SAND2009-3822C

Thermal processes in metal nanosystems after short pulsed laser heating

CINT/LANL Seminar

Thursday, June 18, 2009

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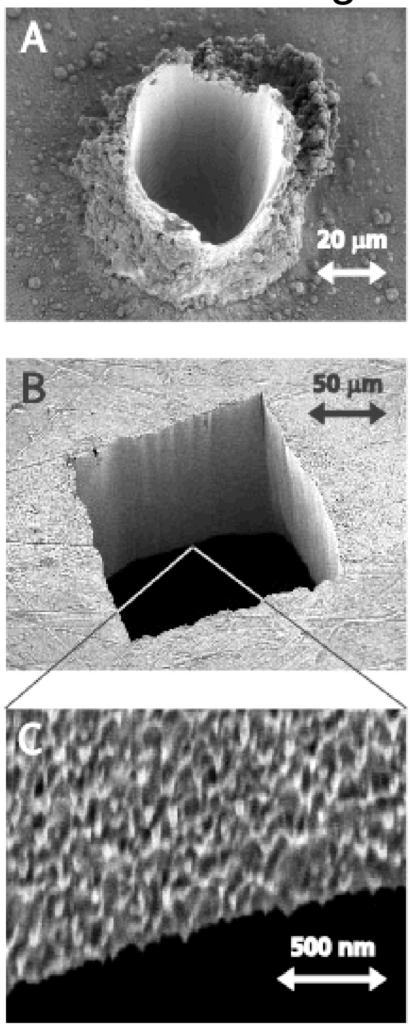
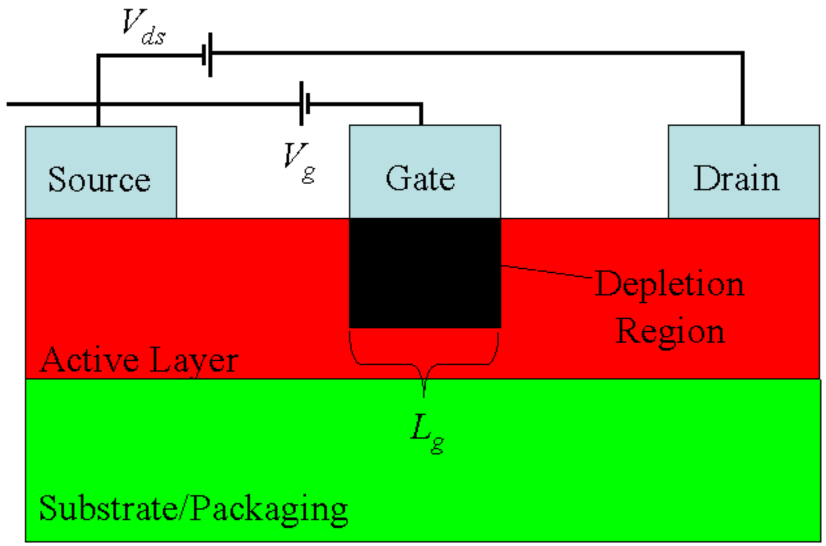
under contract DE-AC04-94AL85000.

Thermal processes in metals

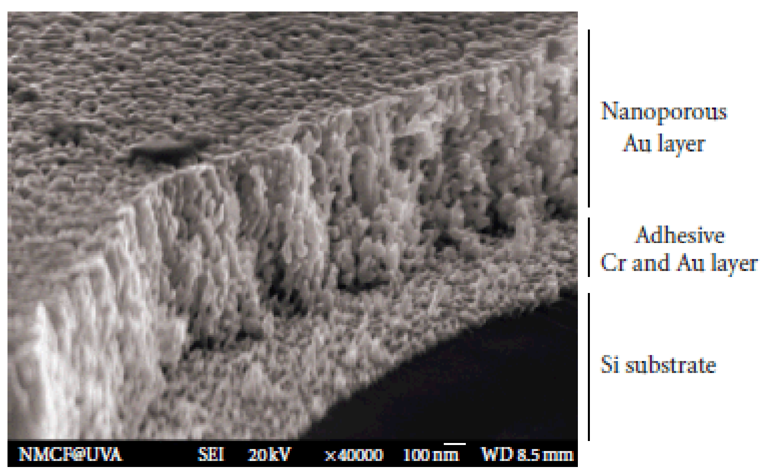
Laser machining and manufacturing



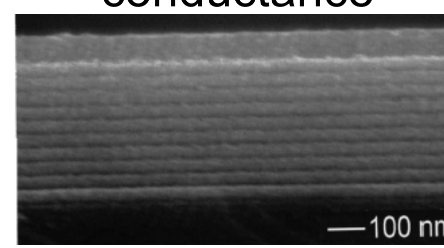


Heat generated at metal contacts in transistors

Nanoporous metals



Multilayered systems – understanding interface conductance



$$ZT = \frac{S^2 \sigma T}{k}$$

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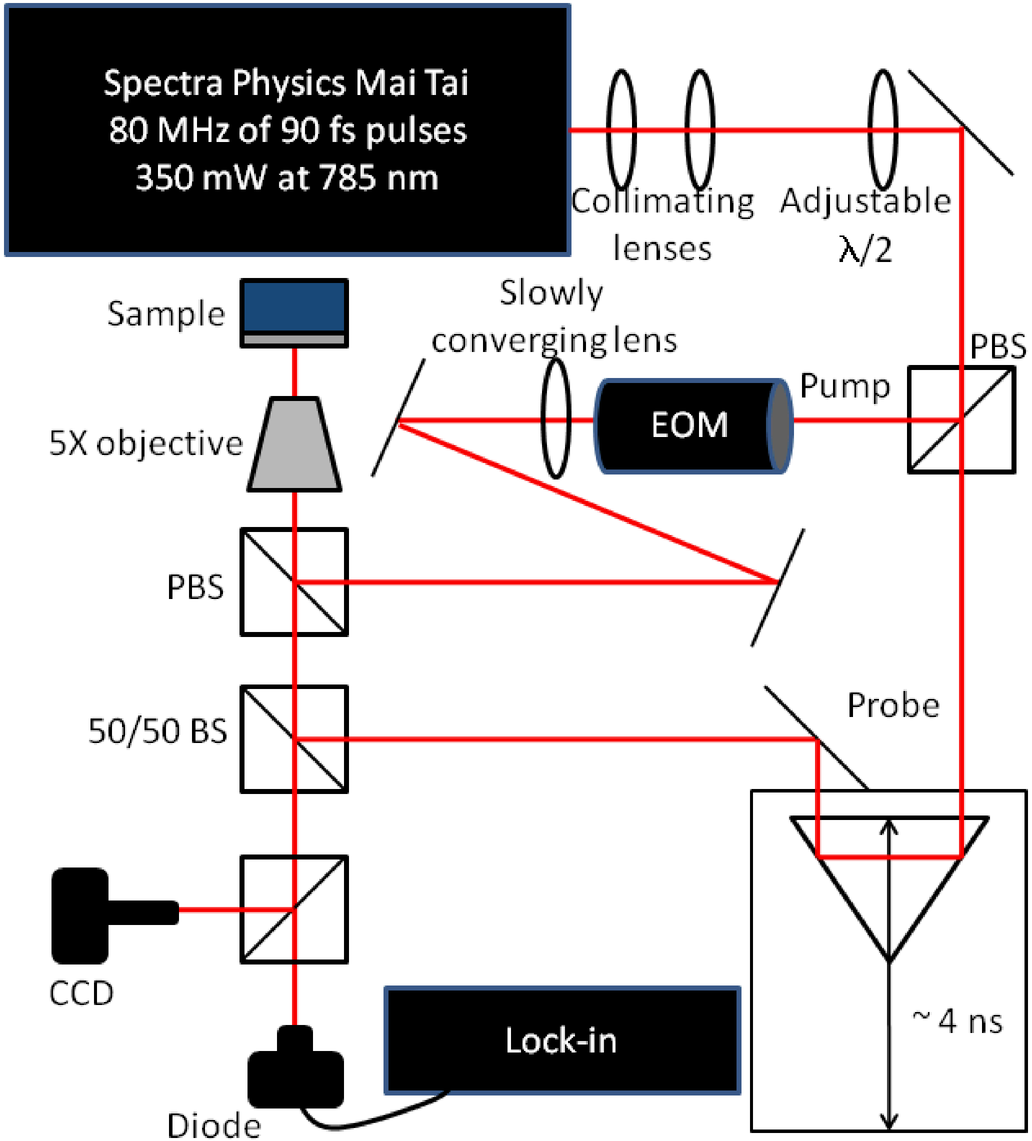
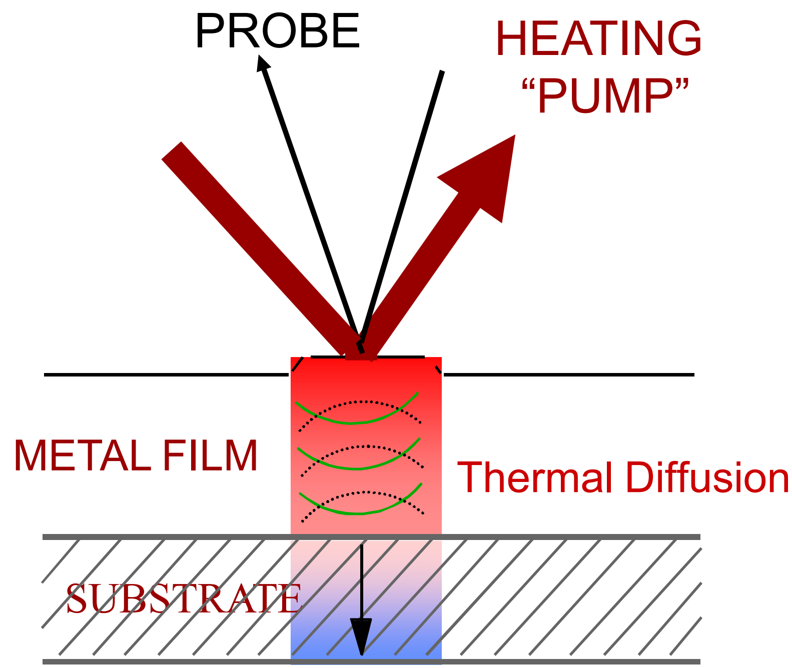
Outline

- The transient thermoreflectance technique (TTR)
  - Thermal conductivity measurements of metalized layers
  - Electron-phonon coupling factor measurements in metal films
  - Effects of interface scattering
- Electron scattering in nanoporous metals
- Electron-phonon coupling enhancement due to d-band excitations
  - Ballistic transport
  - Interface scattering

TTR technique





Thermal processes measured with the TTR technique

Thermal processes in metal films

Free Electrons Absorb Laser Radiation

↓

Electrons Transfer Energy to the Lattice | **Thermal Diffusion by Hot Electrons** → **Electron-Phonon Coupling (~2 ps)**

↓

Thermal Equilibrium

↓

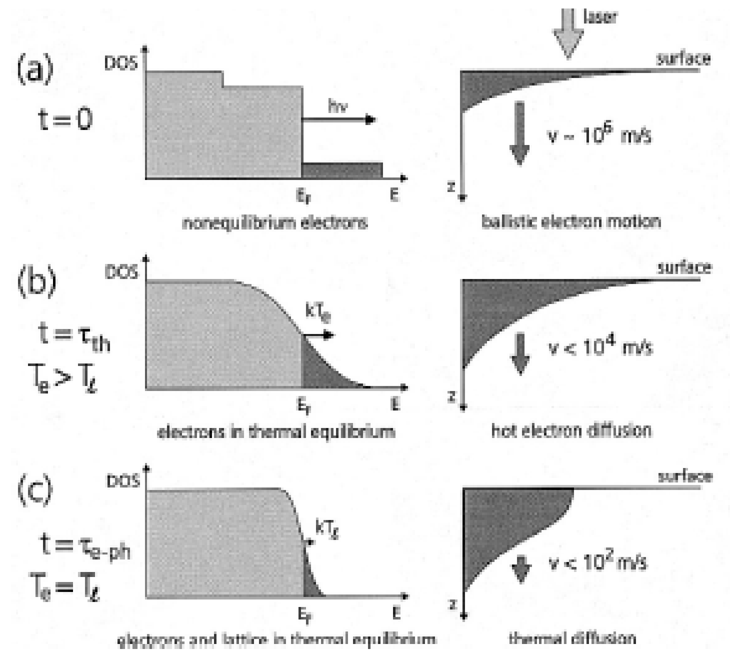
Thermal Diffusion within Thin Film → **Thin film thermal conductivity (~100 ps)**

↓

Thermal Conductance across the Film/Substrate Interface → **Thermal Boundary (~2 ns) Conductance**

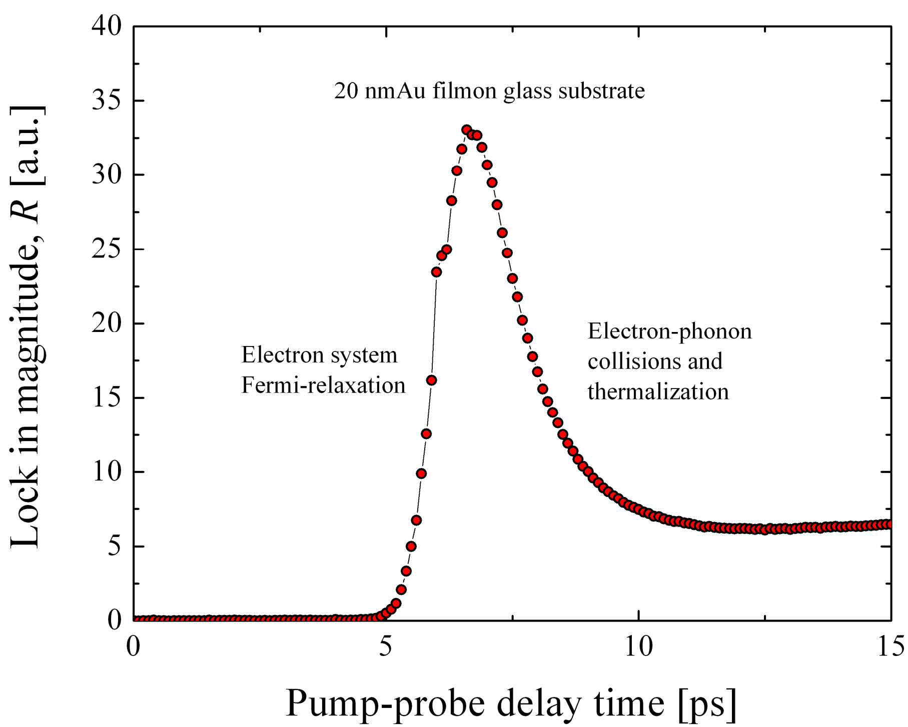
↓

Thermal Diffusion within Substrate → **Substrate Thermal Diffusion (~100 ps – 100 ns)**

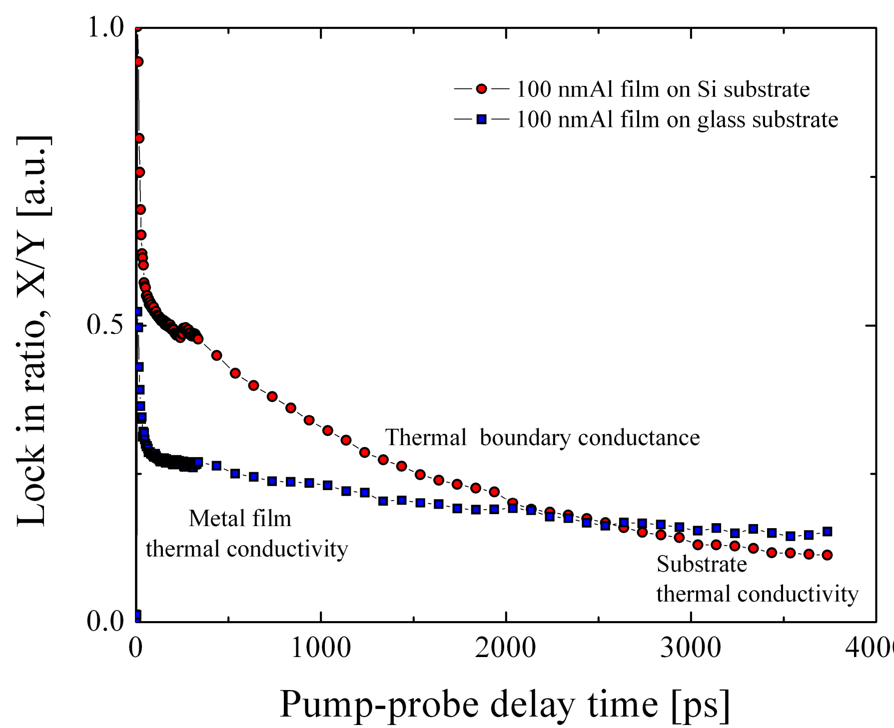


TTR data

Electron-phonon coupling



Thermal diffusion



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Determining the thermal conductivity, Λ

REVIEW OF SCIENTIFIC INSTRUMENTS VOLUME 75, NUMBER 12 DECEMBER 2004

Analysis of heat flow in layered structures for time-domain thermoreflectance

David G. Cahill[a)]
Department of Materials Science and Engineering and Frederick Seitz Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801

REVIEW OF SCIENTIFIC INSTRUMENTS 79, 114902 (2008)

Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance

Aaron J. Schmidt,[a)] Xiaoyuan Chen, and Gang Chen
Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139–4307, USA

Utilize lock-in signals to deduce thermal diffusion phenomena

$$Z(\omega_0) = \frac{(2\pi)^2 \chi}{\omega_s^2} \sum_{M=-\infty}^{\infty} \theta(\omega_0 + M\omega_s) \exp[iM\omega_s \tau]$$

Must know analytical form of surface temperature rise

$$\theta(\omega) = \frac{A}{2\pi} \int_0^{\infty} F(k) \exp\left[\frac{-k^2 (w_0^2 + w_1^2)}{8}\right] k\,dk$$

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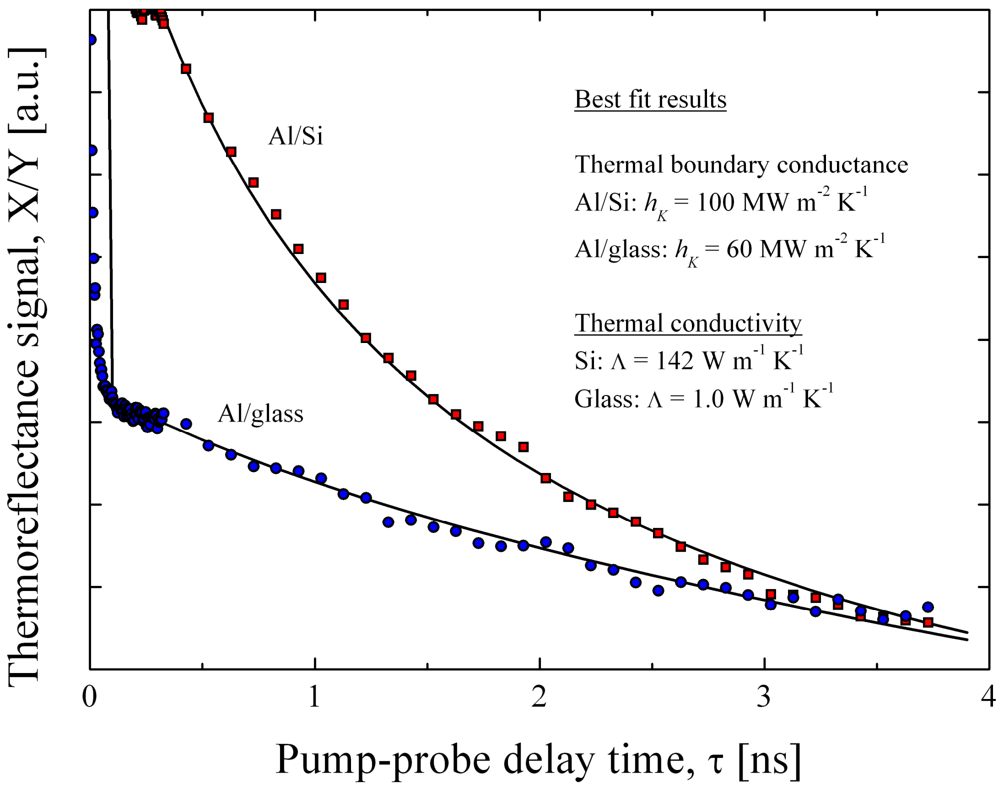
Determining the thermal conductivity, Λ



$$\frac{X}{Y} = \frac{\mathrm{Re}[Z(\omega_0)]}{\mathrm{Im}[Z(\omega_0)]}$$

Extended to measure thermal conductivity of multilayer films

IMECE2009-12238

EFFECTS OF THERMAL ANALYSIS ON THERMAL CONDUCTIVITY MEASUREMENTS IN PUMP-PROBE THERMOREFLECTANCE TECHNIQUES

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To be presented at ASME IMECE in November

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Determining the electron-phonon coupling factor, *G*

REVIEW OF SCIENTIFIC INSTRUMENTS 77, 084901 (2006)

Signal analysis and characterization of experimental setup for the transient thermoreflectance technique

Robert J. Stevens[a)]
Mechanical Engineering Department, Rochester Institute of Technology, Rochester, New York 14623

Andrew N. Smith
Department of Mechanical Engineering, United States Naval Academy, Annapolis, Maryland 21402

Pamela M. Norris
Department of Mechanical and Aerospace Engineering, University of Virginia, Charlottesville, Virginia 22904

$$R = a\Delta T \longrightarrow \Delta T$$

Lock in magnitude is proportional to temperature rise through a (thermoreflectance coefficient)

Temperature rise determined from thermal model

$$\phi$$

Pulse accumulation from lock in phase

Advantage: Do not need to know analytical form of ΔT

Disadvantage: *R* contains instrument noise where *X*/*Y* noise cancels out

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Determining the electron-phonon coupling factor, *G*

The two temperature model (TTM)

Energy stored in *e* system — Energy conducted through *e* system — Energy transferred from *e* system to *p* system

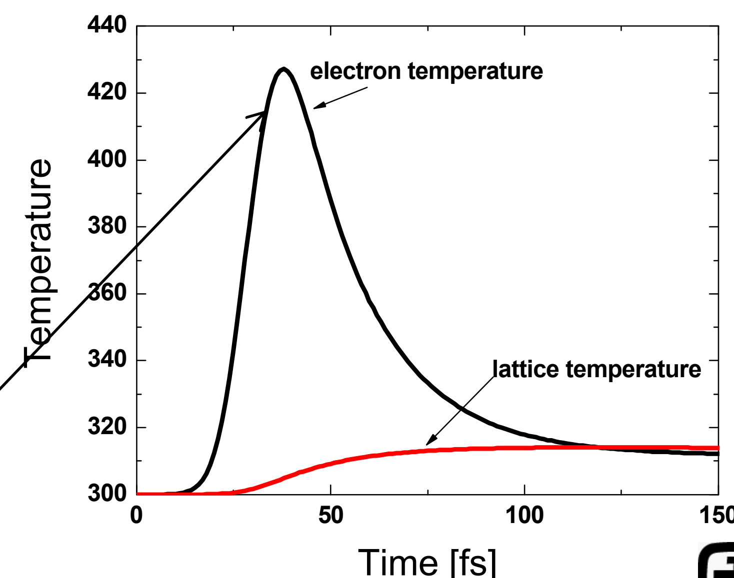
$$\gamma_e T_e \frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}\left(k_e(T_e,T_p)\frac{\partial T_e}{\partial z}\right) - G\left[T_e - T_p\right] + S(z,t)$$

Energy deposited into *e* system

Electron-phonon coupling factor

$$C_p \frac{\partial T_p}{\partial t} = G\left[T_e - T_p\right]$$

Energy stored in *p* system — Energy gained by *p* system from *e* system



Note large change in temperature at early pump-probe time delays

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Determining the electron-phonon coupling factor, *G*

The thermoreflectance coefficient

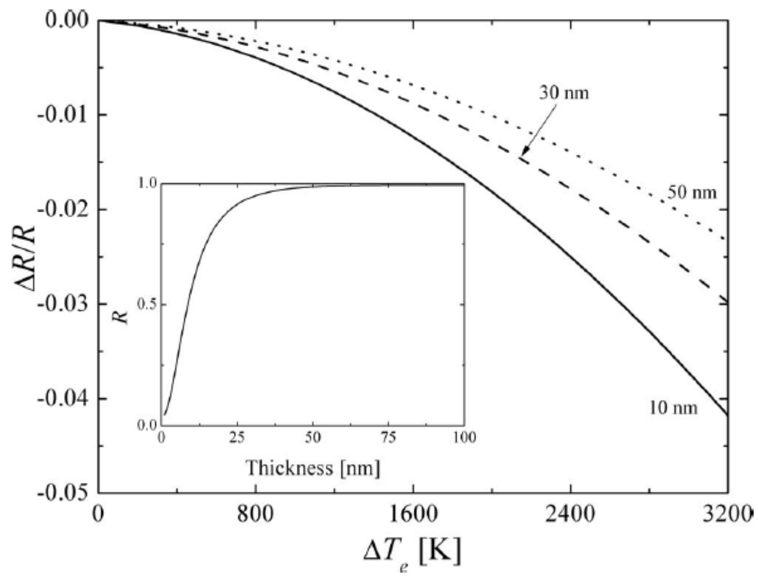
From TTM

$$\Delta T_e \,\&\, \Delta T_p$$

$$R = a\Delta T_e + b\Delta T_p$$

Phase corrected data

Linear thermoreflectance relation only valid when $\Delta T < 150$ K



Consider Drude model

$$n = n_1 + in_2 = \sqrt{\varepsilon_f} = \sqrt{1 - \frac{\omega_p^2}{\omega\left(\omega + \frac{i}{\tau}\right)}}$$

Nonlinear thermoreflectance relation for free electron metals

$$R = f\left(\Delta T_e\right) + b\Delta T_p$$

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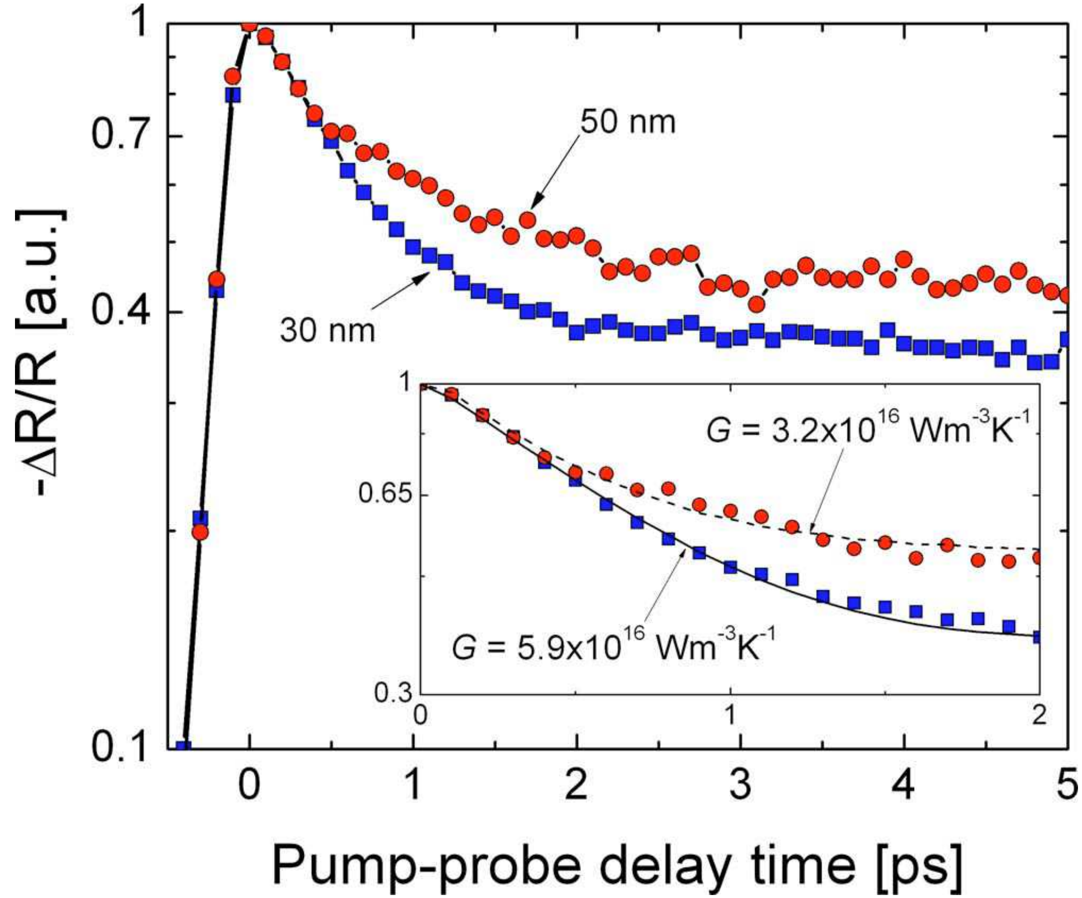
Determining the electron-phonon coupling factor, *G*



G is a material property. For Au, $G = 3\text{x}10^{16}$ W m^{-3} K^{-1}.
Measured *G* changes with film thickness. Why?

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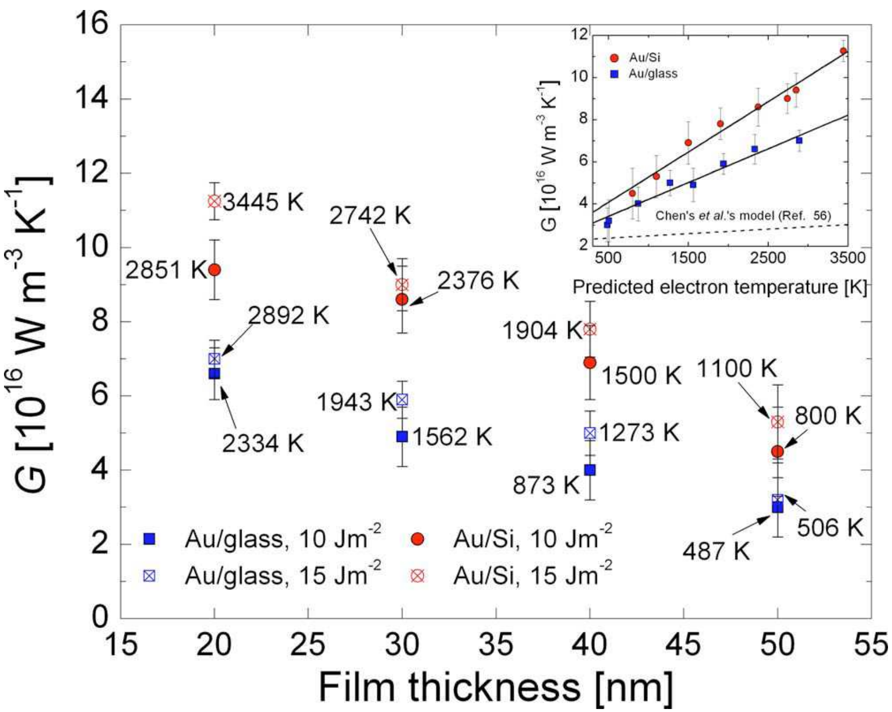
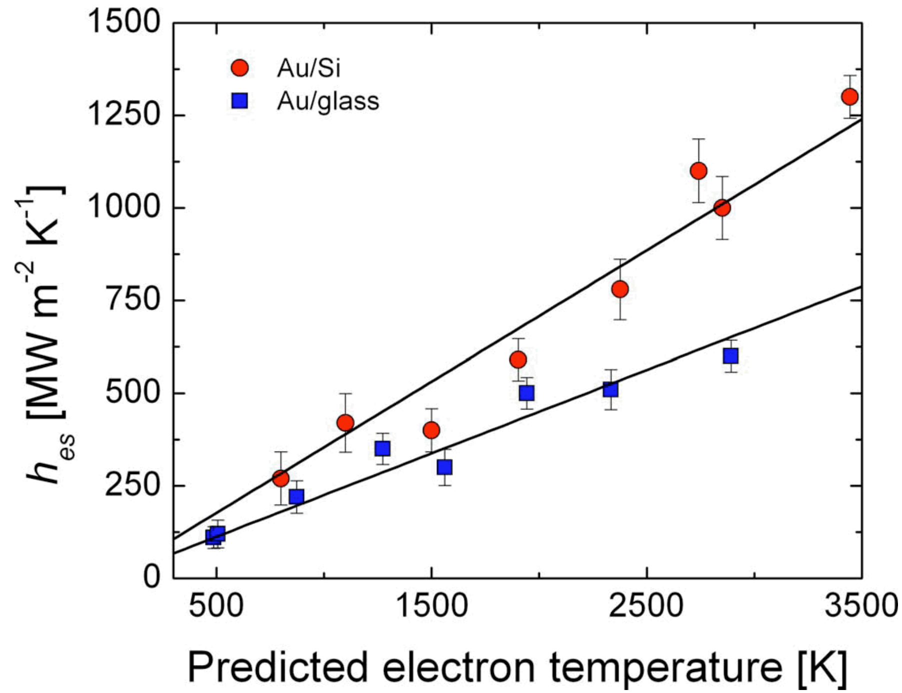
Influence of electron-interface scattering on *G*





Film thickness and substrate dependence suggests electron-interface scattering.

Use 3 temperature model (3TM) to determine h_{es}

Hopkins *et al.*, Journal of Applied Physics, **105**, 023710 (2009)

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Influence of electron-interface scattering on thermoreflectance

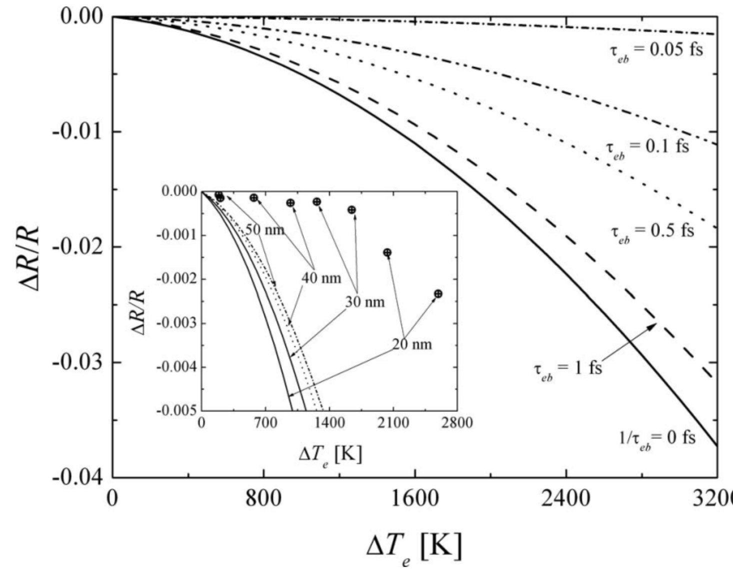
The thermoreflectance coefficient

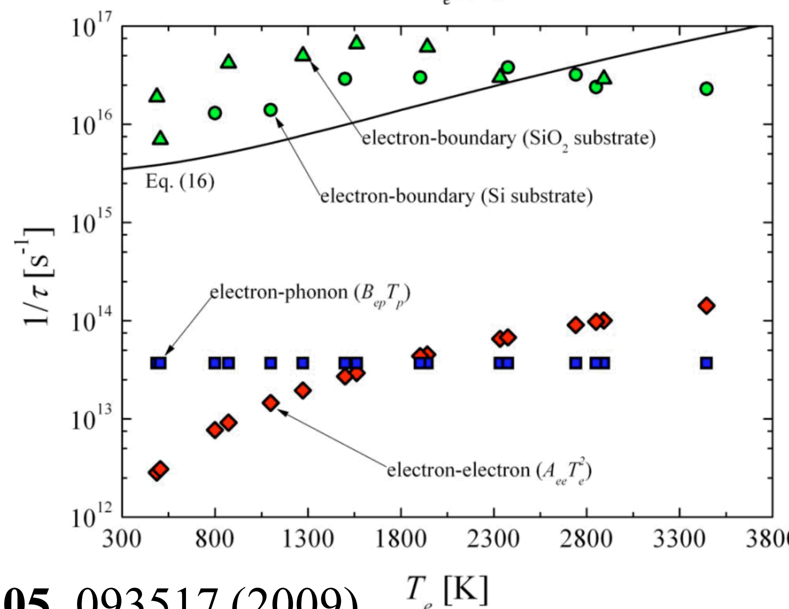
Consider Drude model that accounts for electron-interface scattering

$$n = n_1 + in_2 = \sqrt{\varepsilon_f} = \sqrt{1 - \frac{\omega_p^2}{\omega\left(\omega + \frac{i}{\tau}\right)}}$$

$$\frac{1}{\tau} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_{eb}}$$

$$\frac{1}{\tau_{eb}} = \frac{3\pi\left(\frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}}\right)^2}{35\zeta(3) q_{th} v_L}\left[1 + 2\left(\frac{v_L}{v_T}\right)^3\right]$$





Hopkins, Journal of Applied Physics, **105**, 093517 (2009)

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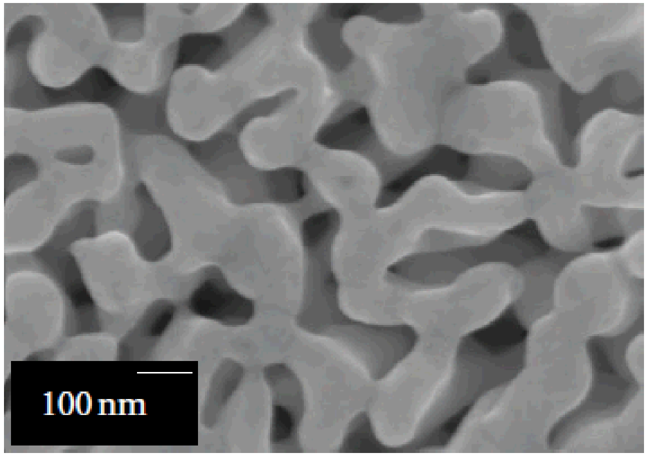
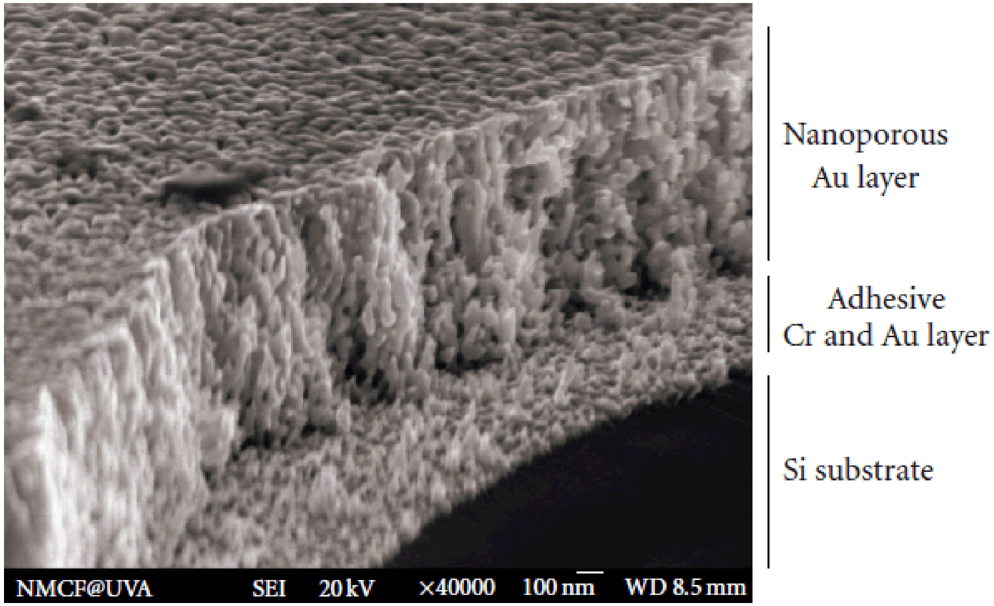
Thermal properties of nanoporous metals

"Matrix" of nanowires





Thermal properties should show influence from ligament-boundary scattering

Hopkins *et al*., Journal of Nanomaterials, **2008**, 418050 (2008)

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Effect of electron-phonon nonequilibrium and boundary scattering on thermal conductivity

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

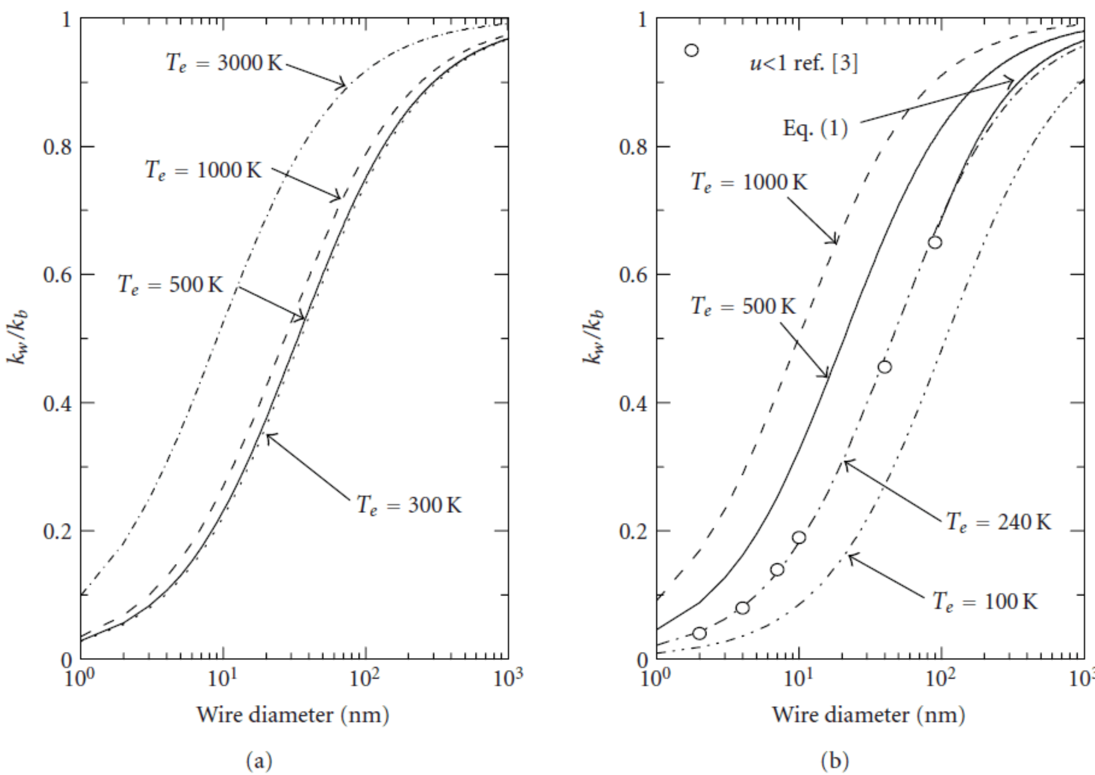
ep non-equilibrium

$$\Lambda = \frac{1}{3}\gamma T_e v_F^2 \frac{1}{AT_e^2 + BT_p + \frac{v_F}{d}}$$

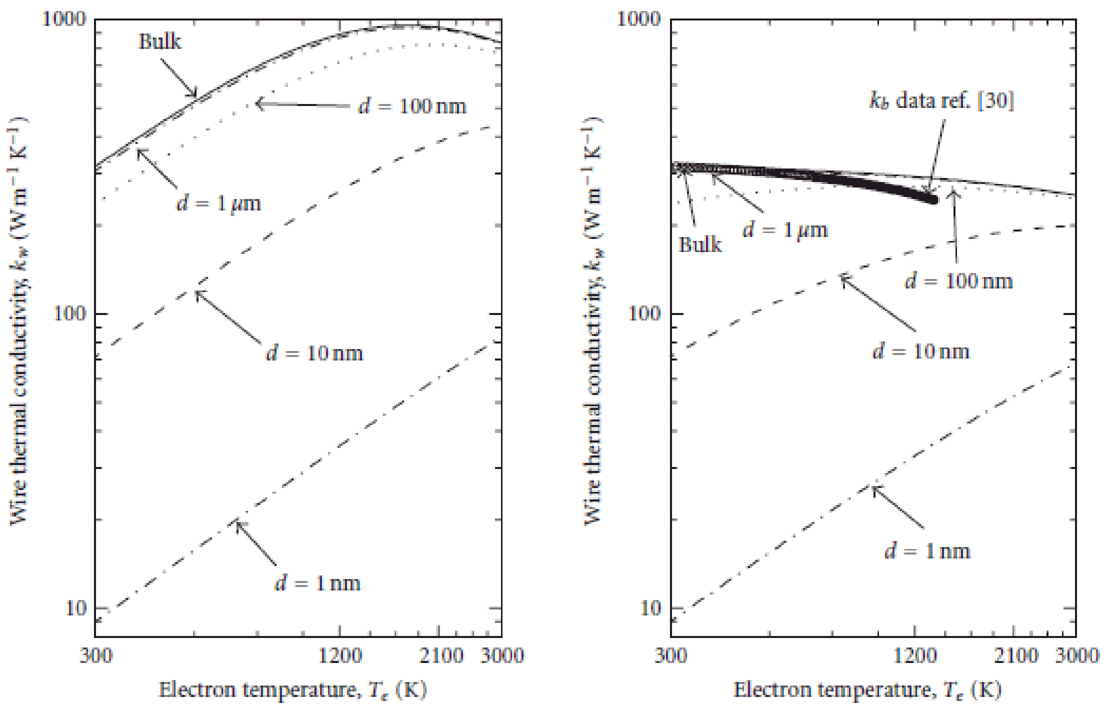
ep equilibrium

$$\Lambda = \frac{1}{3}\gamma T v_F^2 \frac{1}{AT^2 + BT + \frac{v_F}{d}}$$

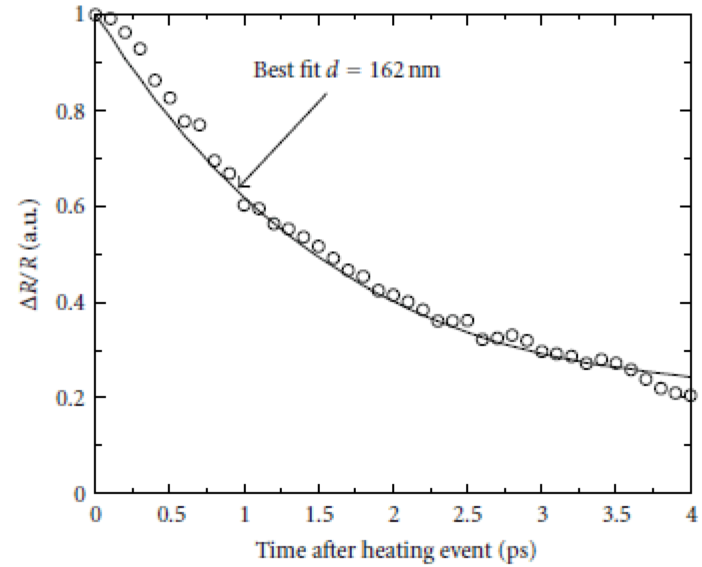
$$\frac{1}{\tau} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{ep}} + \frac{1}{\tau_{eb}}$$

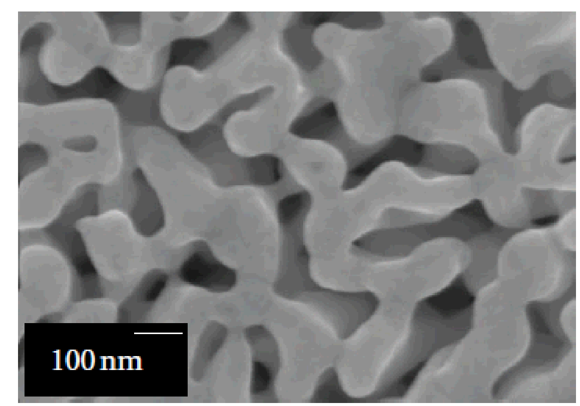


Effect of electron-phonon nonequilibrium and boundary scattering on thermal conductivity



(a) (b)





Hopkins *et al*., Journal of Nanomaterials, **2008**, 418050 (2008)

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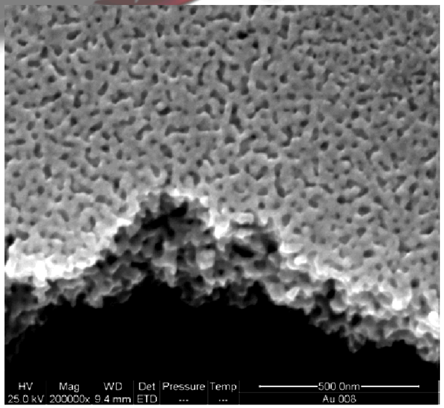
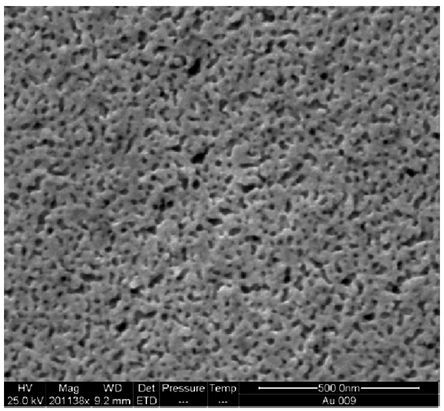
G in nanoporous metals

Nanoporous metal fabrication at CINT

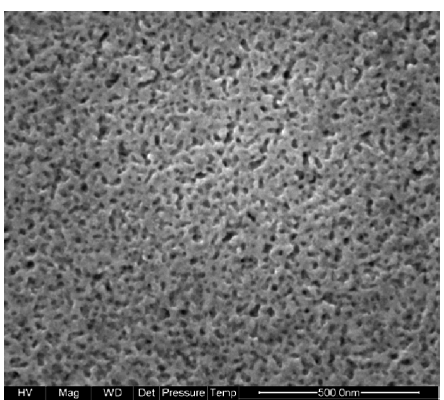
Collaborators: Amit Misra and Hongqi Li



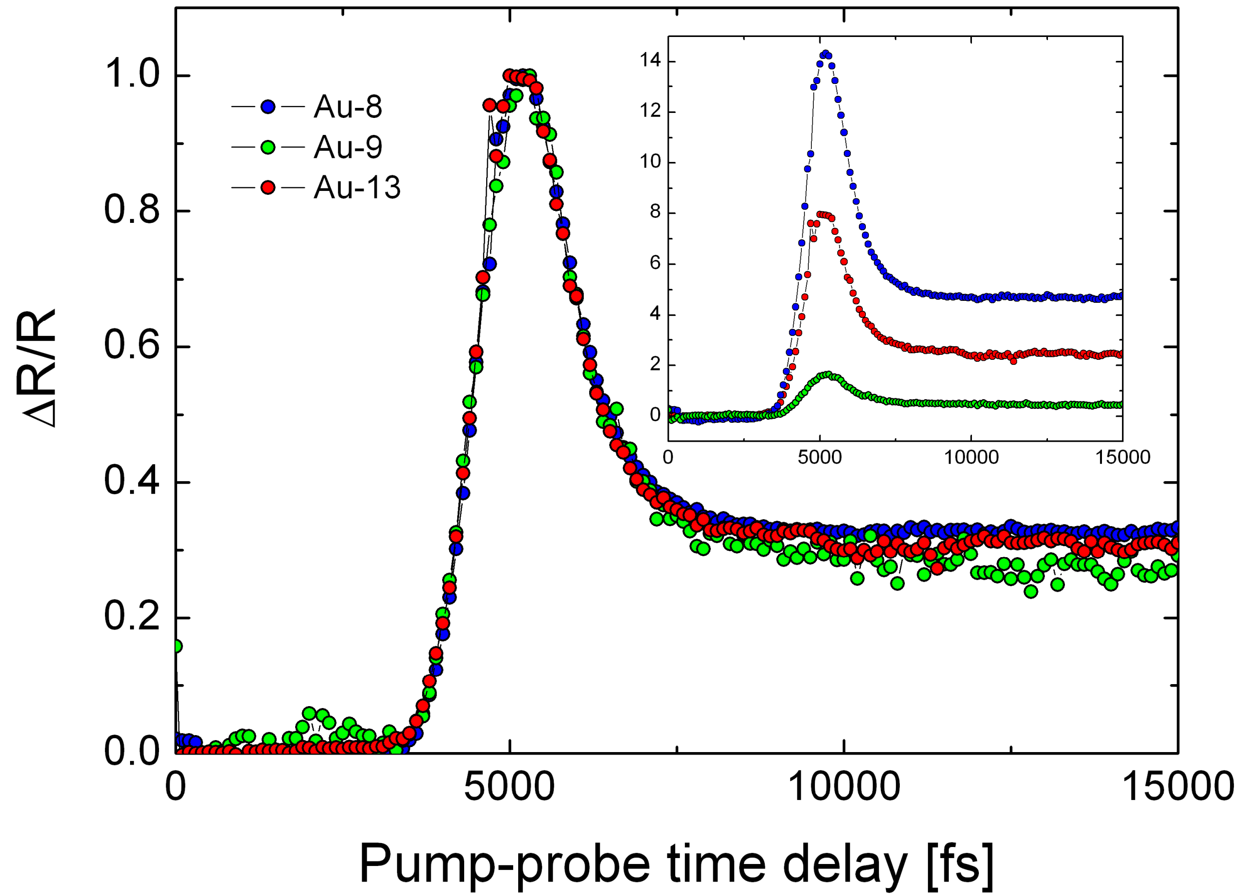
Au-8



Au-9



Au-13



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