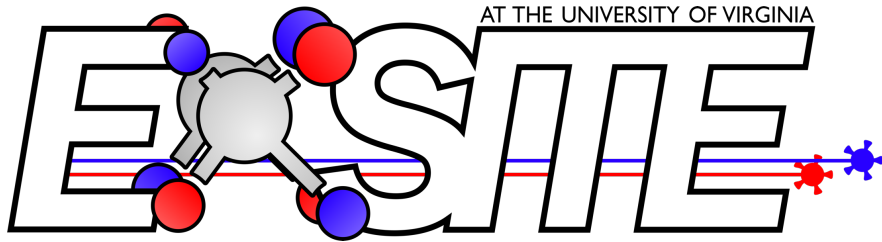




UVA

SCHOOL *of* ENGINEERING & APPLIED SCIENCE

Pushing the limits of thermal transport in ceramics with wurtzite ferroelectric nitrides, entropy stabilized oxides, and high mobility cadmium oxide



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Rost, Prof. VT



Donovan, Prof. USNA



Hutchins, PD TAMU



Gaskins, CEO LT



Makarem, PhD



Tomko, Diamond Foundry



Giri, Prof. UMD



Foley, VP LT



Zare, PhD



Braun, VP LT



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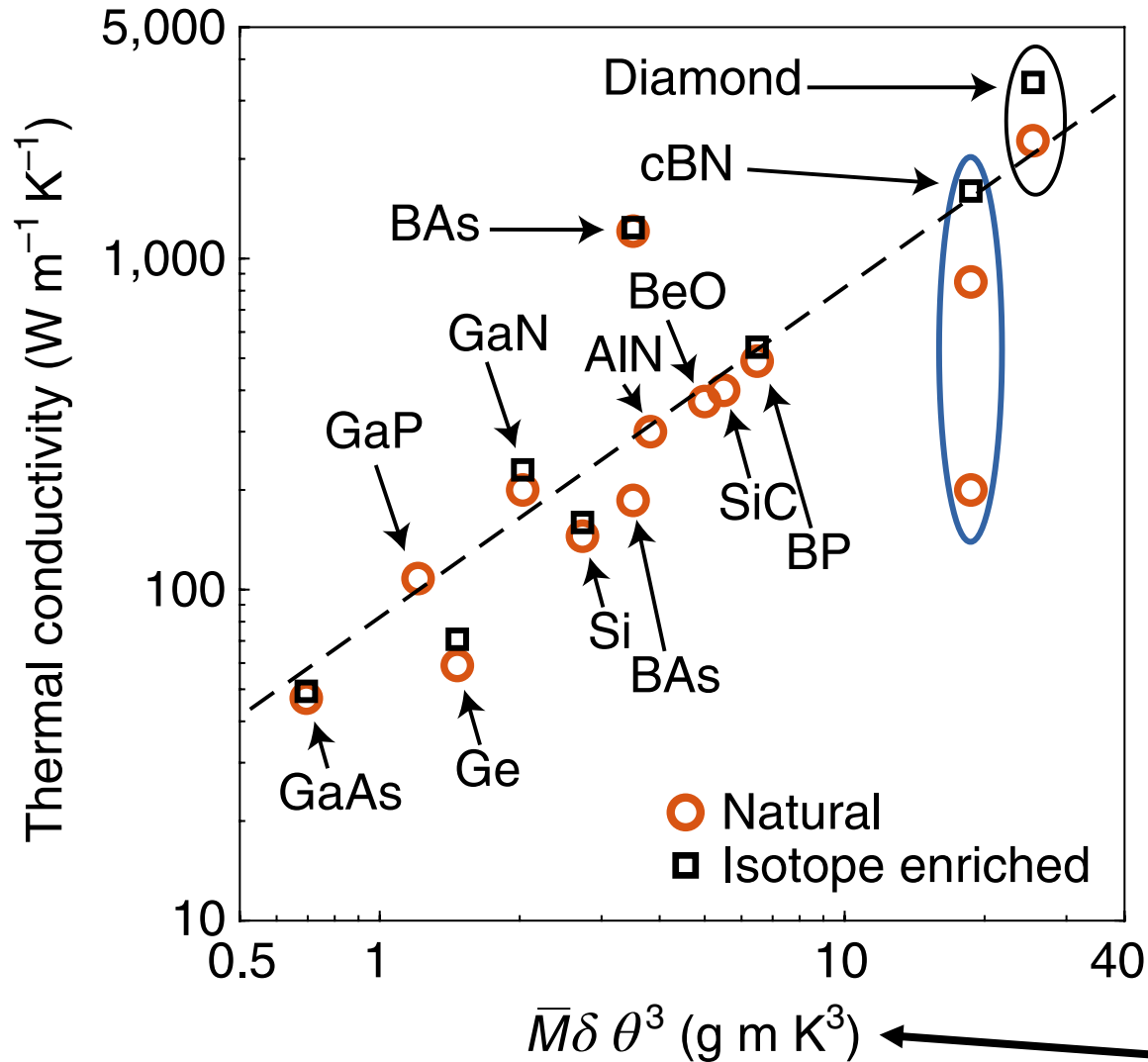
SCHOOL *of* ENGINEERING & APPLIED SCIENCE

**How Professor JP Maria has impacted and redefined
thermal transport in materials: building bridges
and creating new directions**



Stiffer crystalline materials - Larger λ - Larger ν - Larger κ

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



- What makes a high κ material?
- Slack Criteria:
 - Strong bonds
 - Light masses
 - Small unit cell
 - Minimize defects!

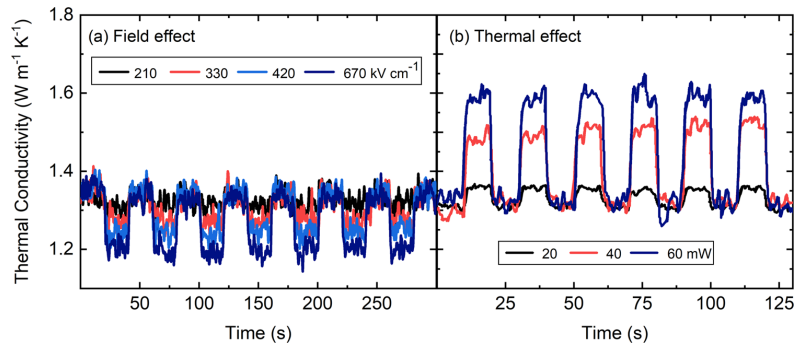
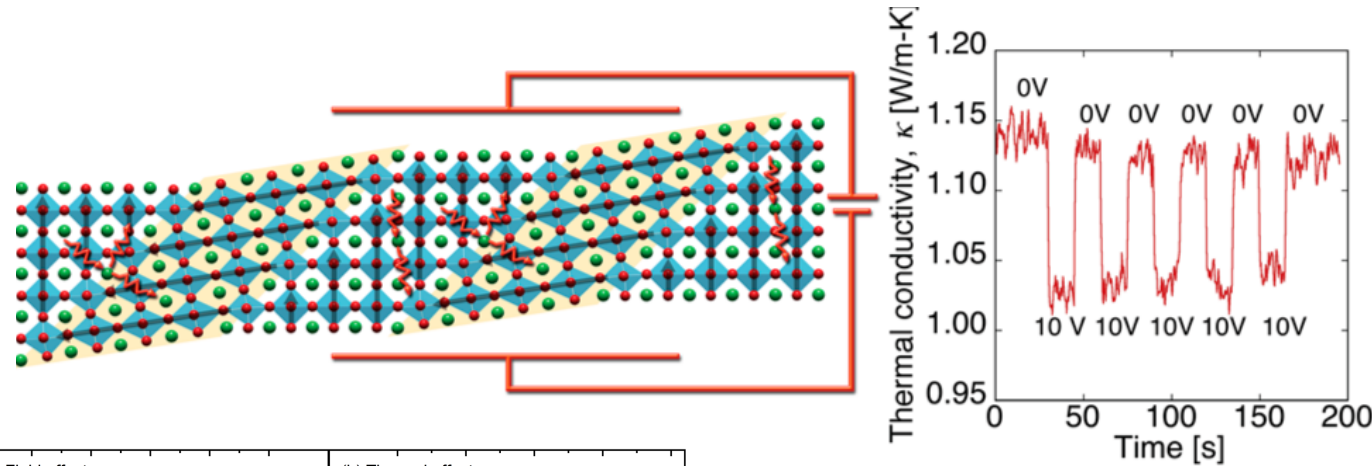
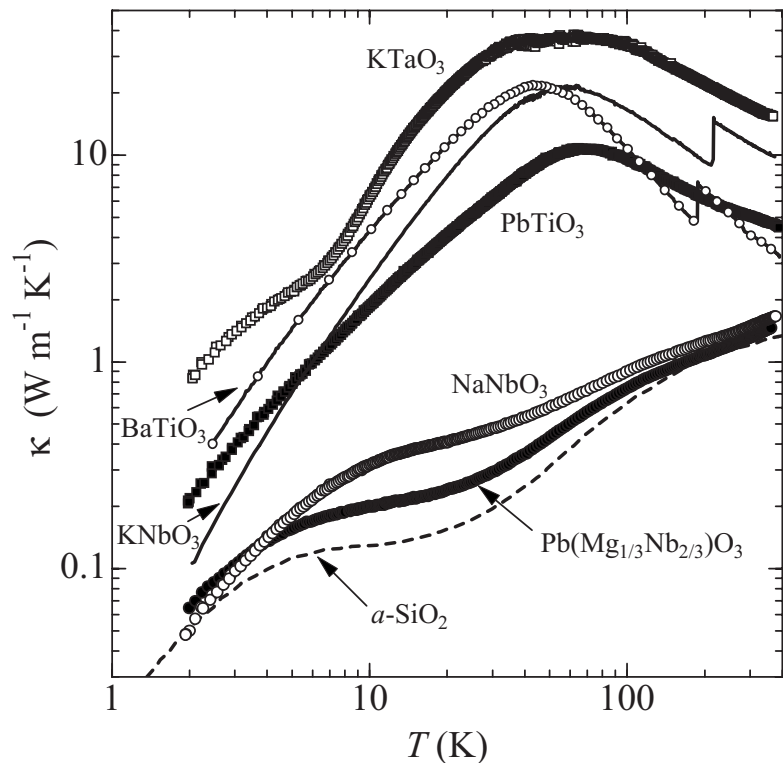
Nat. Mat. **19**, 481 (2020)

“The Slack parameter”

Thermal conductivity in ferroelectrics: Typically low

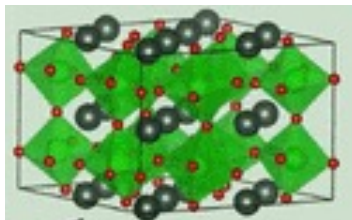
Perovskite Ferroelectrics
APL **93**, 092902

But remarkable thermal functionality!



Nano Lett **15**, 1791
Nat. Comm. **13**, 1573

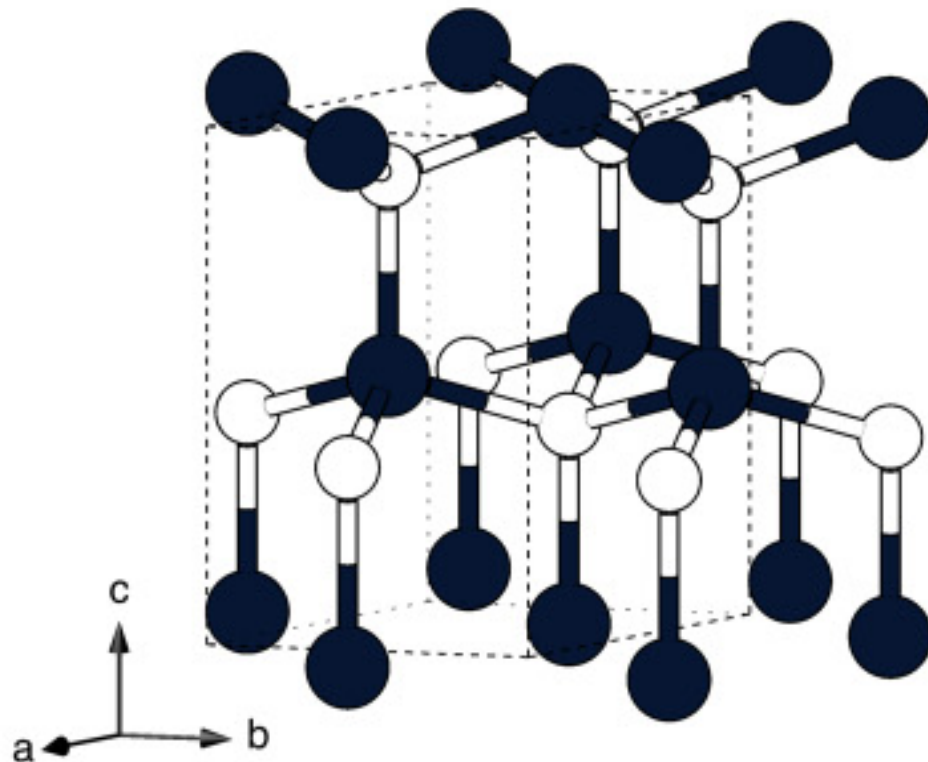
How can we improve thermal conductivity of ferroelectrics?



Remember the Slack Criteria? AlN Ferroelectrics

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

- What makes a high κ material?
- Slack Criteria:
 - Strong bonds
 - Light masses
 - Small unit cell
 - Minimize defects!



PHYSICAL REVIEW MATERIALS 5, 044412 (2021)

Ferroelectricity in boron-substituted aluminum nitride thin films

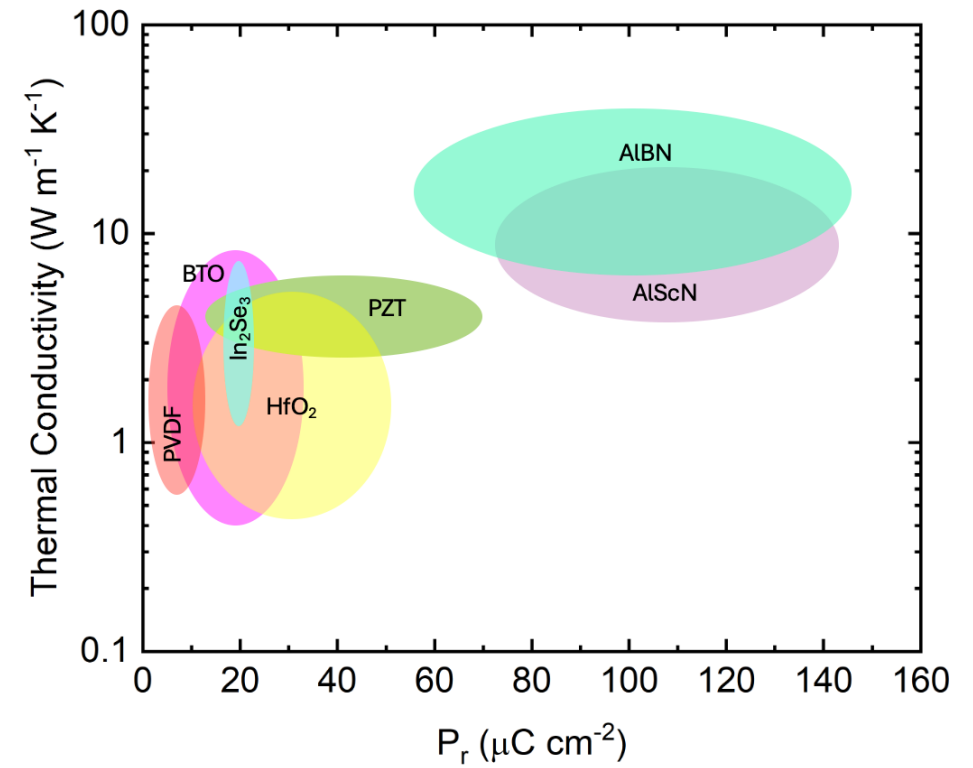
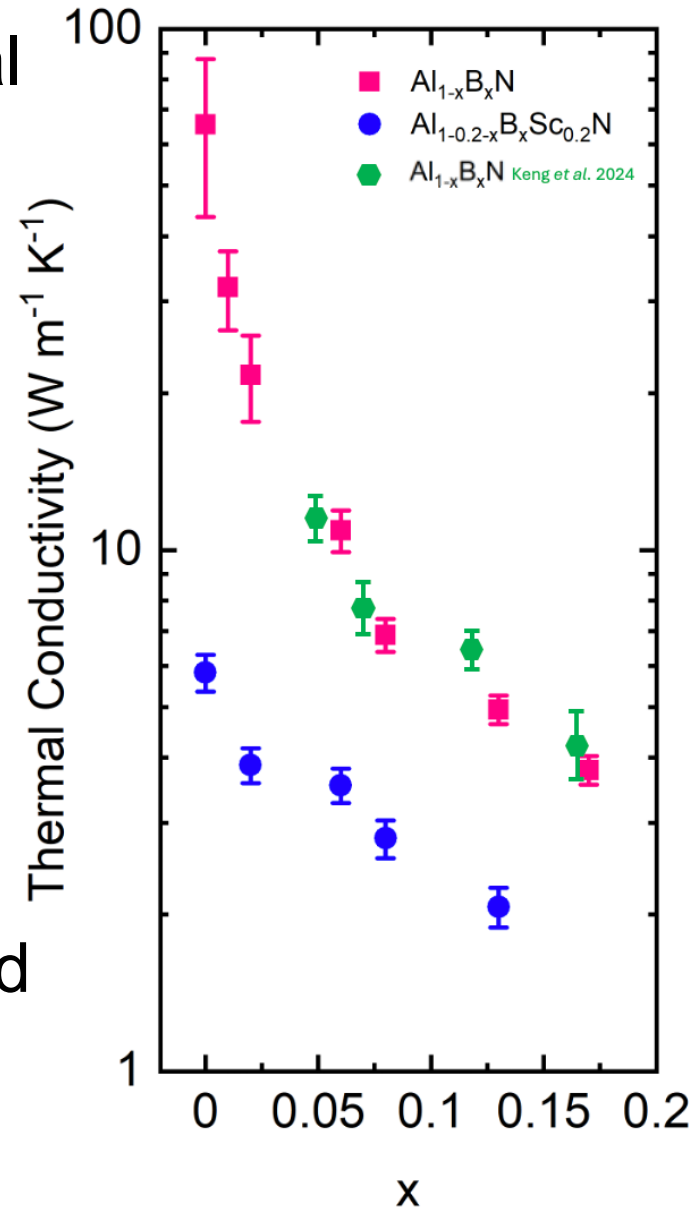
John Hayden^{1,*}, Mohammad Delower Hossain,¹ Yihuang Xiong,¹ Kevin Ferri,¹ Wanlin Zhu,¹ Mario Vincenzo Imperatore,² Noel Giebink,² Susan Trolier-McKinstry,¹ Ismaila Dabo,¹ and Jon-Paul Maria¹

- Aluminum nitride
 - Strong bonds
 - Light masses
 - Small unit cell

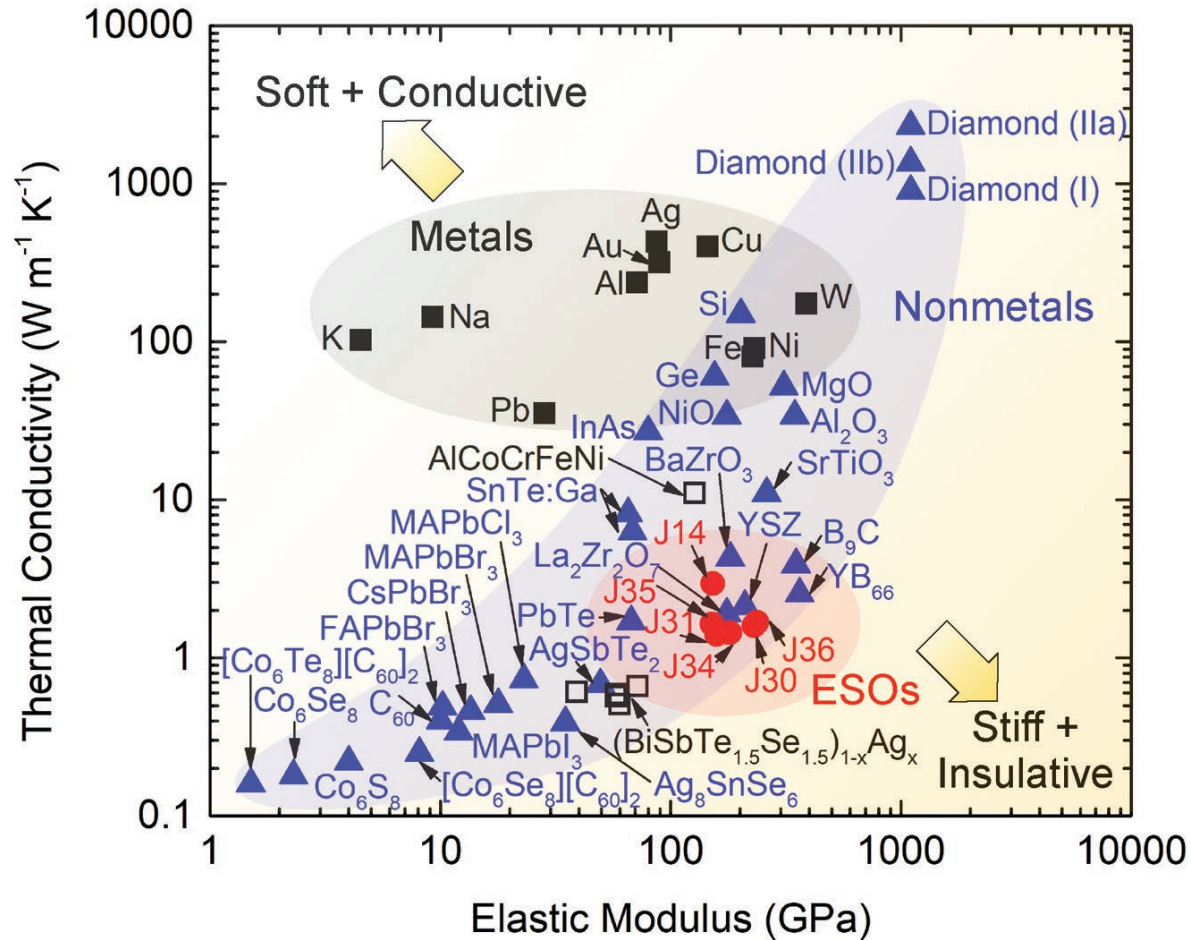


Highest thermal conductivity ferroelectric: $\text{Al}_{1-x}\text{B}_x\text{N}$

- $\text{Al}_{1-x}\text{B}_x\text{N}$ highest thermal conductivity of any ferroelectric to date
- Defects reduce thermal conductivity, but light atomic mass of boron helps with maintaining high κ compared to Sc
- “Type” of defect can be used to tune both thermal conductivity and ferroelectric response for “co-design”



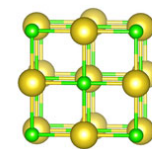
Softer disordered materials - shorter λ - smaller v - smaller κ



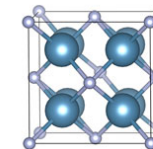
Adv. Mater. **30**, 1805004

How can we make a low κ material w/o sacrificing modulus?

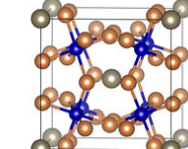
- What makes a low κ material?
- Opposite of Slack Criteria:
 - Weak bonds
 - Heavy masses
 - Large unit cell
 - Increase “defects”!



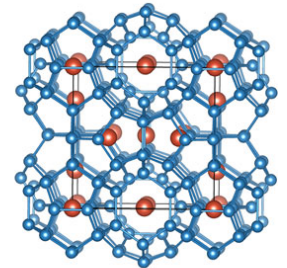
(a) NaCl-type (AB)



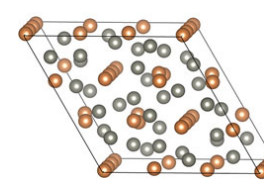
(b) fluorite (AB₂)/anti-fluorite (A₂B)



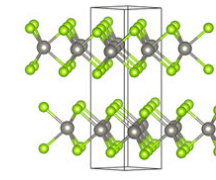
(c) filled skutterudite (AB₄C₁₂)



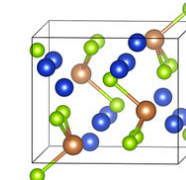
(d) clathrate-I (A₈B₁₆C₃₀)



(e) Zn₄Sb₃



(f) WSe₂



(g) Cu₃SbSe₃

Crys. Res. Tech. **52**, 1700114

The entropy stabilized oxide

nature COMMUNICATIONS

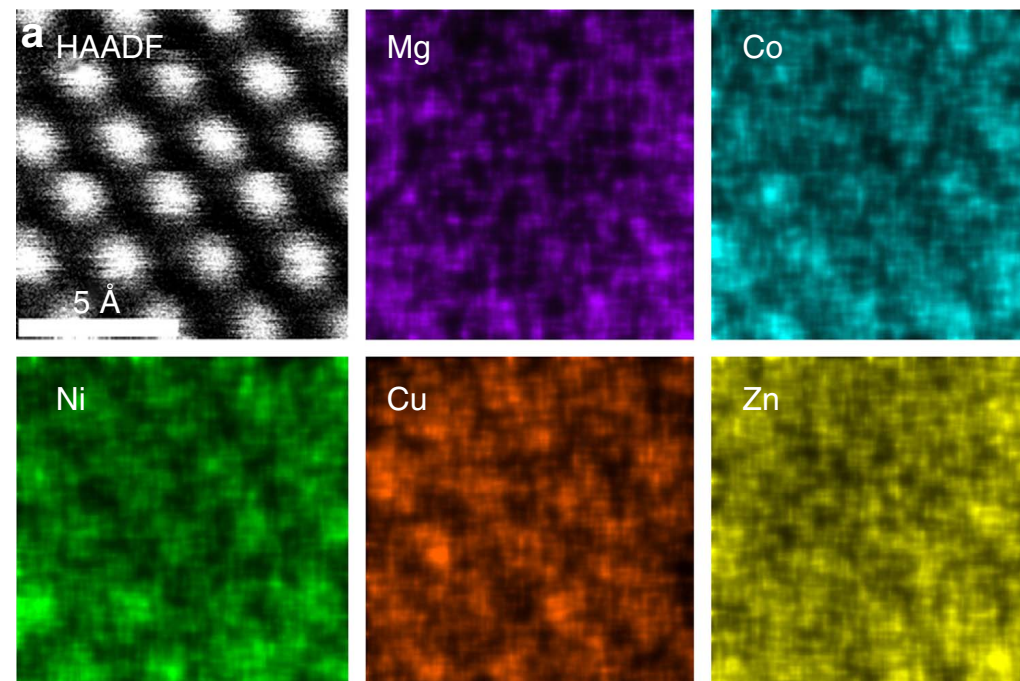
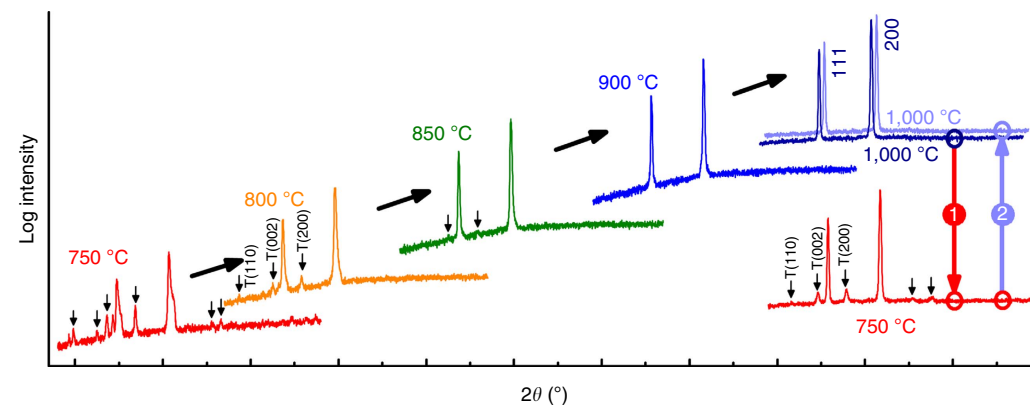
ARTICLE

Received 8 Apr 2015 | Accepted 25 Aug 2015 | Published 29 Sep 2015

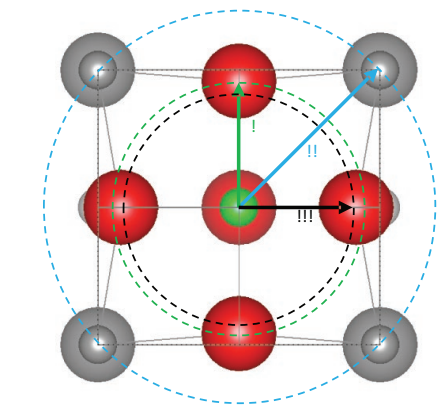
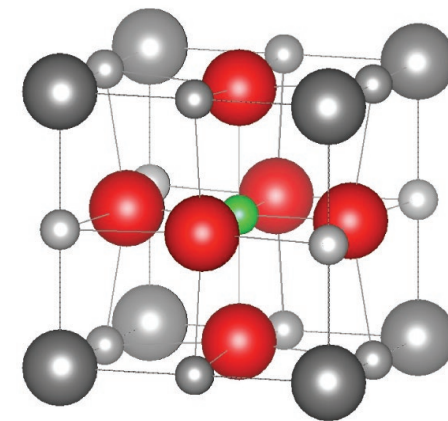
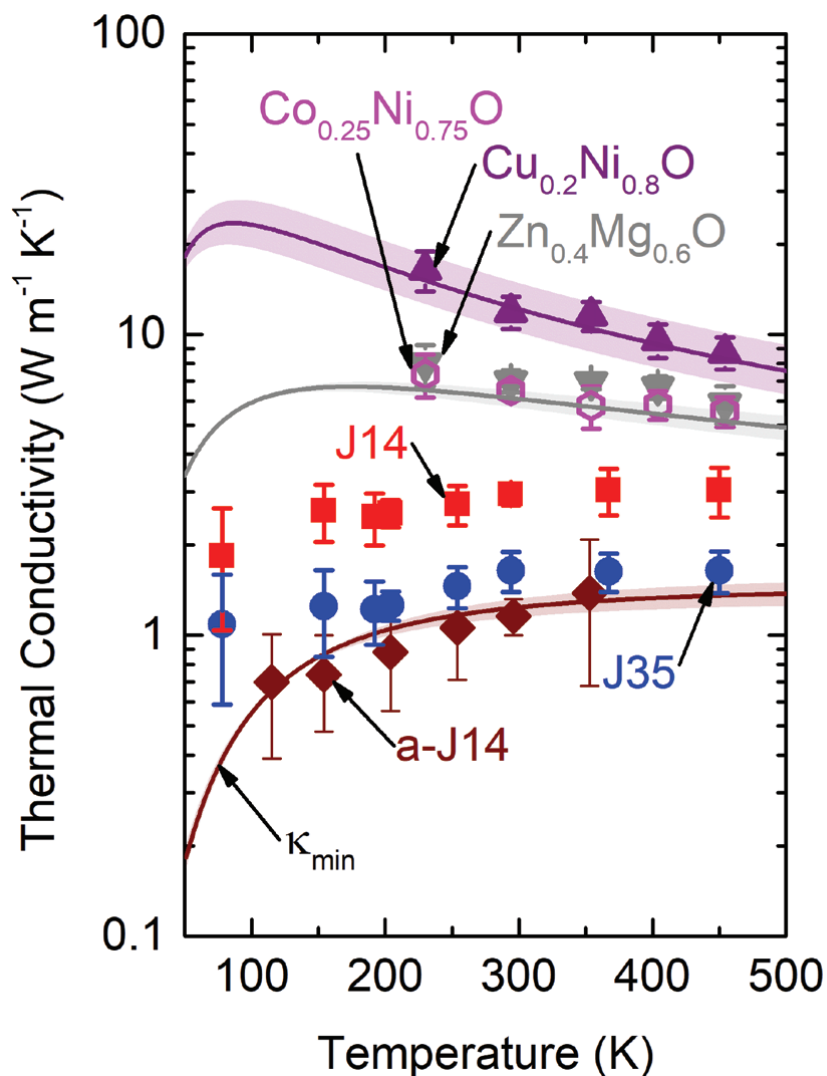
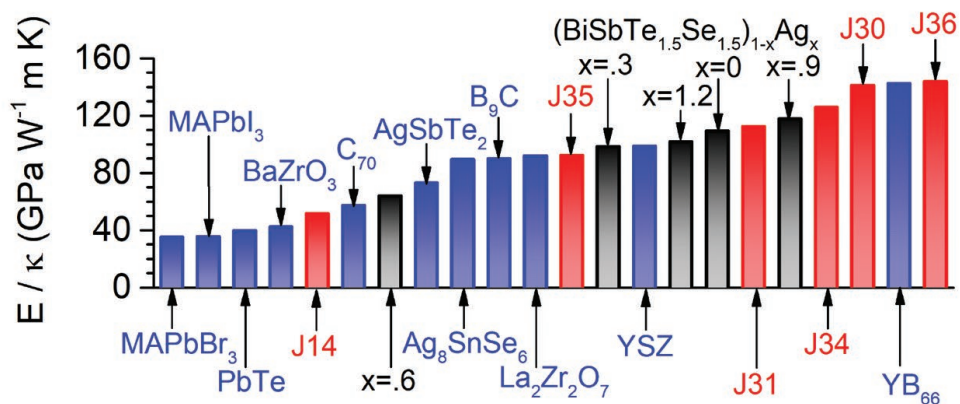
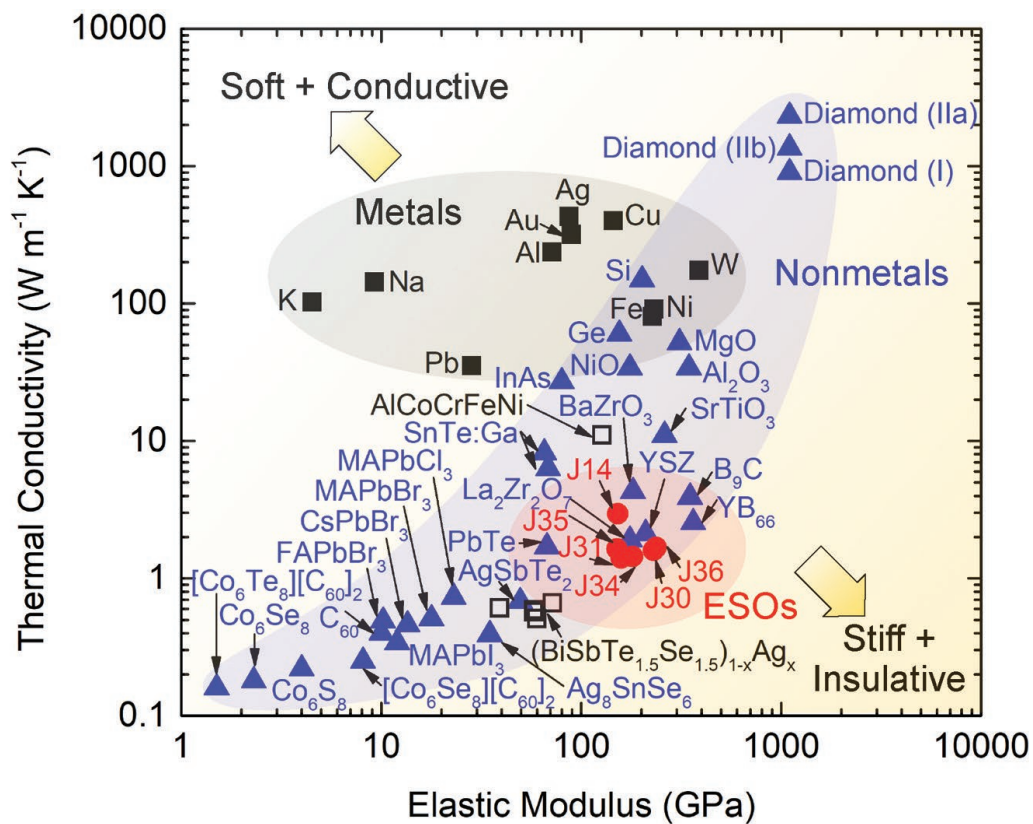
DOI: 10.1038/ncomms9485 OPEN

Entropy-stabilized oxides

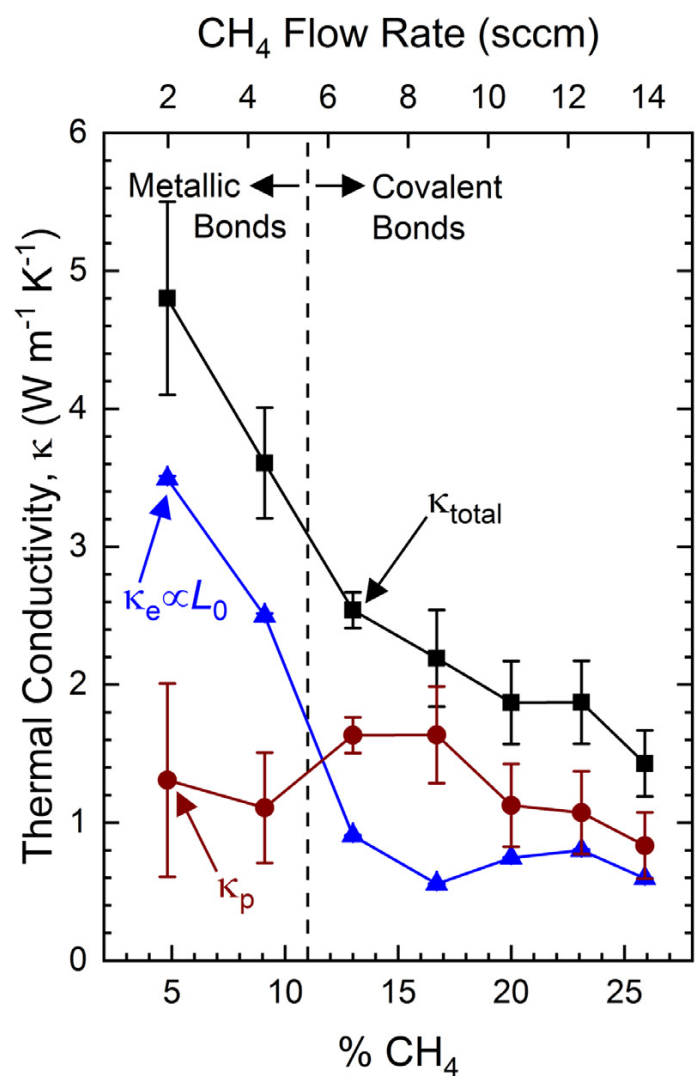
Christina M. Rost¹, Edward Sachet¹, Trent Borman¹, Ali Moballeg¹, Elizabeth C. Dickey¹, Dong Hou¹, Jacob L. Jones¹, Stefano Curtarolo² & Jon-Paul Maria¹



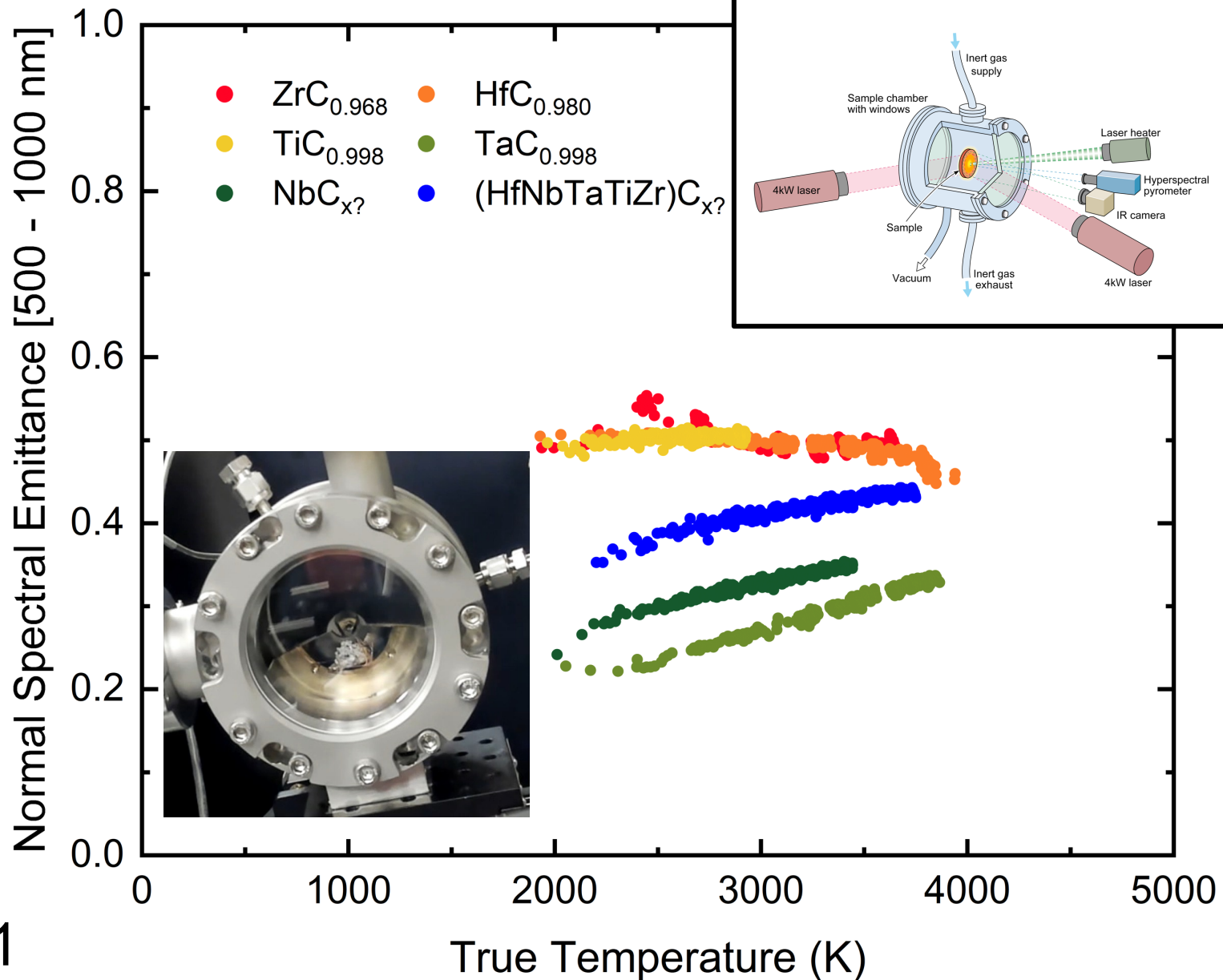
Entropy stabilized oxides: Cation disorder + local distortions



HE carbides: stoichiometry tunable thermal properties

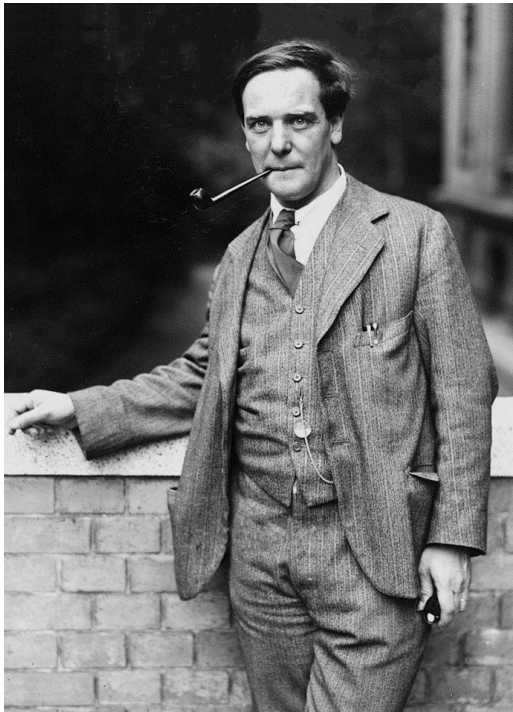


Acta Mat. **196**, 231

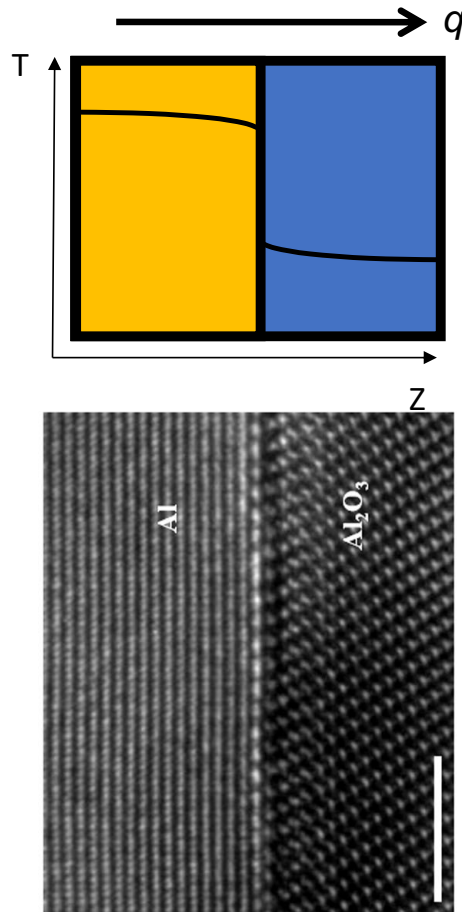


Thermal boundary conductance across interfaces

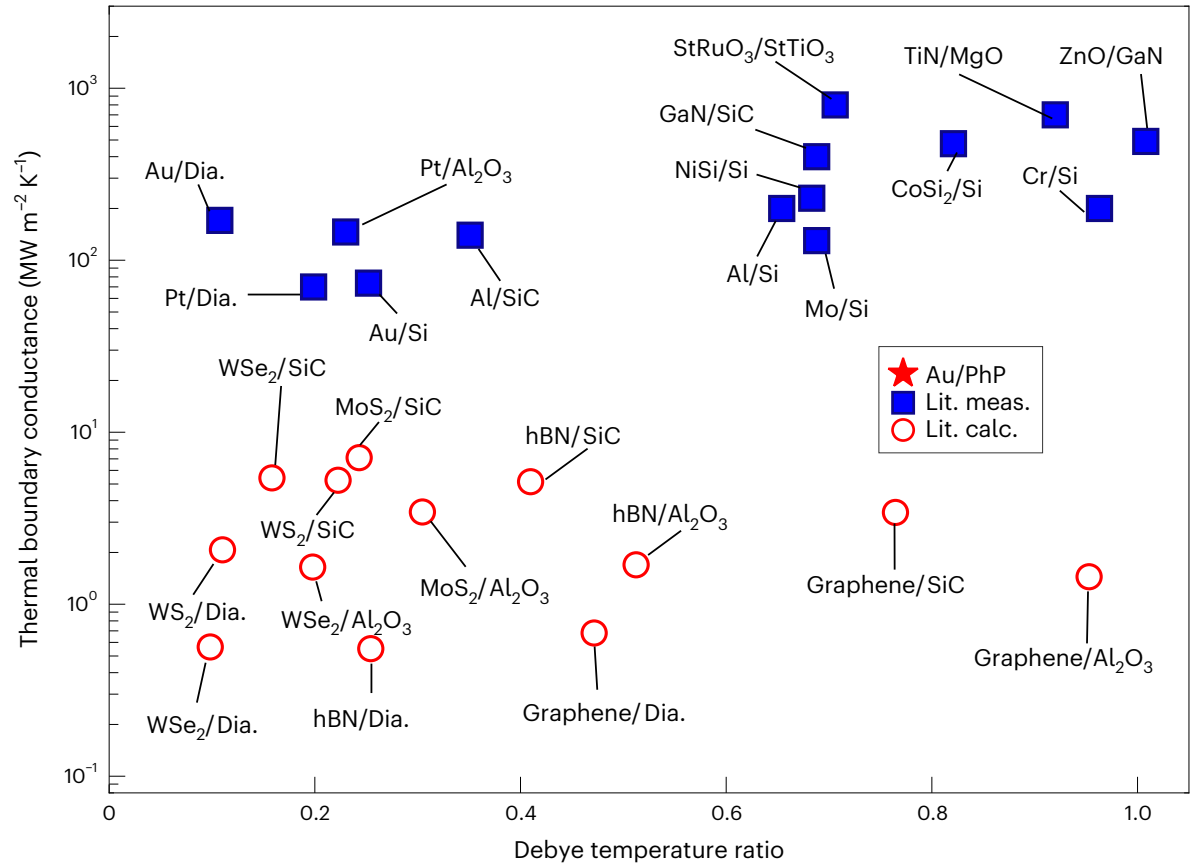
Better overlap of phonon DOS improves TBC at interfaces dominated by phonon transport (does not include 2D interfaces due to weak bonding)



Pyotr Kapitsa

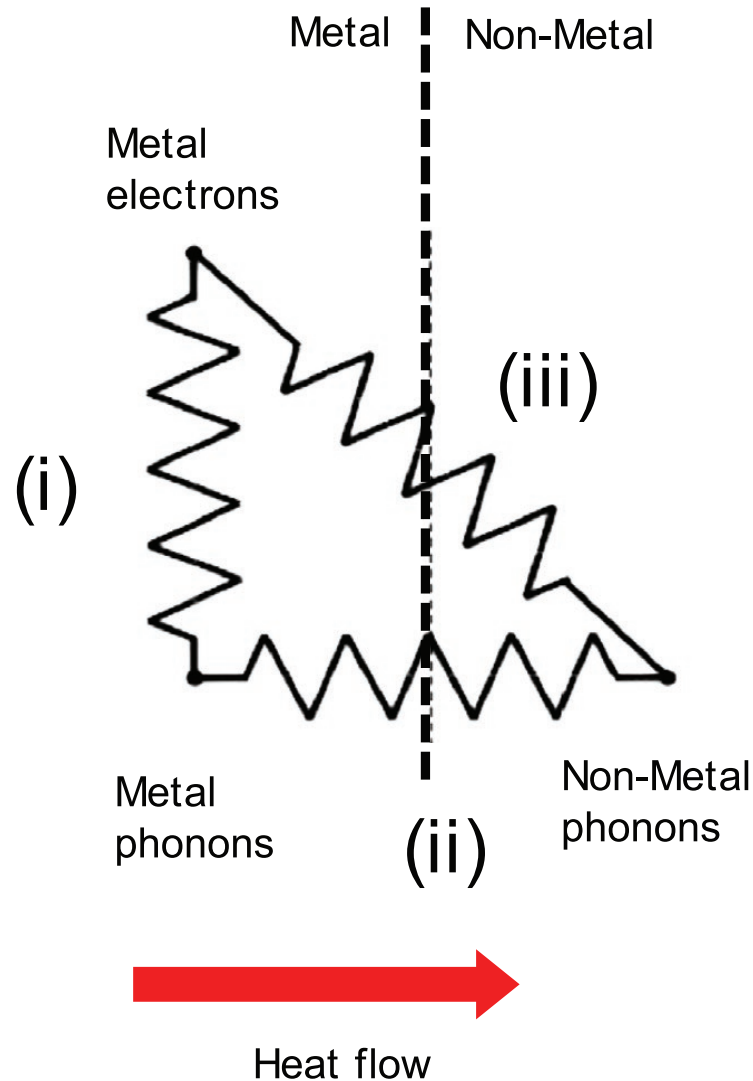
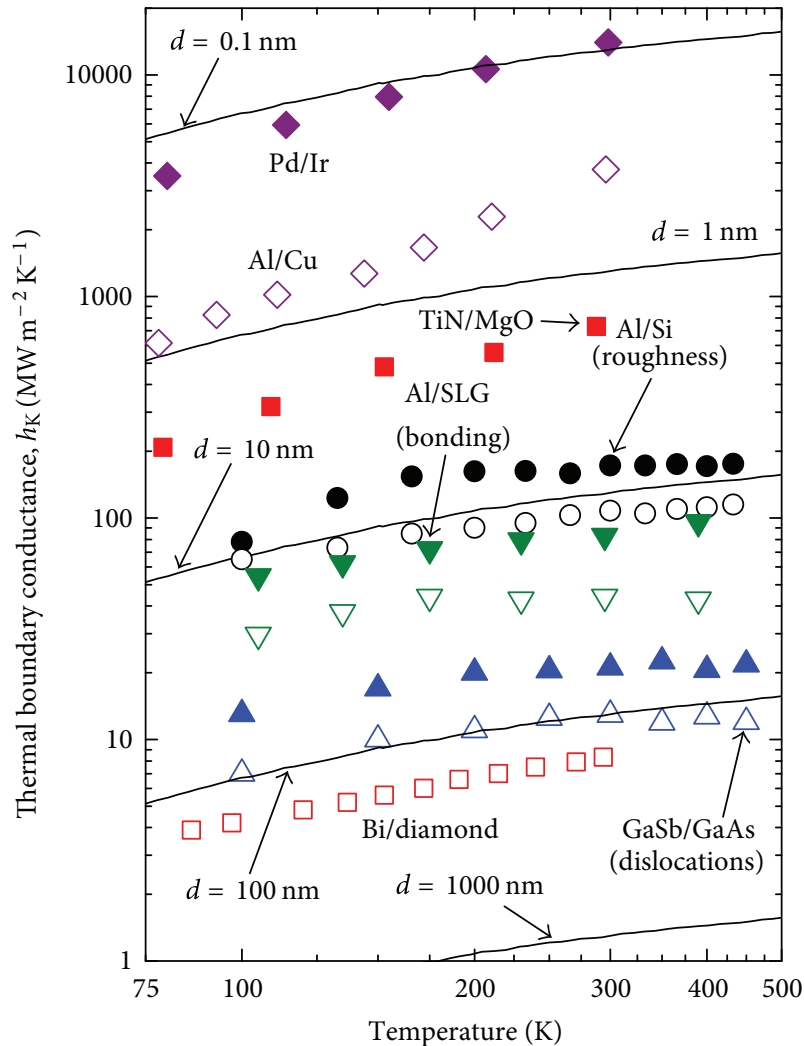


Comm. Physics **3**, 115



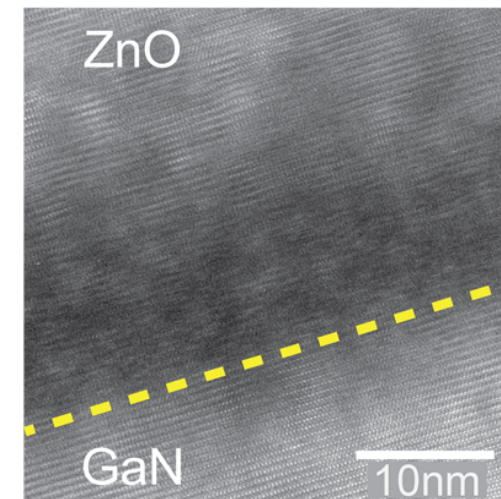
Nature Materials **24**, 698

TBC at metal/non-metal interfaces: phonons vs. electrons



Metal/non-metal

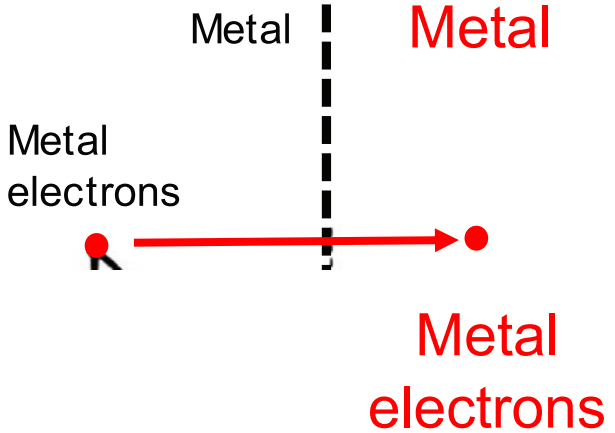
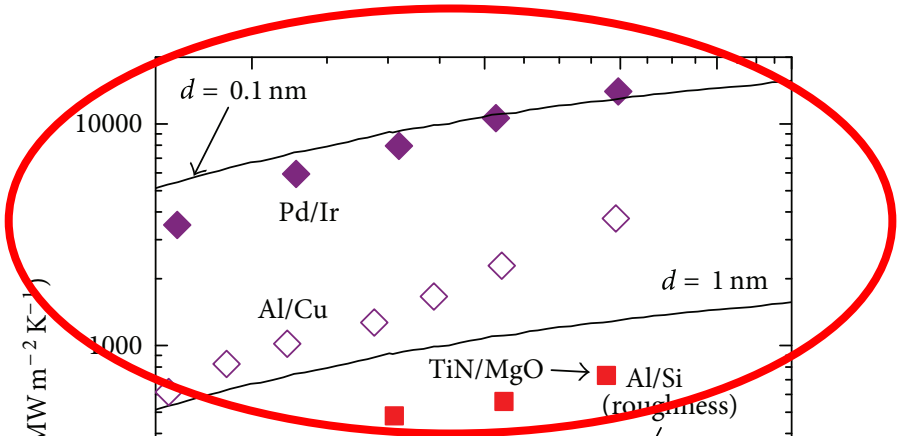
- Conduction across interface driven by phonons
- Temp. dependence \sim phonon C
- Heat flux \sim phonon velocity $\times C$



Nano Lett
18, 7469



TBC at metal/non-metal interfaces: phonons vs. electrons



$$C_e v_F > C_p v_p$$

Metal/metal

- Conduction across interface driven by free electrons
- Temp. dependence ~ electron C
- Heat flux ~ Fermi velocity x electron C

How can we access this high electronic heat flux to improve TBC at interfaces with non-metals??

Highly conducting, high carrier density cadmium oxides

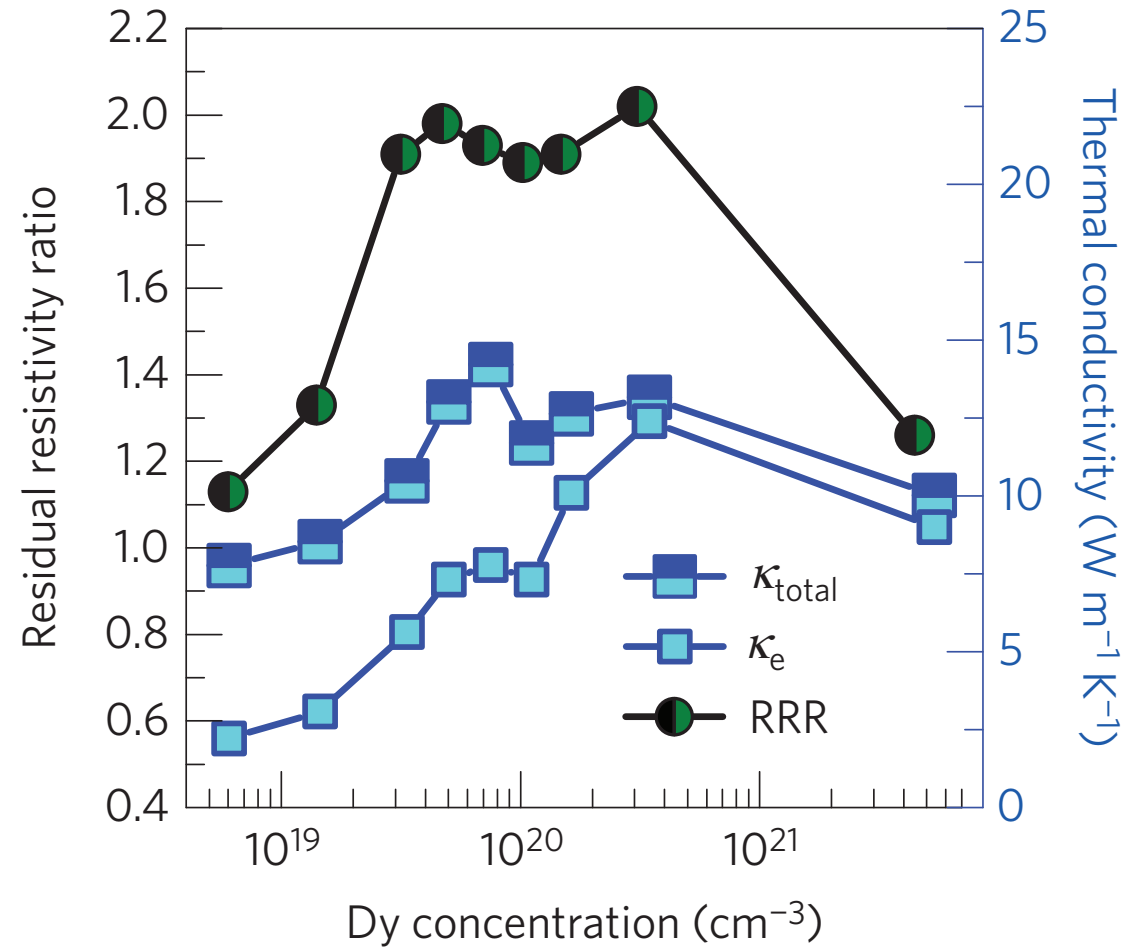
ARTICLES

PUBLISHED ONLINE: 16 FEBRUARY 2015 | DOI: 10.1038/NMAT4203

nature
materials

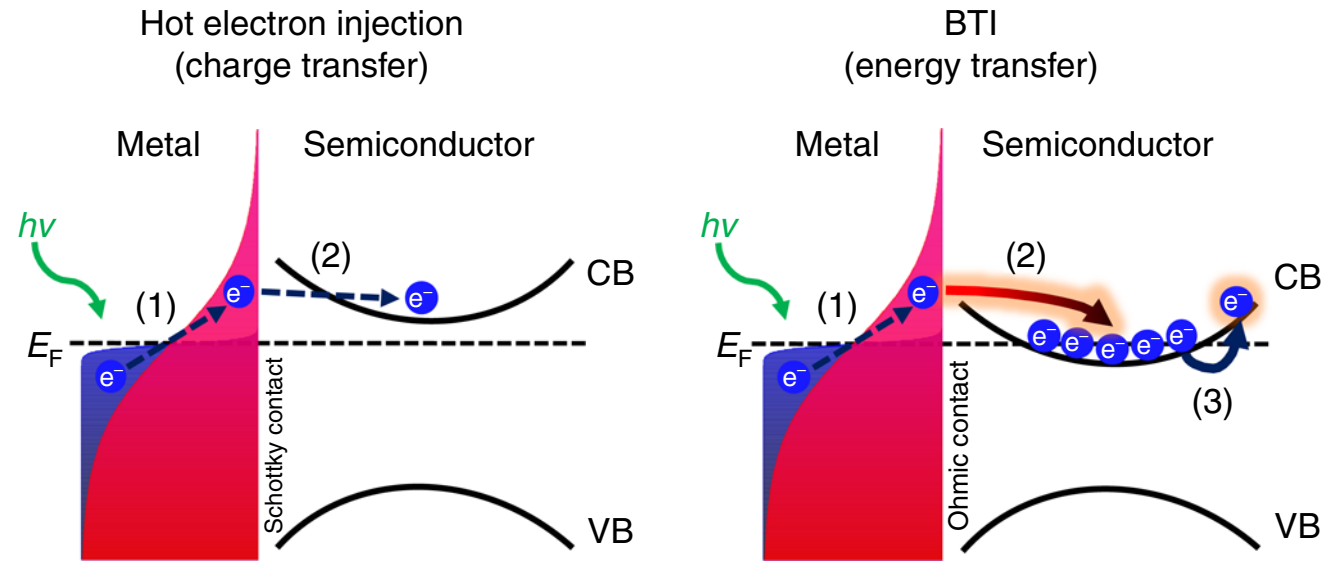
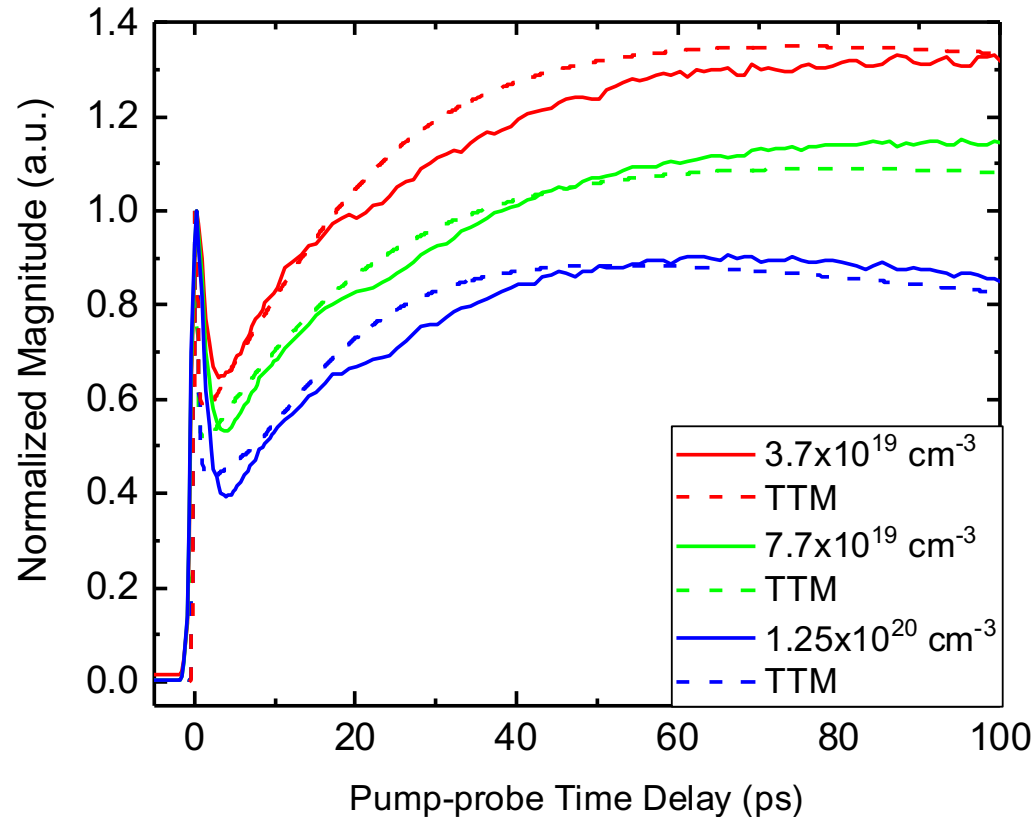
Dysprosium-doped cadmium oxide as a gateway material for mid-infrared plasmonics

Edward Sachet¹, Christopher T. Shelton¹, Joshua S. Harris¹, Benjamin E. Gaddy¹, Douglas L. Irving¹, Stefano Curtarolo^{2*}, Brian F. Donovan³, Patrick E. Hopkins⁴, Peter A. Sharma⁵, Ana Lima Sharma⁵, Jon Ihlefeld⁵, Stefan Franzen⁶ and Jon-Paul Maria^{1*}

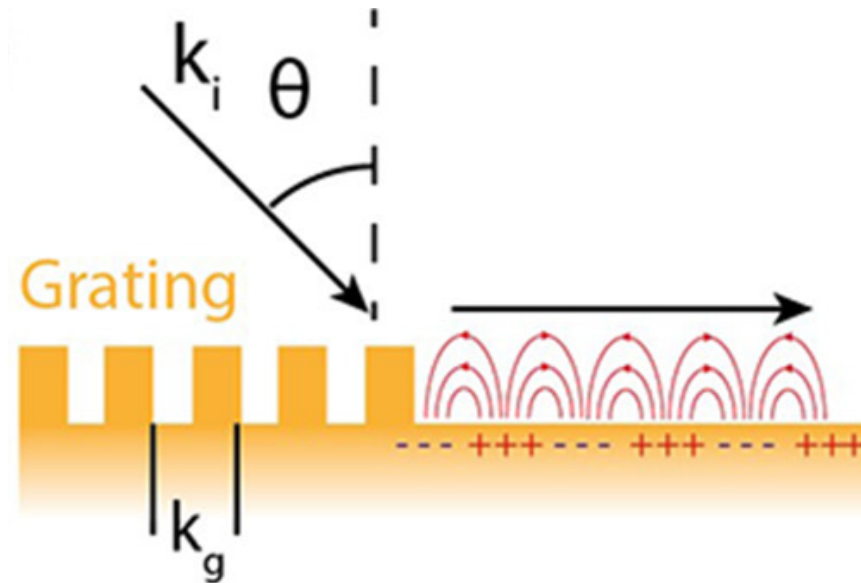
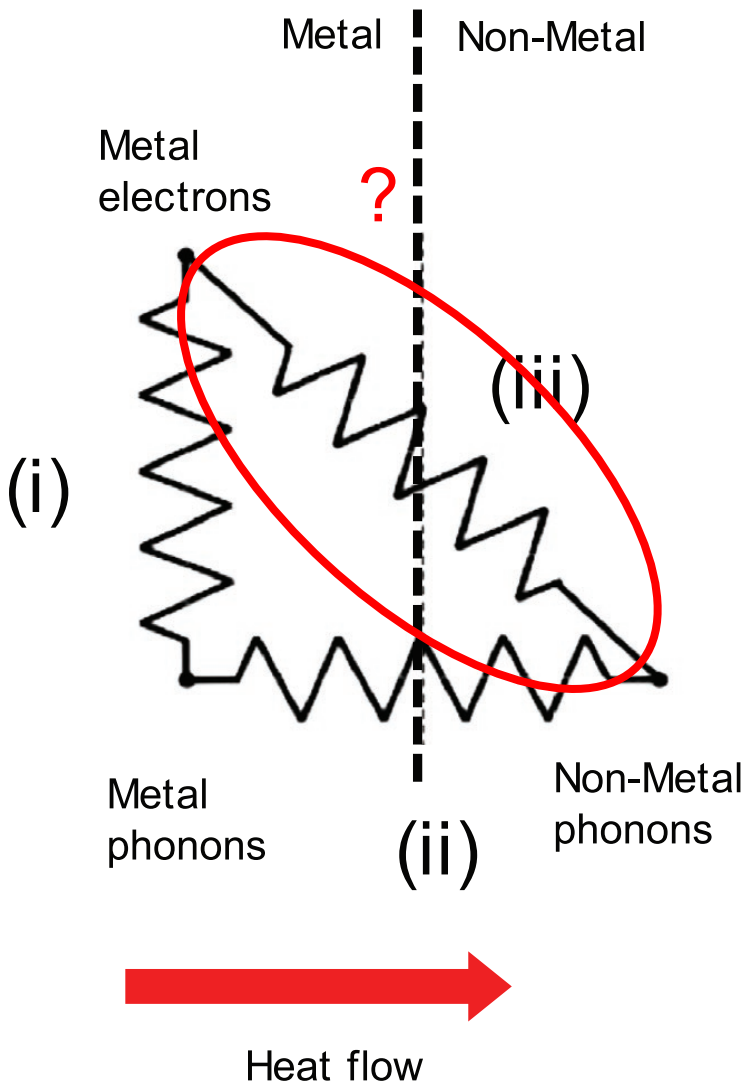


Electron mediated TBC at metal/non-metal interfaces

- Energy easily transmitted across Au/doped CdO interface when out of equilibrium with phonons, like metal/metal
- This “ballistic thermal injection” is different than charge injection

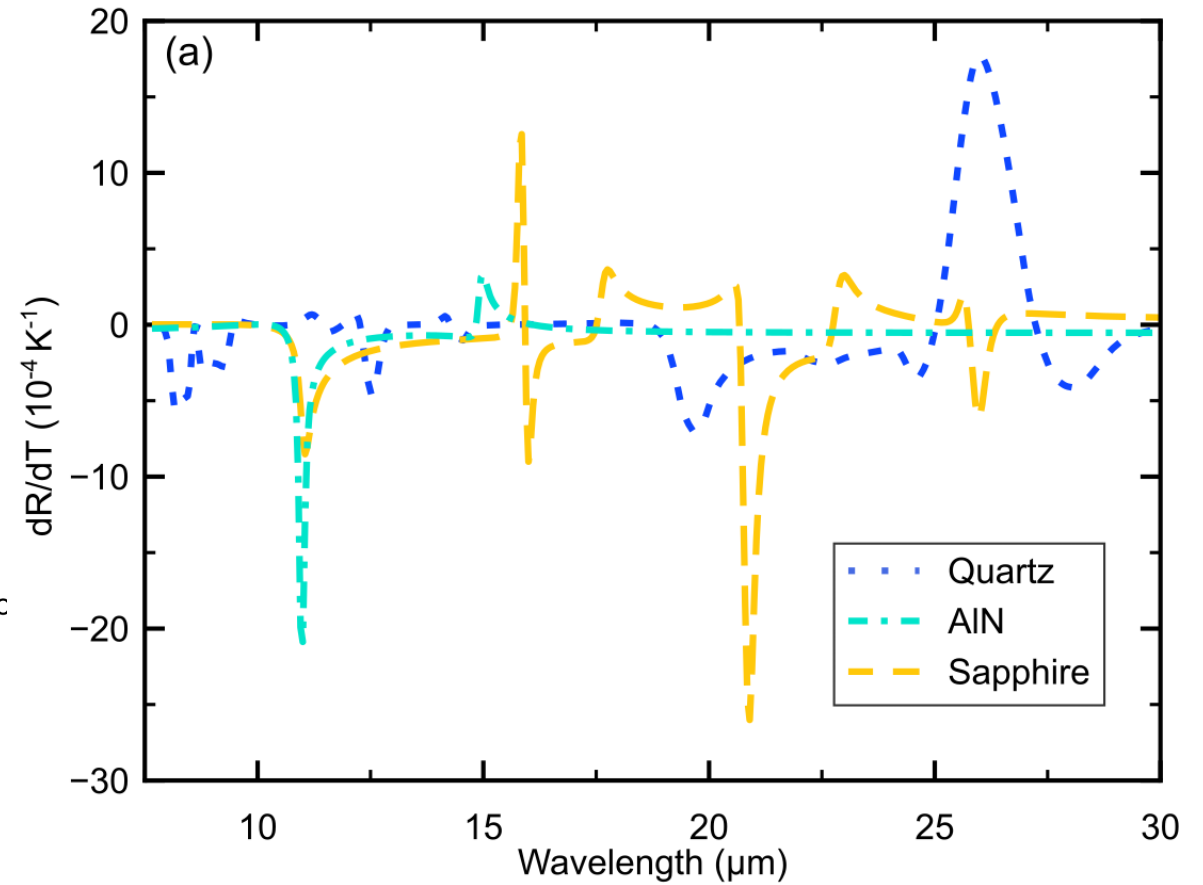
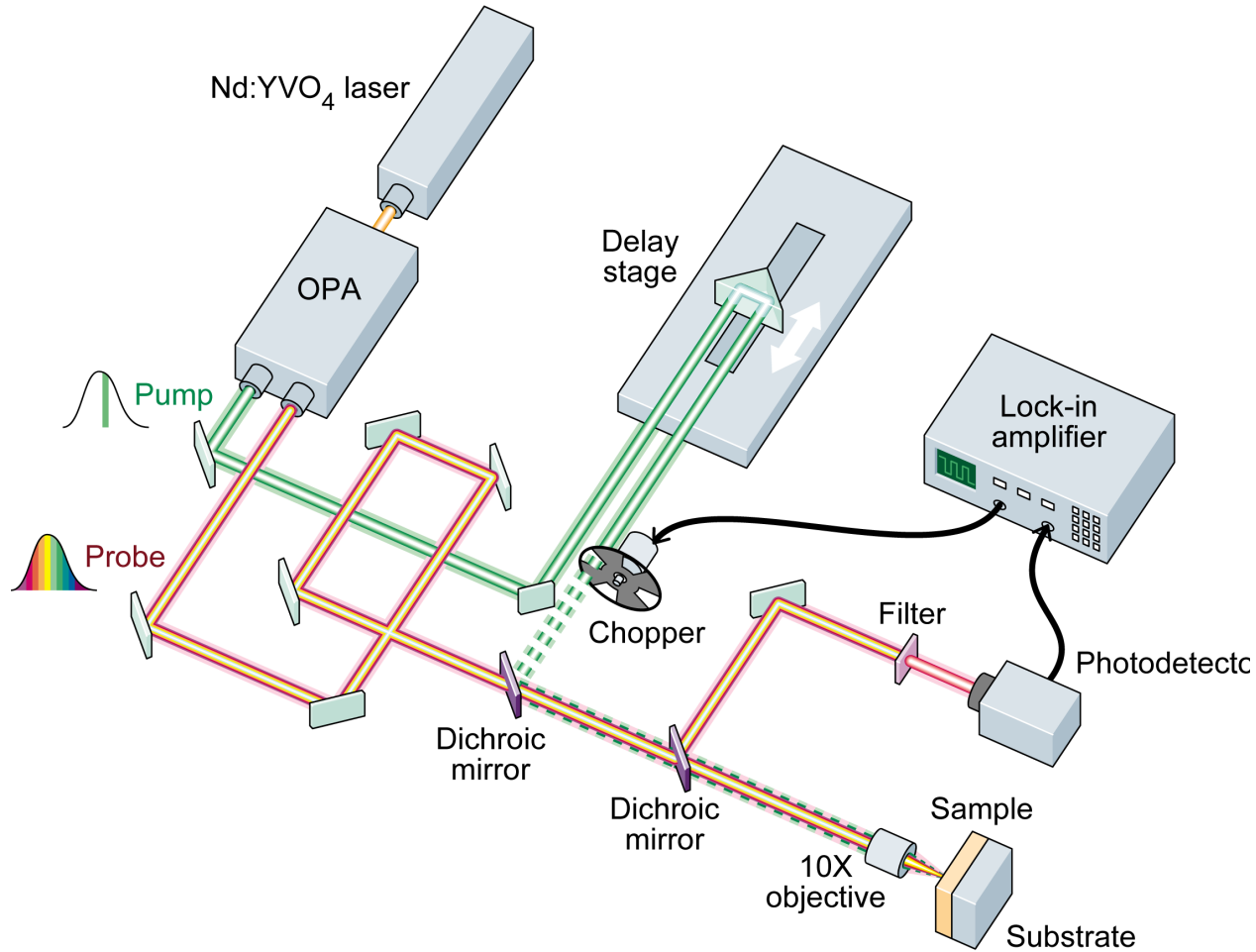


Non-metal interfaces: can we overcome sluggish phonons?



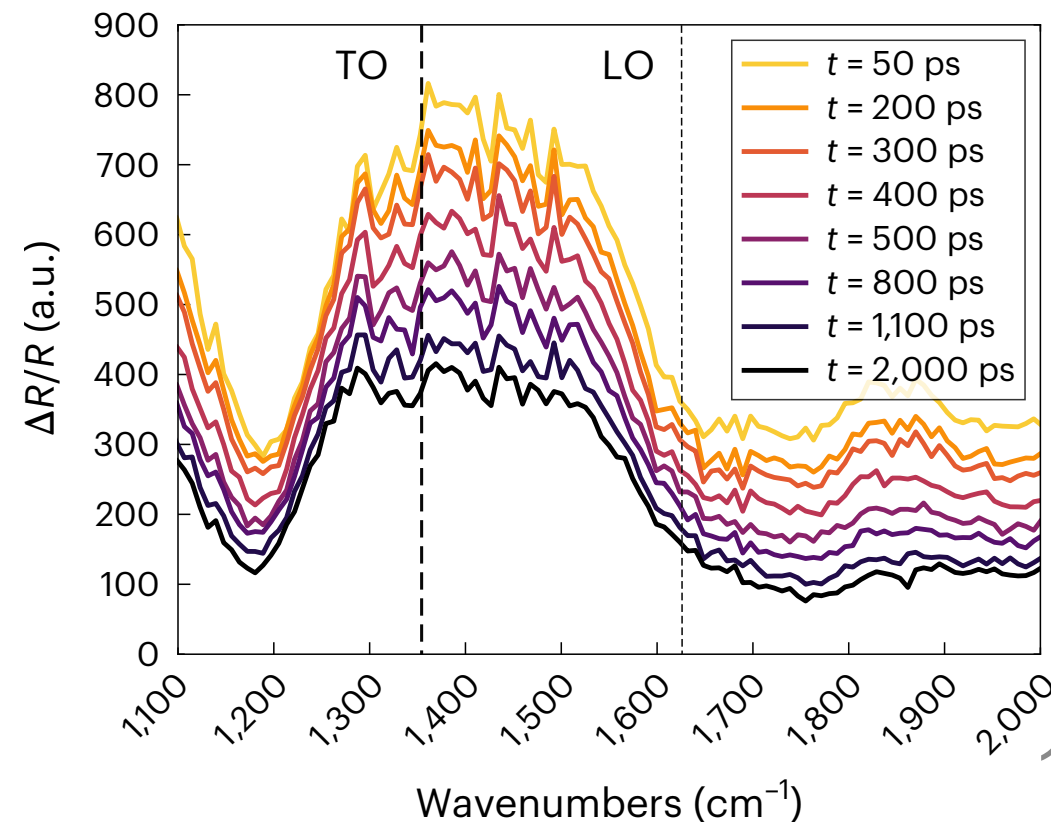
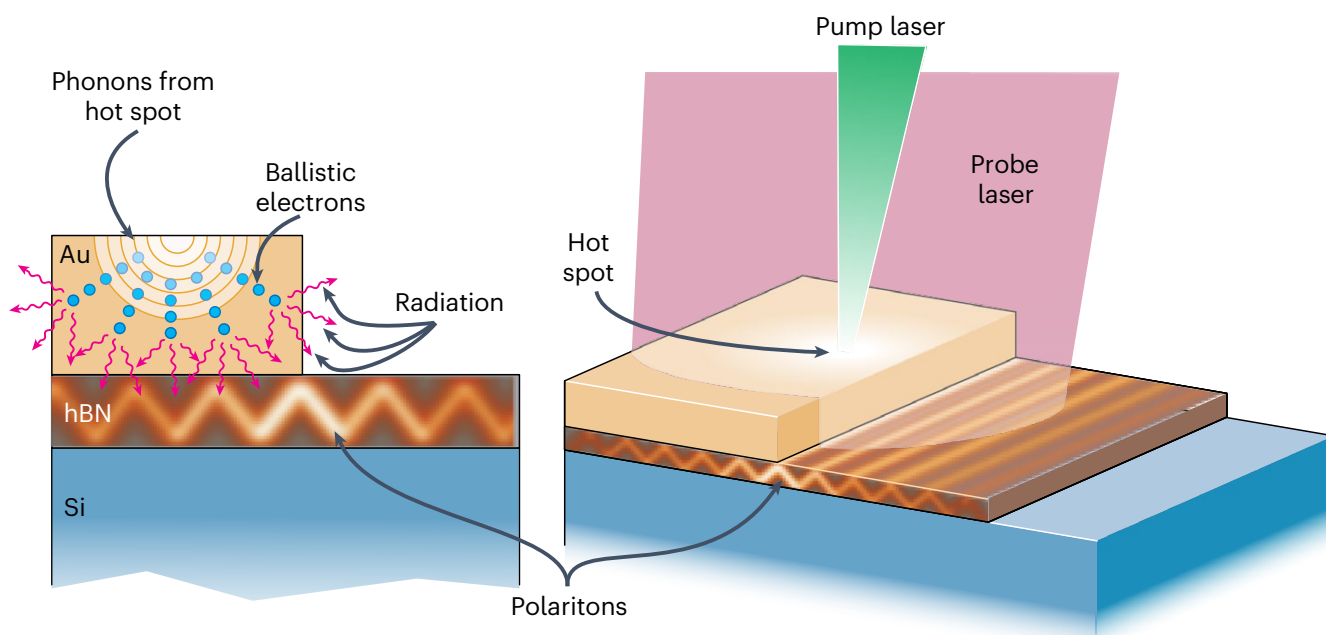
- Light couples with electric dipole creating quasiparticle
- At IR wavelengths: Phonon polariton!
- PhP quasiparticles can propagate at $\sim 1\%$ of the speed of light!
- Prior evidence of thermal conductivity enhancement during excitation of PhPs

Phonon thermorelectance: phonons to measure temperature



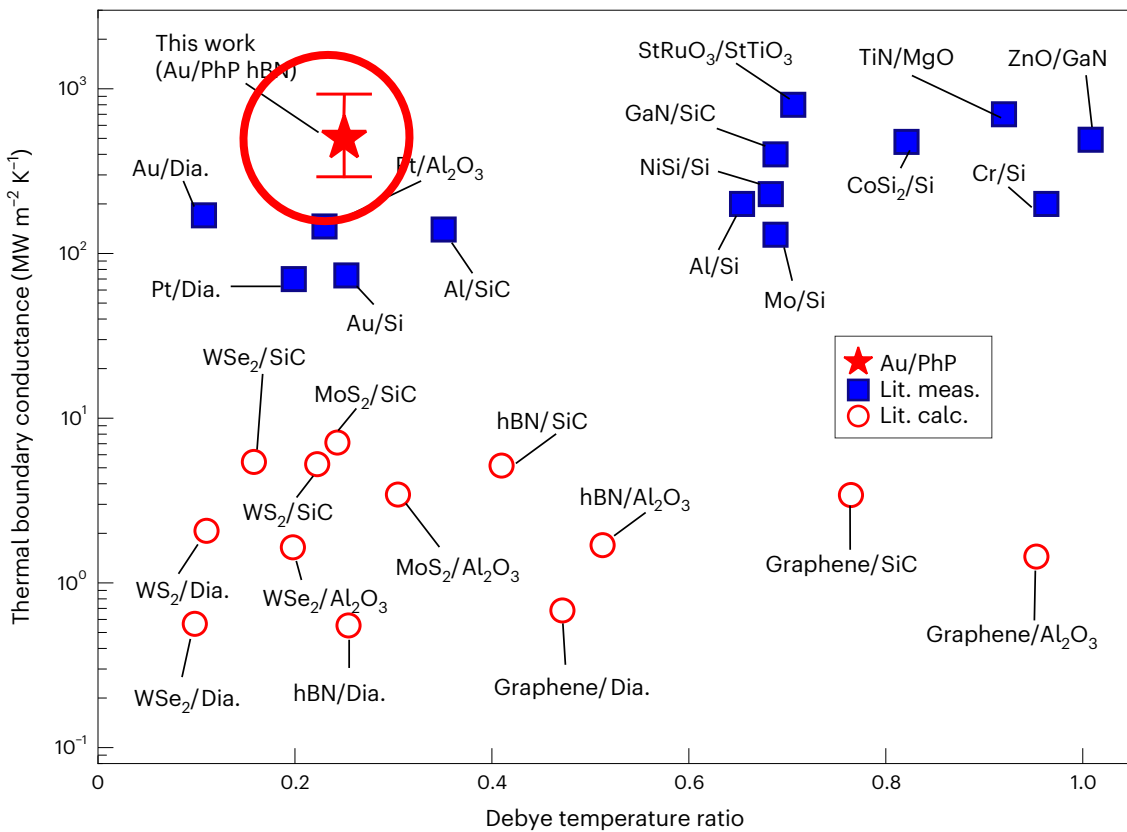
IR pump-probe using *phonon* thermorefectane

- Heating of Au pad will indirectly heat hBN
- Two heat transfer pathways across Au/hBN interface: phonon-phonon, NFRT from Au to hBN via coupling with hBN phonon polaritons

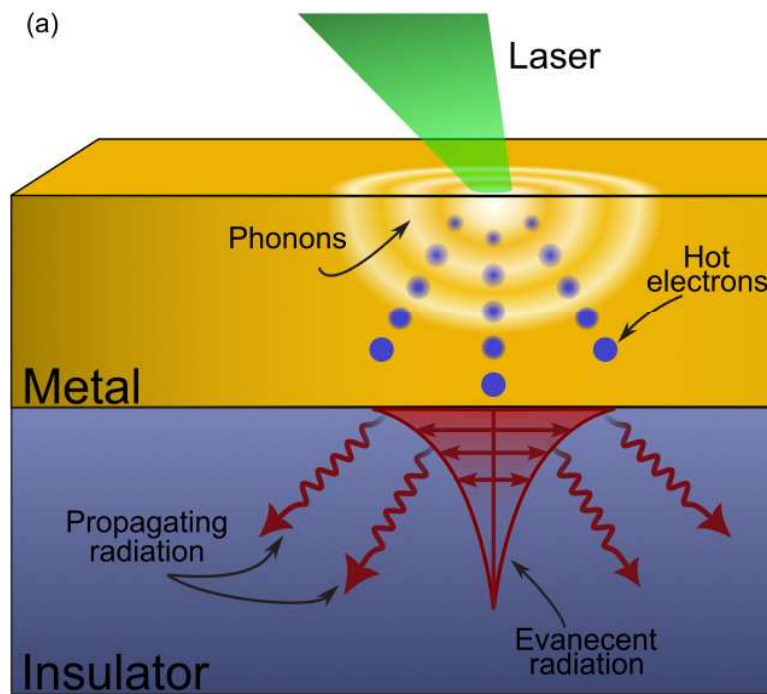


Mechanism: Non-equilibrium interfacial heat transfer

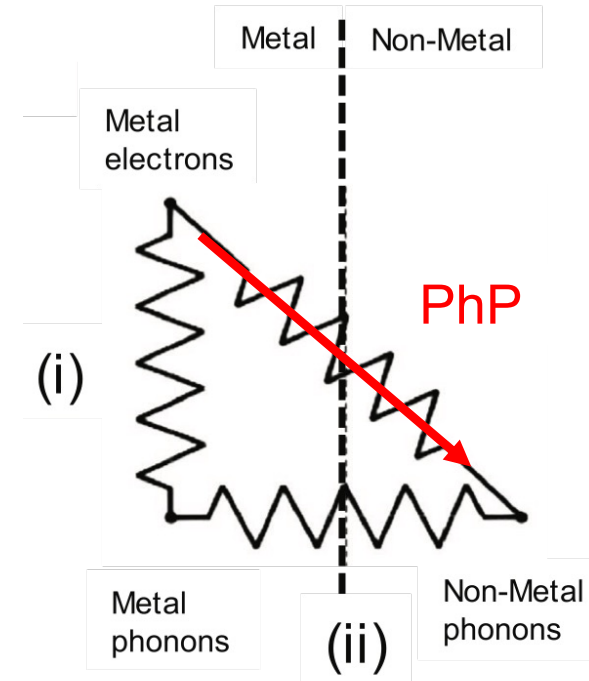
- Not enough NFRT at 300 K, electrons need to be “hot”
- Hotter electrons at boundaries = more NFRT



Nature Materials **24**, 698

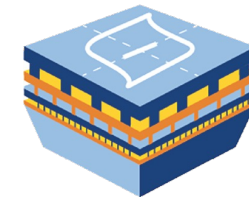


PRL **134**, 186302



Professor JP Maria's research in novel functional materials has built research bridges and spawned new directions in material heat transfer

- New regimes for high thermal conductivity functional materials
- New materials for ultralow thermal conductivity materials
- New mechanisms for transferring heat across interfaces and developing “non-equilibrium thermal polaritonics”



3D
FeM²

