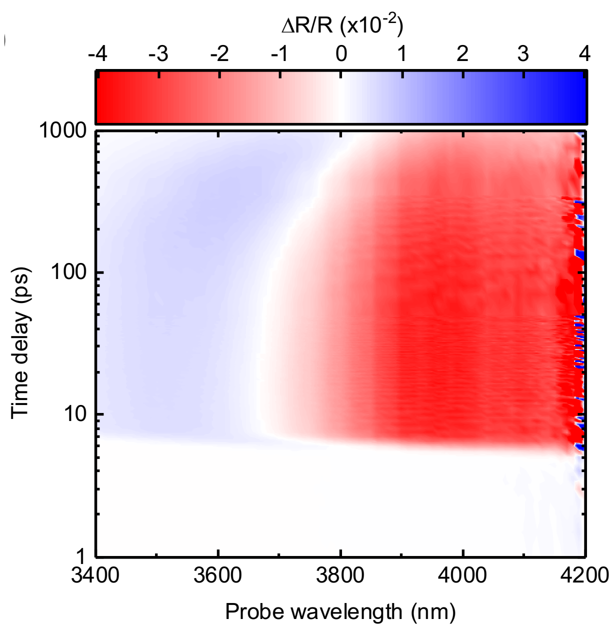
And it's not an optical artifact

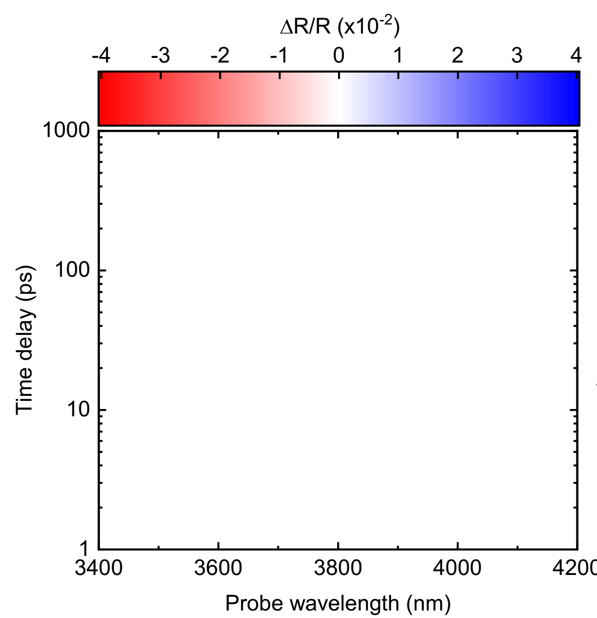


15 nm HfO_2 layer prevents any electron energy from moving from Au to CdO, resulting in no measurable response



Au/CdO



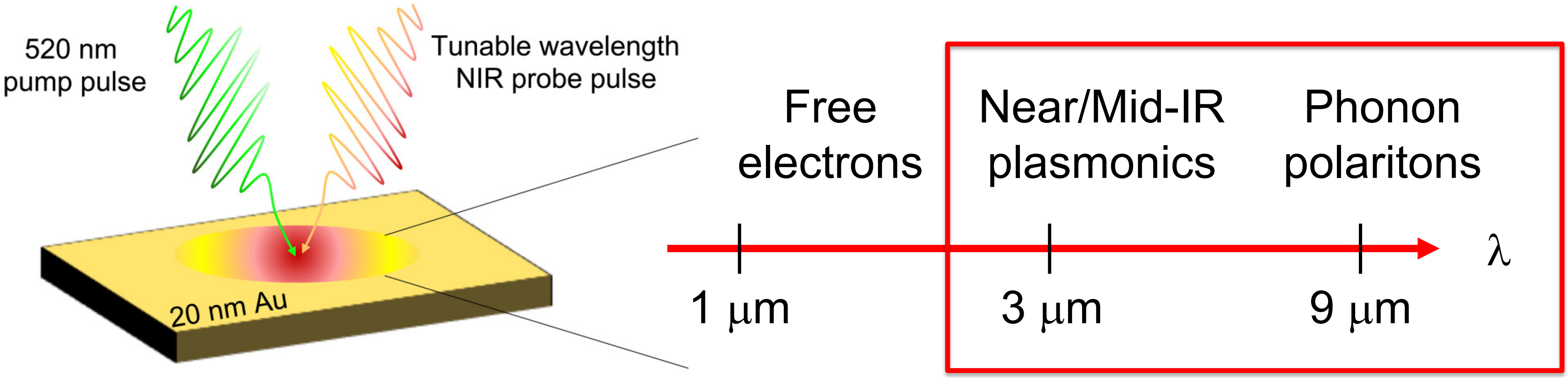


Au/HfO_2/CdO (same scale)

Tomko *et* al. *Nature Nano.* **16**, 47

Collaboration: Caldwell (Vanderbilt), Maria (PSU), Prezhdo (USC)

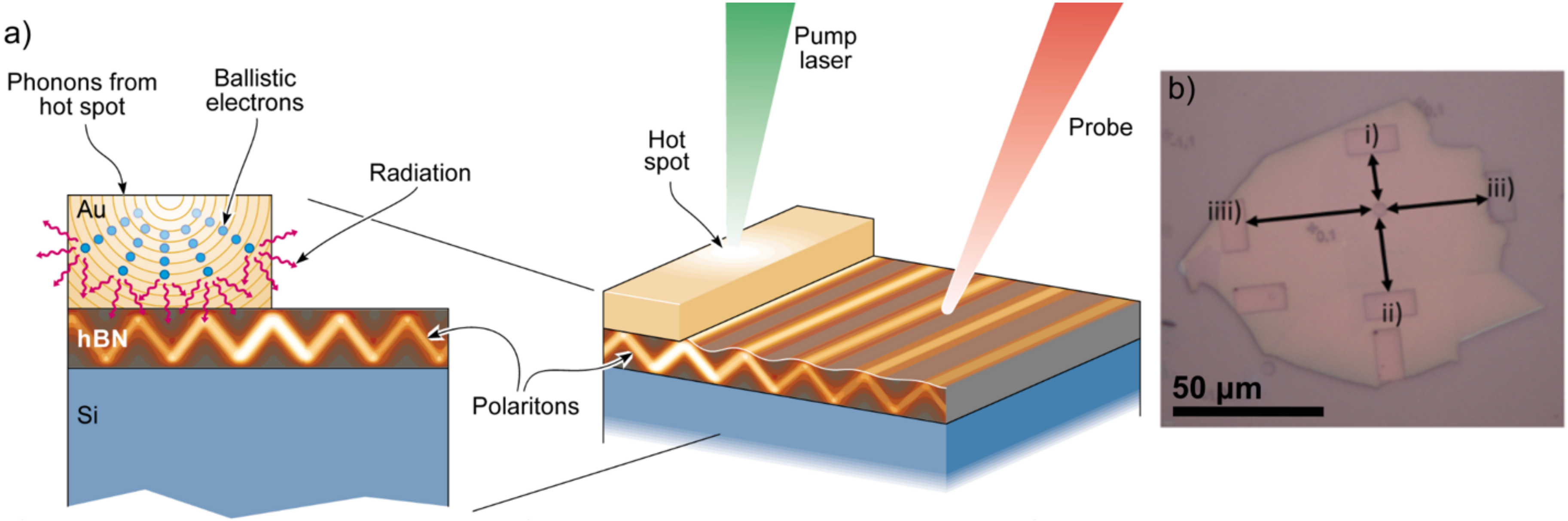
Pumping with heat, probing in IR away from e⁻ transitions



- **Thermally modulated plasmon-polariton in CdO**
  - Electron TBC controls plasmon response
- **Thermally modulated phonon-polariton in h-BN**
  - Broadband radiative flux from Au heat PhP

Can we modulate and control PhP lifetimes with heat?

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



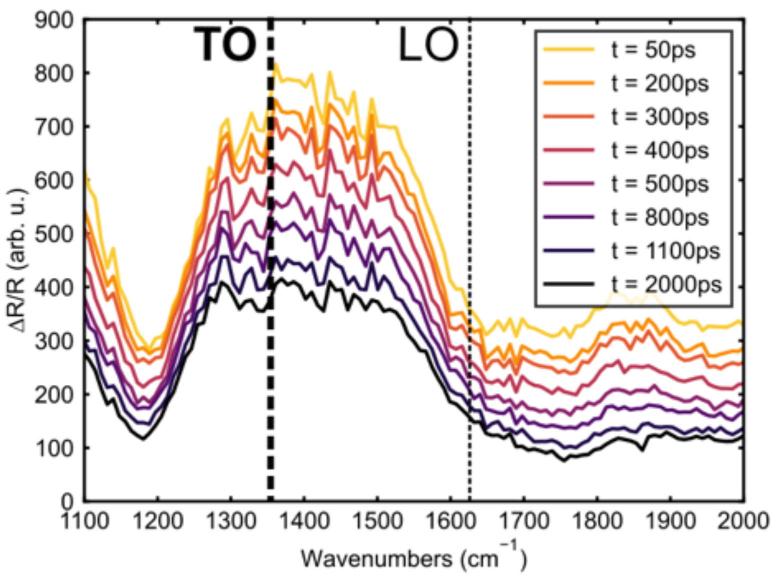
Ultrafast evanescent heat transfer across solid interfaces via hyperbolic phonon-polaritons in hexagonal boron nitride

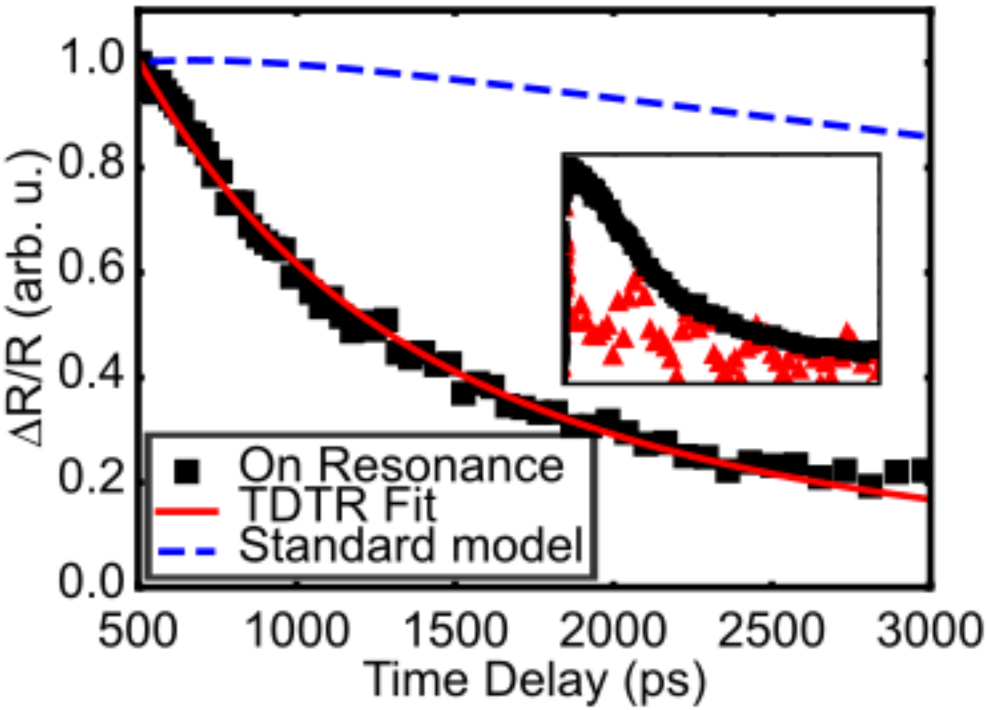
William Hutchins,[1] John A. Tomko,[1] Dan M. Hirt,[1] Saman Zare,[1] Joseph R. Matson,[2] Katja Diaz-Granados,[2] Mingze He,[3] Thomas Pfeifer,[1] Jiahan Li,[4] James Edgar,[4] Jon-Paul Maria,[5] Joshua D. Caldwell,[2,3,&] Patrick E. Hopkins[1,6,7,%]

arXiv: 2401.09594

Enhanced heat sinking with PhP coupling

- Temperatures of PhP modes decay an order of magnitude faster than non-PhP modes

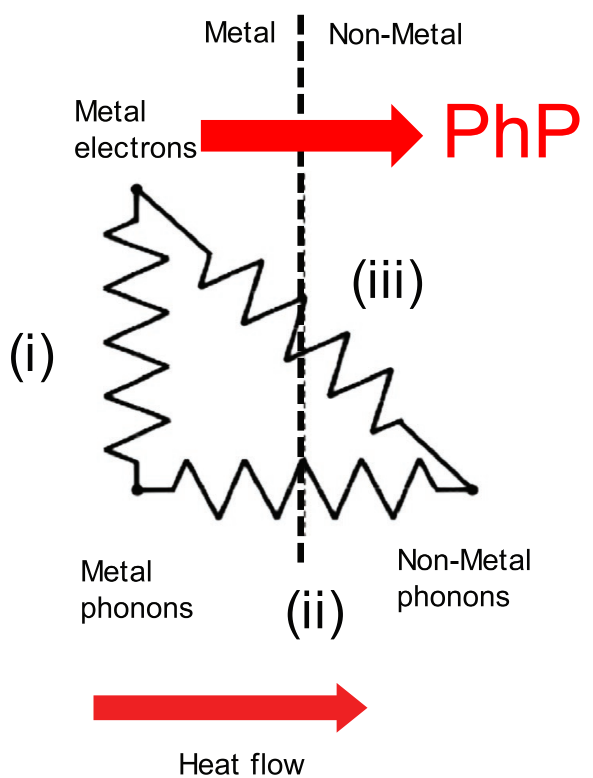


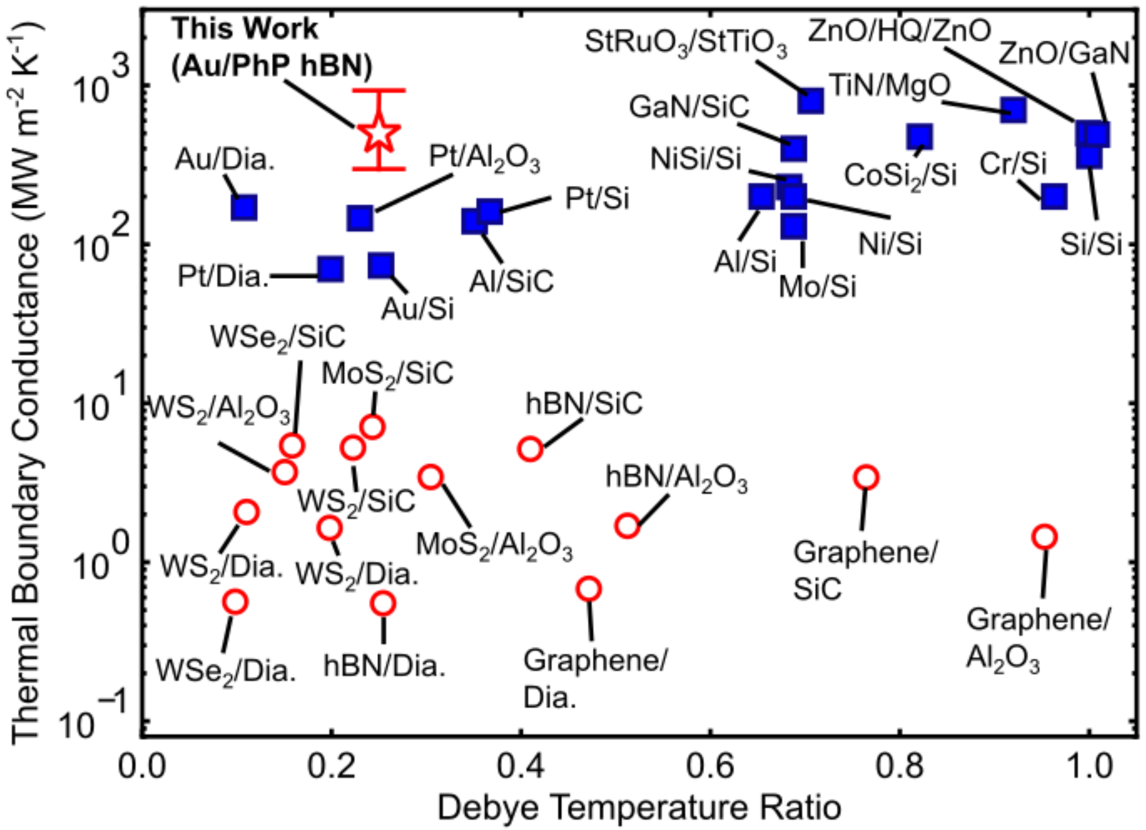


arXiv: 2401.09594

Enhanced heat sinking with PhP coupling

- Intensely heated nonequilibrium electrons emit energy in near field to HPhP's in hBN
- Provide additional heat transfer mechansim across Au/hBN interface to enhance TBC





arXiv: 2401.09594

Outline

Theme: Coupling of carriers across interfaces lead to unique energy states and modes of heat transfer

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  - Minimum thermal conductivity and creating "crystals of interfaces"
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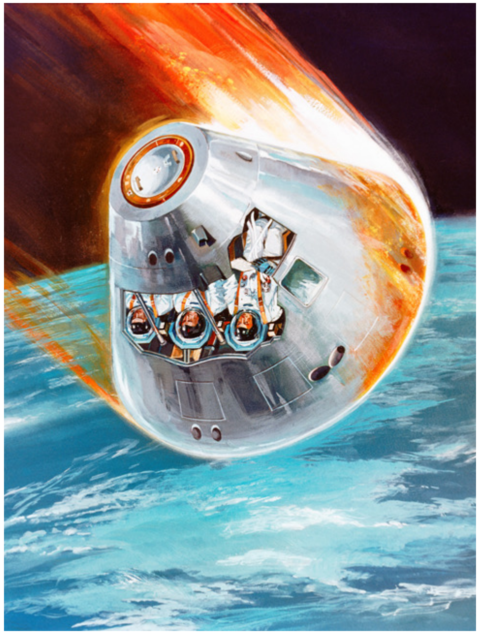
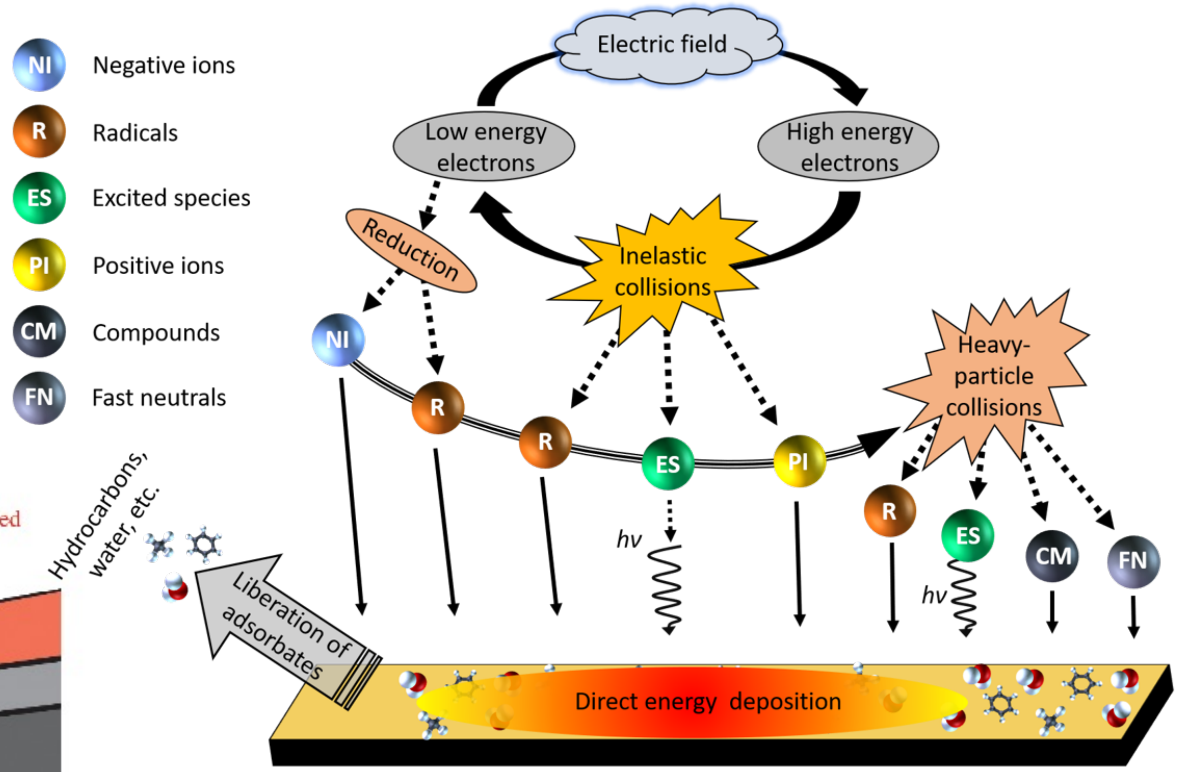
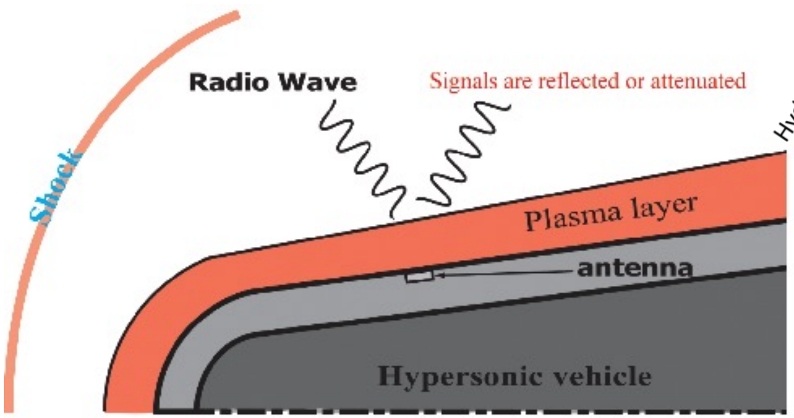
- **Transient temperature changes during plasma-surface interactions**
  - **"Plasma cooling"**

Plasma surface interactions



Re-entry vehicle: “All you can do is pray for it”
J. App. Phys. news referencing: **117** 233301 (2015)

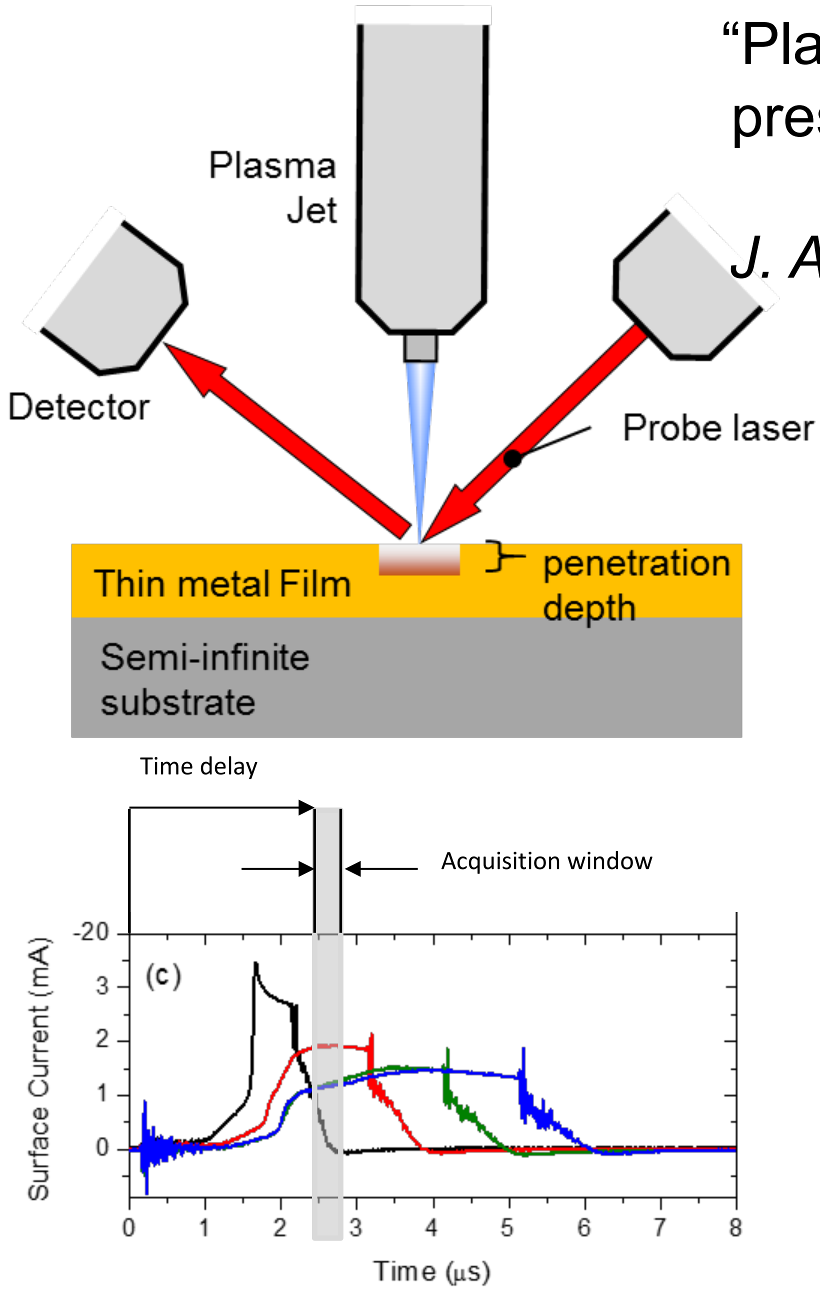




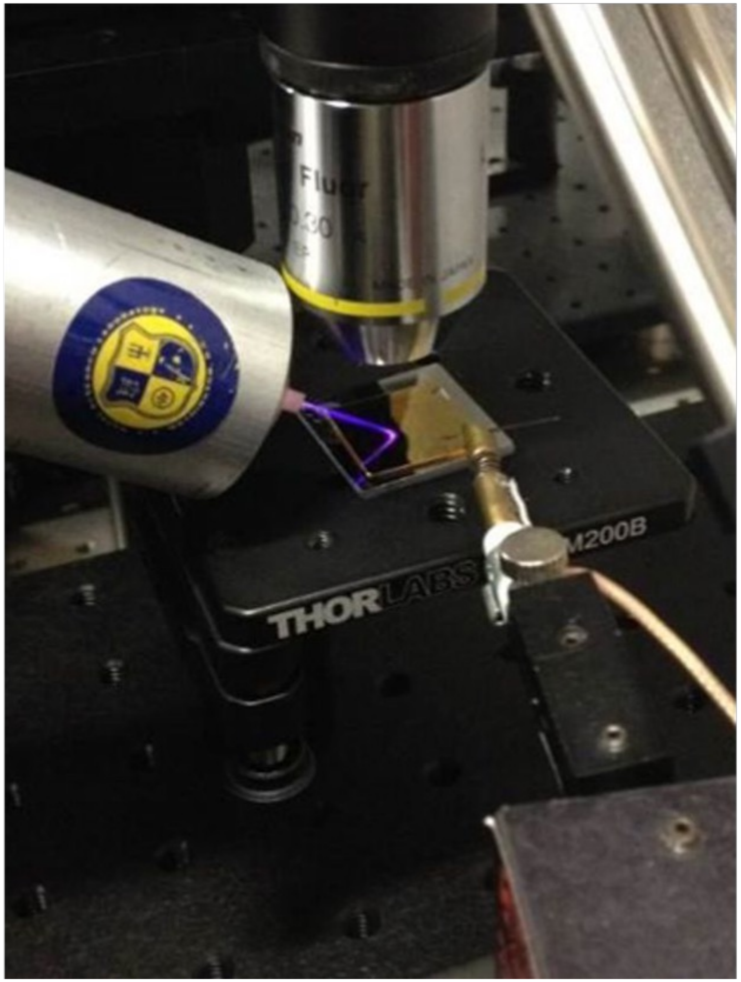
Kim: “Plasma communication for radio blackout during hypersonic flight”

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

Thermoreflectance for plasma diagnostics



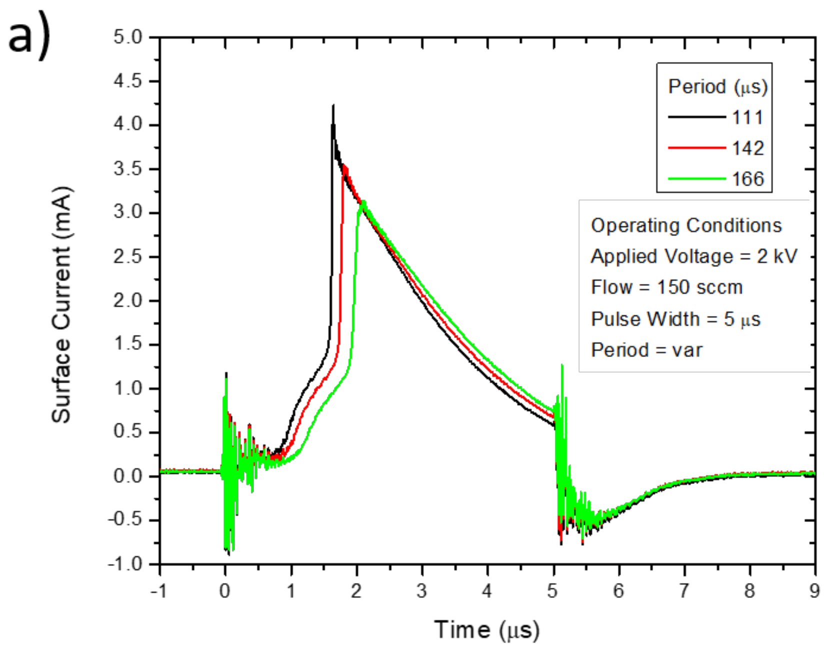
"Plasma-surface interactions in atmospheric pressure plasmas: *In situ* measurements of electron heating in materials"
J. Appl. Phys. **124**, 043301 (**Editor's Pick**)

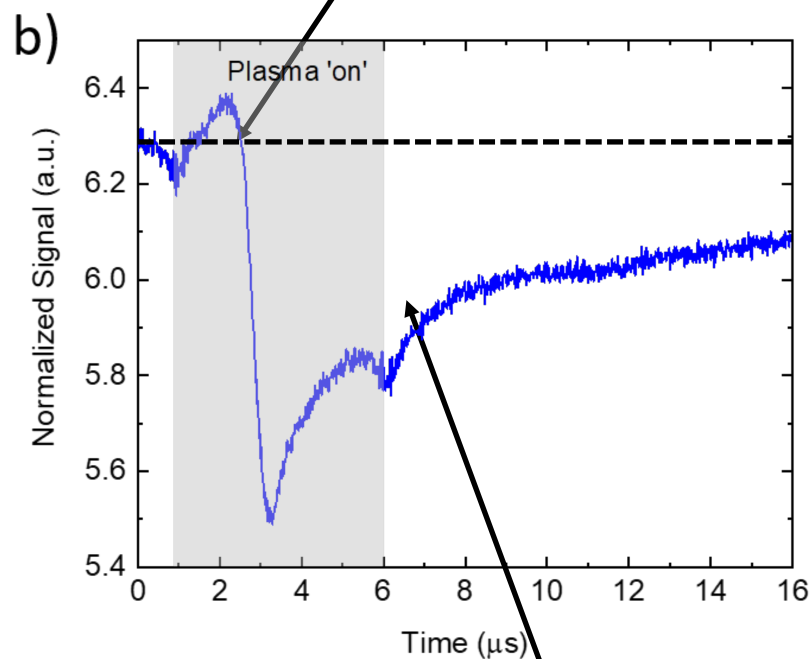


Tracking surface temperature during plasma jet irradiation

Surface current temporal profile for different DC pulse widths

Note sign inflection….why?

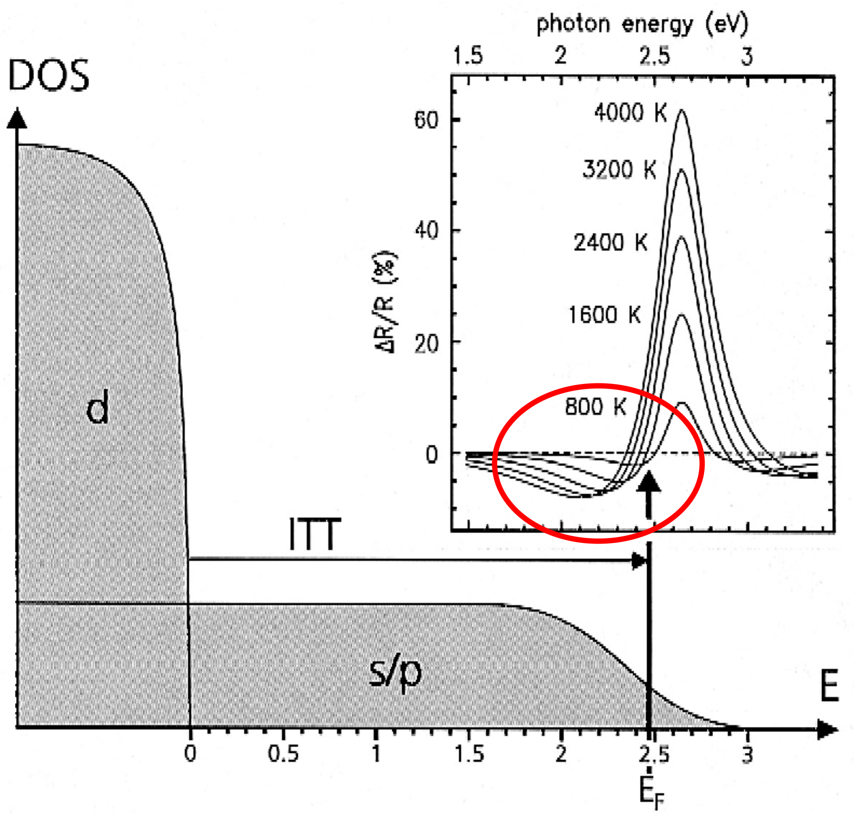




"Accommodation" from neutrals?

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

Thermoreflectance of gold



ELSEVIER

Chemical Physics 251 (2000) 237–258

Chemical Physics

www.elsevier.nl/locate/chemphys

Electron and lattice dynamics following optical excitation of metals

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

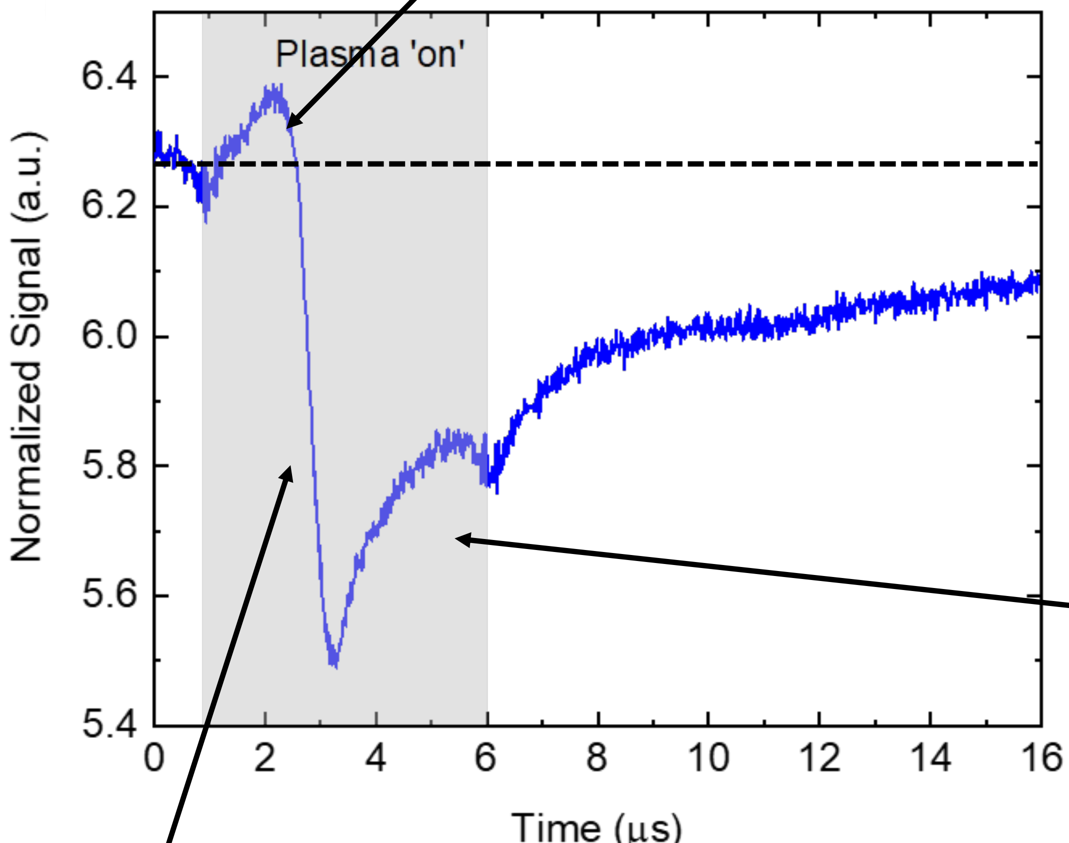
Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

Received 13 May 1999

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

Plasma cooling of surface

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

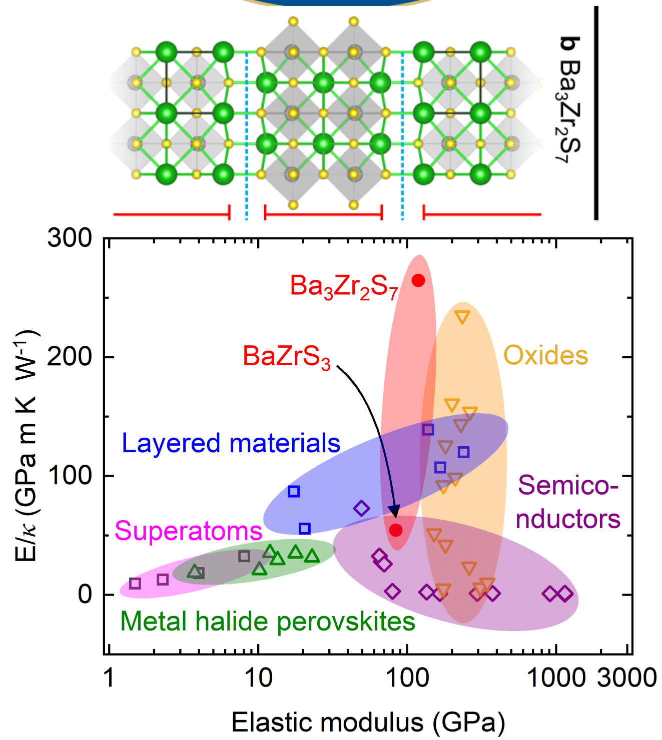


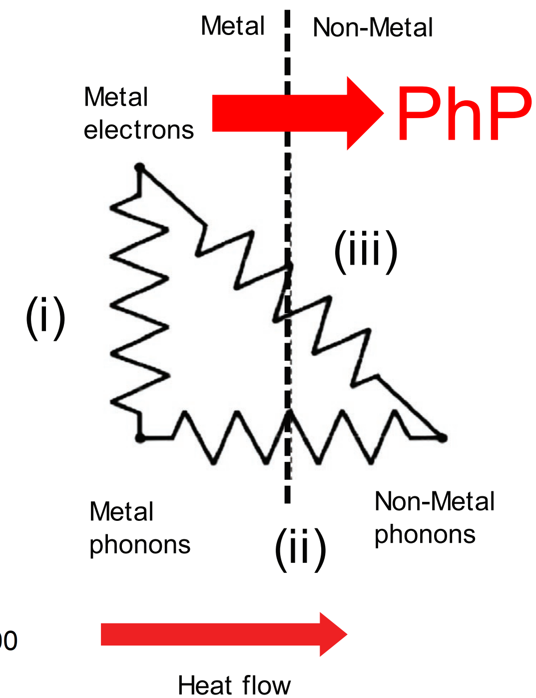
Cooling of surface from decrease in surface current or pulse turning off

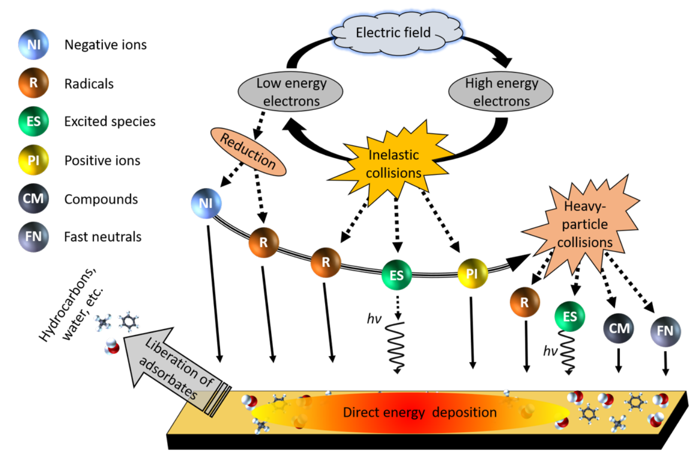
-ΔR = increase in temperature from energy transfer to gold

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

Theme: Coupling of carriers across interfaces lead to unique energy states and modes of heat transfer







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ExSITE AT THE UNIVERSITY OF VIRGINIA

Recent review and perspective: *ACS Nano* 17, 14253 (2023)

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Ultrafast and Nanoscale Energy Transduction Mechanisms and Coupled Thermal Transport across Interfaces

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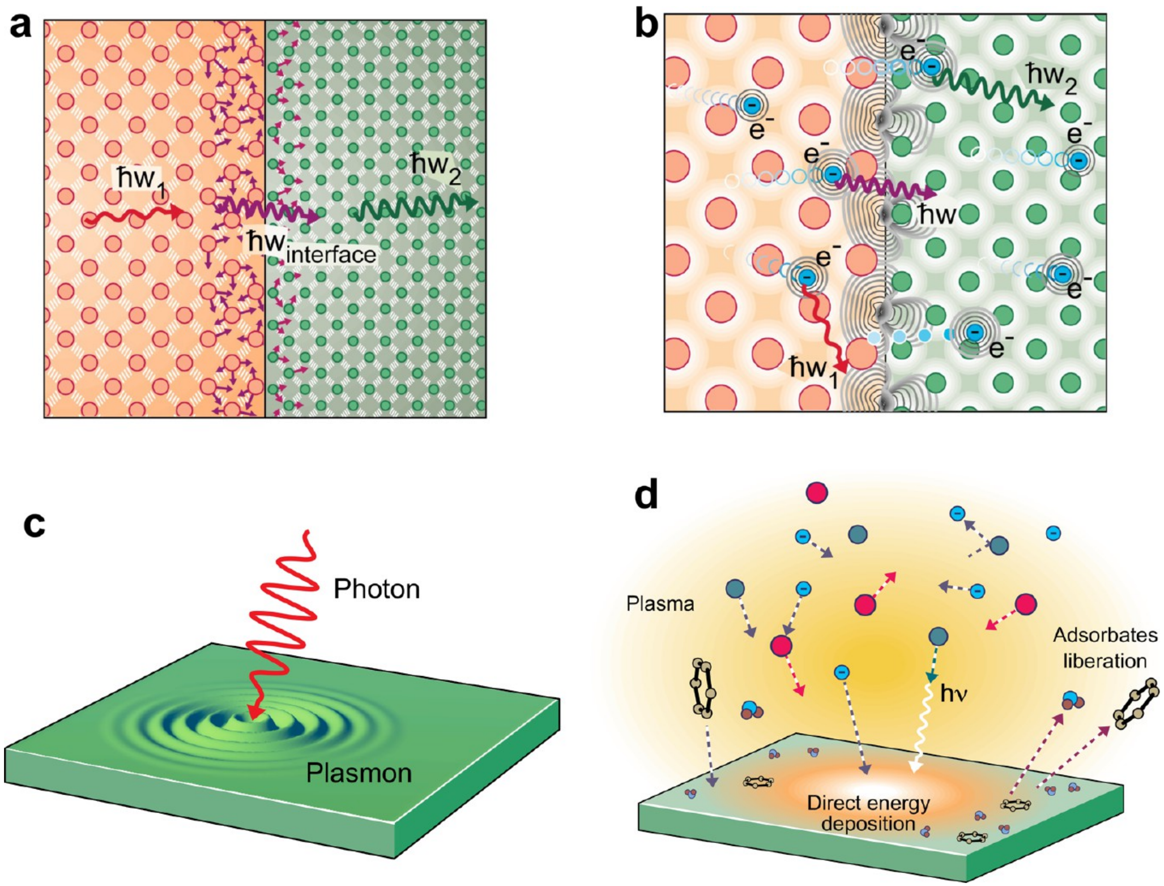
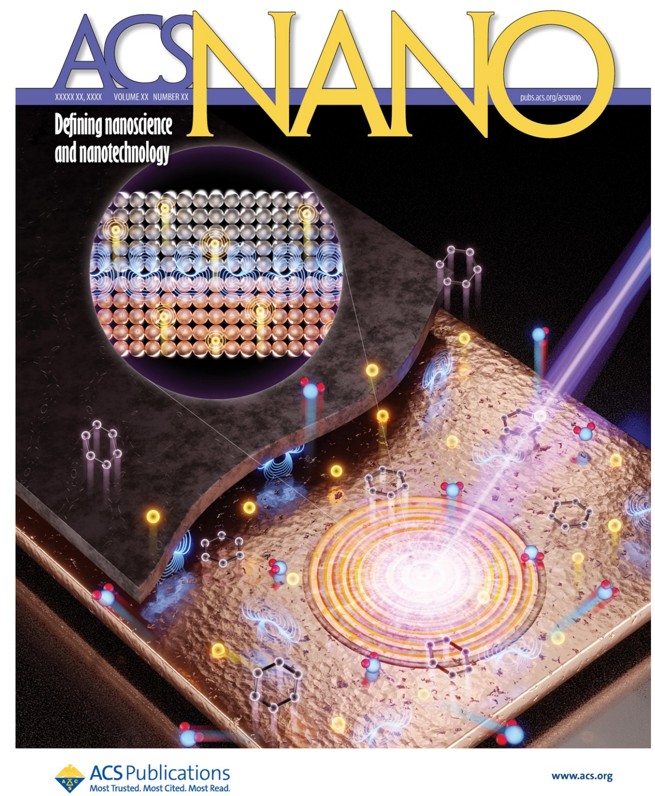
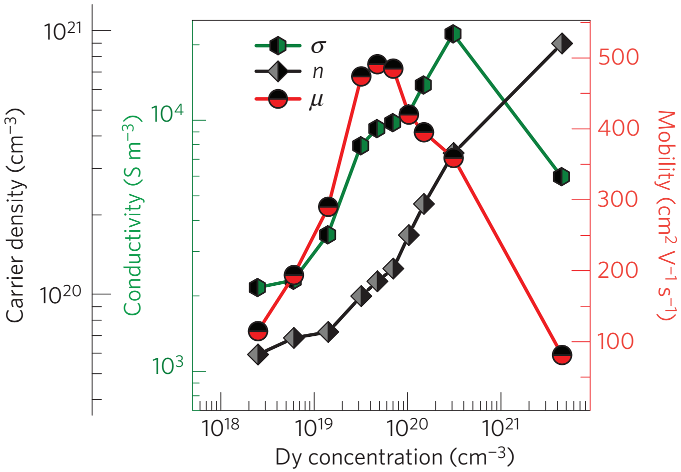
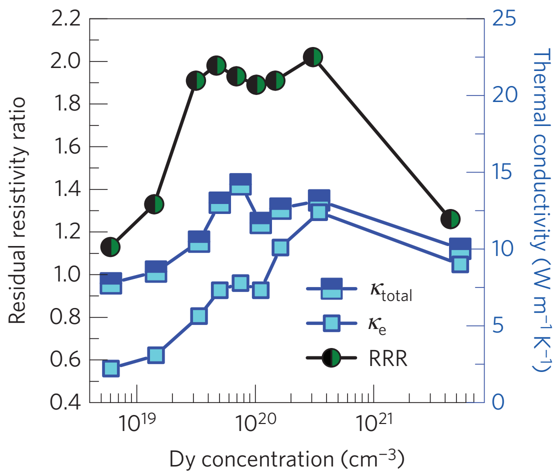




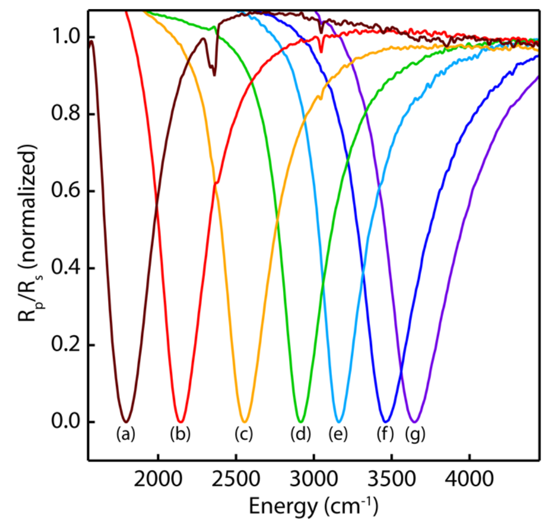
CdO – a gateway for mid-IR plasmonics





Nat. Mat. **14**, 414

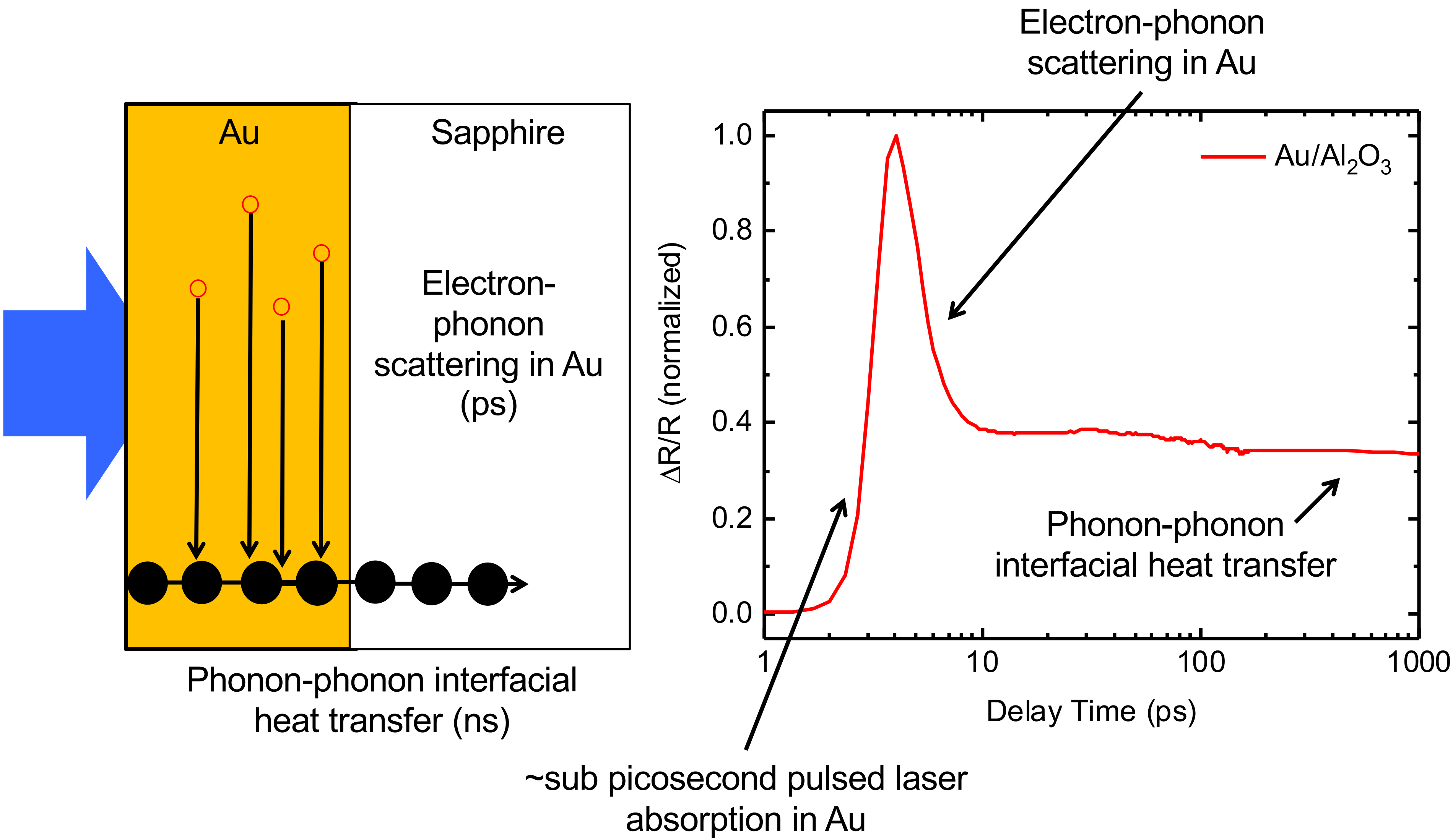
Appl. Phys. Lett. **108**, 021901



ACS Photonics **4**, 1885

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

Nonequilibrium processes at Au/CdO interfaces



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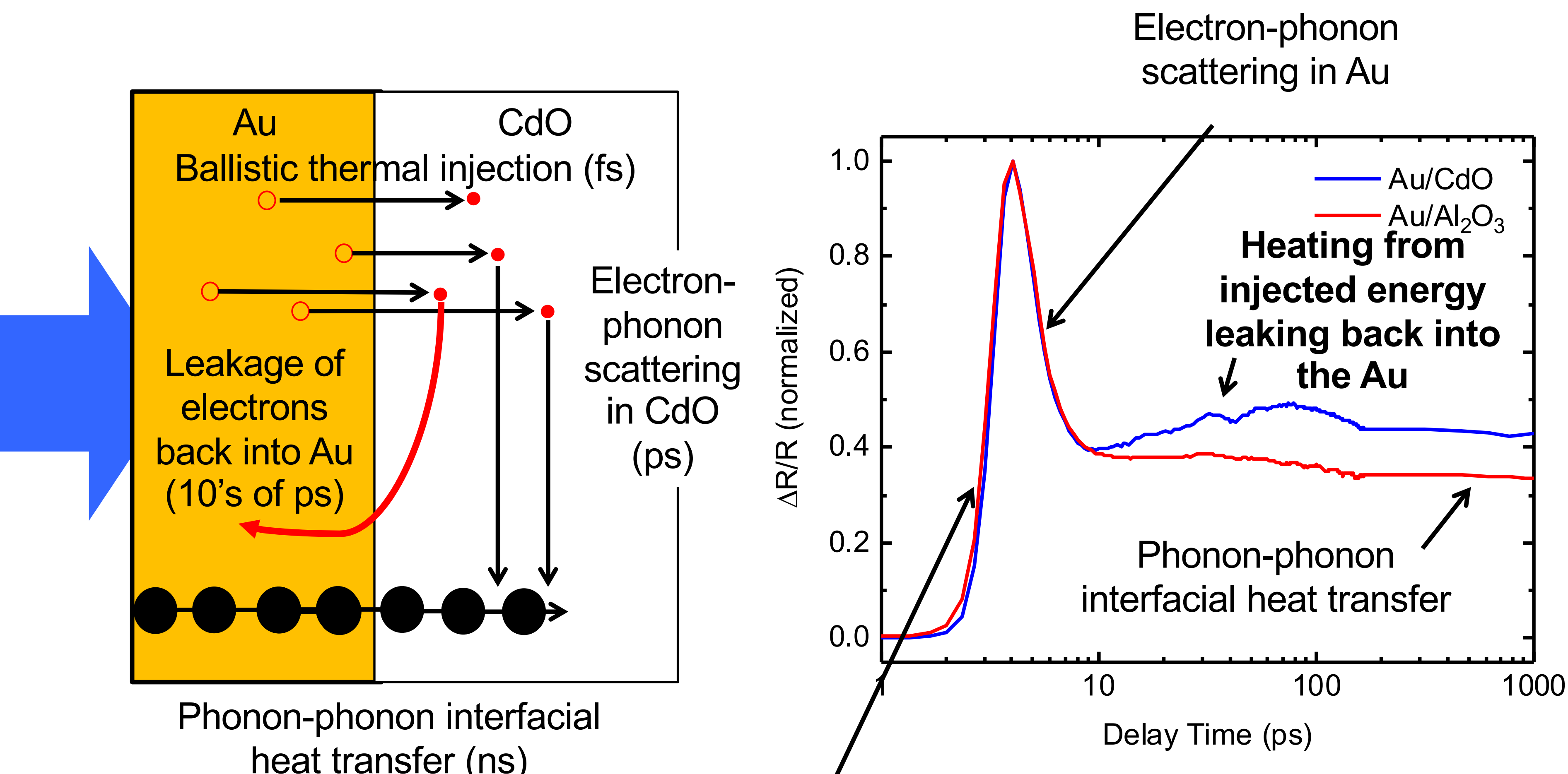
Nonequilibrium processes at Au/CdO interfaces



~sub picosecond pulsed laser absorption in Au **+ charge injection into CdO**

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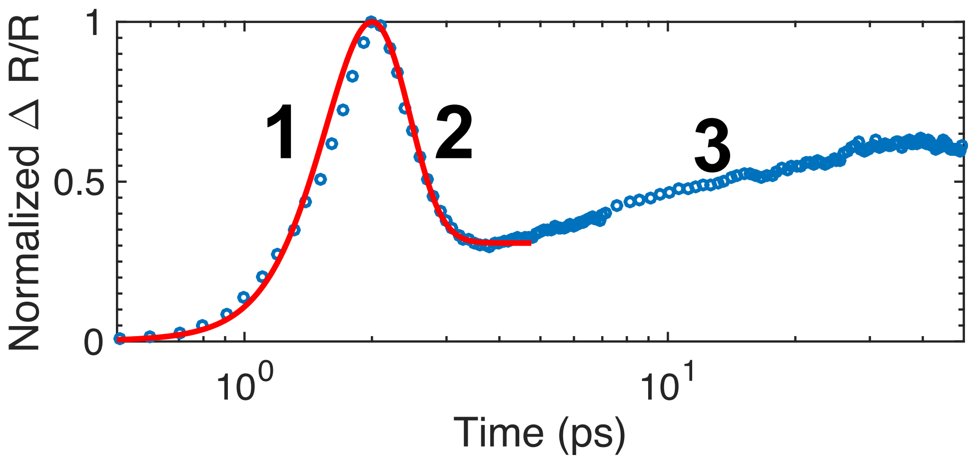
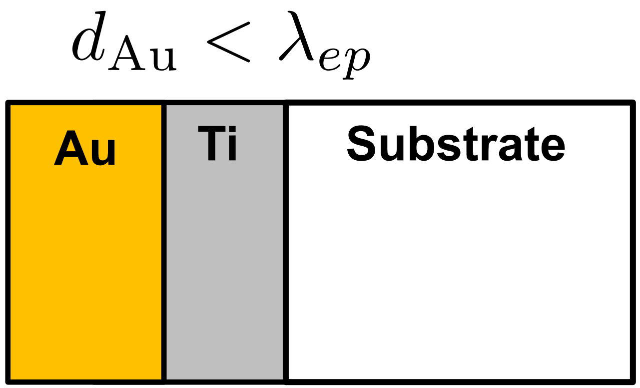
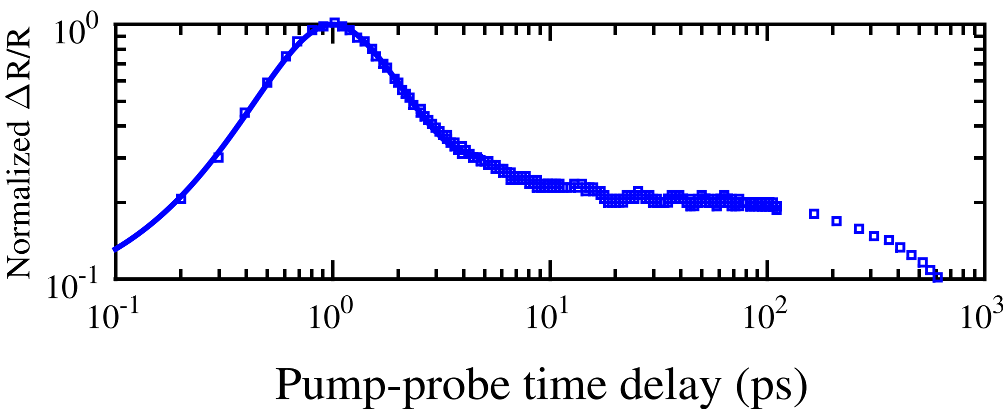
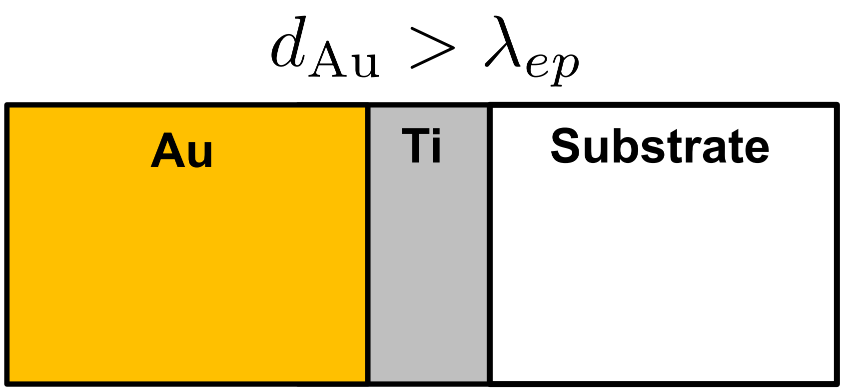
TDTR measurements of time scales of noneq. transport

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



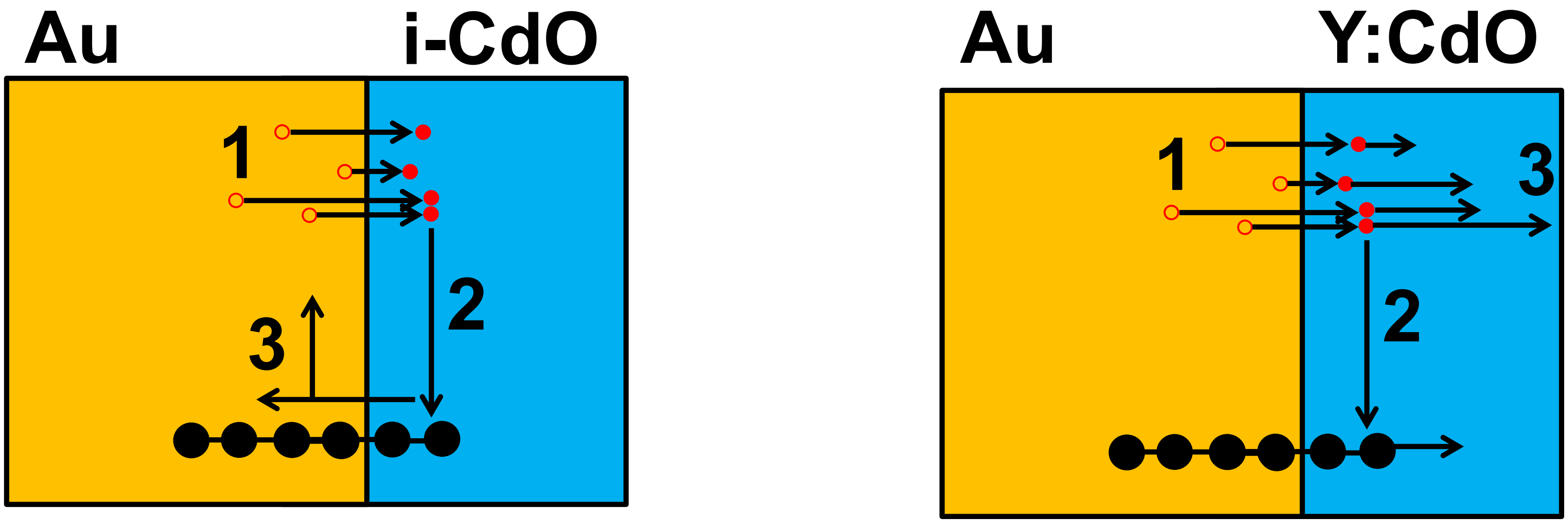






Nonequilibrium processes at Au/CdO interfaces

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



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