



SCHOOL of ENGINEERING & APPLIED SCIENCE
UNIVERSITY of VIRGINIA

Young **I**nvestigator **P**rogram (Year 1 Summary)

Electron dynamics during high-power, short-pulsed laser interactions with solids and interfaces

US AFOSR Grant #FA9550-13-1-0067



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Introduction



- Ph.D. in Mech. E., U.Va., 2008
- Truman Postdoc, Sandia ABQ, 2008 – 2011
- Joined U.Va. Faculty 11/2011



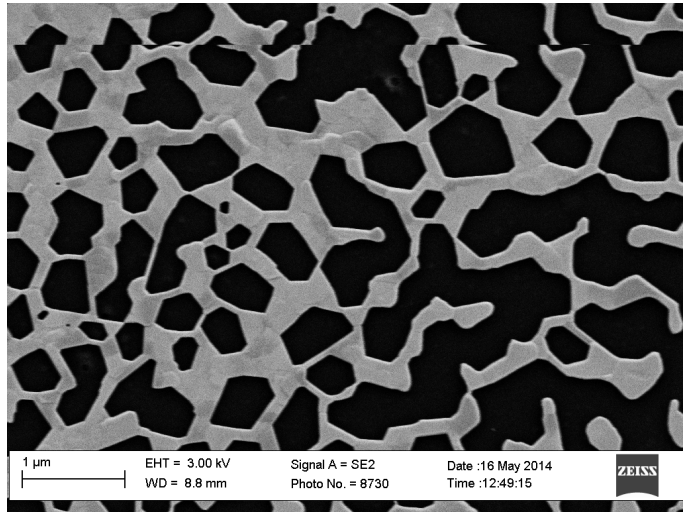
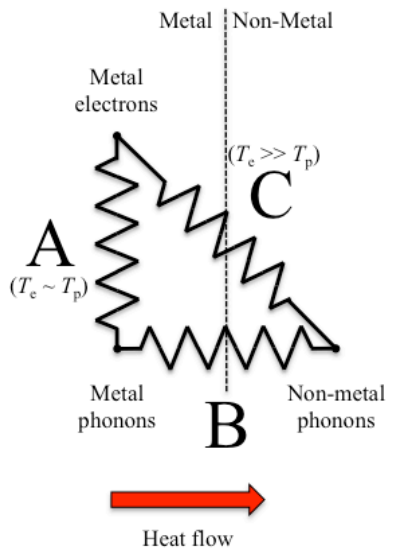
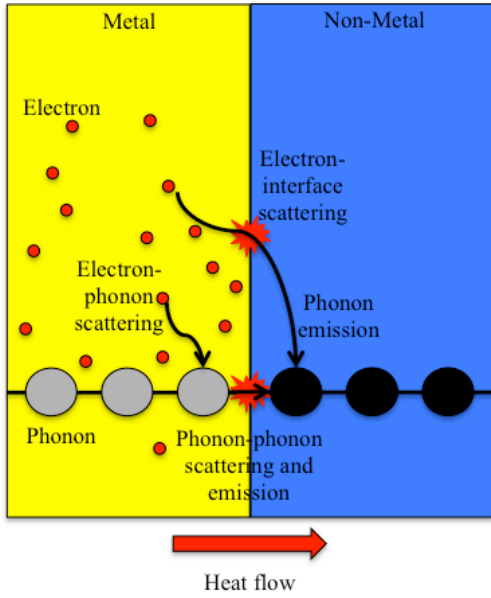
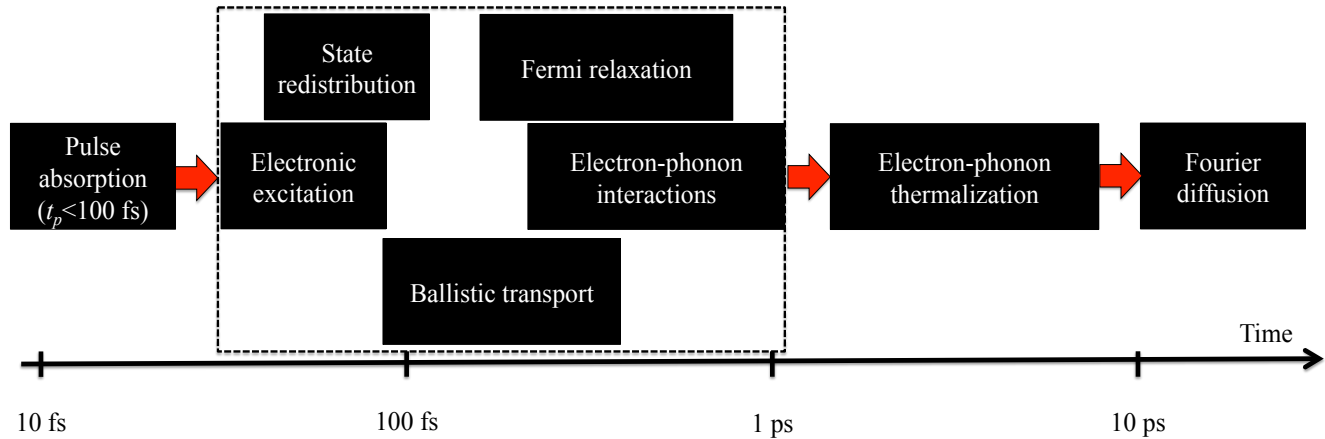
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“Themes” of program: fs excitations meet nanoscale HX

One sentence goal of YIP

Material engineering of nanosystems and interfaces to manipulate electron relaxation, heat transfer and damage from femtosecond pulses



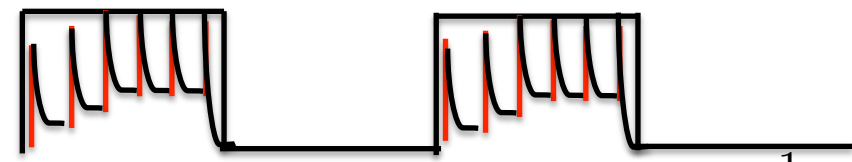
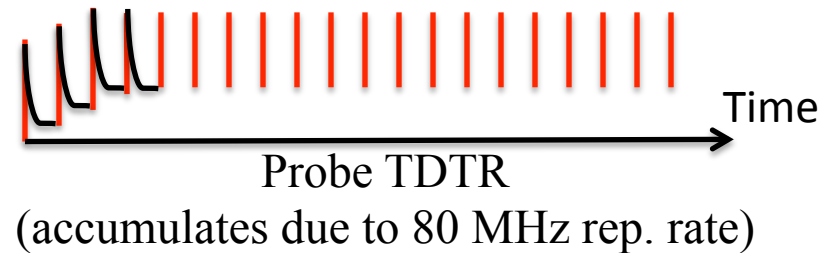
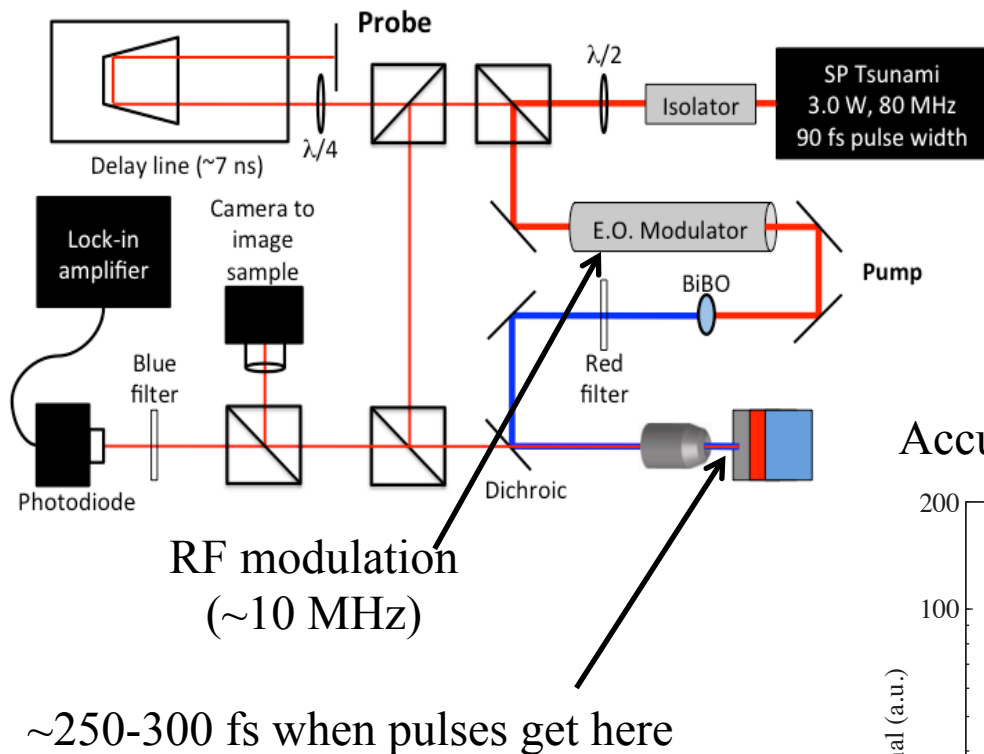
**Year 1 experimental approach (Femtosecond oscillator (nJ/pulse))
pulse accumulation, interfacial heat transfer and material damage**

- 1) Temperature dependence of nonequilibrium electron dynamics in Au**
- 2) Thermal conductivity of “hot” electrons ($T_e \neq T_p$)**
- 3) Substrate influence and “DC” heating (steady state laser heating from pulse accumulation)**
- 4) Electron-interface scattering during electron nonequilibrium – experiments and quantum/semi-classical modeling**
- 5) DC heating induced damage and role of electron-interface scattering**
- 6) Looking ahead to Year 2**

Year 1 – Pump-probe with oscillator

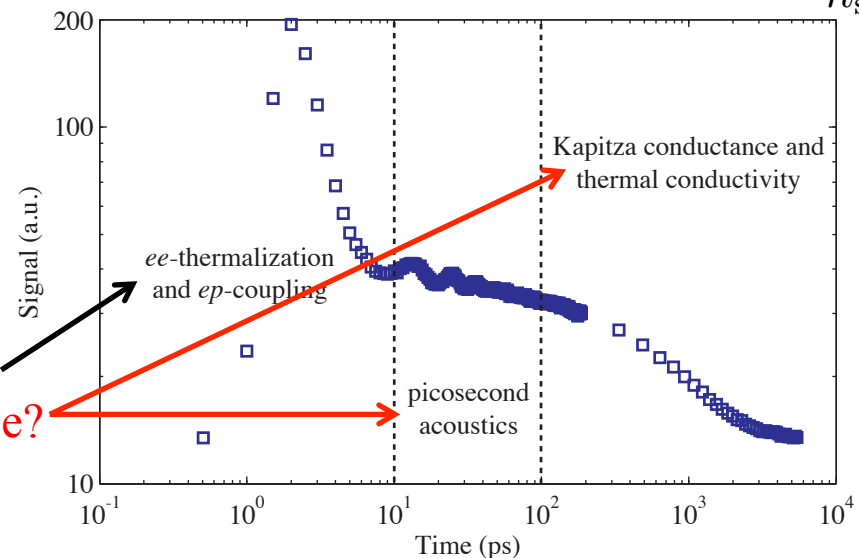
Why start here?

- Small perturbations/low fluences to simplify experiments
 - Pulse accumulation and heat transfer



Accumulation = “DC” heating $\Delta T_{DC} \propto \frac{1}{\kappa_{\text{substrate}}}$

How is this related to these?



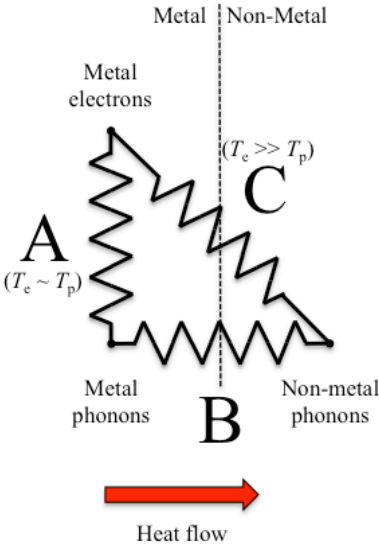
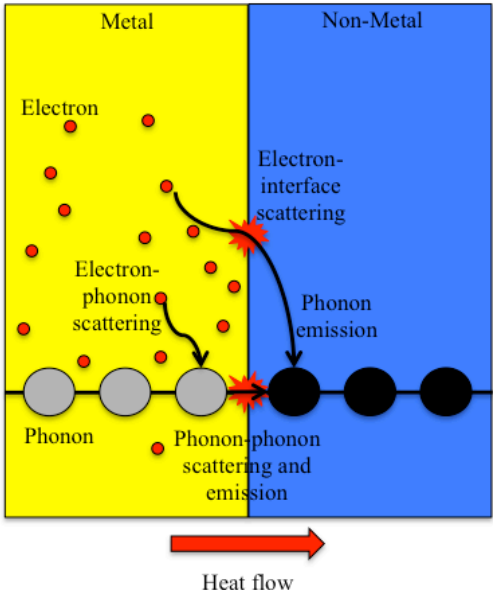
Electron-phonon coupling factor and interface conductance

Thermal quantities that we consider

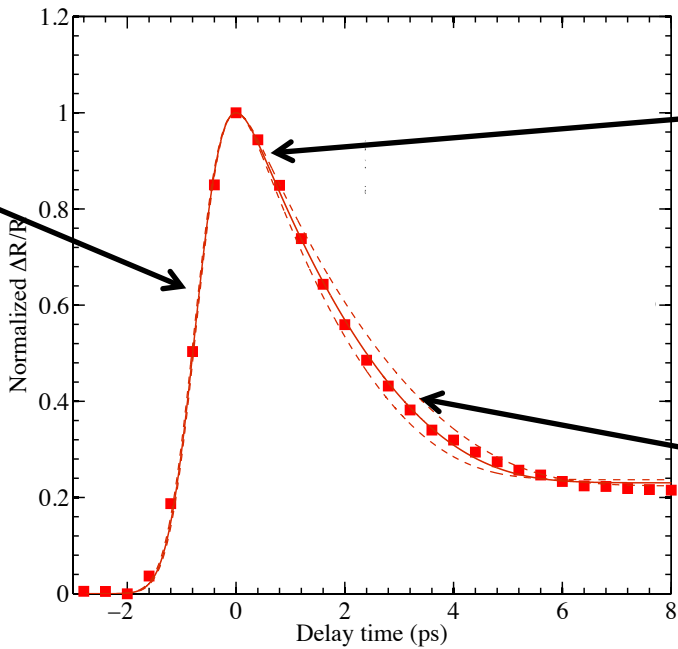
G – Electron-phonon coupling factor
(W/m³/K)

h – Thermal boundary conductance
(W/m²/K)

Quantify with two temperature model
and heat equation



Pulse excitation and
extended electron
thermalization



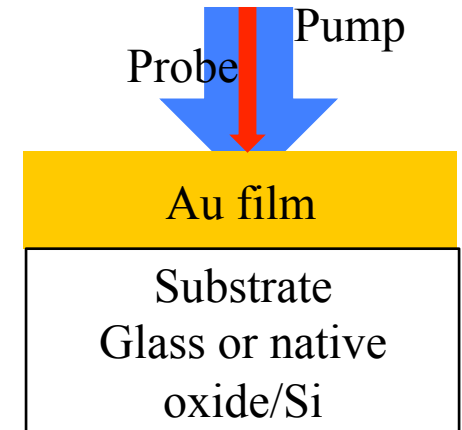
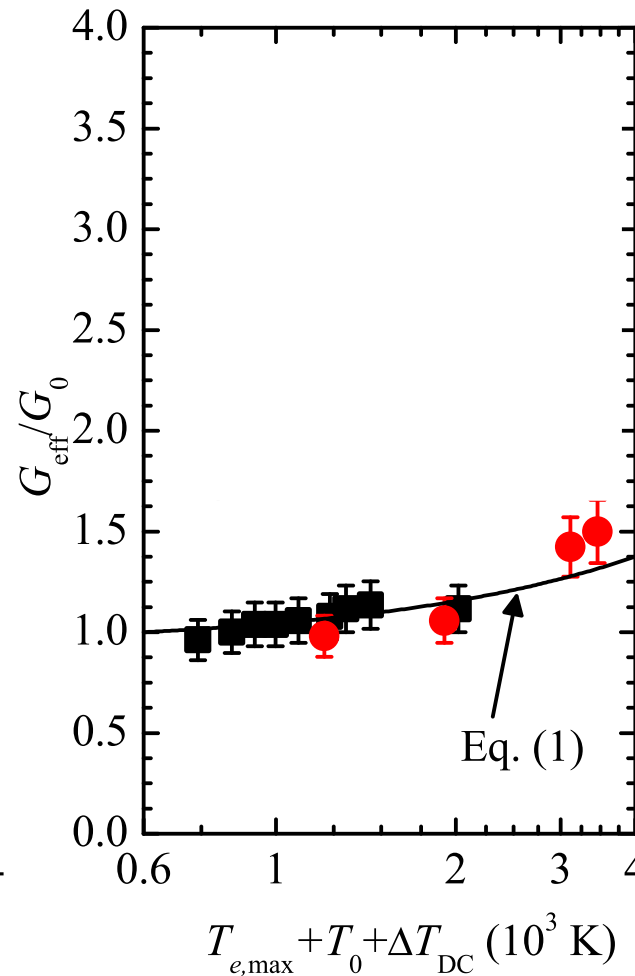
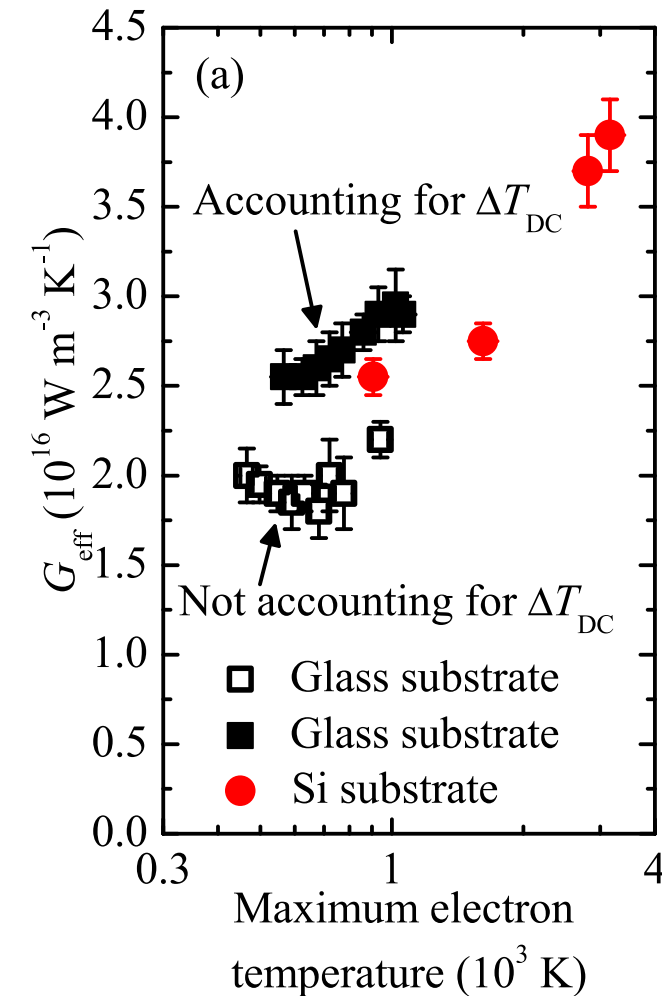
Competing non-thermal/
thermal effects

“Hot” electron transport and
relaxation with lattice and
interface (?)

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- 6) Looking ahead to Year 2**

“DC” heating from substrate “heat sink” – 20 nm Au films



Take-aways

- Substrate matters due to “DC” heating
- Electron relaxation affects EP heat transfer
- Limitation in fluence for Au/glass due to damage (more to come...)

$$\text{Eq. 1 : } G_{eff} = G_0 (A_{\text{electron-electron}} (T_e + T_p) + B_{\text{electron-phonon}})$$

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Electron-electron scattering effects on G – gold films

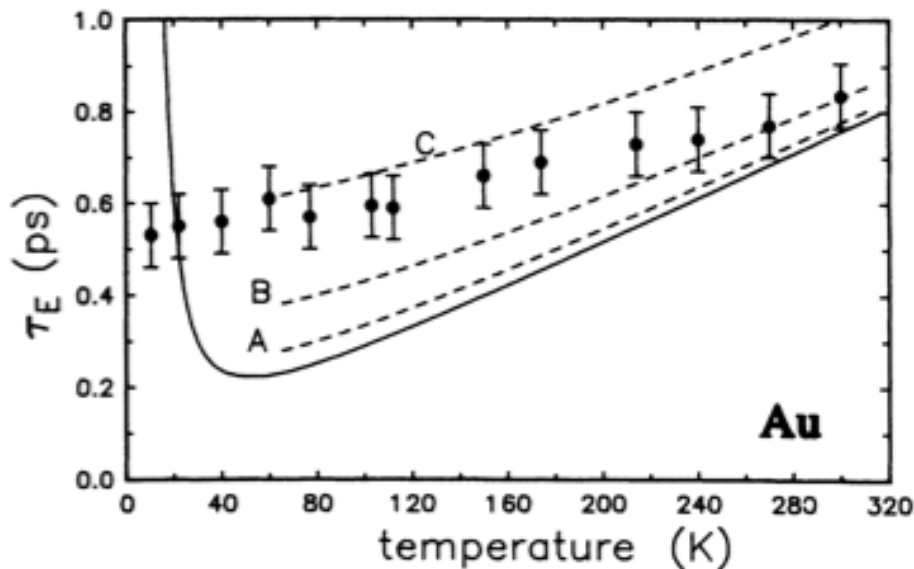
Can electron-electron scattering explain 20 years of discrepancies in reported values of G ?

PHYSICAL REVIEW B VOLUME 51, NUMBER 17 1 MAY 1995-I

Femtosecond spectroscopy of electron-electron and electron-phonon energy relaxation in Ag and Au

Rogier H. M. Groeneveld* and Rudolf Sprik
*Van der Waals-Zeeman Laboratorium der Universiteit van Amsterdam,
Valckenierstraat 65-67, 1018 XE Amsterdam, The Netherlands*

Ad Lagendijk
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and Fundamenteel Onderzoek der Materie-Instituut voor Atoom-en Molecuulfysica,
Kruislaan 407, 1098 SJ Amsterdam, The Netherlands*
(Received 12 October 1994)

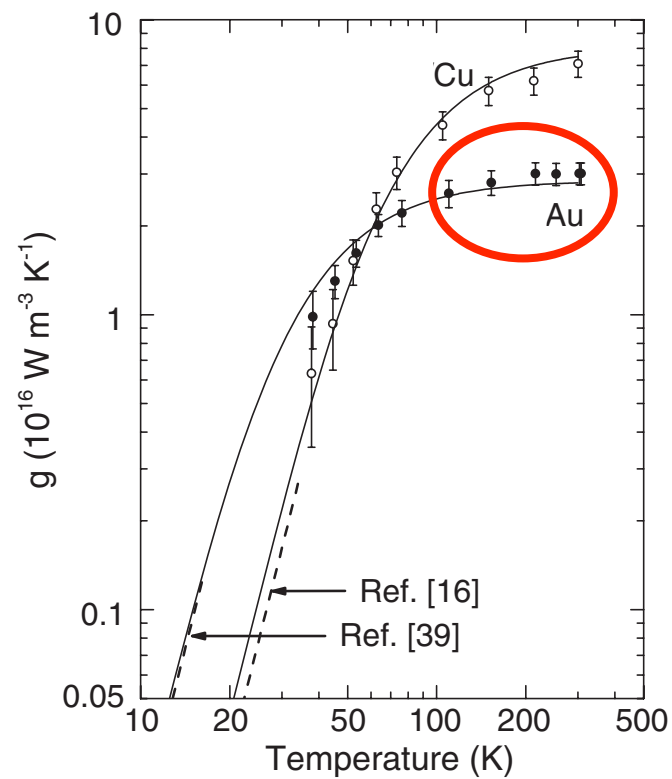


Weak perturbation = linear change in relaxation time (correlates to linear change in G)

PRL 109, 175503 (2012) PHYSICAL REVIEW LETTERS week ending 26 OCTOBER 2012

Limits to Thermal Transport in Nanoscale Metal Bilayers due to Weak Electron-Phonon Coupling in Au and Cu

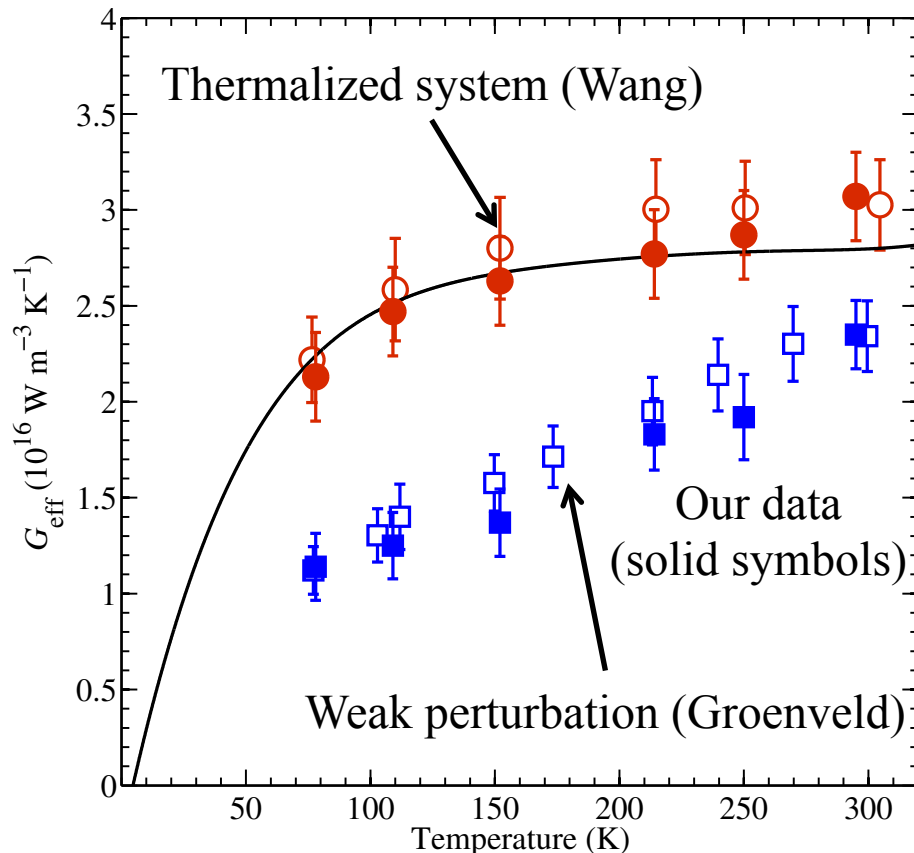
Wei Wang (汪维)* and David G. Cahill



However, G should be constant with T above Debye temperature

Electron-electron scattering effects on G – 20 nm Au film

Insight from varying T_p and T_e outside of “linear response” regime



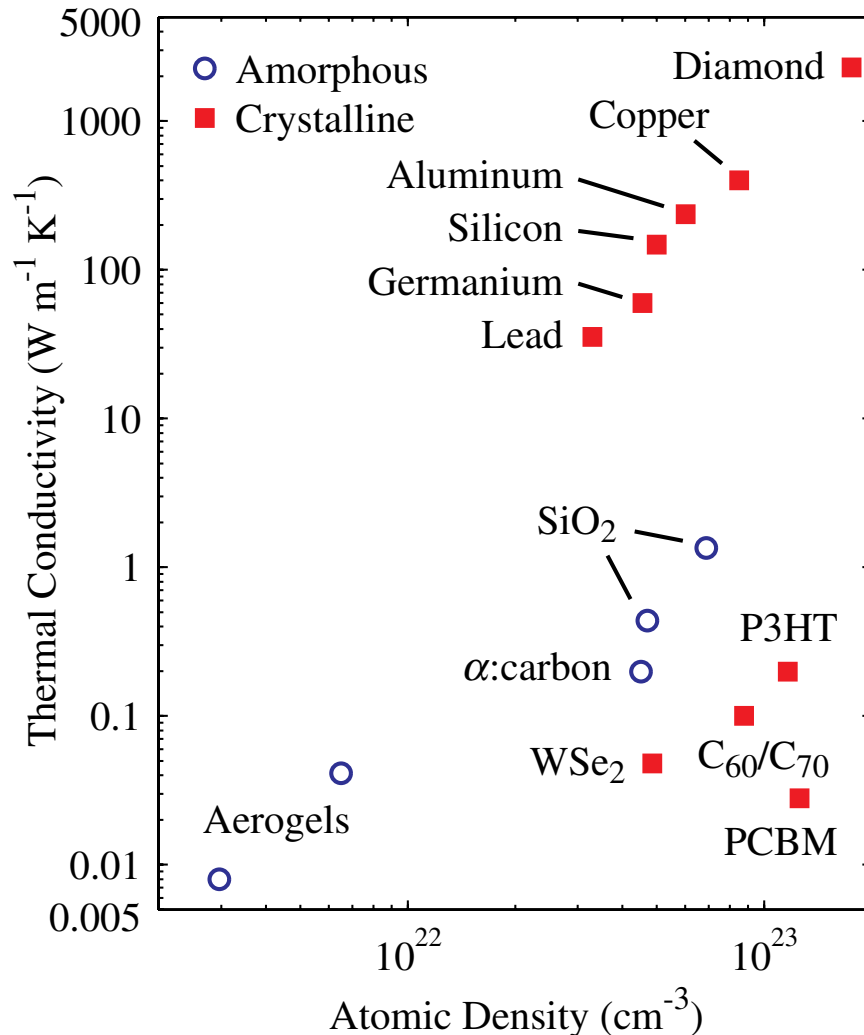
Take-aways

- After sub-picosecond short pulsed heating, gold electrons are NOT fully thermalized
- Agreement between our data and thermalized systems is coincidental
- Electron-electron scattering (“Fermi” relaxation) governs electron-lattice heat transfer

So does the “degree” of nonequilibrium drive G ?
Yes, which means G IS NOT A MATERIAL CONSTANT,
but $G = G(n(\varepsilon), T_p)$ even in free electron regime

What about diffusion effects in a thicker Au film?

Setting the stage: Thermal conductivity of solids



- Thermal conductivity always reported when electron temperature is same as phonon temperature (room temperature shown at left)

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

- What about the regime when electrons are at different “temperature” than phonons?

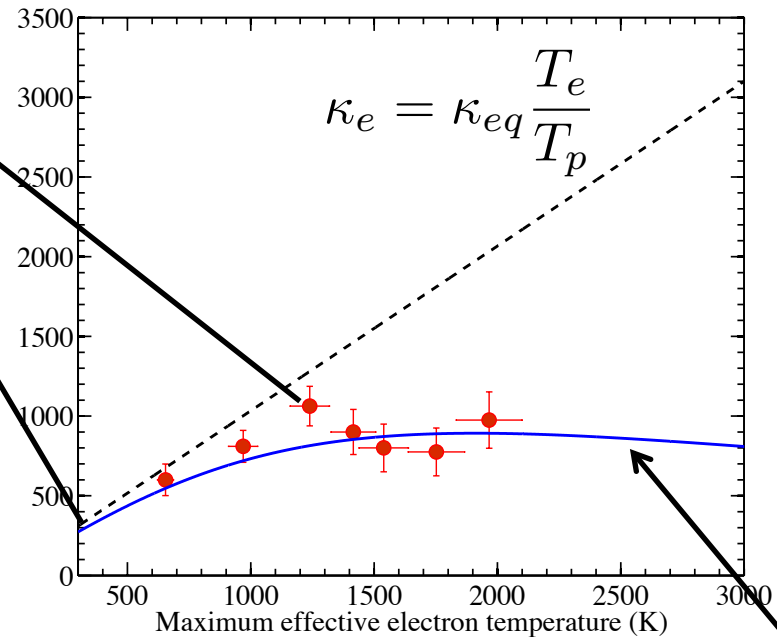
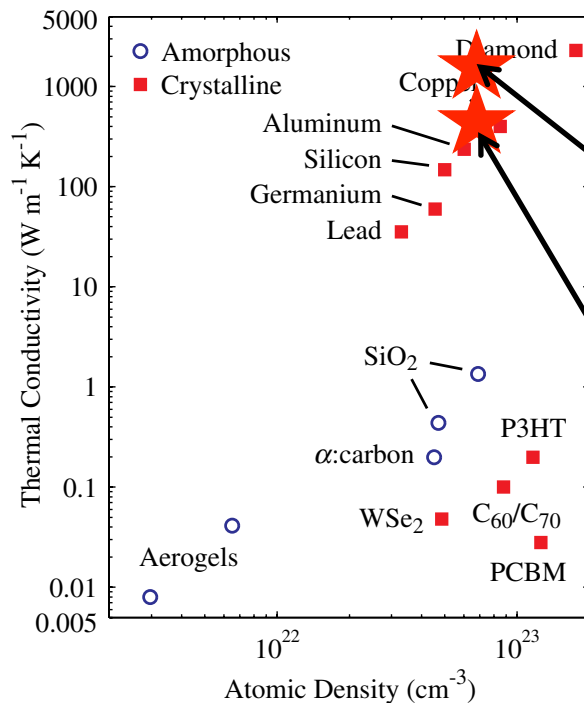
$$\frac{1}{\tau} = \frac{1}{\tau_{e-e}(T_e)} + \frac{1}{\tau_{e-p}(T_p)}$$

What about diffusion effects in a thicker Au film?

We know $G = G(n(\epsilon), T_p)$ from 20 nm film measurements, so for thicker films only unknown is diffusion of energy

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

$$\frac{1}{\tau} = \frac{1}{\tau_{e-e}(T_e)} + \frac{1}{\tau_{e-p}(T_p)}$$



First experimental validation of theory that $\kappa = \kappa(n(\epsilon), T_p)$

$$\kappa_e = \frac{1}{3} v_F \frac{\gamma T_e}{A T_e^2 + B T_p}$$

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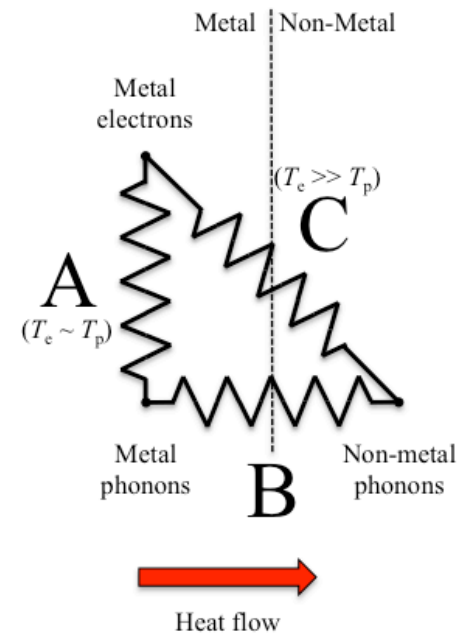
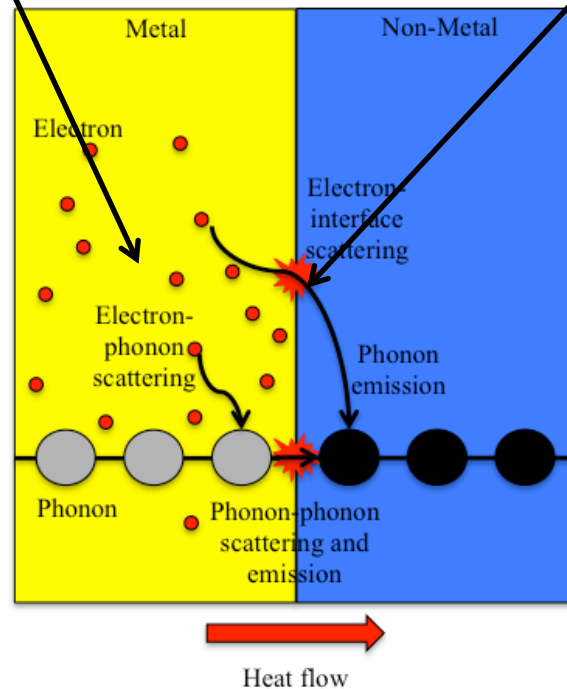
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What about conductance across metal/nonmetal interface?

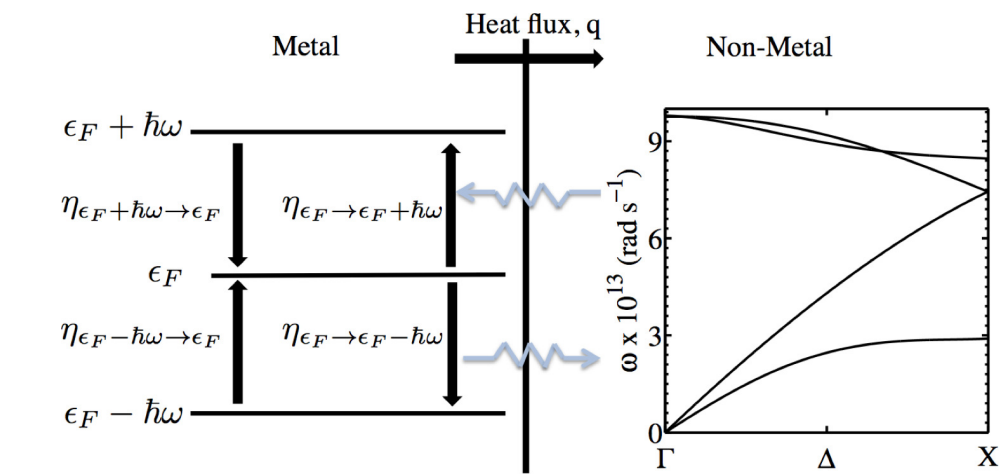
This is the ultimate goal....

For Au, we now have a good handle of this in the low power regime

When is this important at metal/nonmetal interfaces?

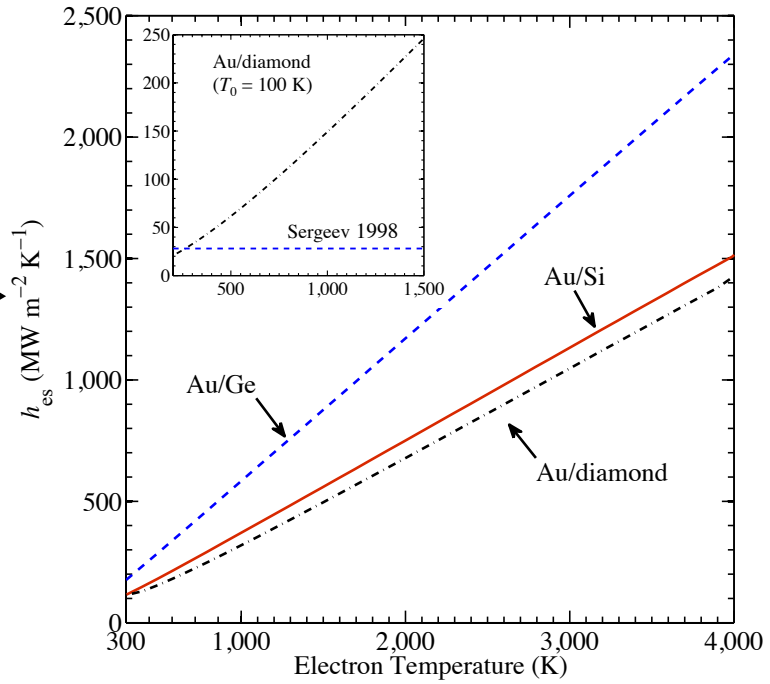
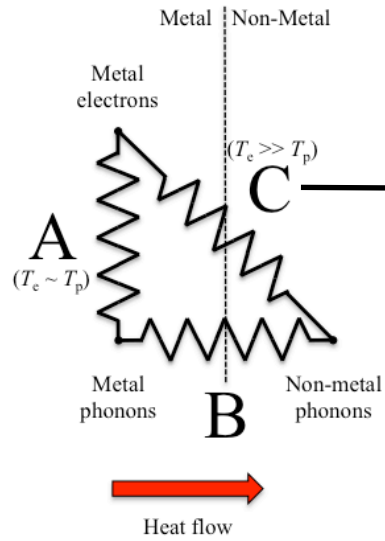
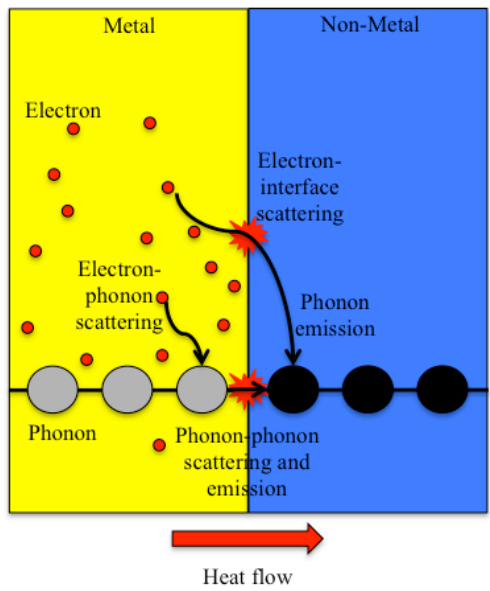


Quantum mechanical/semi-classical model



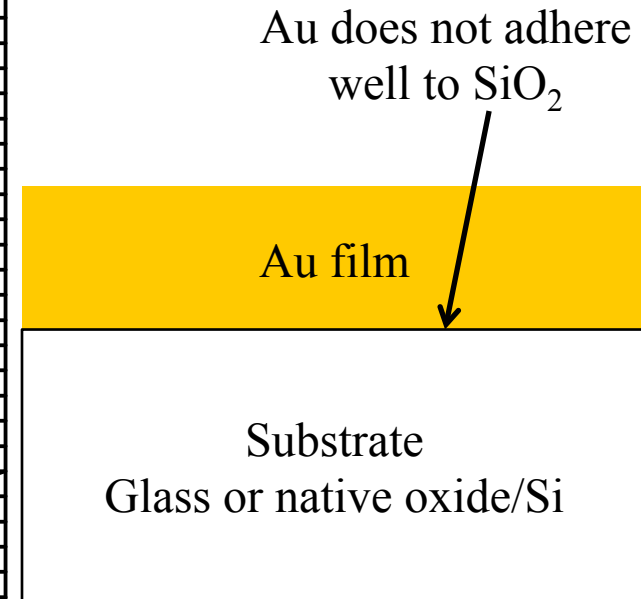
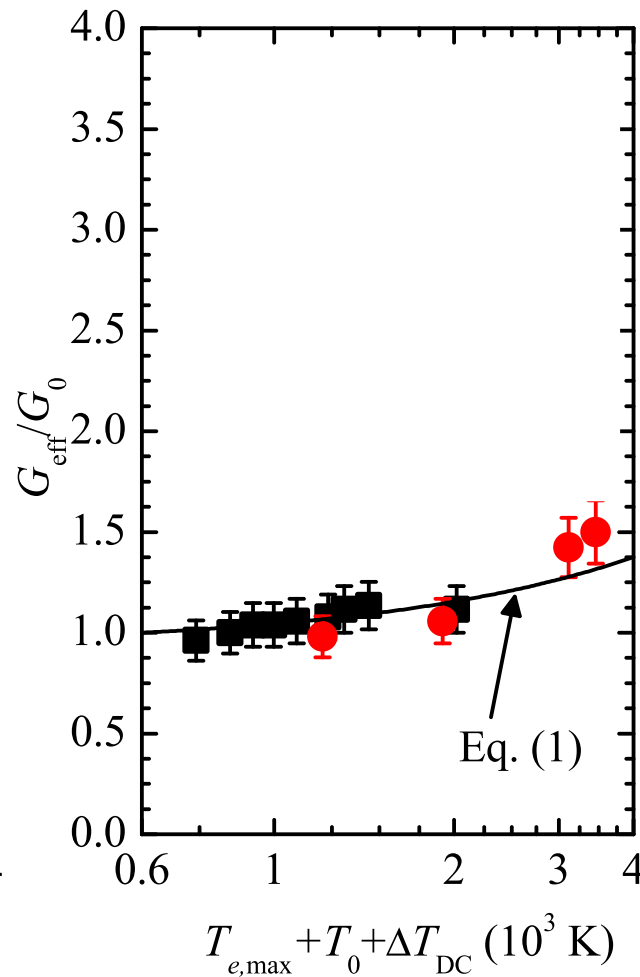
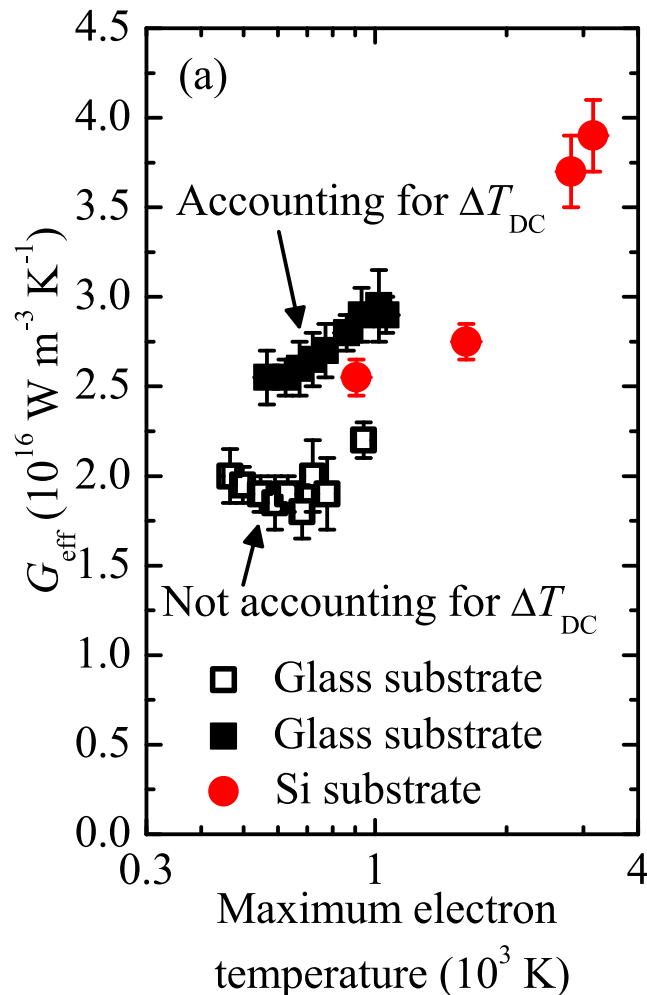
Giri *et al.* to appear in *J. Heat Trans.*

From our model, electron-interface scattering SHOULD be a possible thermal pathway



Au/substrate electron-interface thermal conductance

So why didn't we see it????

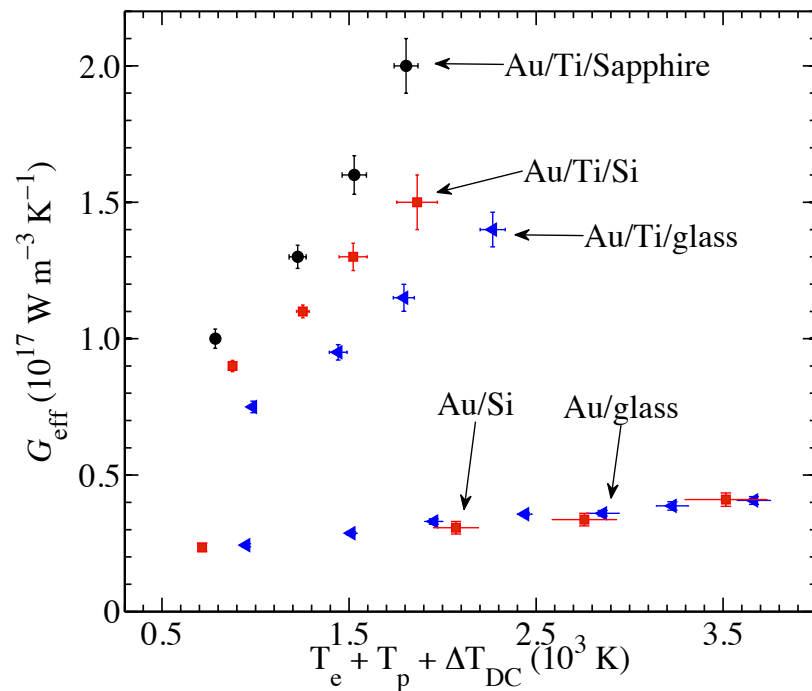


Weak bonding leads to larger interface resistance (small h)

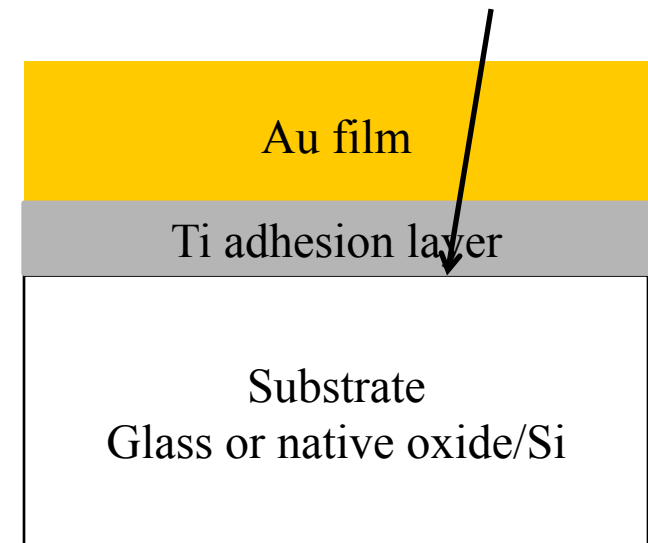
Hopkins *et al. Nano Lett.*
12, 590 (2012)

Au/substrate electron-interface thermal conductance

It comes down to that atomic bond....

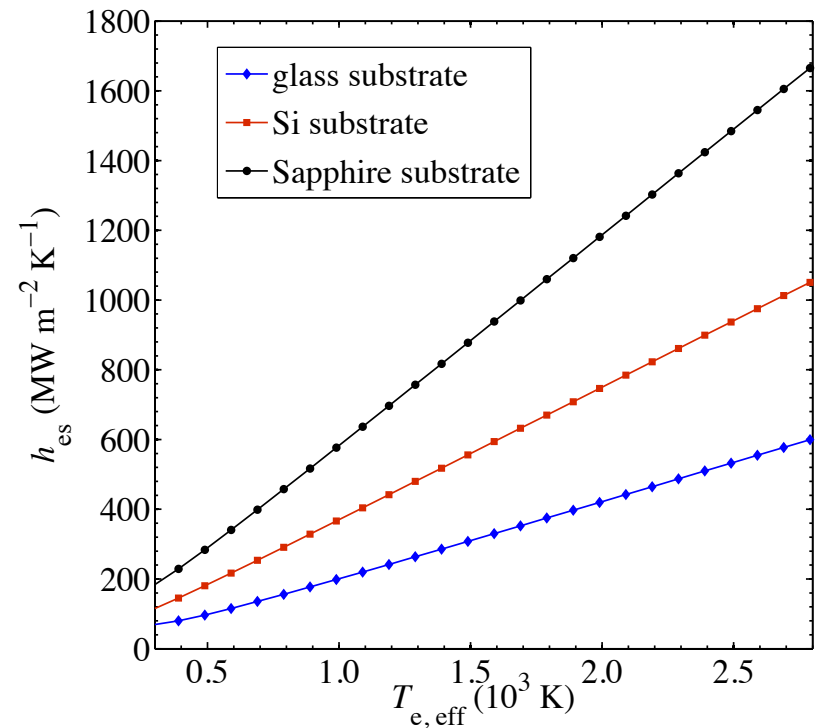
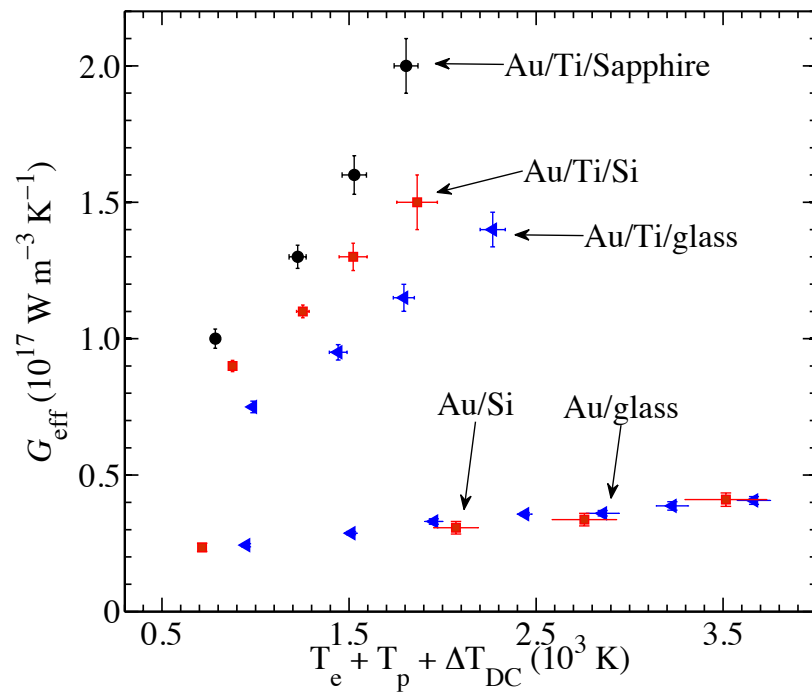


Now it does!



Au/substrate electron-interface thermal conductance

Qualitative agreement with quantum/semi-classical model
Working on more direct quantitative comparison....



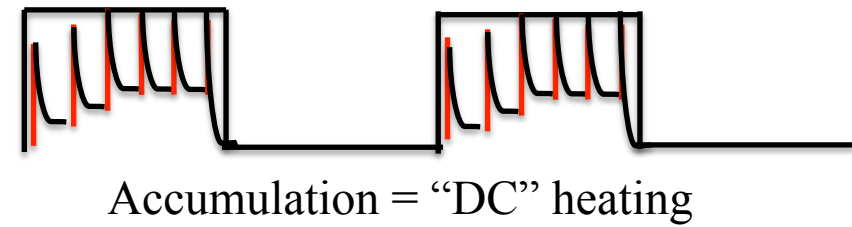
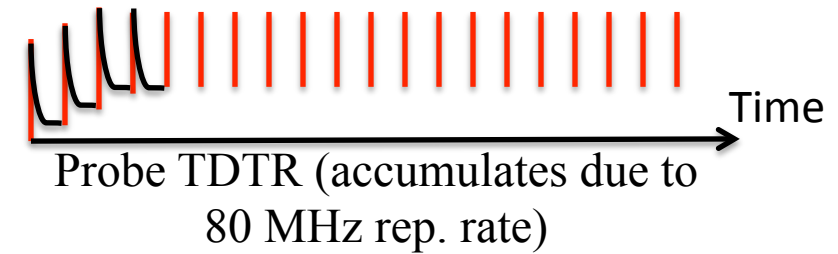
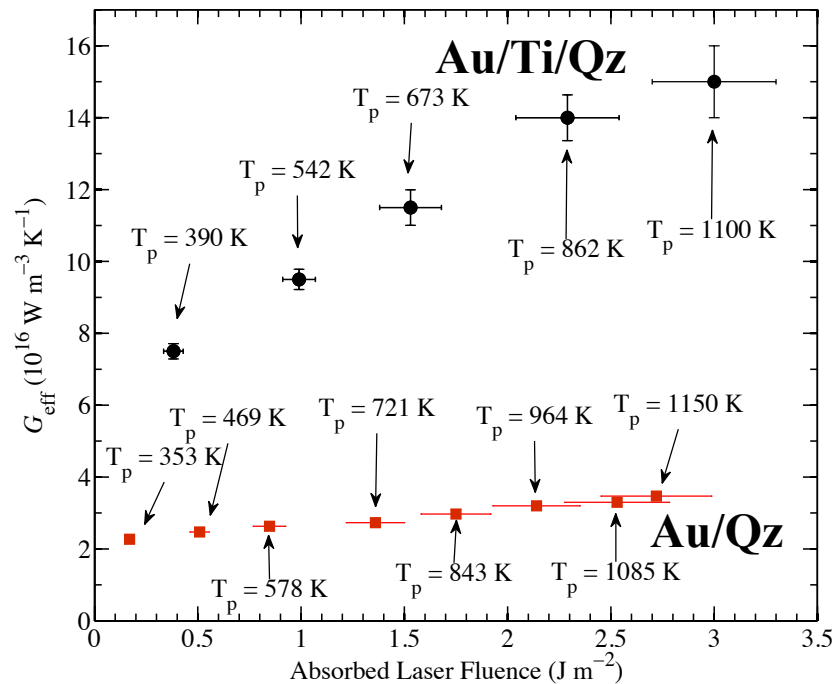
What effect does this have on damage from pulse accumulation?

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The physics sets the stage.....



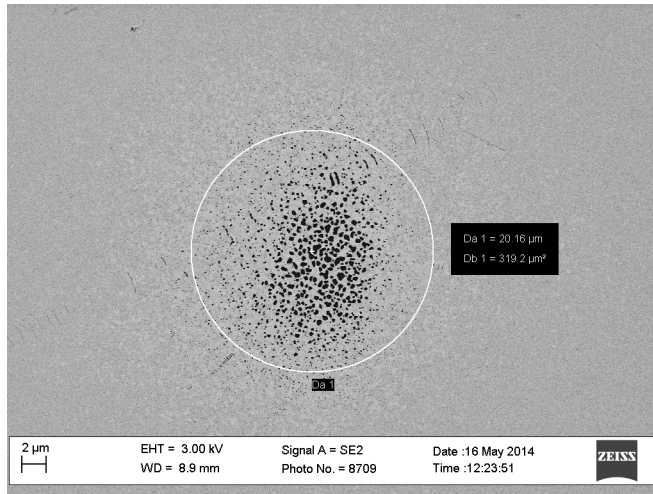
$$\Delta T_{\text{DC}} \propto \frac{1}{\kappa_{\text{substrate}}}$$

Take-aways

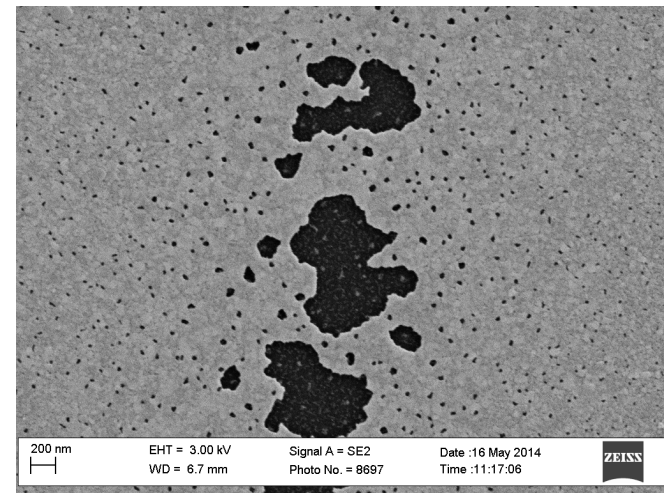
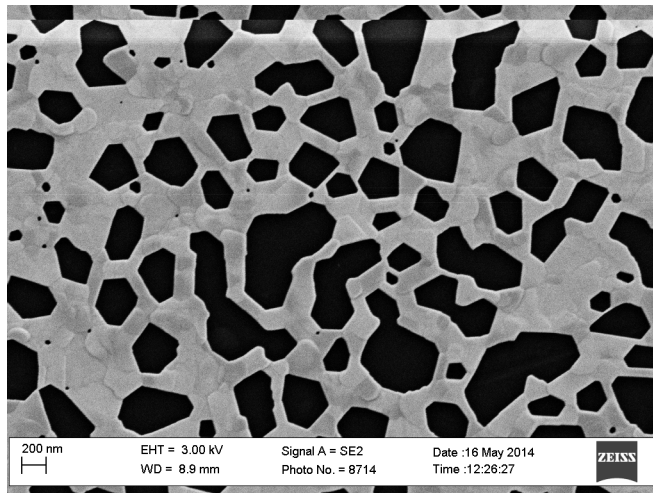
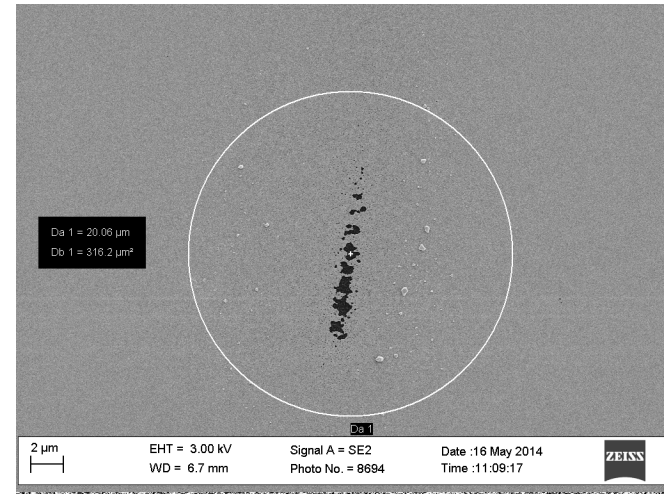
- Au damage at low fluence on Qz substrate (but not on Al_2O_3 or Si substrates)
- Au/Ti/Qz and Au/Qz damage at same fluence
- Indicative of "melting" or "diffusive" damage (NOT high energy short pulse damage)

But the damage is different – only difference is ~ 2 nm Ti

Au/quartz

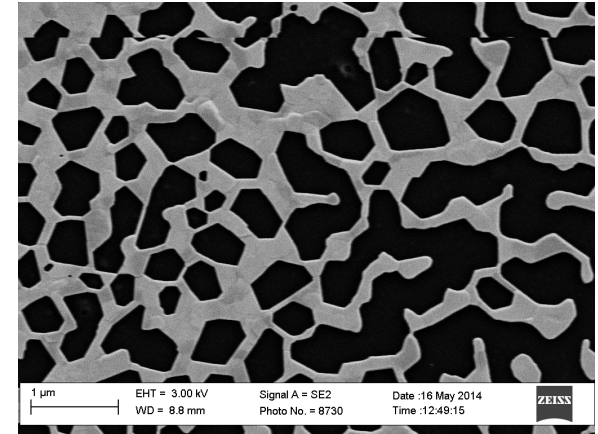
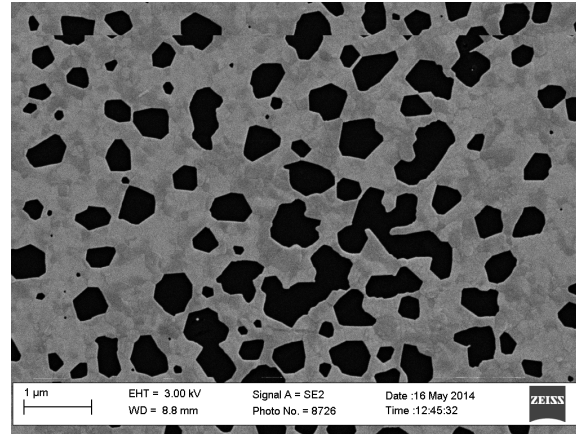
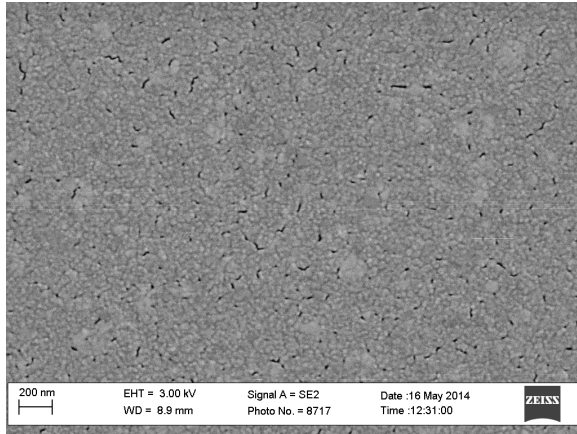


Au/Ti/quartz



Au/quartz damage – damage coarsening at higher fluence

Au/quartz



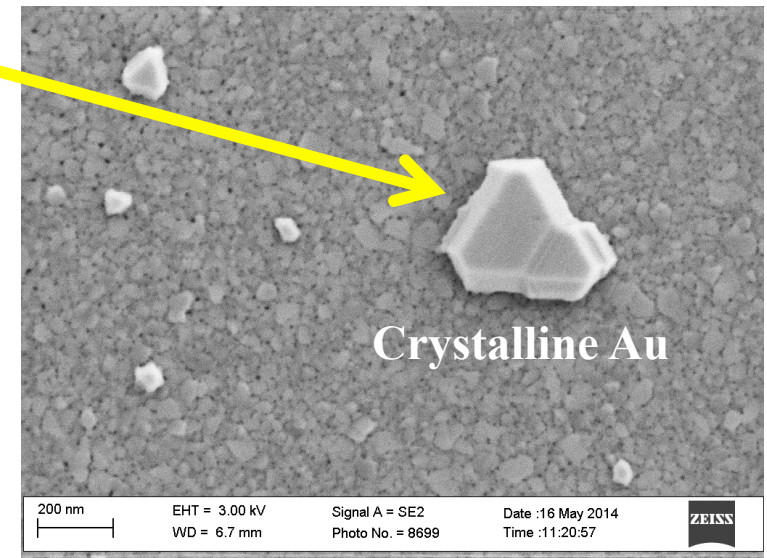
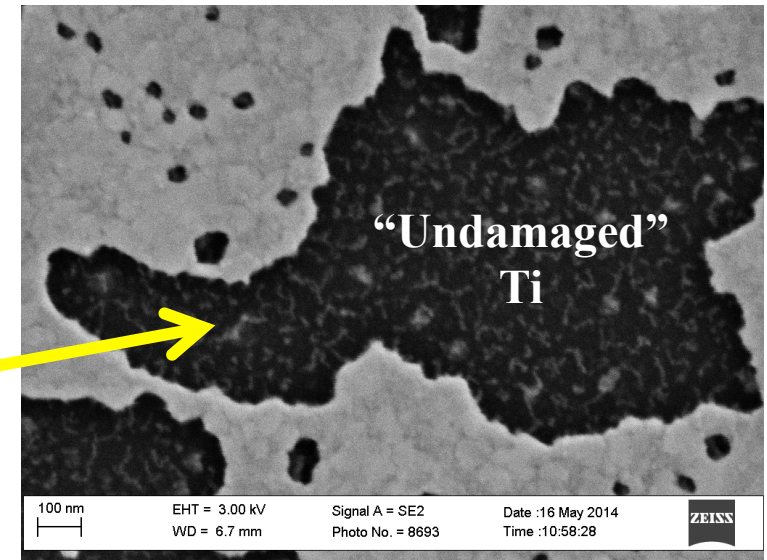
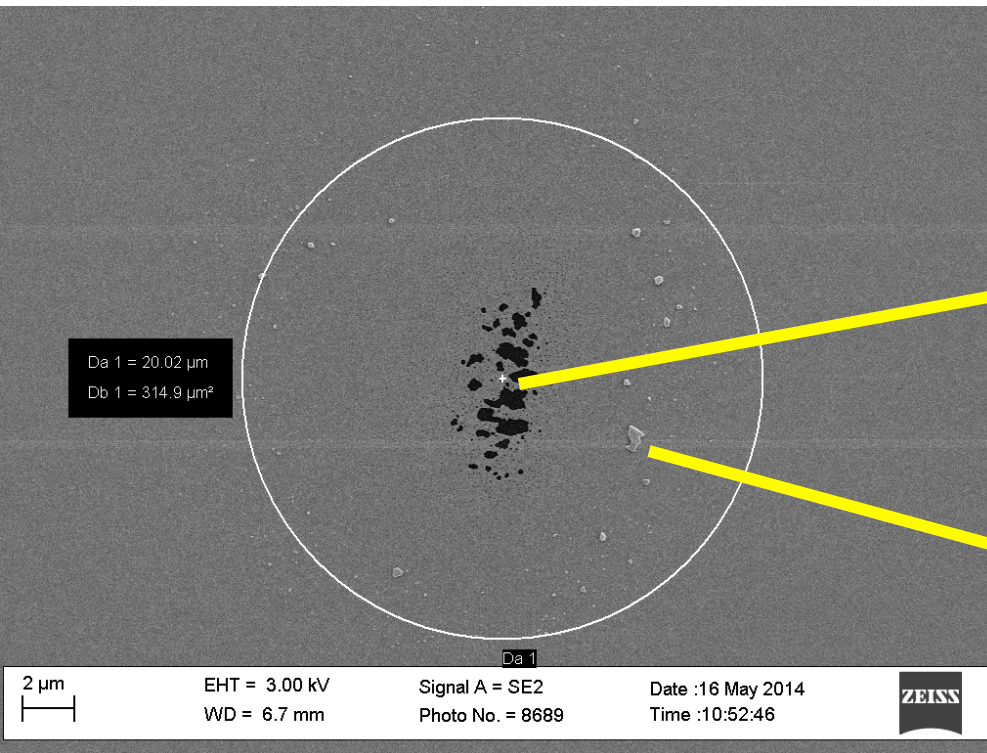
Increased fluence



- **Damage appears that it could be originating at grains/defects (not surprising)**
- **High fluence = higher temperature = larger damage (grain coarsening)**
- **Mechanisms related to homogeneous heating of Au film**

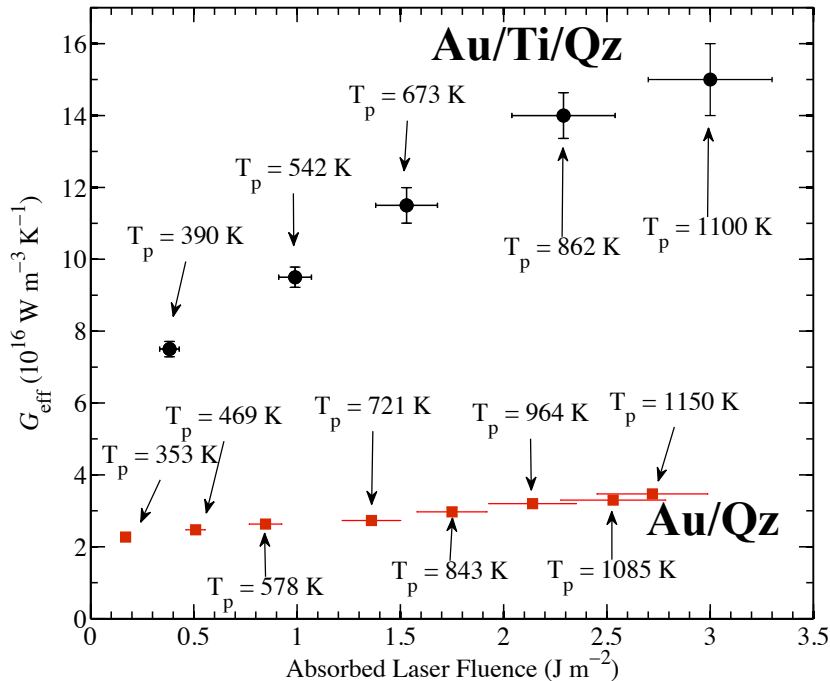
Au/Ti/quartz damage – interface heating and Au ejection

Au/Ti/quartz



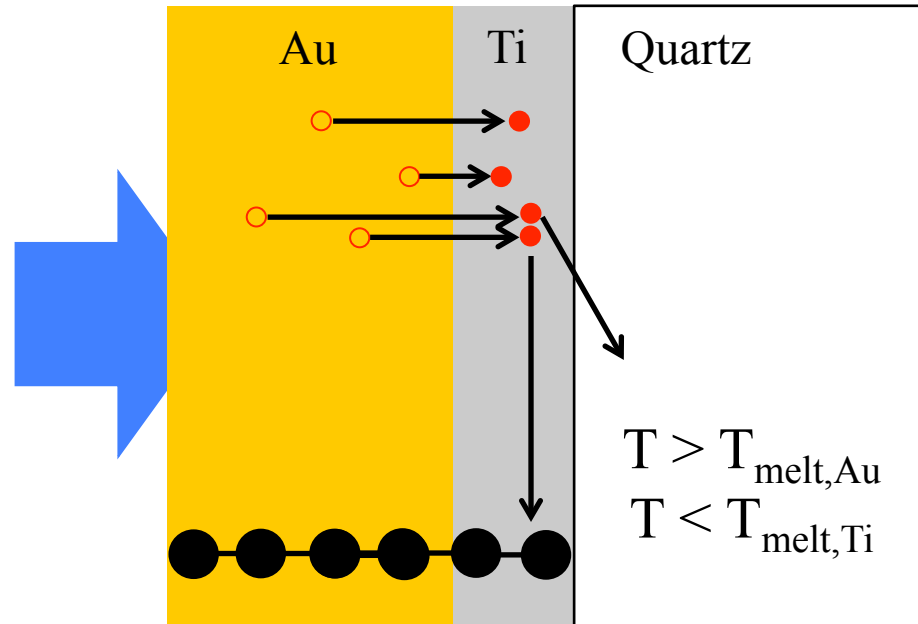
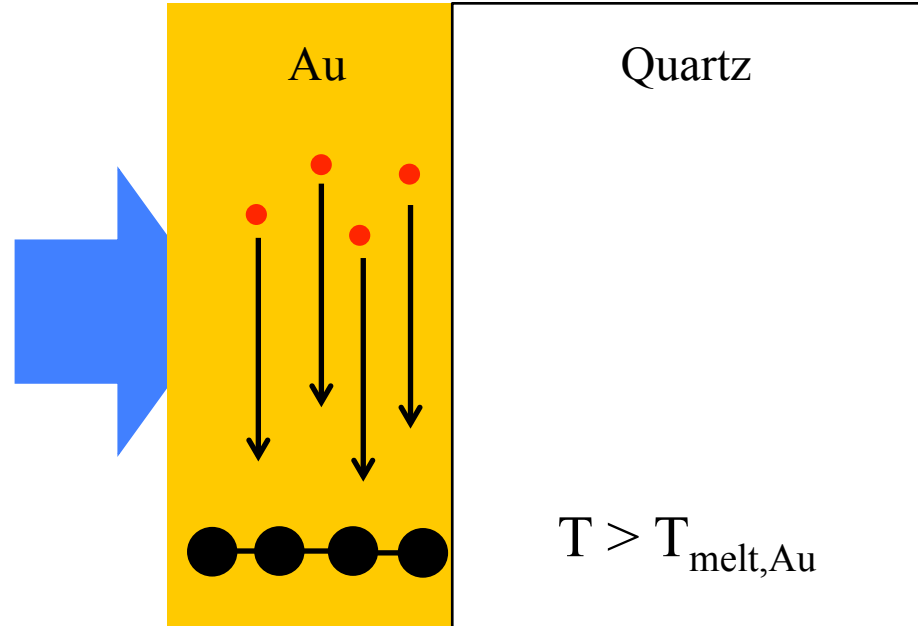
- Does this make sense? Let's go back to our thermal measurements

EP coupling and energy deposition during damage

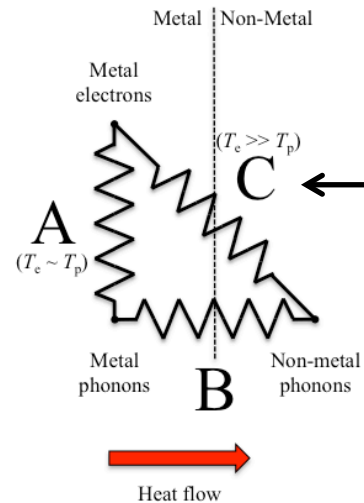
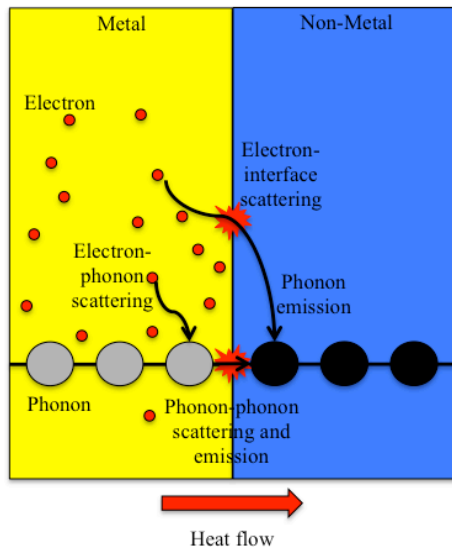


Take-aways

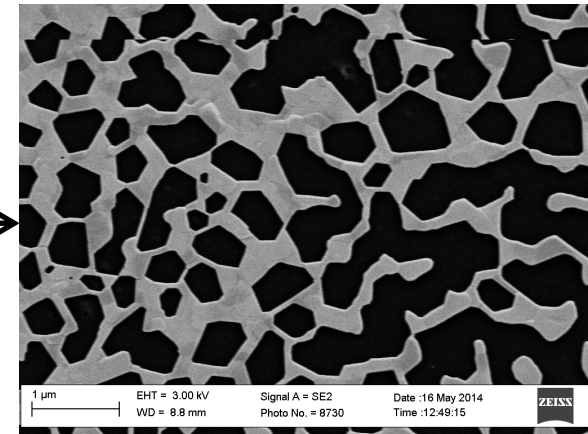
- Electron energy deposited at Ti/interfacial region
- Indirect heating of Au through Ti
- Causes Au damage from the “back” interface
- Different from Au/Quartz
- Perhaps strain mediated in Ti layer? (more work needed...)



Conceptual “take-away”



**This
affects this**



Dissemination of results and breakdown of support – Year 1

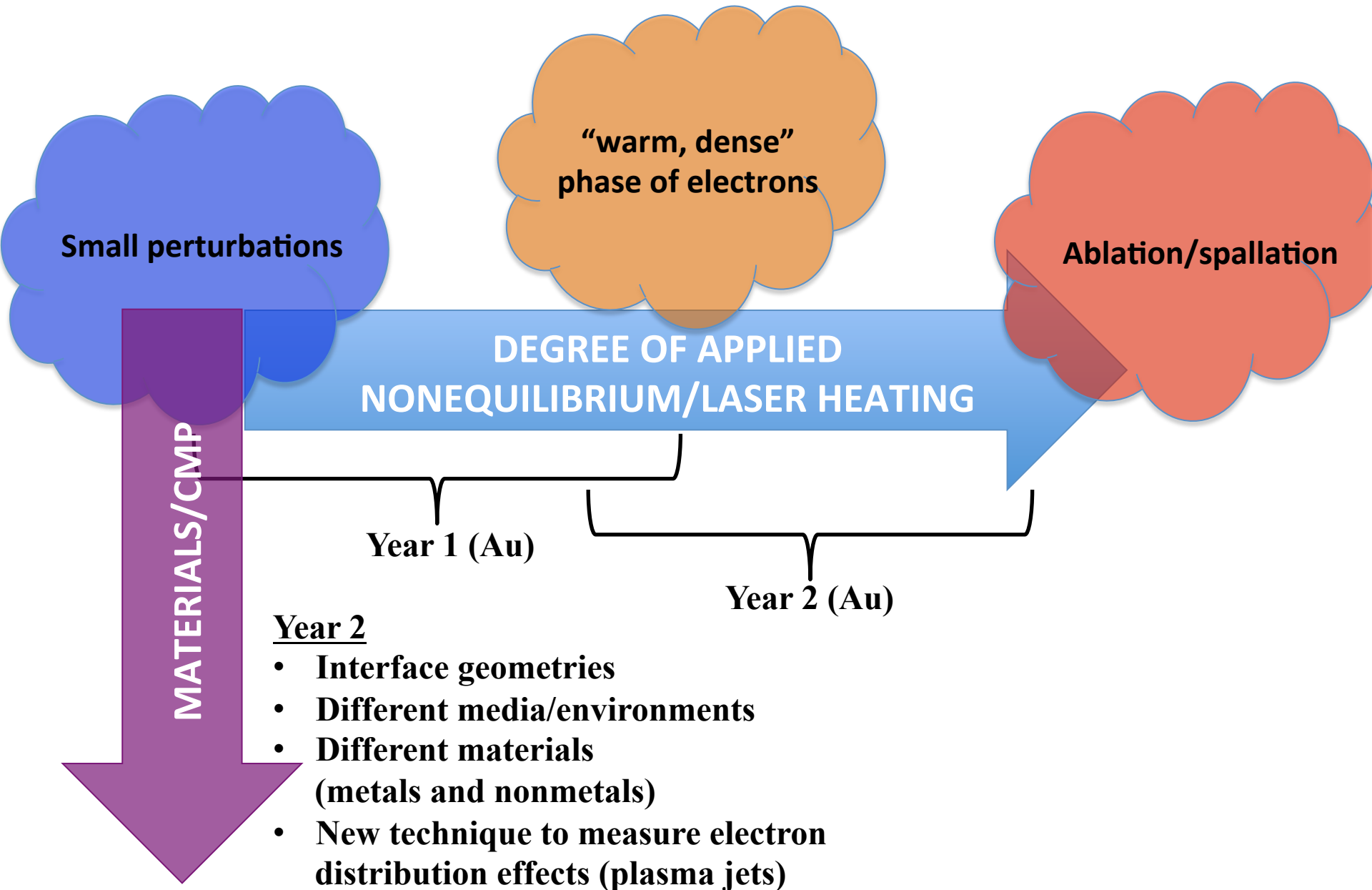
- **Supported 1 graduate student (Ashutosh Giri)** →
- **Will support 1.5 – 2 graduate students in Year 2**
- **Published 5 refereed journal papers**
- **2 additional papers currently under review**
- **1 invention disclosed and applied for patent**
“Single element Raman Thermometry”
- **Presented AFOSR YIP supported work in 11 contributed conference talks and 2 invited talks/seminars**



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Looking ahead



Conclusions, questions and thank you AFOSR YIP

