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Ballistic thermal injection: controlling electron-phonon scattering at interfaces for long-lived plasmonic modulation

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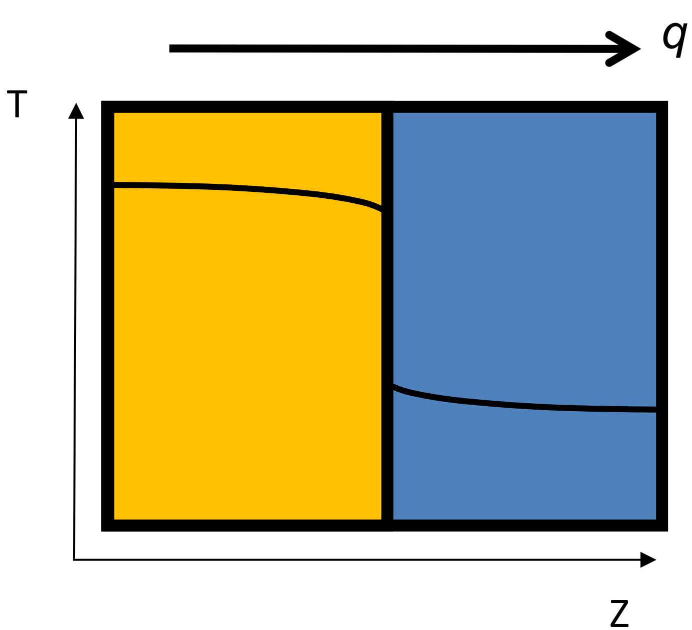
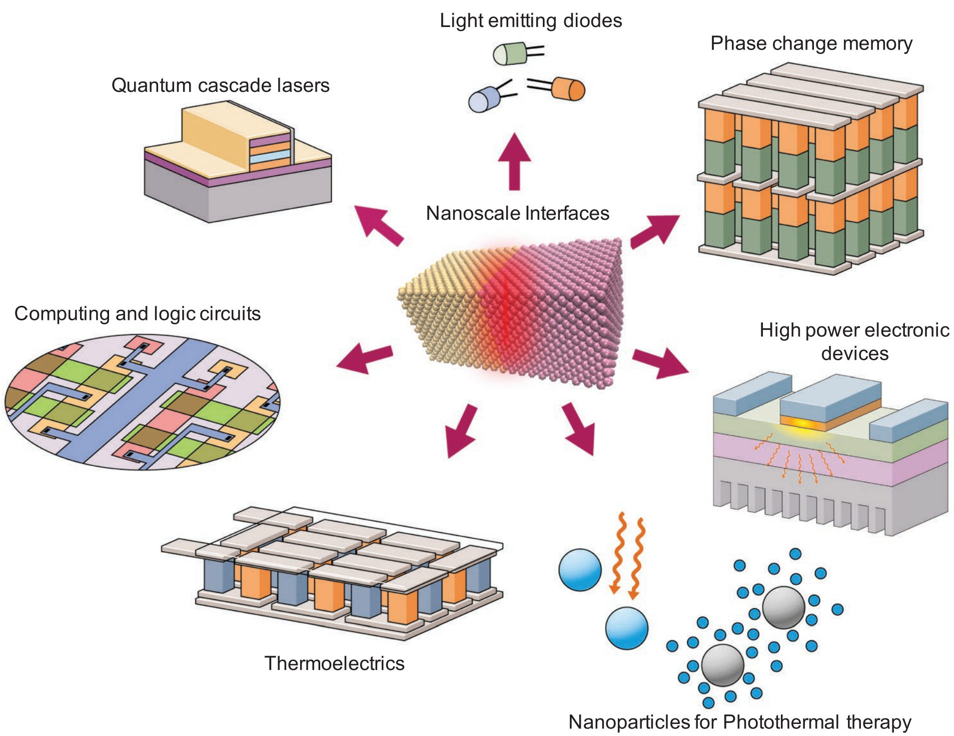
Interfacial heat transport : Thermal boundary conductance

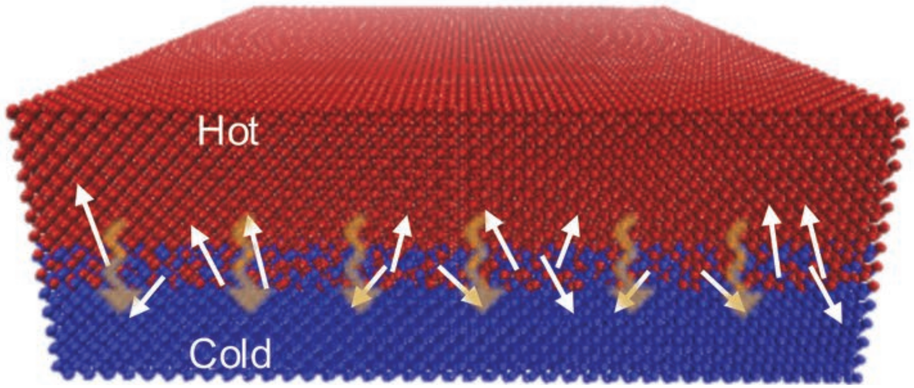
$$q = h_{\mathrm{K}} \Delta T = \frac{1}{R_{\mathrm{K}}} \Delta T$$





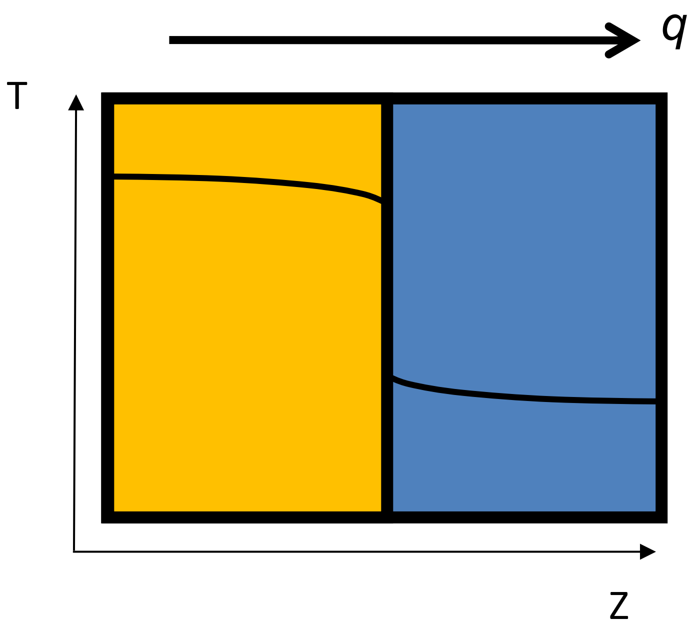
<u>Recent Review</u>

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



Interfacial heat transport : Thermal boundary conductance

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



Kapitza Length

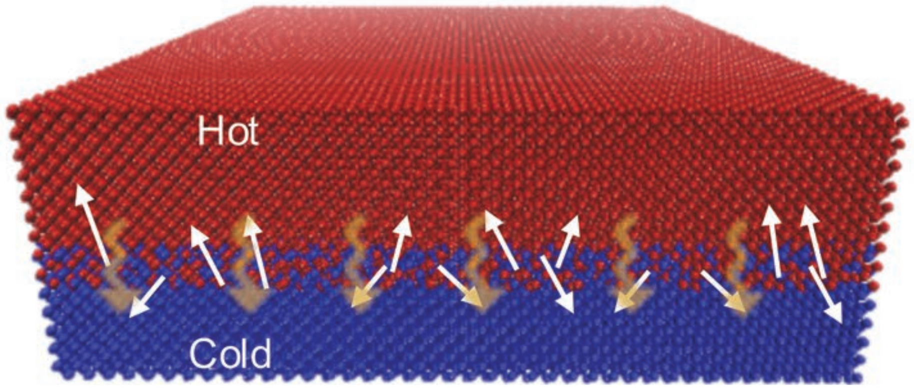
$$L_K = \frac{\kappa}{h_K}$$

κ ~ 1 – 100 W m^{-1} K^{-1}

L_K ~ 10 nm – 1 μm

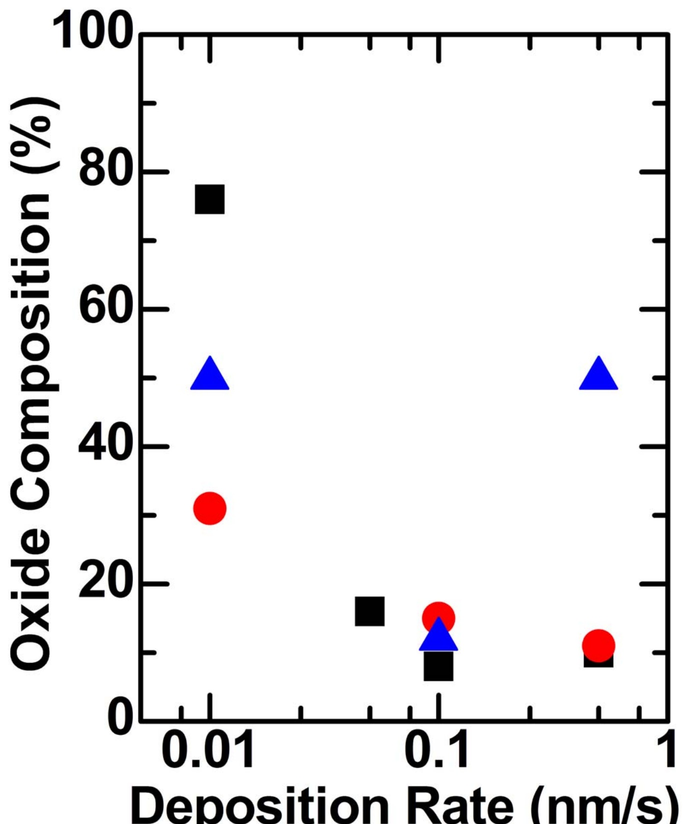
Recent Review

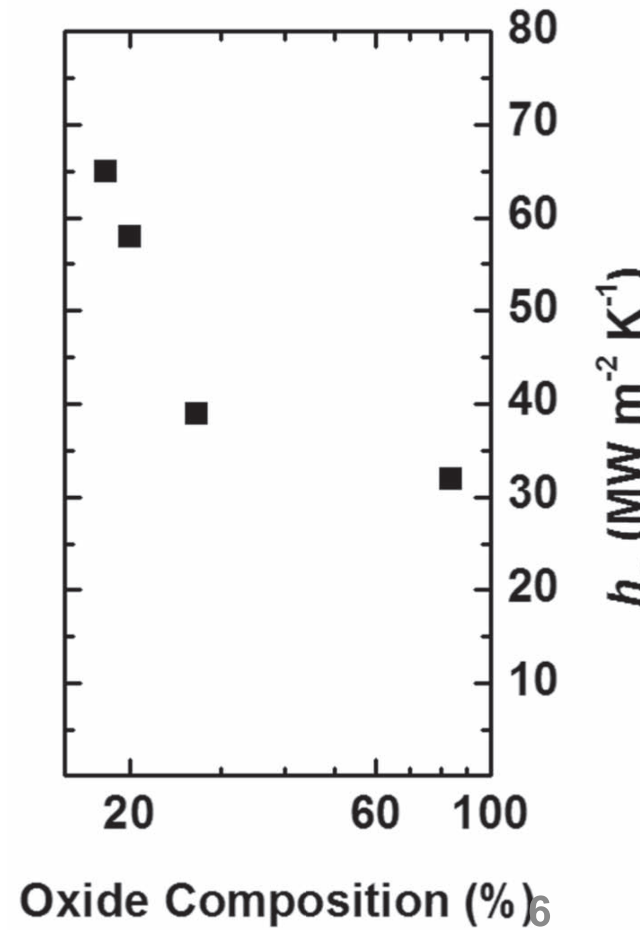
Advanced Functional Materials **30**, 1903857 (2020)



Chemistry & TBC at metal/non-metal interfaces (phonons)

Au/TiO_x/substrate with varying "x" (oxygen stoichiometry) to control thermal boundary conductance

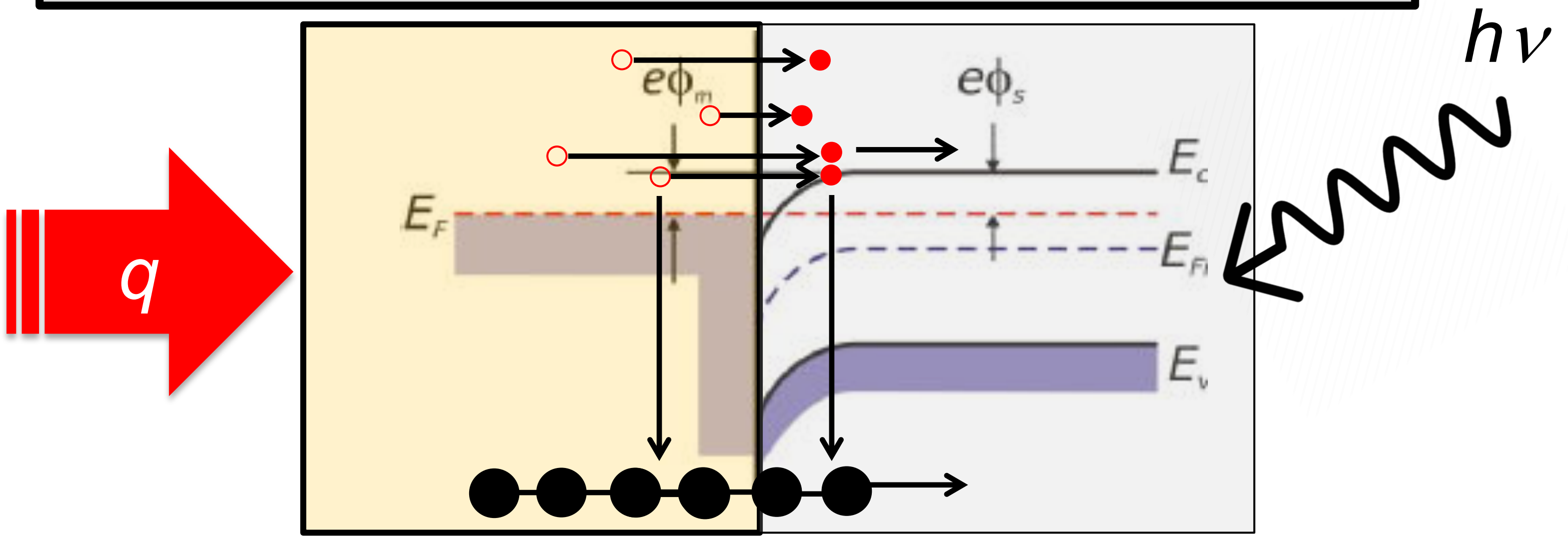




Nanotechnology **29**, 145201

Interfacial heat transport : Thermal boundary conductance

But what are the electron-phonon heat transport mechanisms at interfaces?



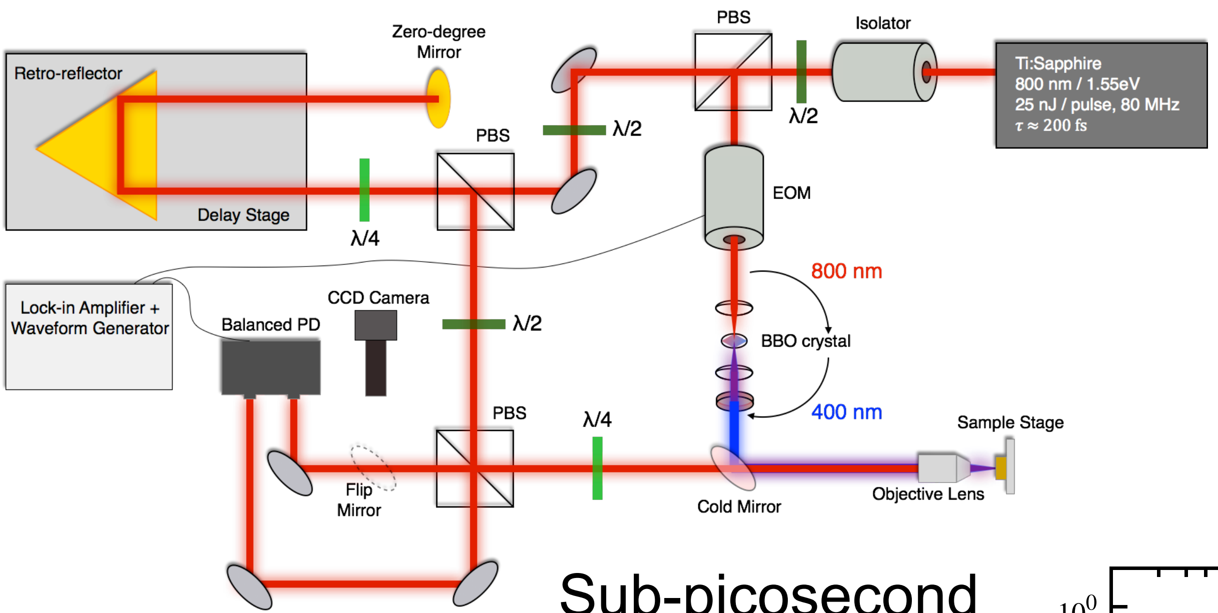
1. Electron energy transmission across metal/metal interfaces and electron-phonon coupling

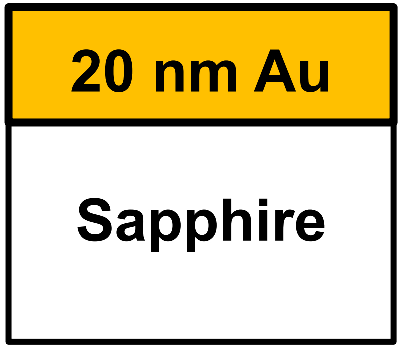
Ex: Au/Ti adhesion layer/insulating substrate

2. Electron energy transmission across metal/non-metal interfaces to control carrier densities in non-metals

Ex: Au/doped CdO to control ultrafast plasmonic absorption in CdO

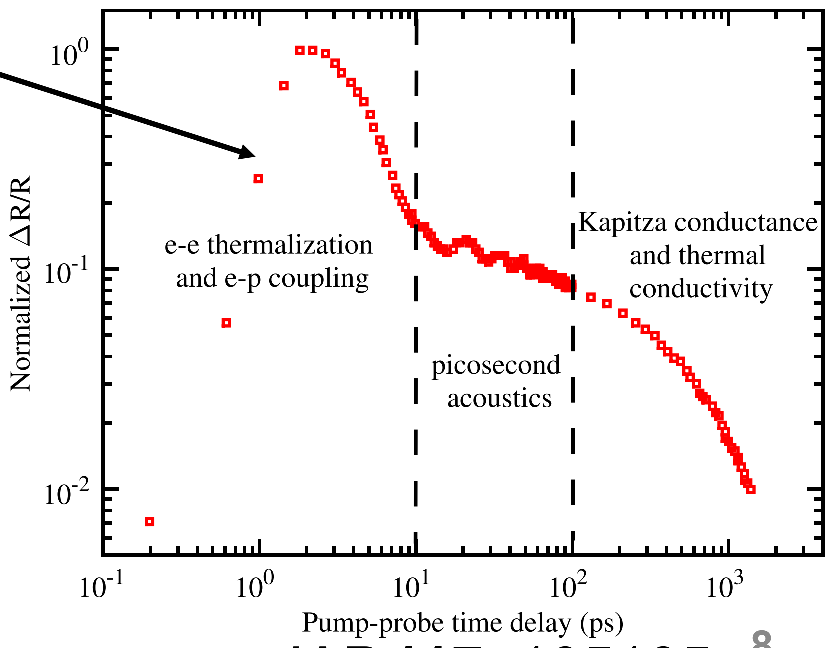
Ultrafast pump-probe to measure EP coupling





Energy no longer deposited in system

Sub-picosecond heat event

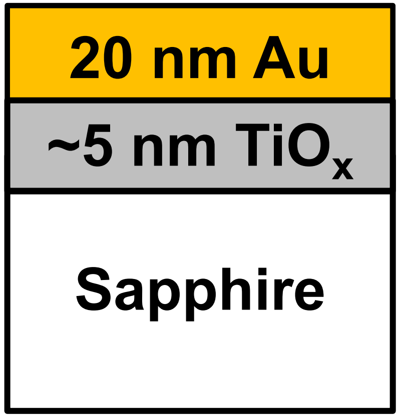


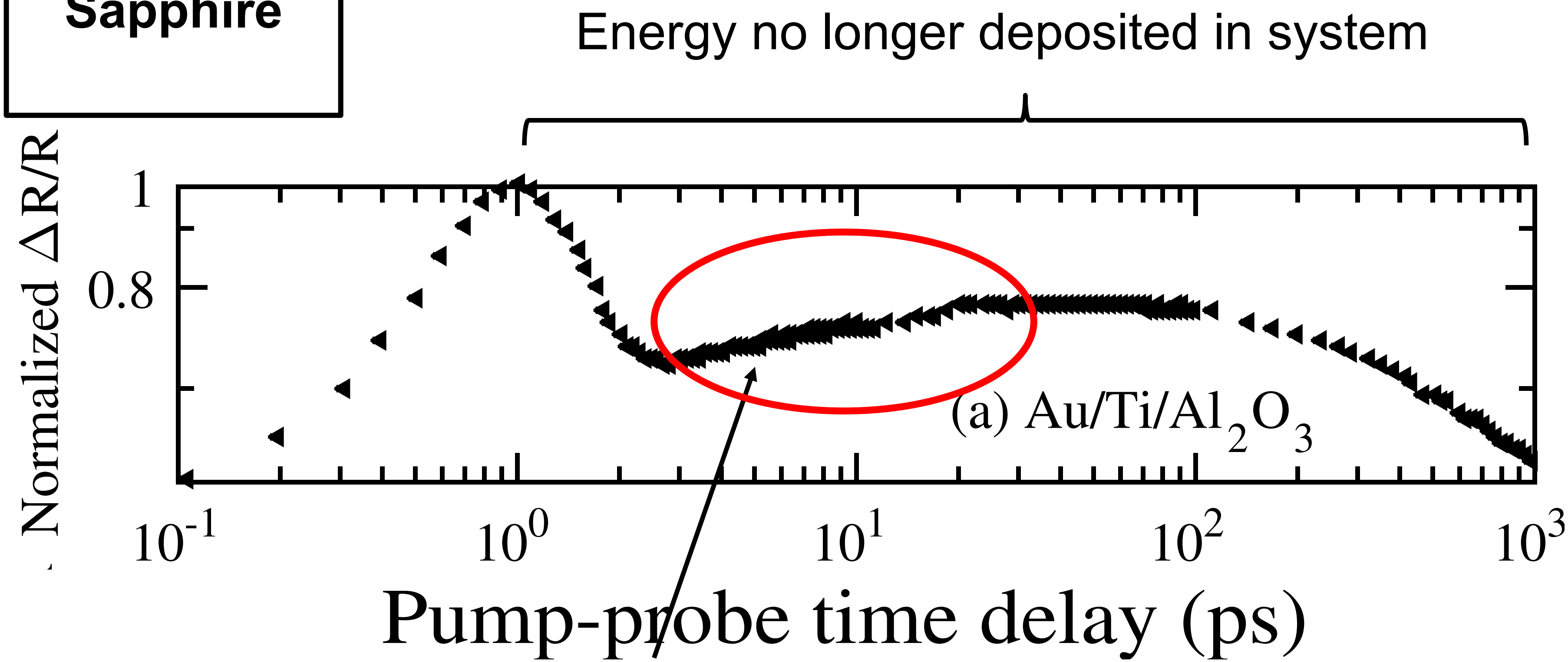
Directly related to temperature (advantage of probing metal)

ACS Photonics **5**, 4880

JAP **117**, 105105

Ultrafast pump-probe to measure EP coupling





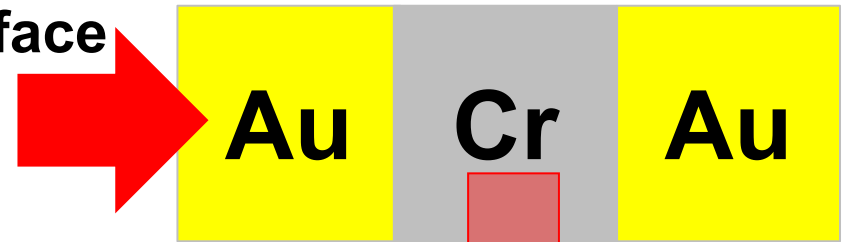
So why does temperature at surface increase when no energy is deposited in the system?

JAP **117**, 105105

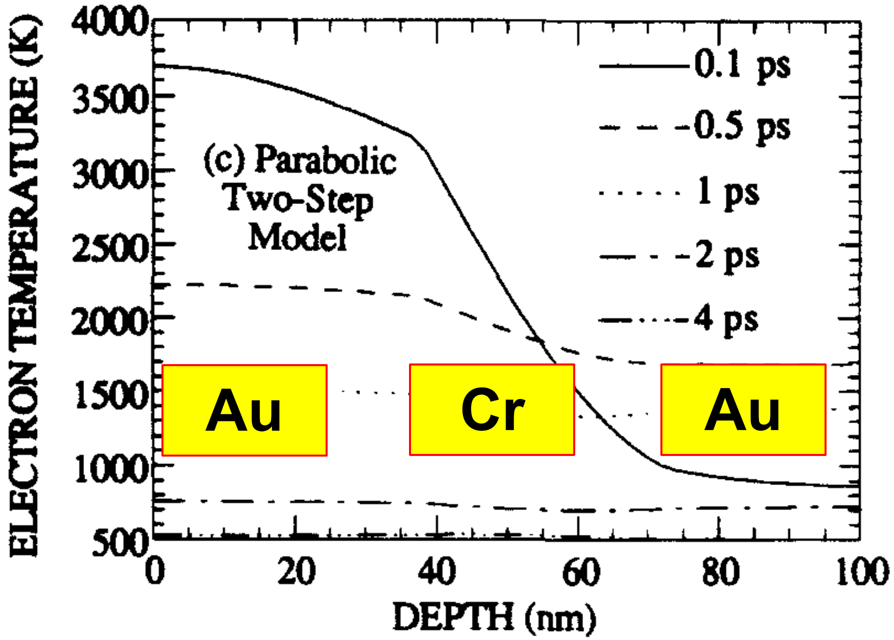
Electron-phonon interactions at metal/metal interfaces

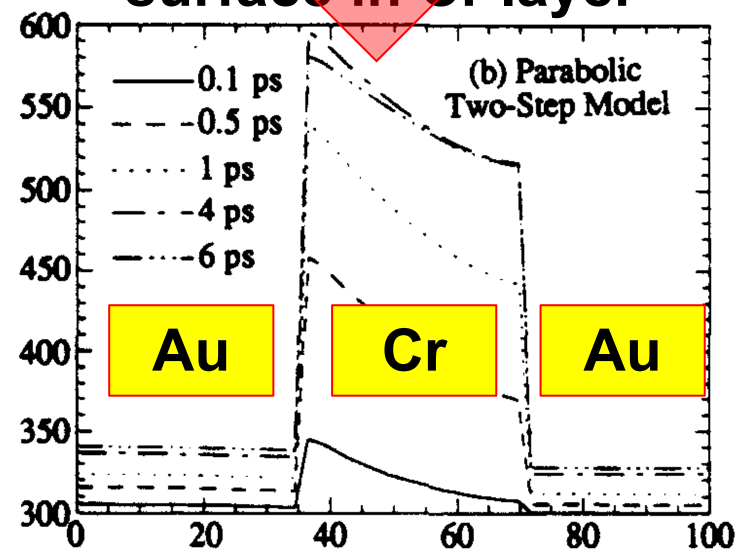
<u>Recall seminal predictions by Tien</u>
Qiu and Tien, *IJHMT* **37**, 2789 (1994)

Laser excites Au surface



Lattice heats up under surface in Cr layer

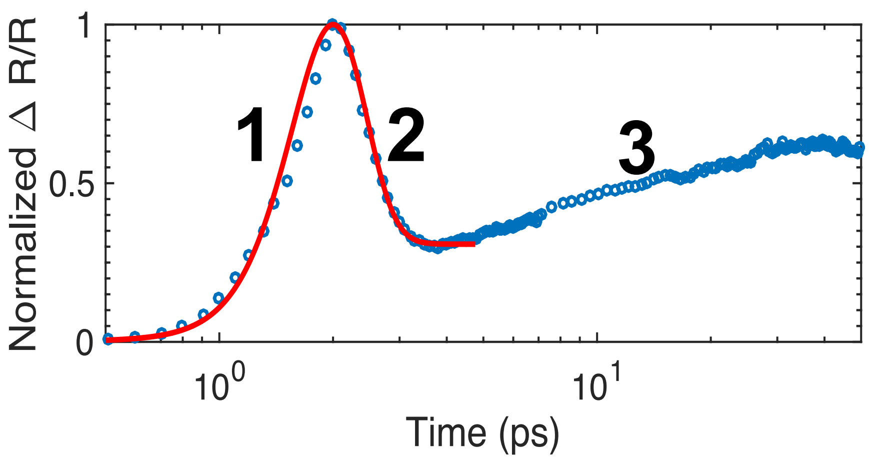


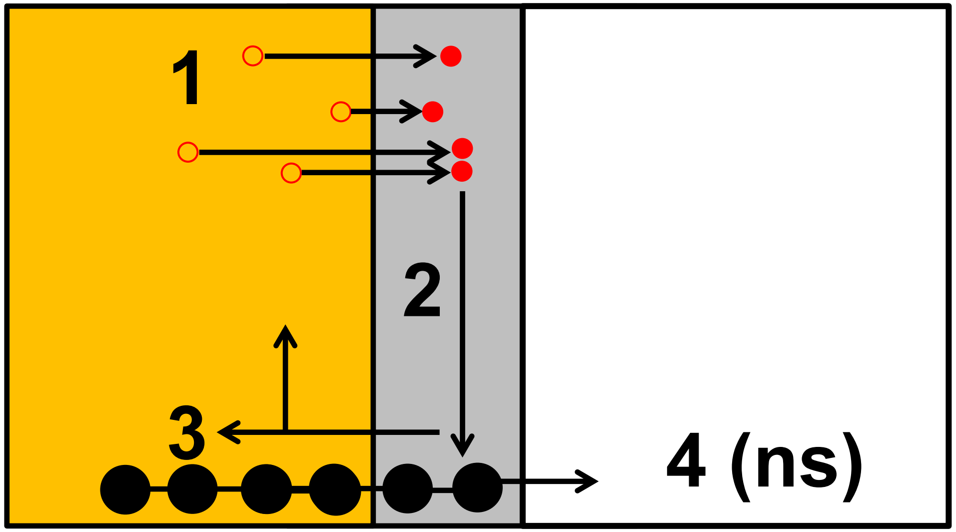


Au lattice slowly responds

Ballistic thermal injection

- Excited electrons in metal from pulse do not thermalize with lattice and deposit their energy to lattice in sub-surface layer
- Ballistic transport of electron energy through gold into titantium



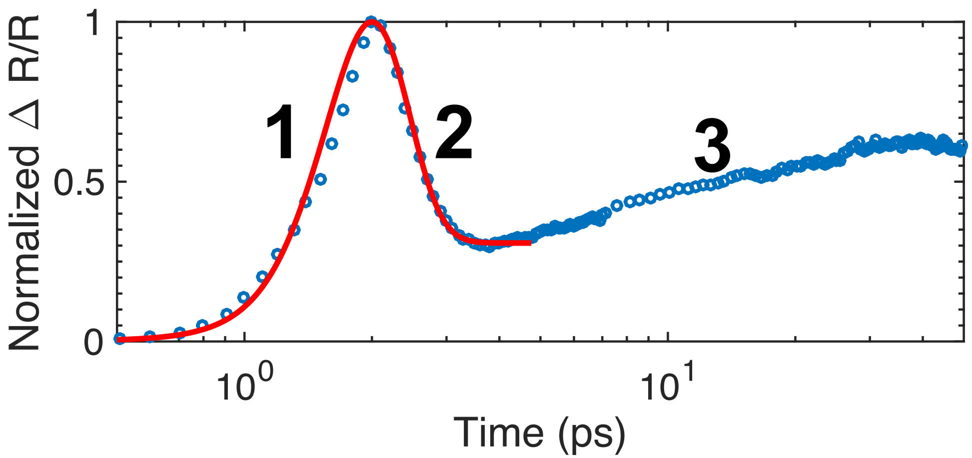


When would we see this effect?

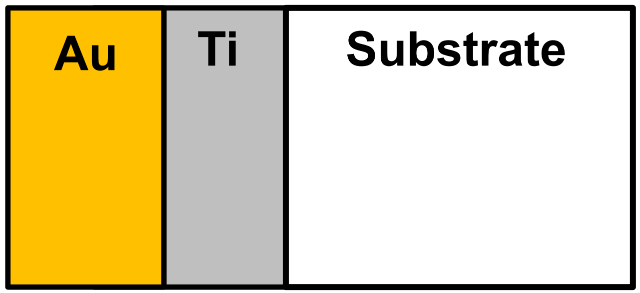
1. Metal/metal or metal/non-metal interfaces with large differences in electron-phonon coupling factor
2. Films with thicknesses less than electron-phonon mean free path
3. Interfaces with very little electron-electron thermal resistance

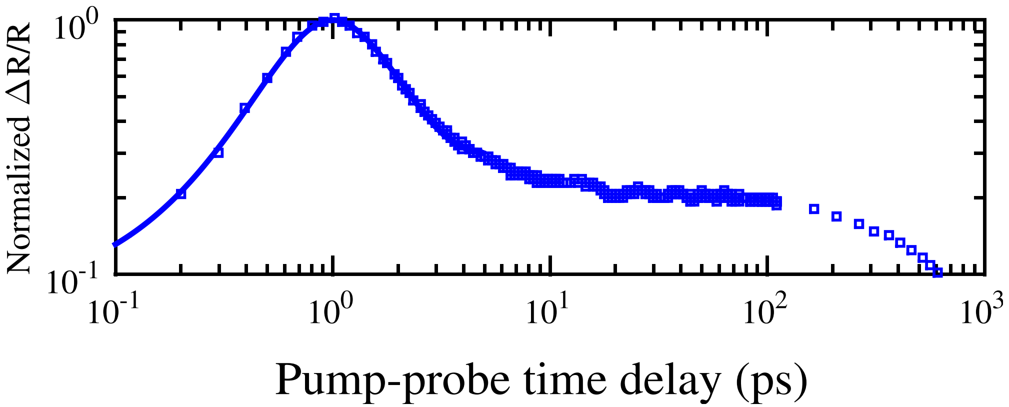
TDTR measurements of time scales of noneq. transport

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



$d_{\mathrm{Au}} < \lambda_{ep}$





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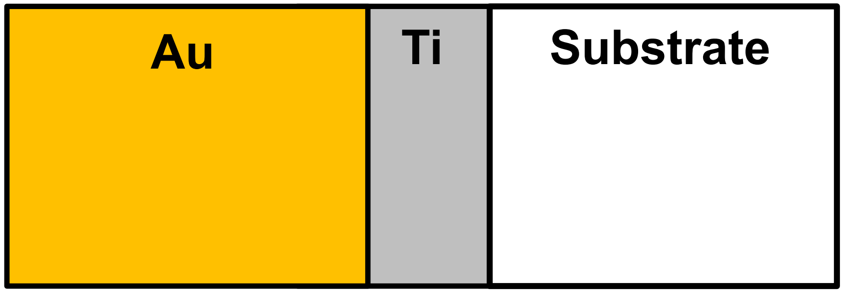
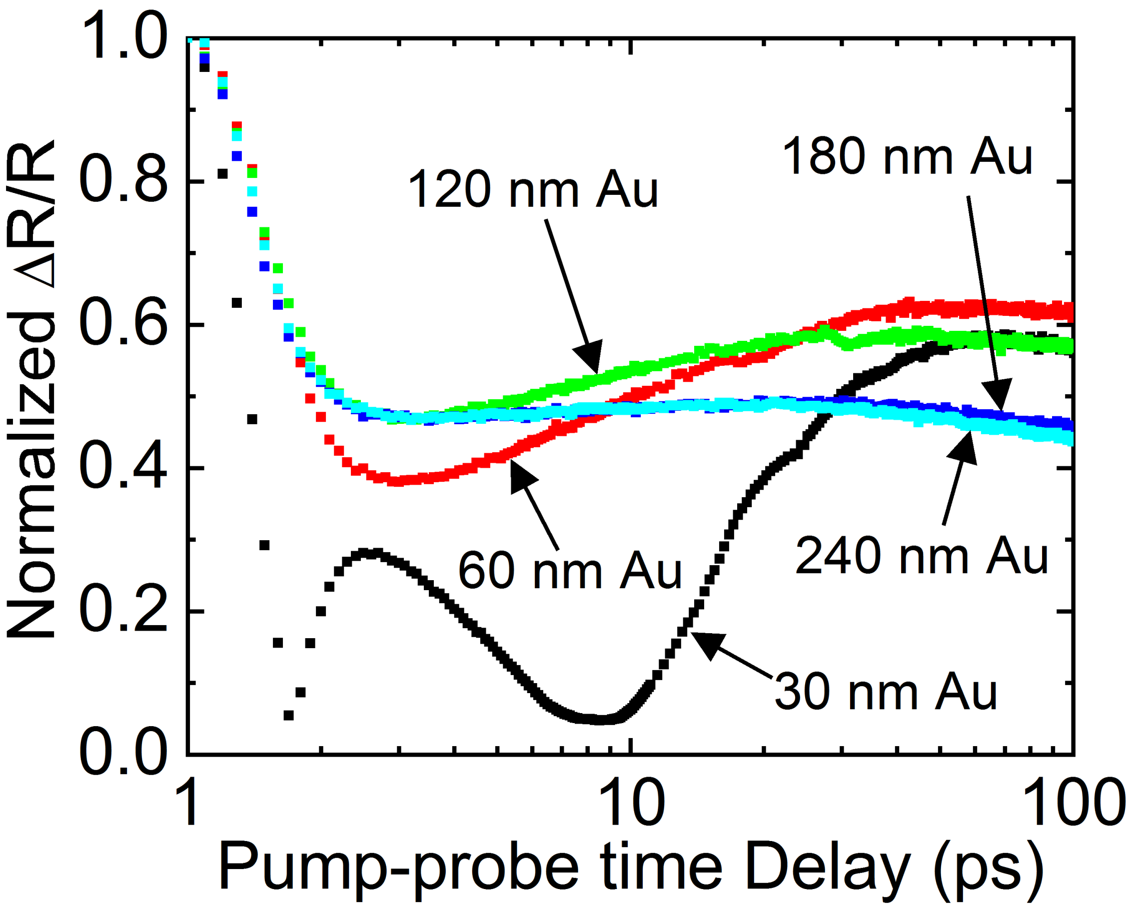


Ballistic electron-phonon mean free path in gold

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



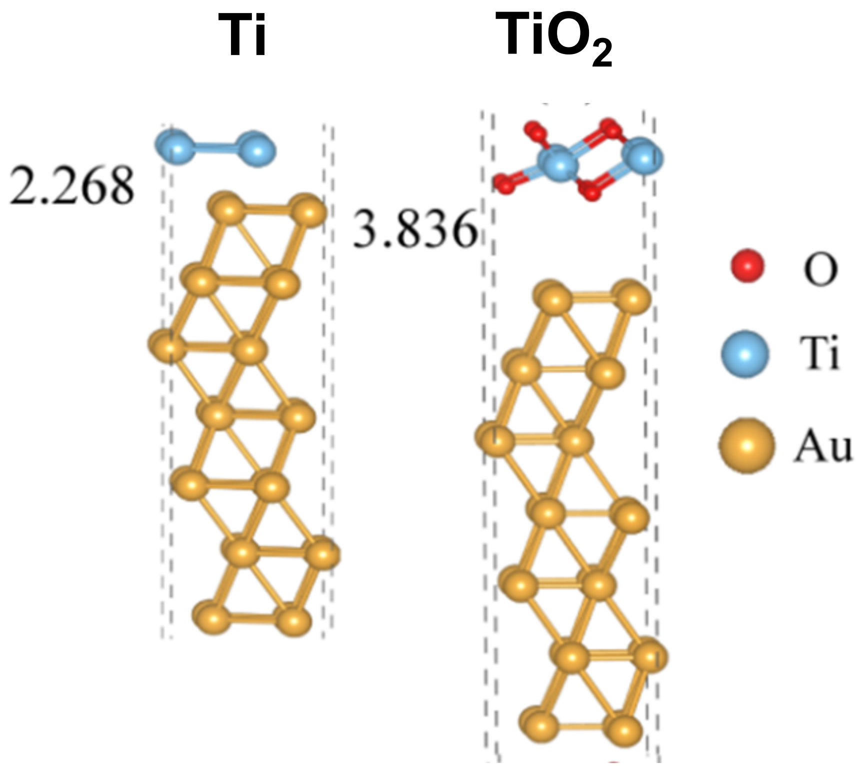
$$\lambda_{\mathrm{ep,Au,calc}} \approx \sqrt{\frac{\kappa_{\mathrm{electron}}}{G}}$$

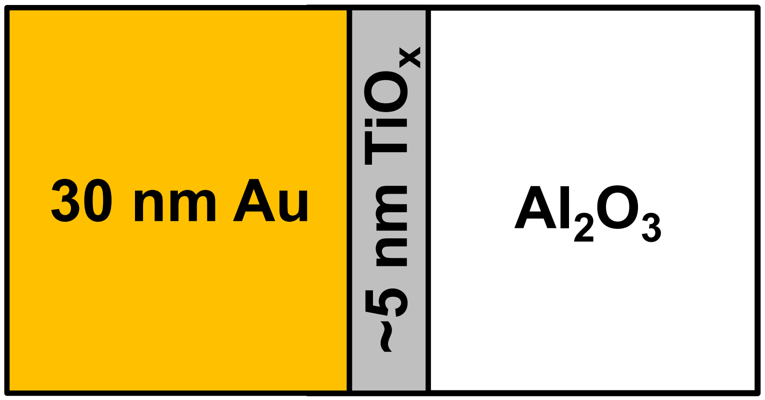
$$\approx 79 - 117\,\mathrm{nm}$$

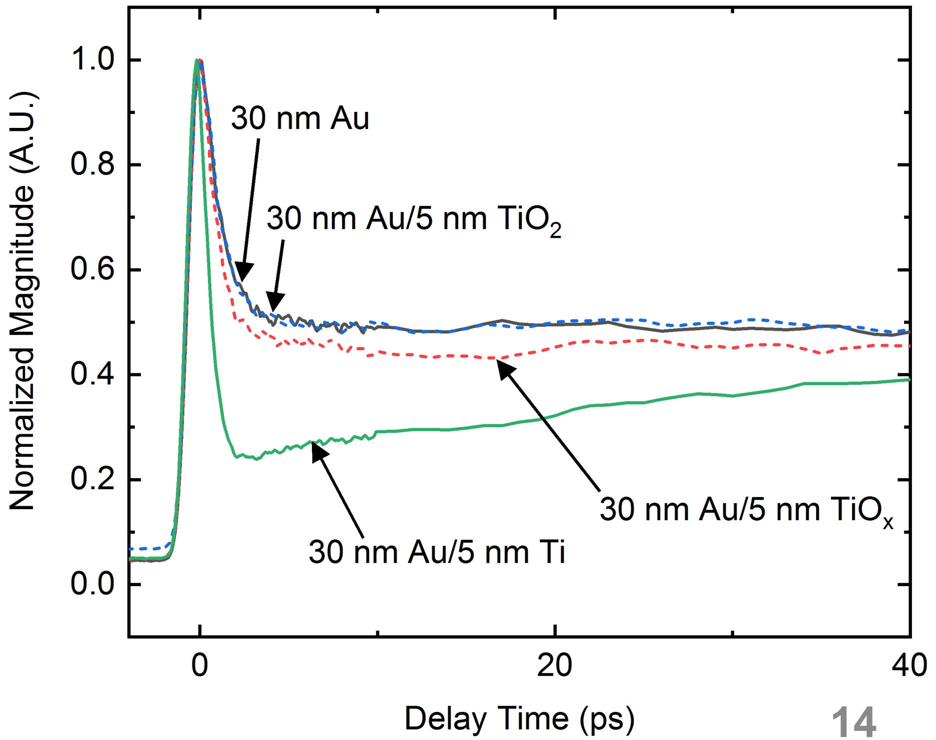
$$\lambda_{\mathrm{ep,Au,meas}} \approx 90 - 130\,\mathrm{nm}$$

Bonding at metal/metal interfaces controls heat injection

- Electron injection from Au to Ti needed to observed “back heating”
- Ti oxygen stoichiometry impacts electron injection to Ti







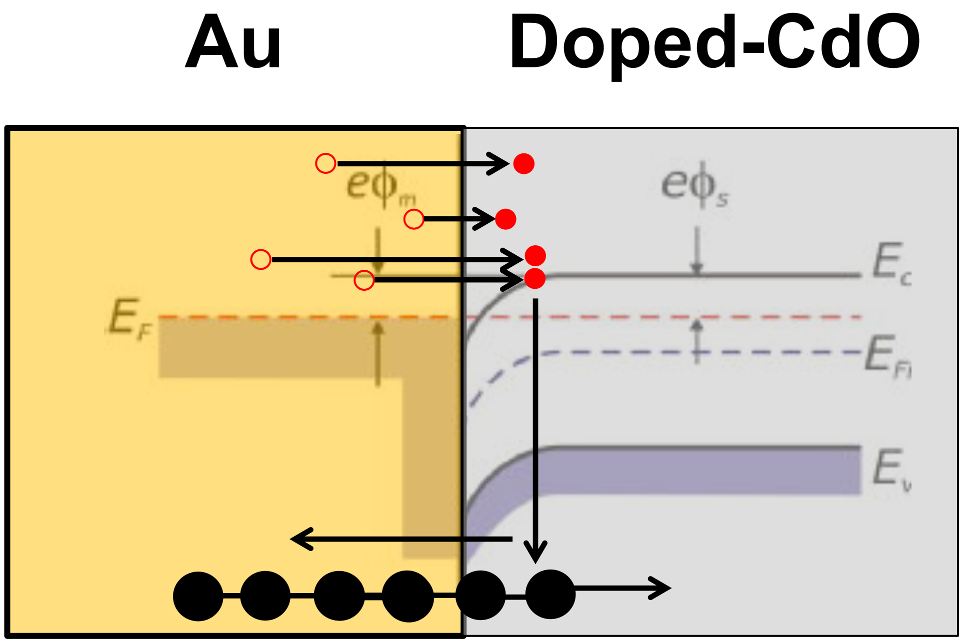
The Journal of Physical Chemistry Letters **11**, 1419

Nonequilibrium at metal/doped non-metal interfaces

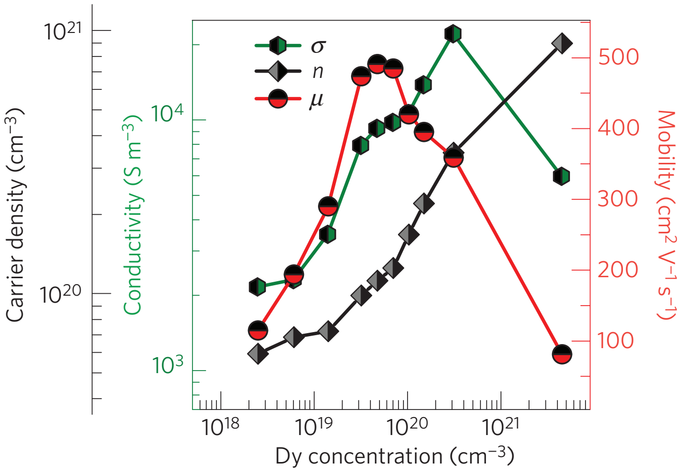
- Consider ohmic contact between metal and doped non-metal
- Vary carrier concentration in non-metal

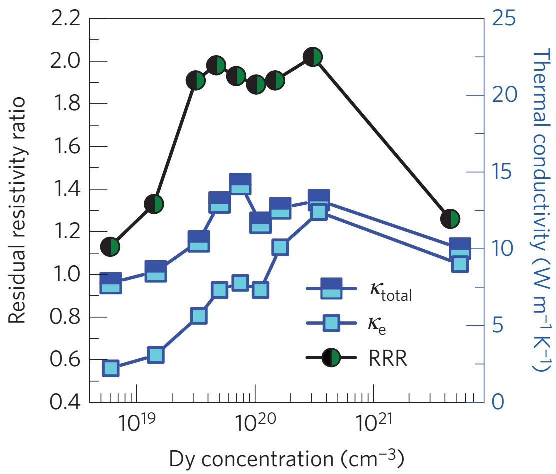
- Electron injection from Au to interfacial layer must occur to observe "back heating" effect

- Will not occur when interface is insulating

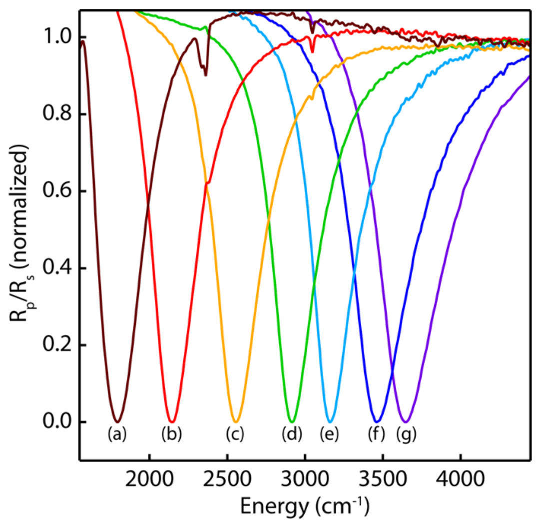


CdO – a gateway for mid-IR plasmonics





Nat. Mat. **14**, 414
Appl. Phys. Lett. **108**, 021901

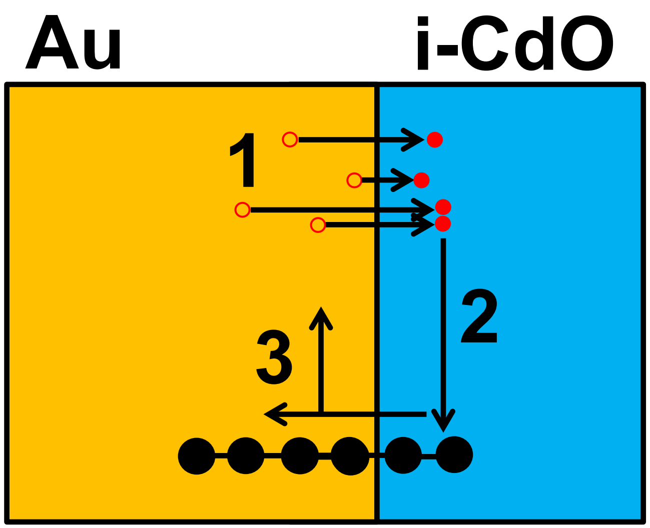


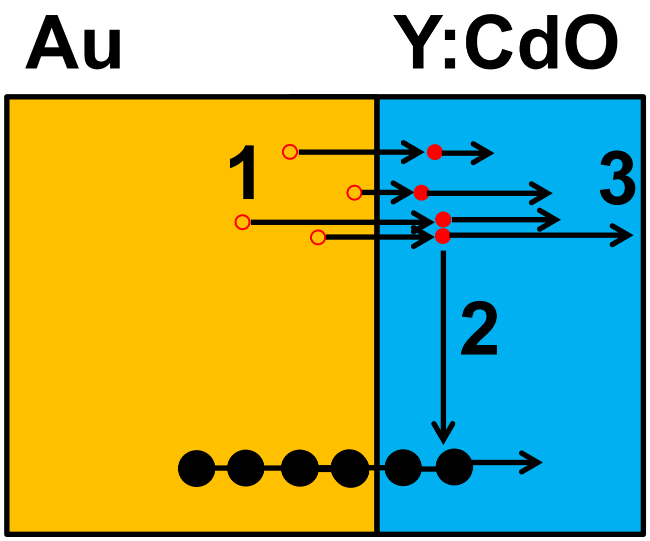
ACS Photonics **4**, 1885

- 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

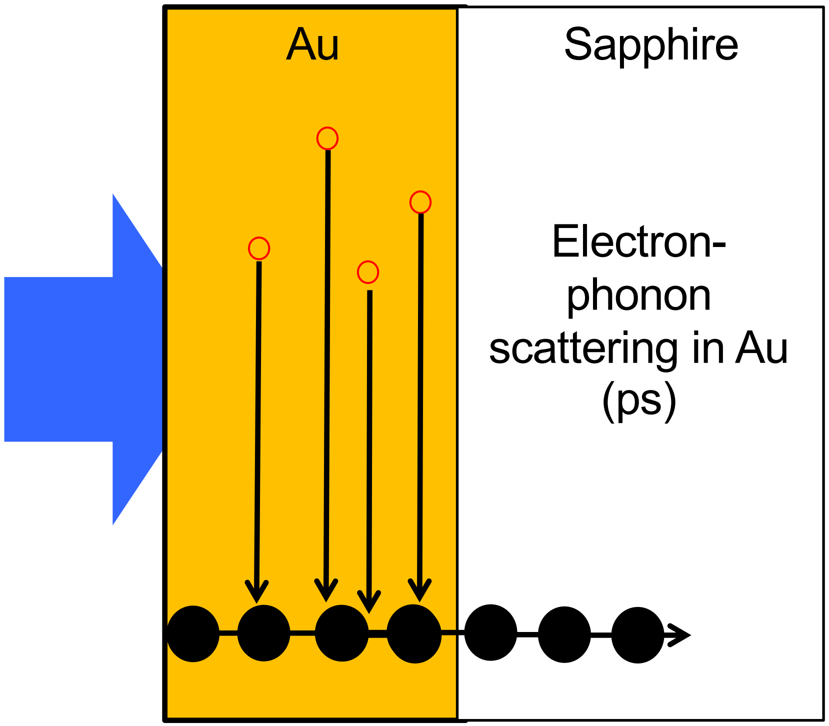
Doping will control electron-electron TBC and electron thermal conductivity in CdO, vary "back heating"

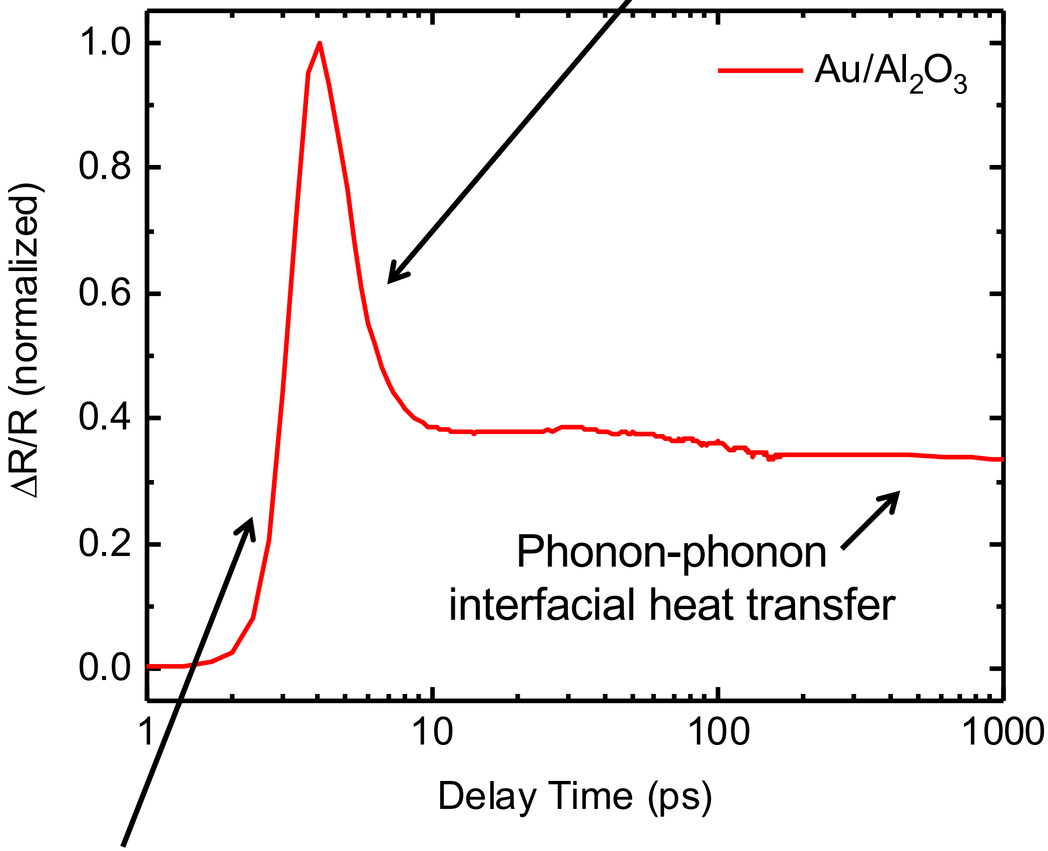




arXiv:2005.10179 *and to appear in Nature Nanotechnology*

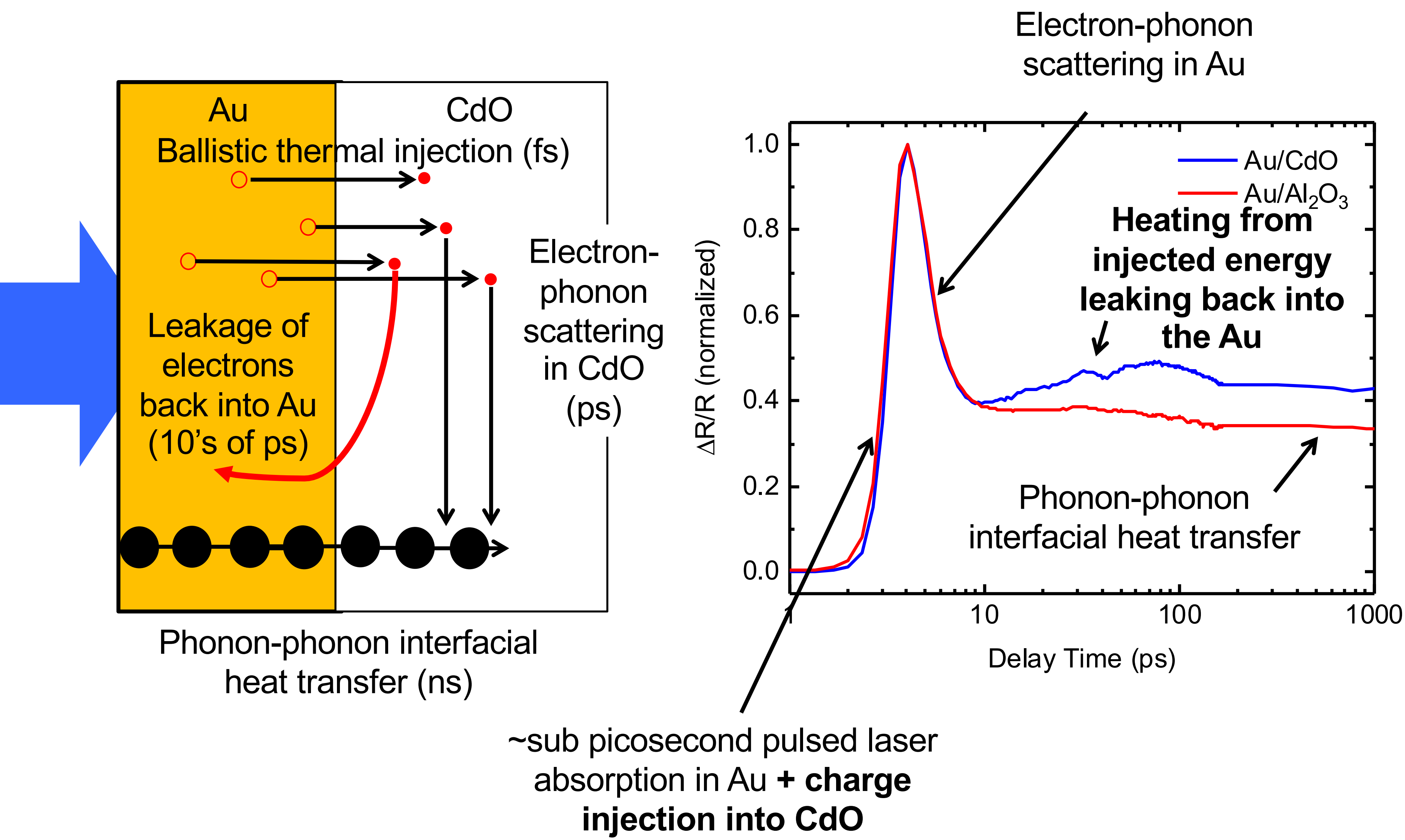
Nonequilibrium processes at Au/CdO interfaces





[arXiv:2005.10179](https://arxiv.org/abs/2005.10179) *and to appear in Nature Nanotechnology*

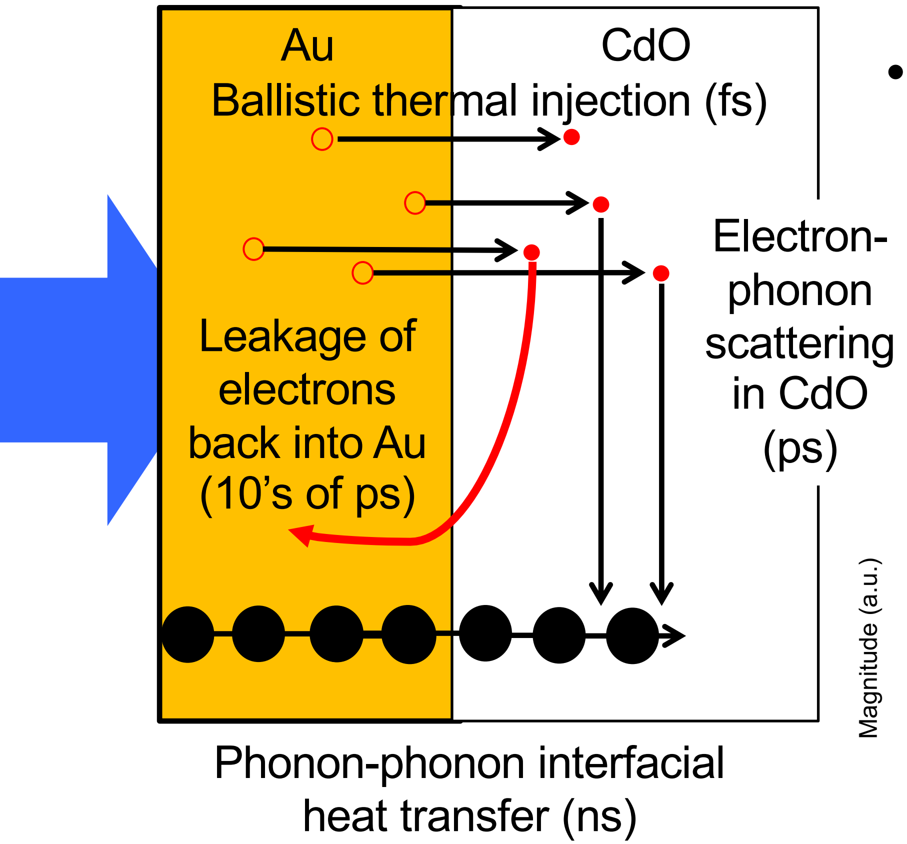
Nonequilibrium processes at Au/CdO interfaces

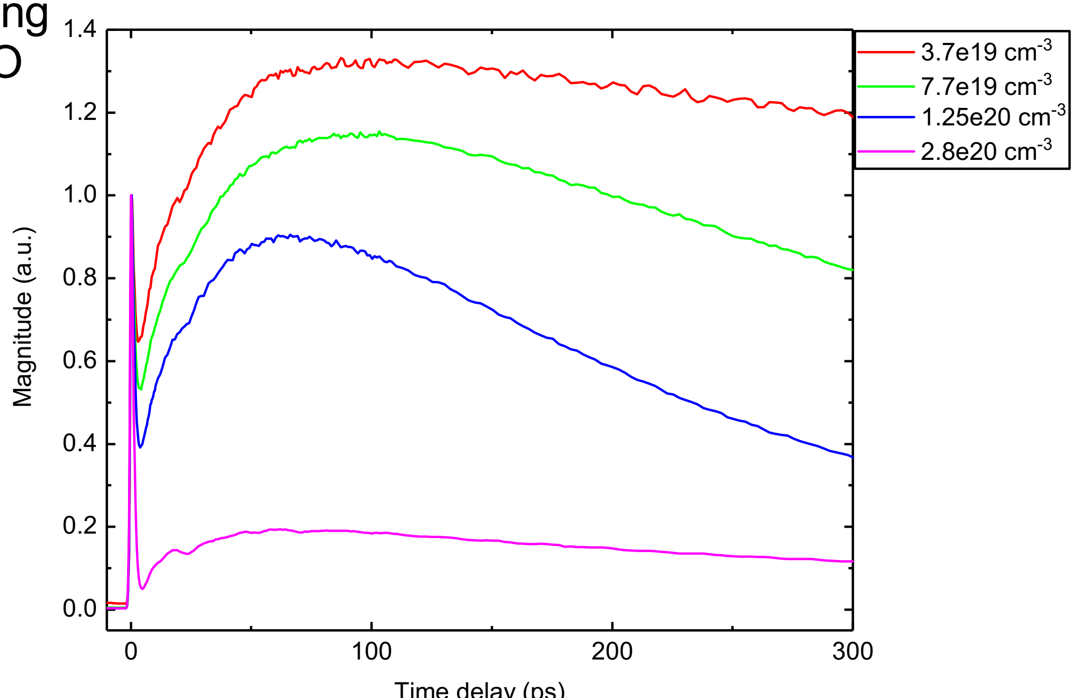


arXiv:2005.10179 *and to appear in Nature Nanotechnology*

Nonequilibrium processes at Au/CdO interfaces

- Doping of CdO dictates amount of "electron leakage" back into Au
- Lower resistivity + higher e-ph coupling in CdO, less electronic back heating into gold contact





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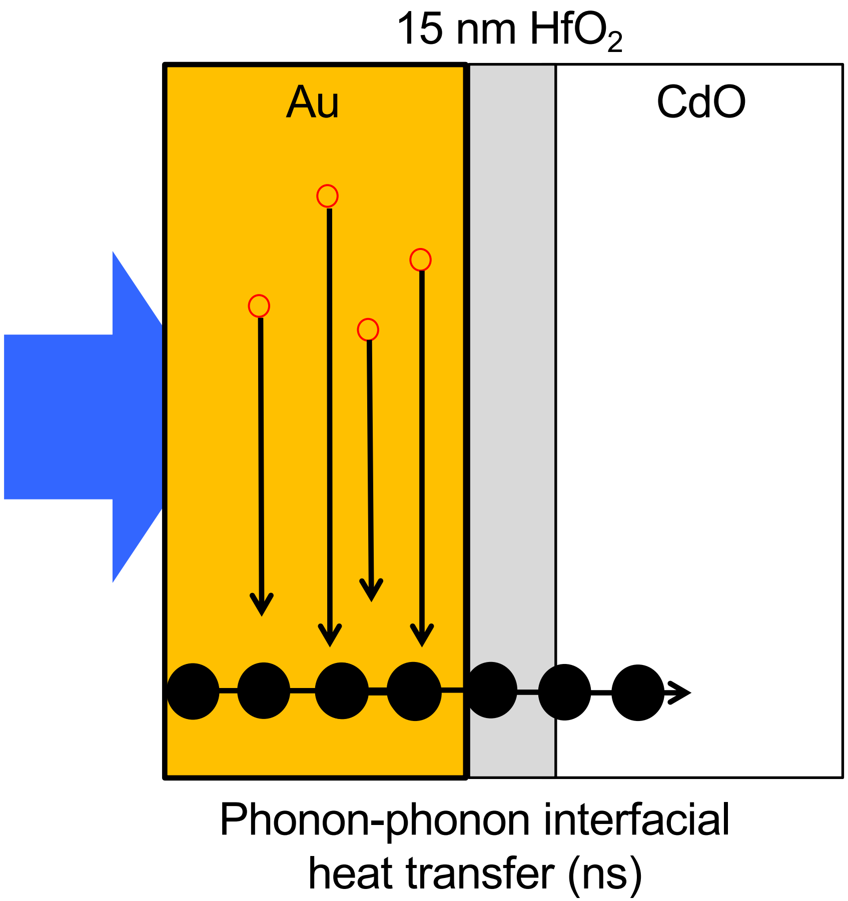
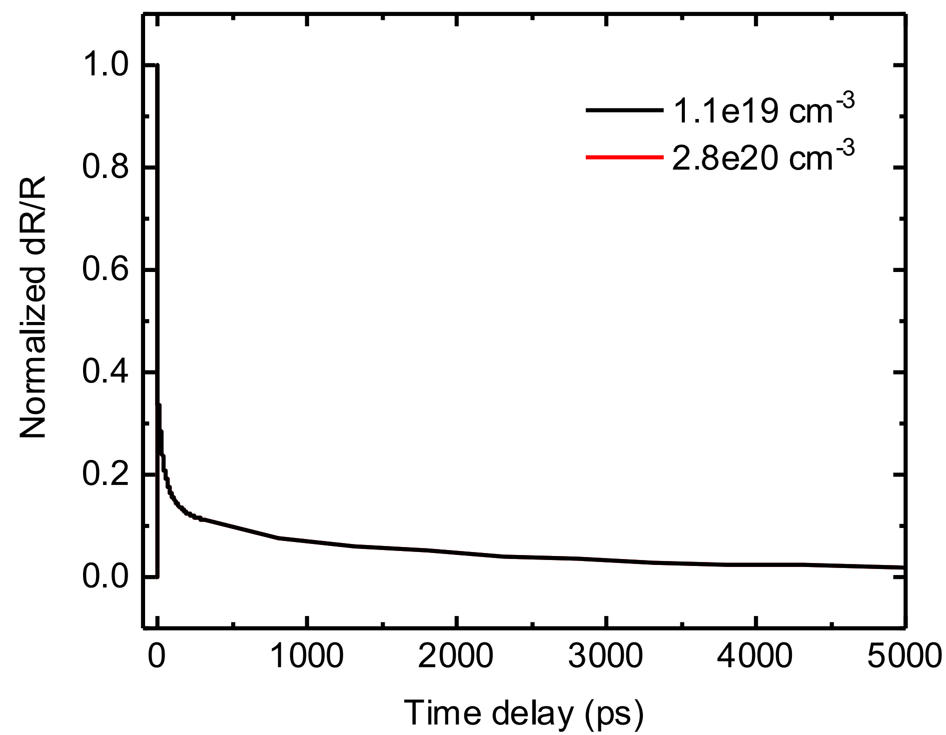
Nonequilibrium processes at Au/CdO interfaces



- Transparent buffer layer stops ballistic electrons, but allows light to transmit
- No back-heating observed for any dopant concentration!



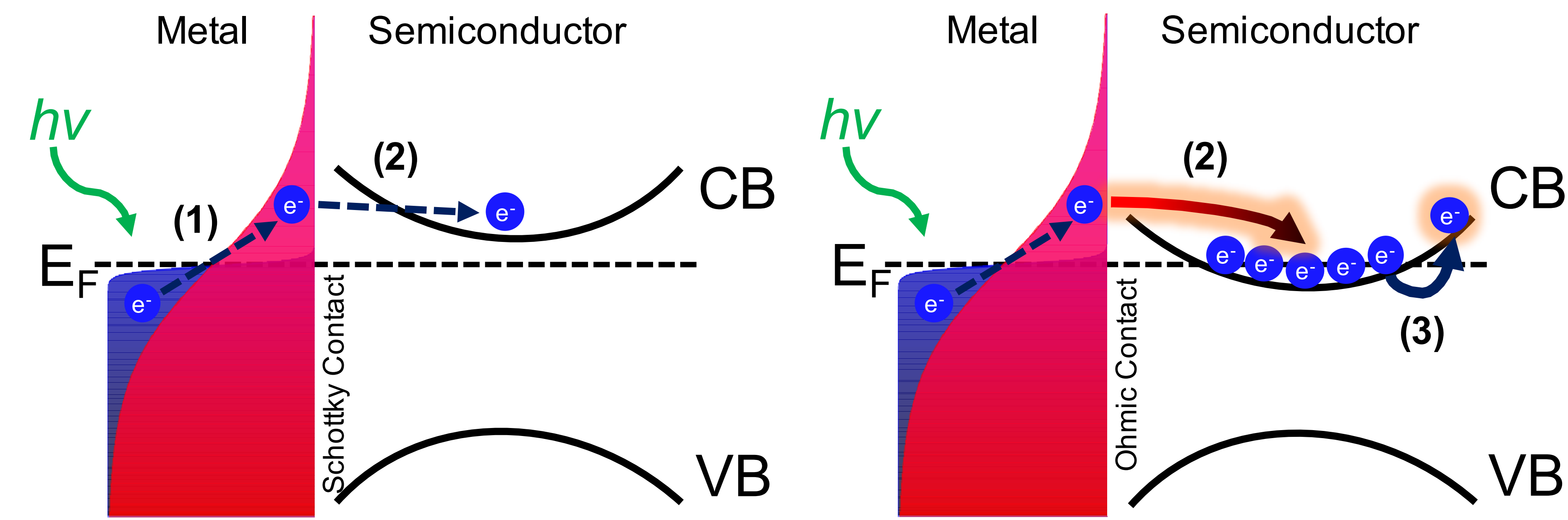
arXiv:2005.10179 *and to appear in Nature Nanotechnology*

Ballistic thermal injection

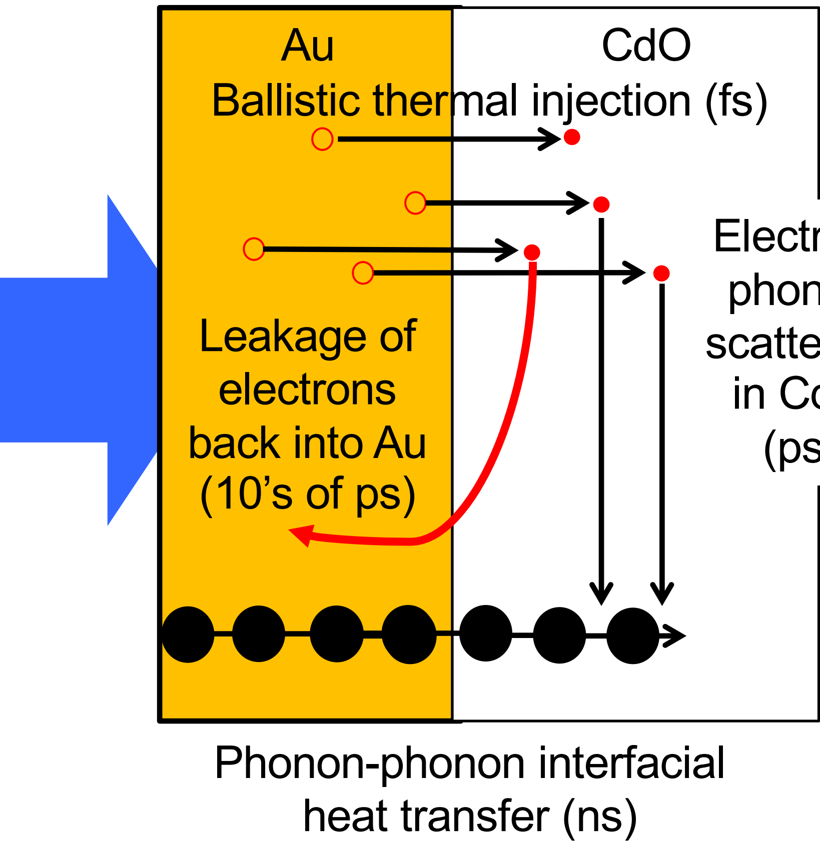
- Can enable a “transient thermal diode” effect
- Energy easily transmitted across interface when traveling ballistically
- Slowly “goes back” across the interface when diffusive
- Is this just hot electron injection (charge)?
  - Too slow of process
  - Can further rule this out by monitoring CdO plasmon response

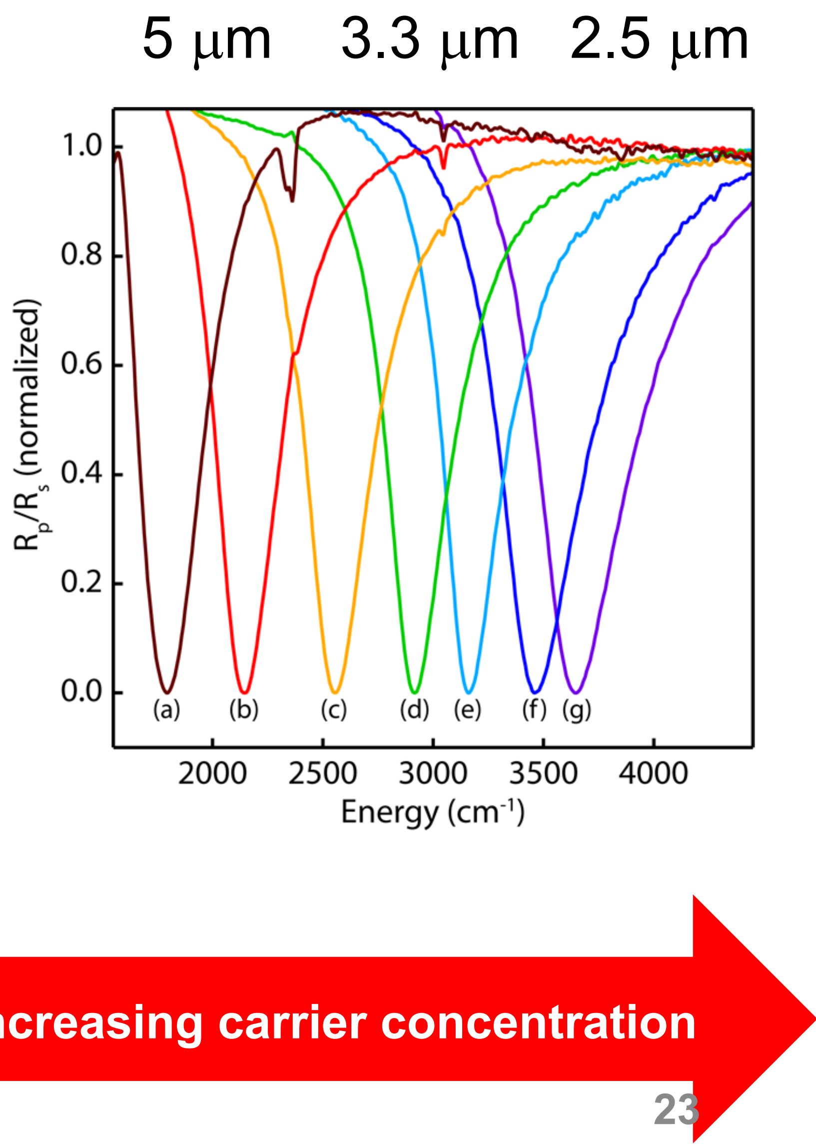
a) Hot electron injection (Charge transfer)

b) Ballistic thermal injection (Energy transfer)

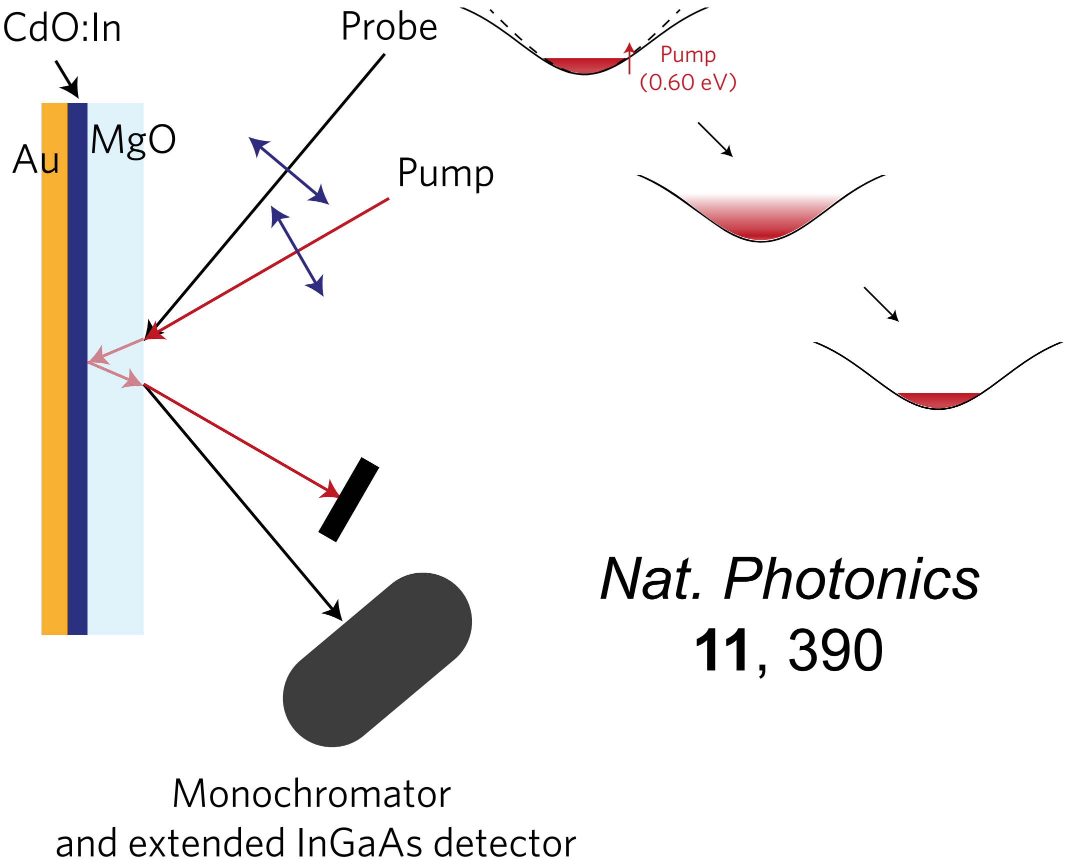


Nonequilibrium electrons to control CdO plasmons

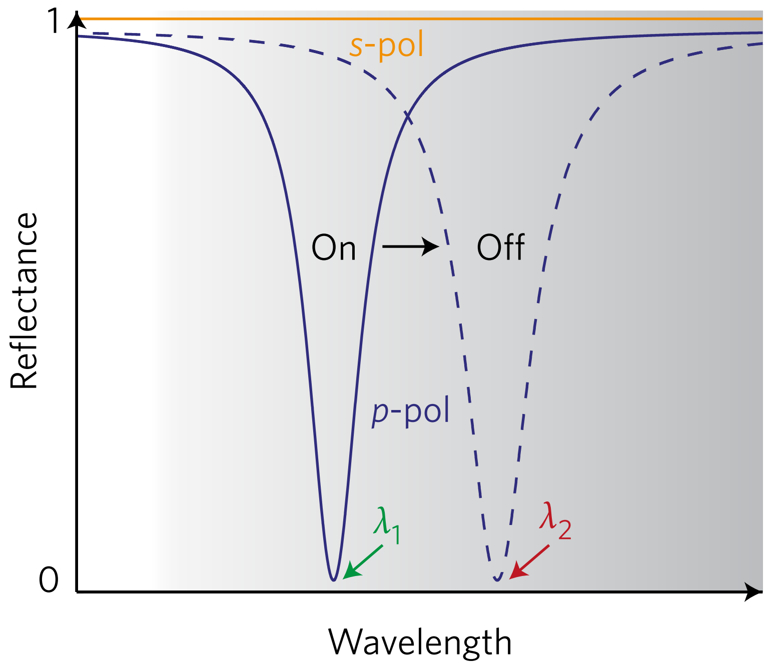




Pumping electrons can impact plasmon response



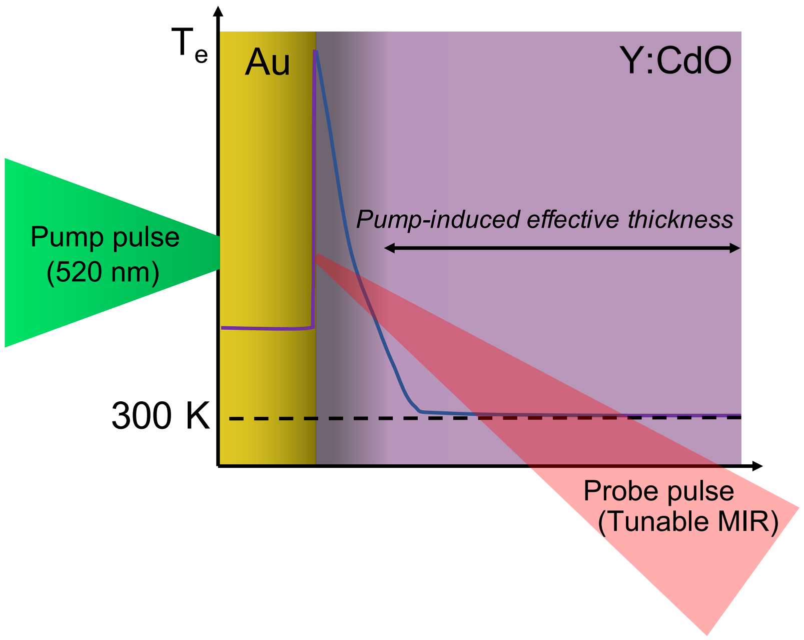
Nat. Photonics
11, 390



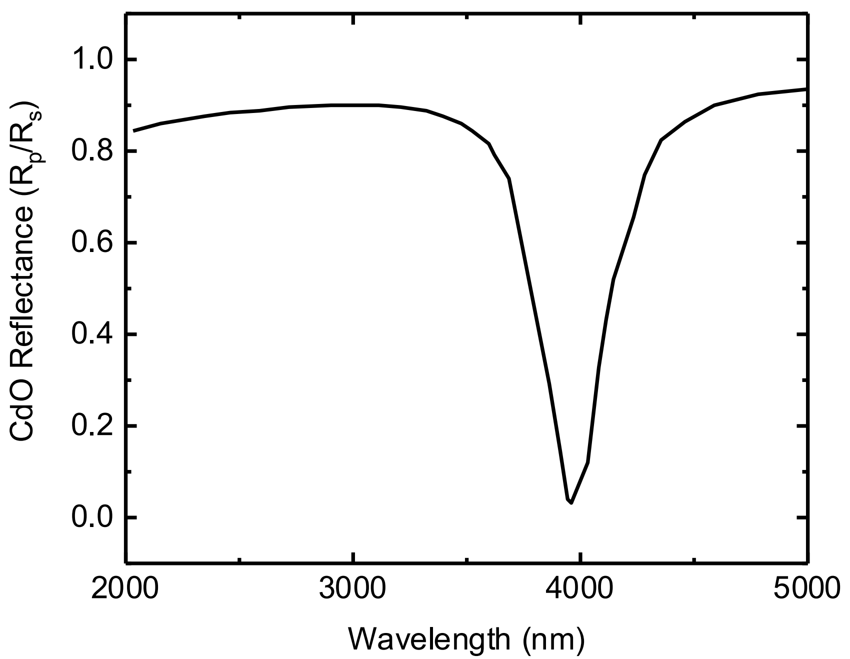
- Exciting electrons in CdO (intraband) can change effective mass and red shift plasmon response
- Requires specific wavelength photons and laser conditions
- **Can we use BTI to inject heat into CdO and impact plasmons?**

Pump electrons in Au, probe plasmon in CdO

How is the IR plasmon response of CdO impacted by ballistic thermal injection?



Au/CdO/sapphire absorption response

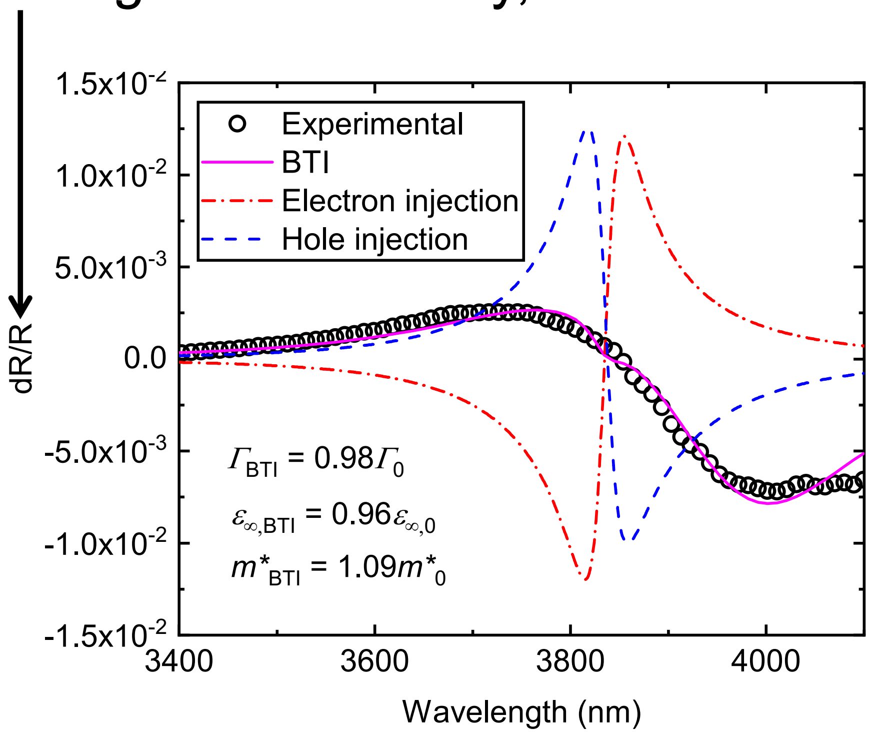


arXiv:2005.10179 *and to appear in Nature Nanotechnology*

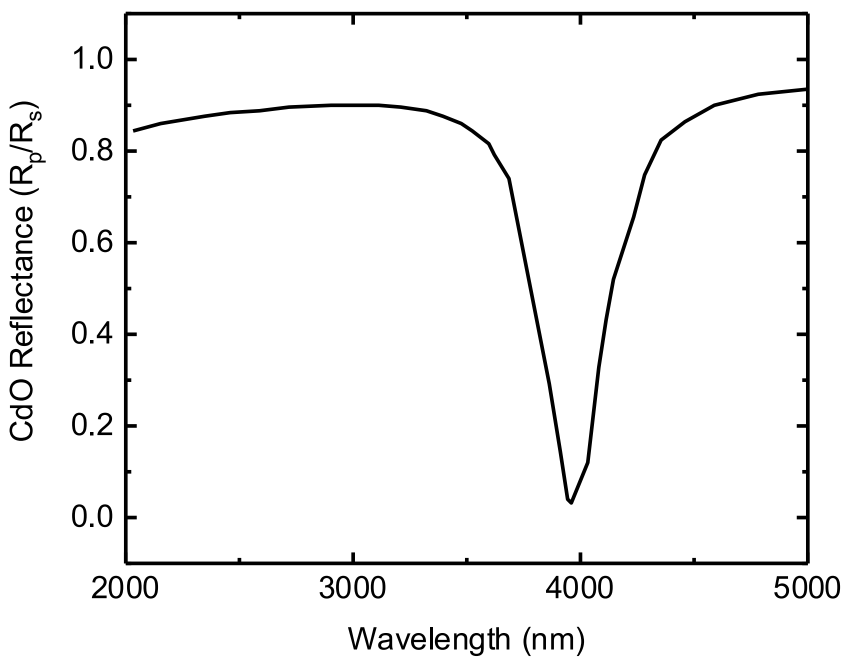
Pump electrons in Au, probe plasmon in CdO

Asymmetric red shift in ENZ plasmon mode due to BTI

Note we are measuring *change* in reflectivity, dR



Au/CdO/sapphire absorption response



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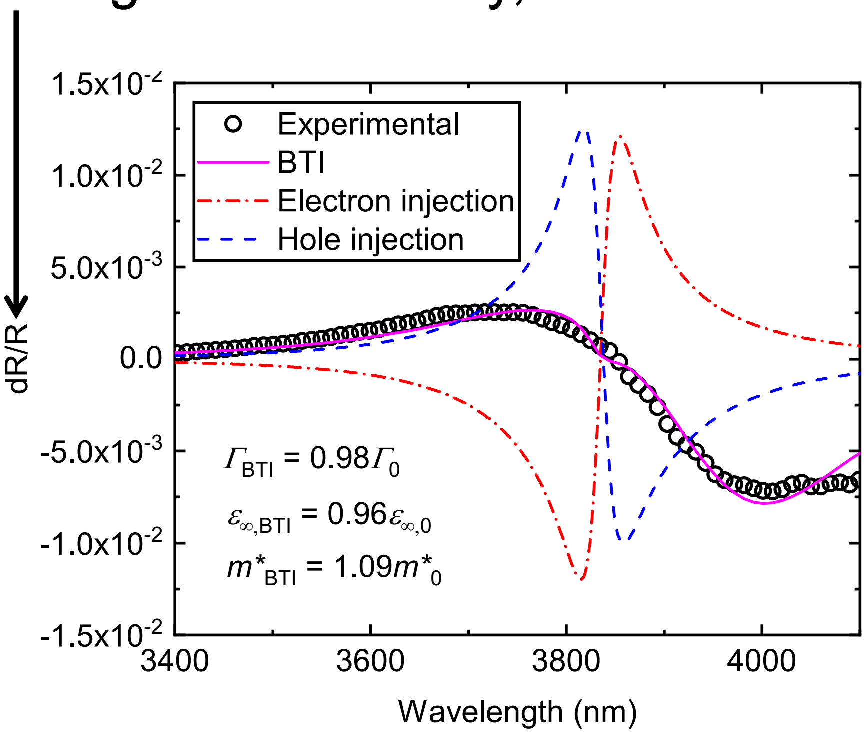

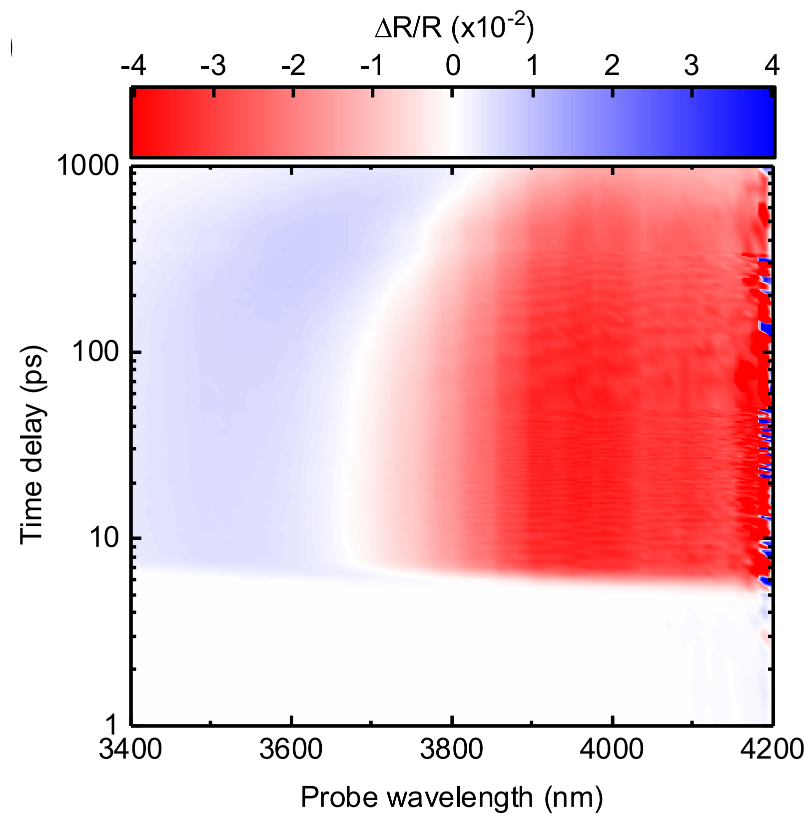

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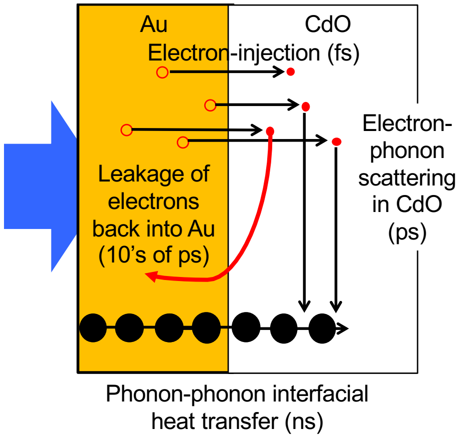
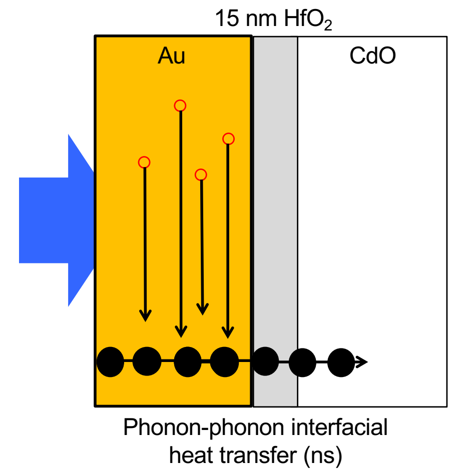
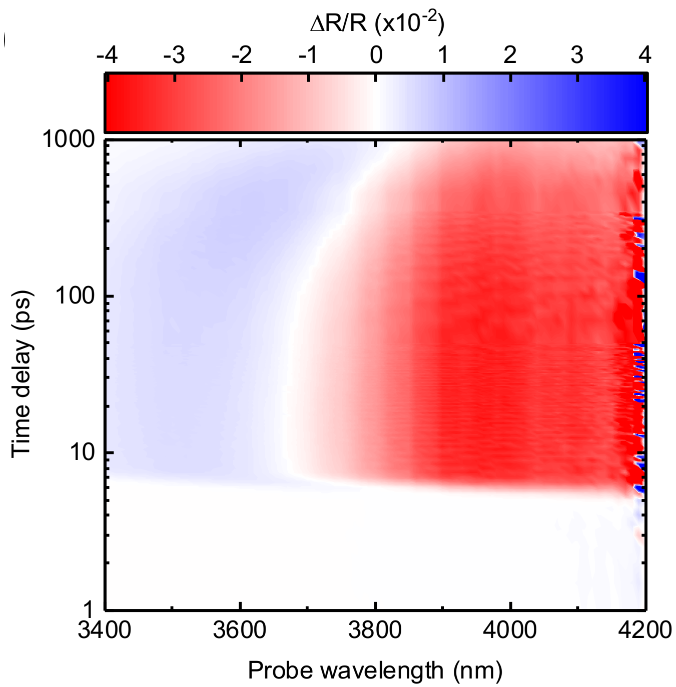
And it's not an optical artifact



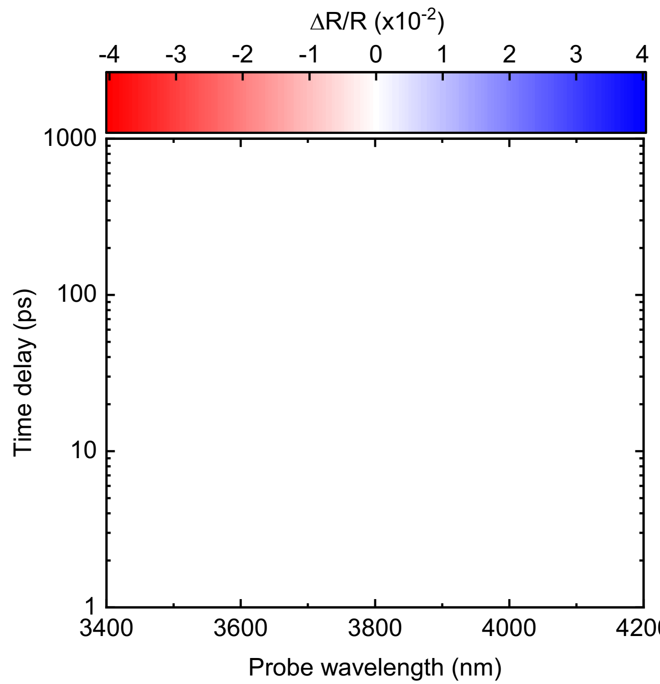
15 nm HfO_2 layer prevents any electron energy from moving from Au to CdO, resulting in no measurable response



Au/CdO



Au/HfO_2/CdO (same scale)



Nonequilibrium electron thermal transport processes control electrons, phonons and plasmons

Ballistic thermal injection (Energy transfer)