Ultrafast laser-based metrology for micron-scale measurements of thermal transport, coefficient of thermal expansion, and temperature

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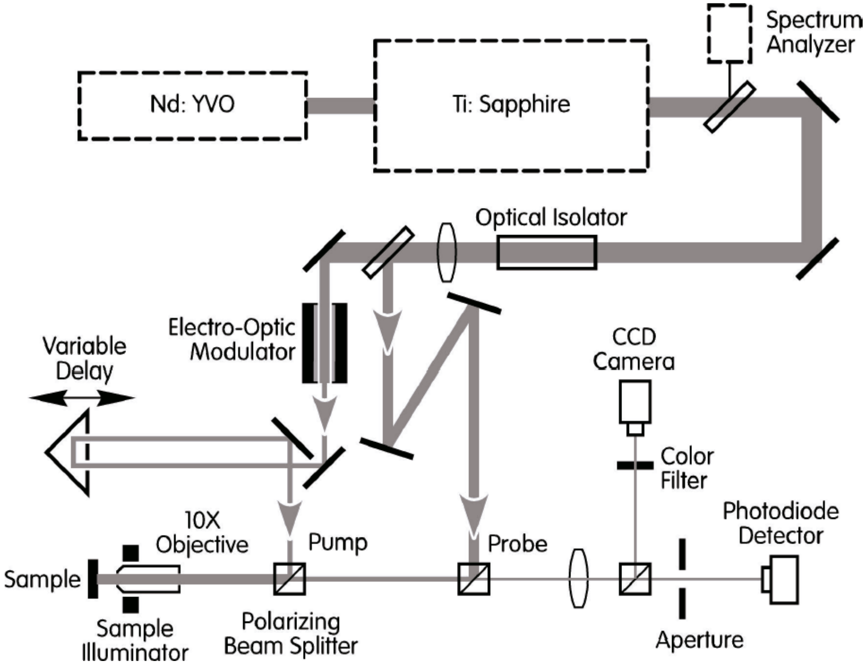
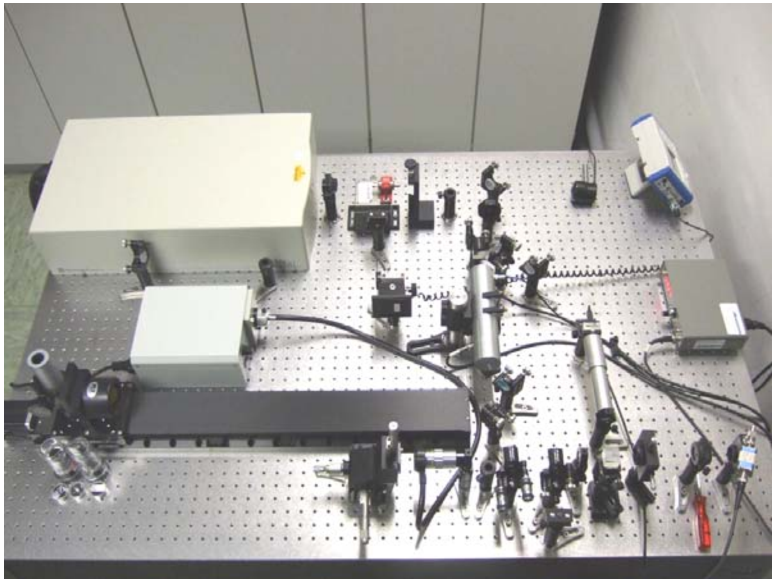
Time domain thermoreflectance since 2003

- Improved optical design
- Normalization by out-of-phase signal eliminates artifacts, increases dynamic range and improves sensitivity
- Exact analytical model for Gaussian beams and arbitrary layered geometries
- One-laser/two-color approach tolerates diffuse scattering

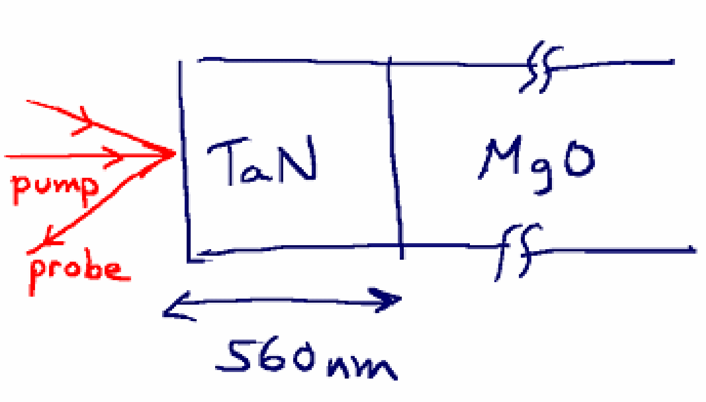


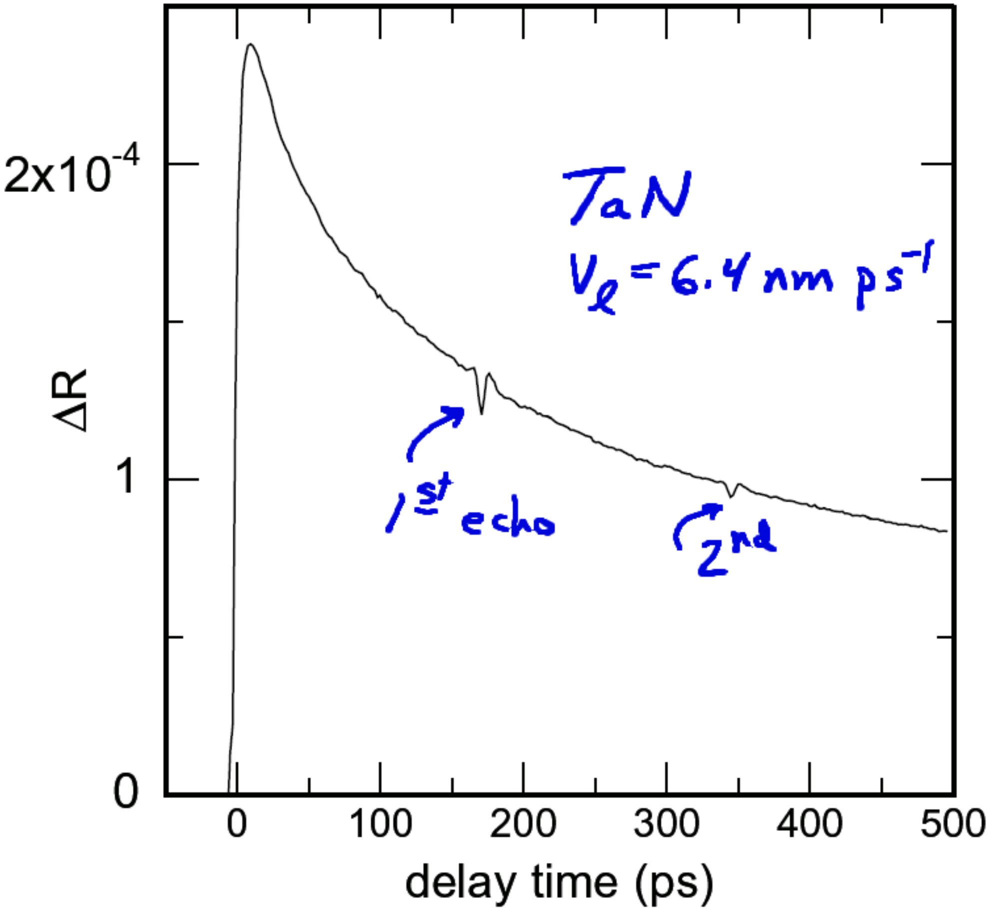
Clone built at Fraunhofer Institute for Physical Measurement, Jan. 7-8 2008



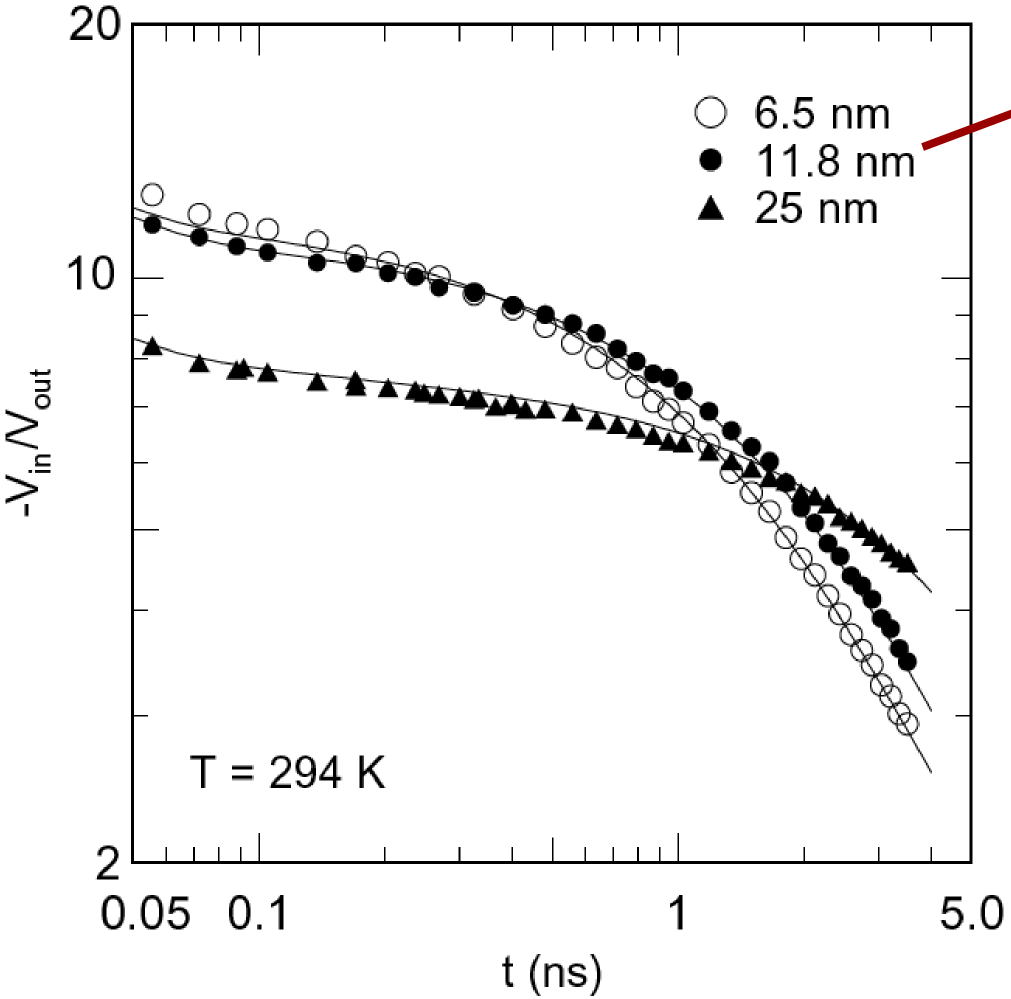
psec acoustics and time-domain thermoreflectance

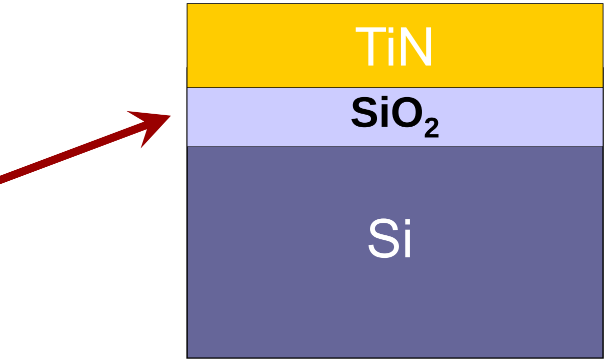
- Optical constants and reflectivity depend on strain and temperature
- Strain echoes give acoustic properties or film thickness
- Thermoreflectance gives thermal properties





Time-domain Thermoreflectance (TDTR) data for TiN/SiO_2/Si



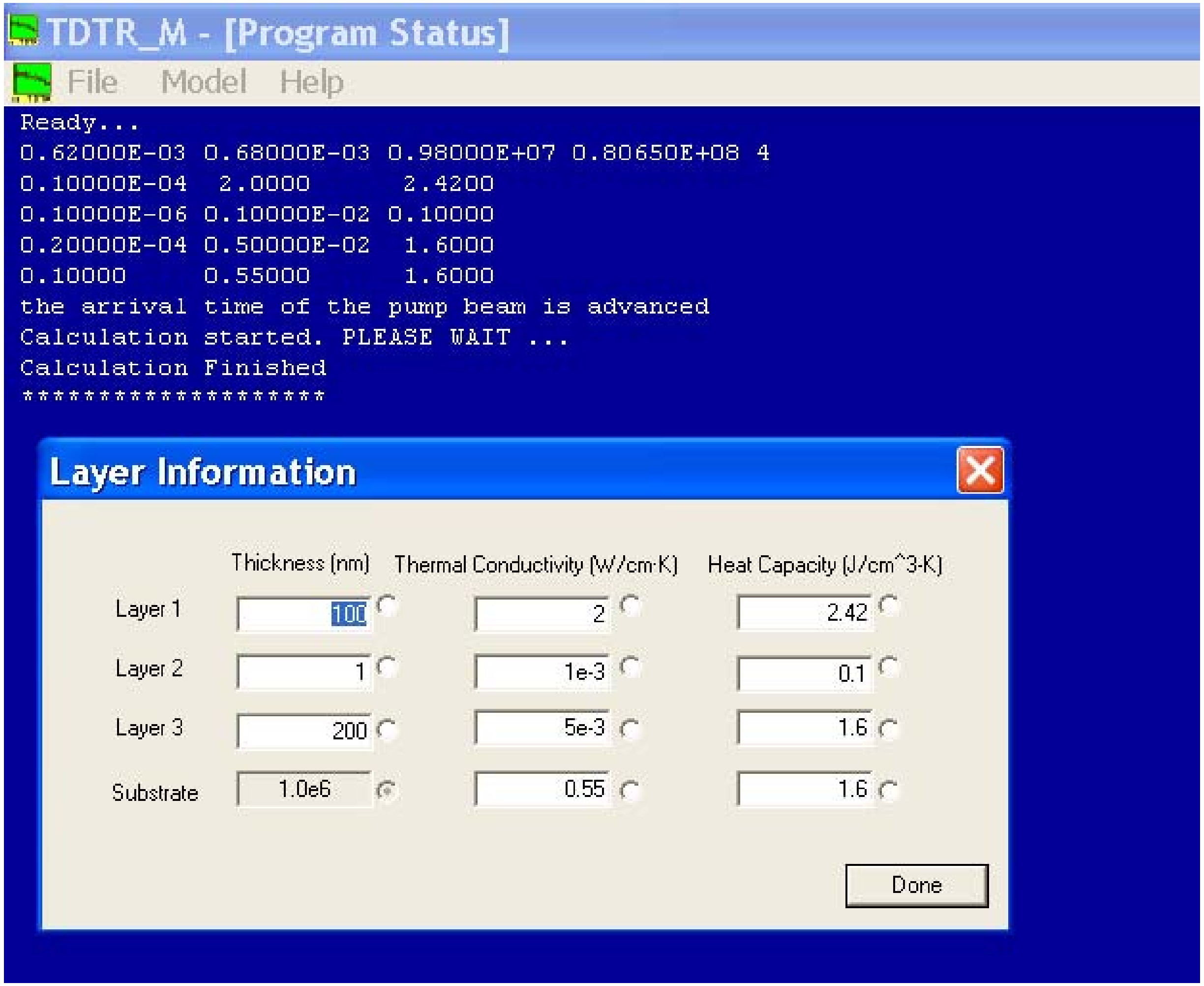


- reflectivity of a metal depends on temperature
- one free parameter: the “effective” thermal conductivity of the thermally grown SiO_2 layer (interfaces not modeled separately)

Windows software

author: Catalin Chiritescu,

users.mrl.uiuc.edu/cahill/tcdata/tdtr_m.zip



```
Ready...
0.62000E-03 0.68000E-03 0.98000E+07 0.80650E+08 4
0.10000E-04  2.0000       2.4200
0.10000E-06 0.10000E-02 0.10000
0.20000E-04 0.50000E-02   1.6000
0.10000      0.55000      1.6000
the arrival time of the pump beam is advanced
Calculation started. PLEASE WAIT ...
Calculation Finished
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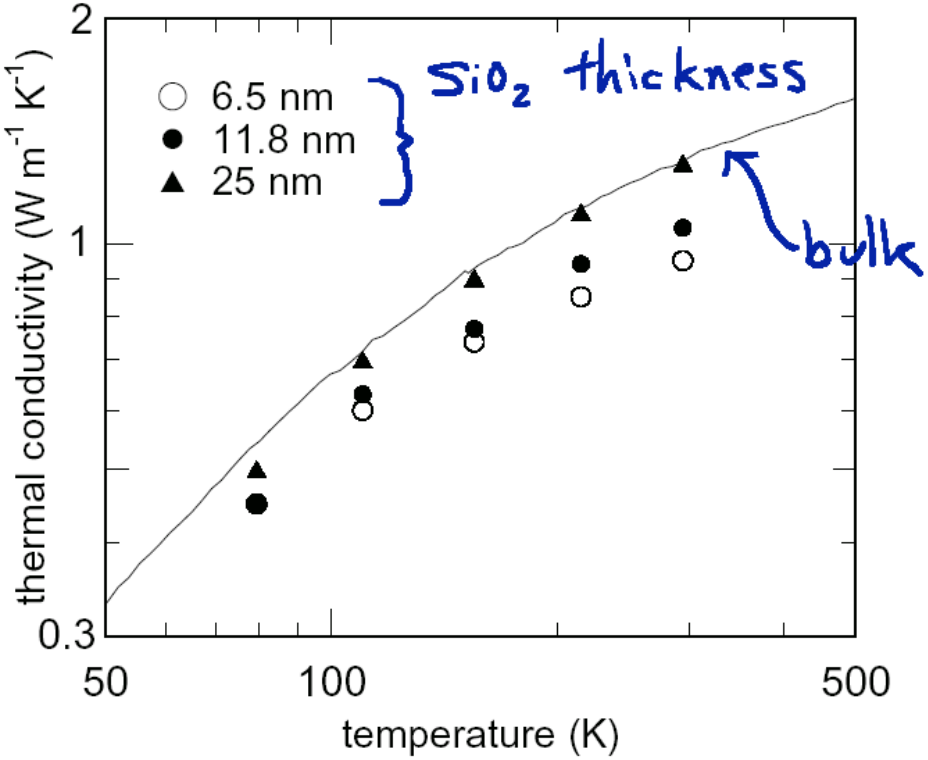
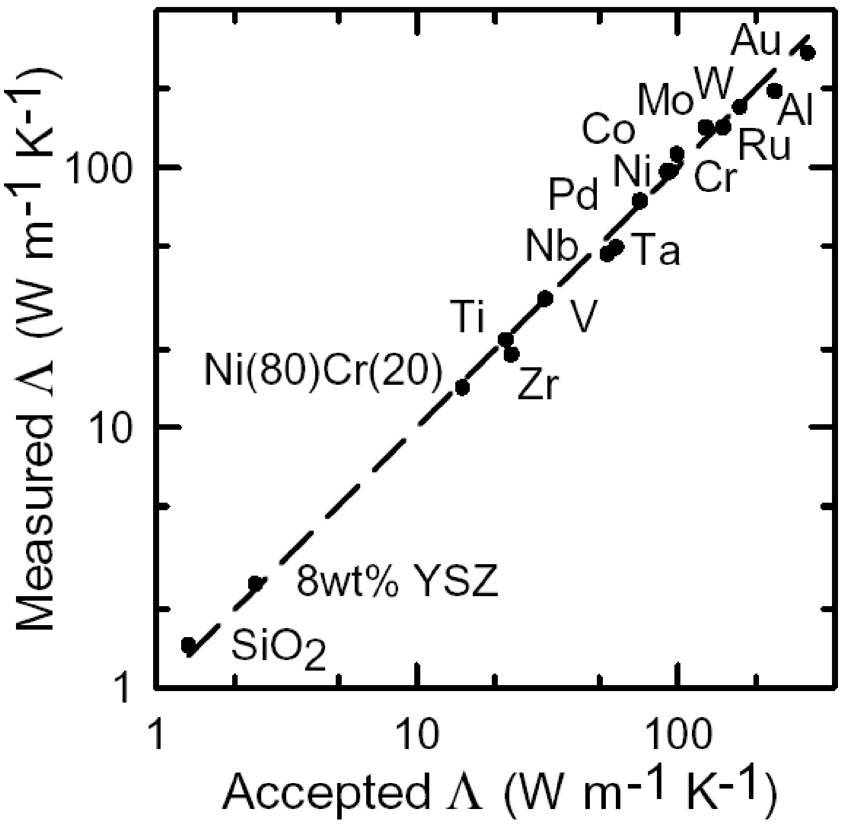
	Thickness (nm)	Thermal Conductivity (W/cm·K)	Heat Capacity (J/cm^3-K)
Layer 1	100 ○	2 ○	2.42 ○
Layer 2	1 ○	1e-3 ○	0.1 ○
Layer 3	200 ○	5e-3 ○	1.6 ○
Substrate	1.0e6 ●	0.55 ○	1.6 ○

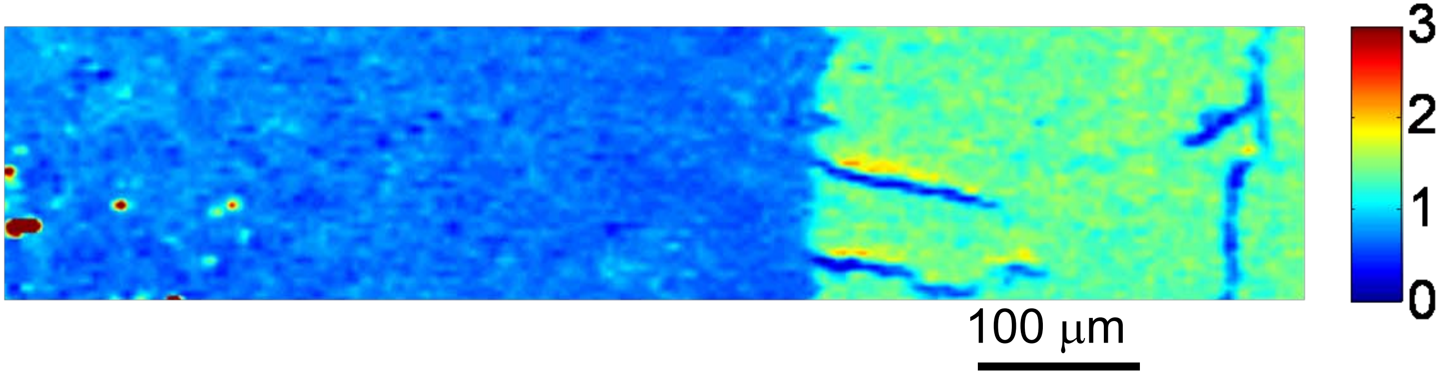
TDTR: Flexible, convenient, and accurate

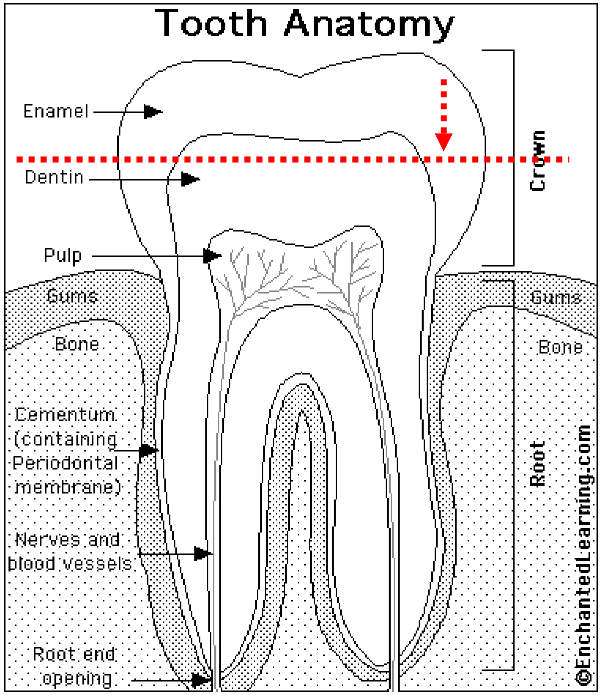




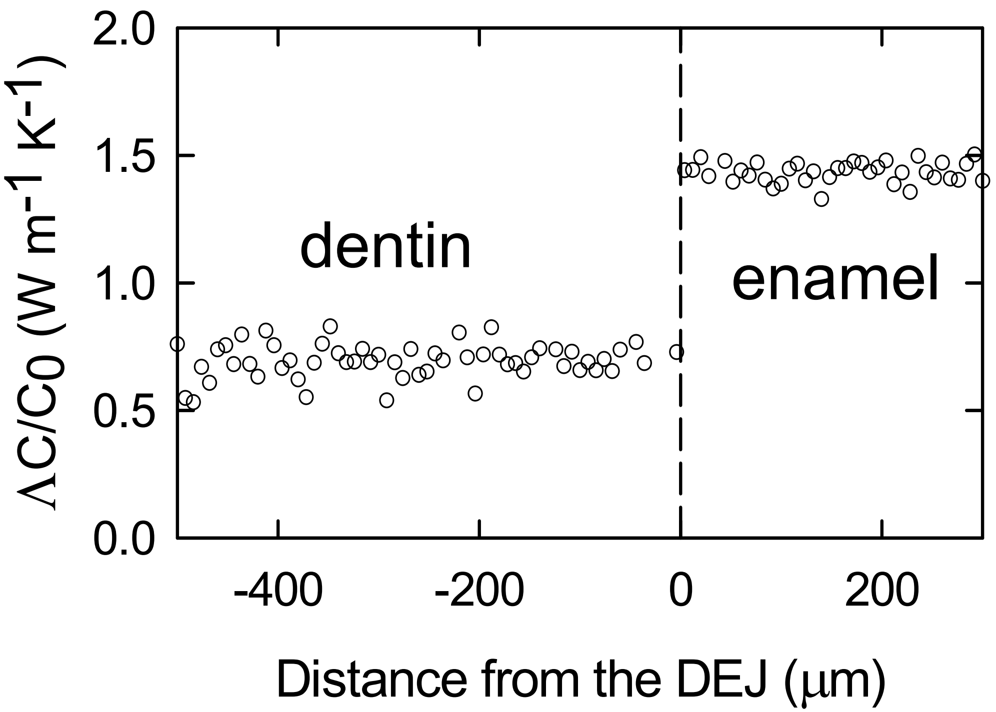
- ...with 3 micron resolution...

Thermal conductivity map of a human tooth

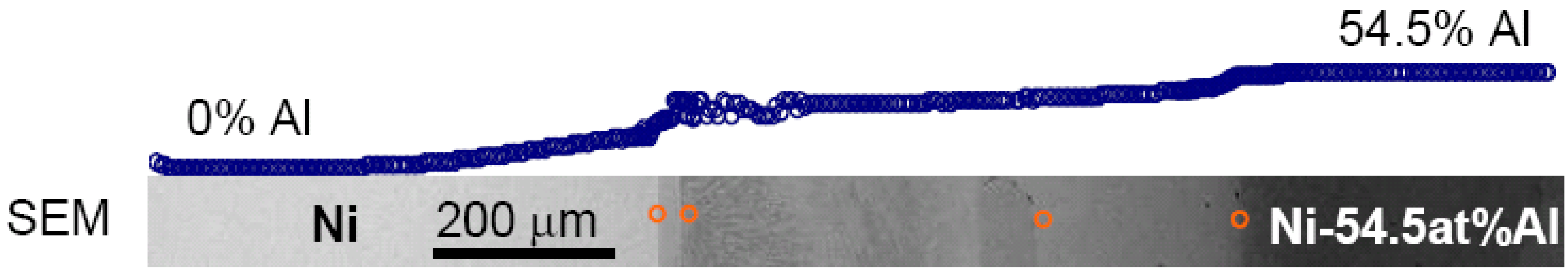


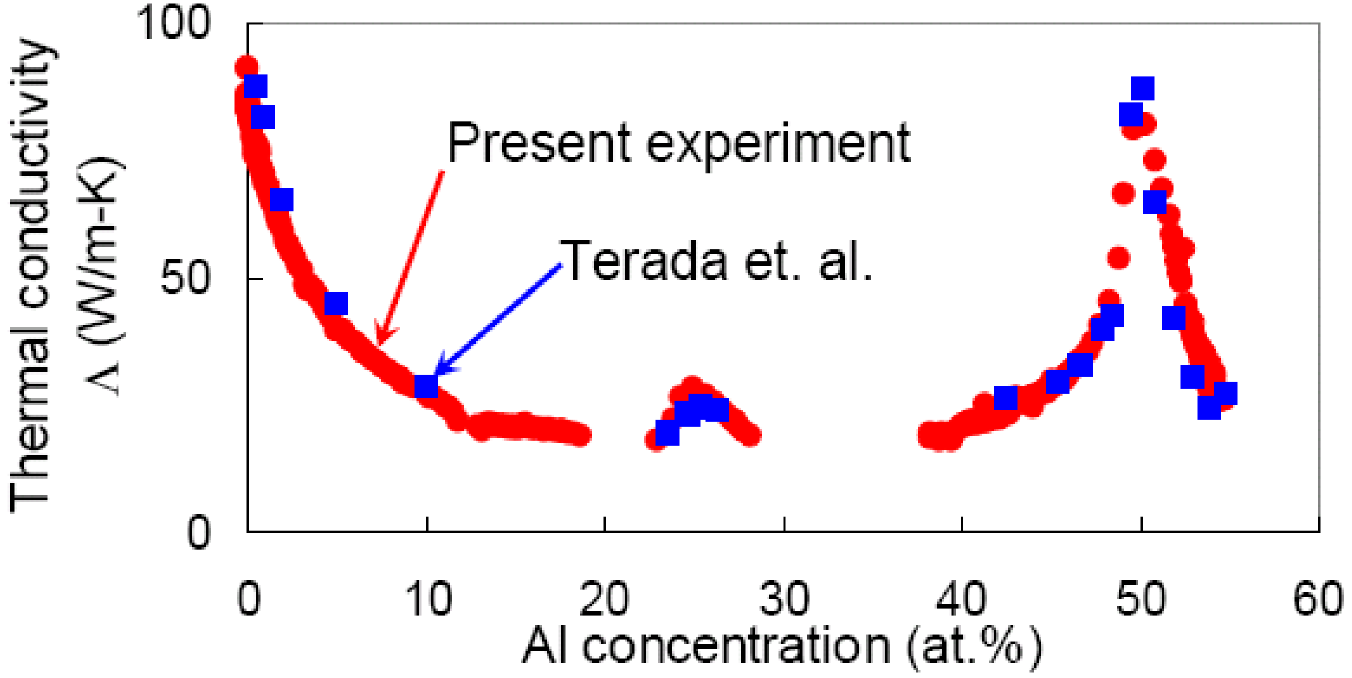


www.enchantedlearning.com/



High throughput data using diffusion couples

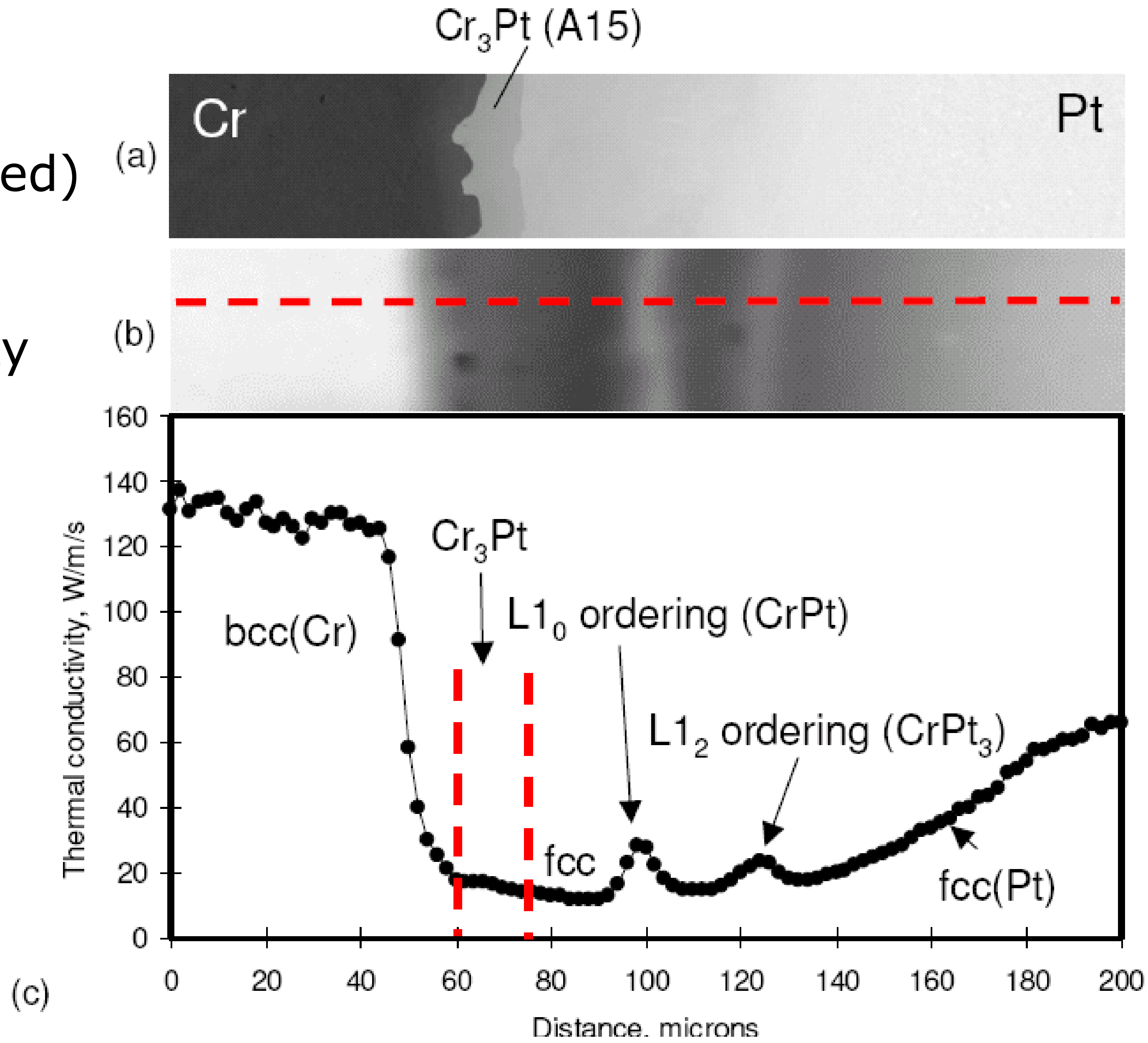




Mapping of a metallurgical diffusion couple

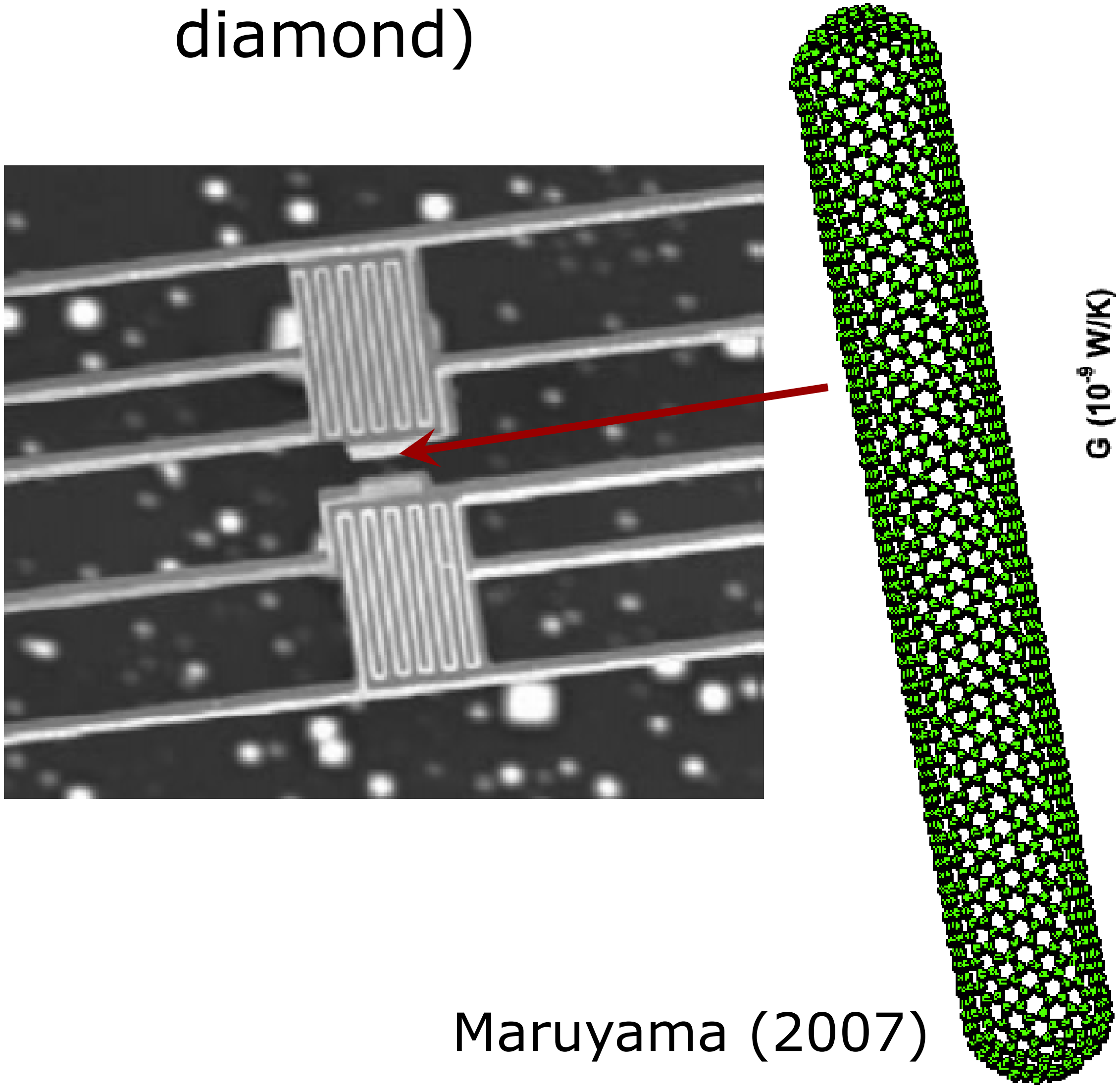
SEM
(backscattered)

thermal
conductivity

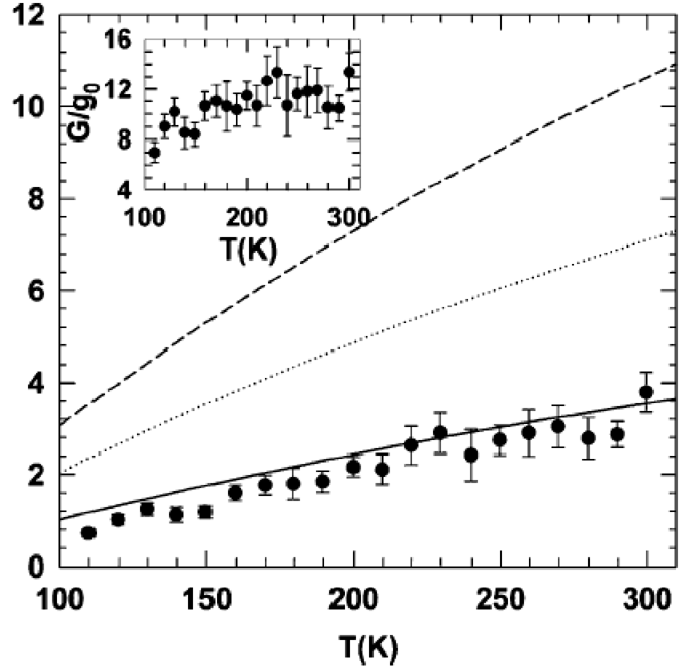


Carbon nanotubes

- Evidence for the highest thermal conductivity any material (higher conductivity than diamond)



Maruyama (2007)

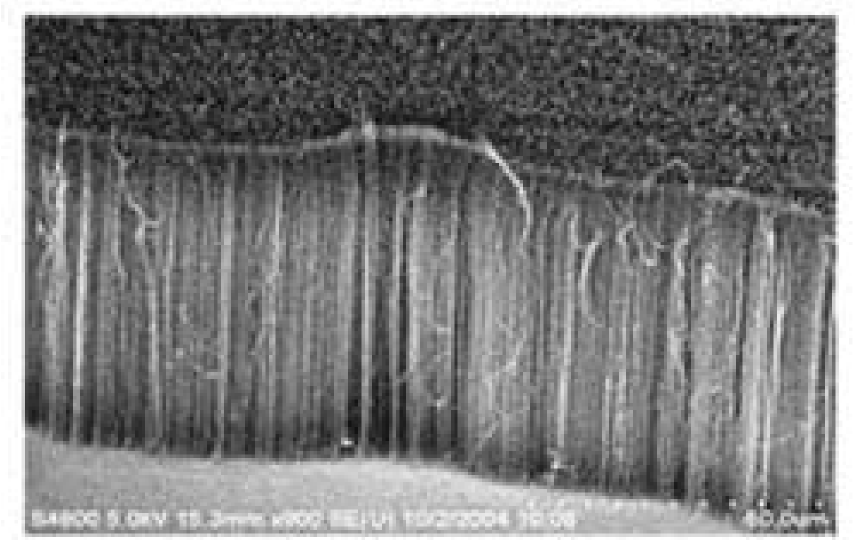


Yu et al. (2005)

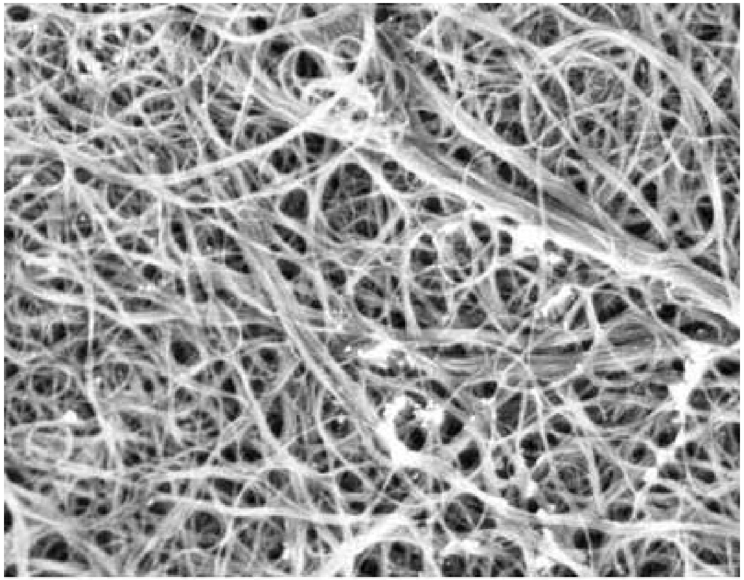
Can we make use of this?

- Much work world-wide:
    - thermal interface materials
    - so-called "nanofluids" (suspensions in liquids)
    - polymer composites and coatings

Fischer (2007)



Oriented carbon nanotube array.

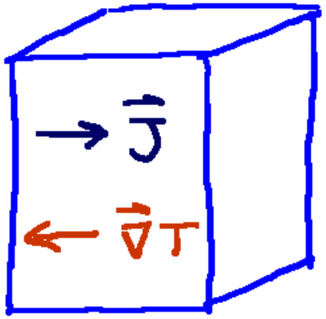


Lehman (2005)

Thermal conductivity and interface thermal conductance

- Thermal conductivity Λ is a property of the continuum

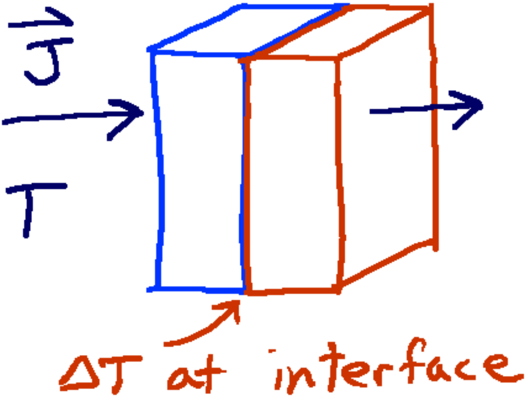
$$\vec{J} = -\Lambda \vec{\nabla} T$$



$$\Lambda = \frac{1}{3Vk_BT^2}\int_0^\infty \langle \vec{j}(t)\cdot\vec{j}(0)\rangle\, dt$$

- Thermal conductance (per unit area) *G* is a property of an interface

$$J = G\Delta T$$

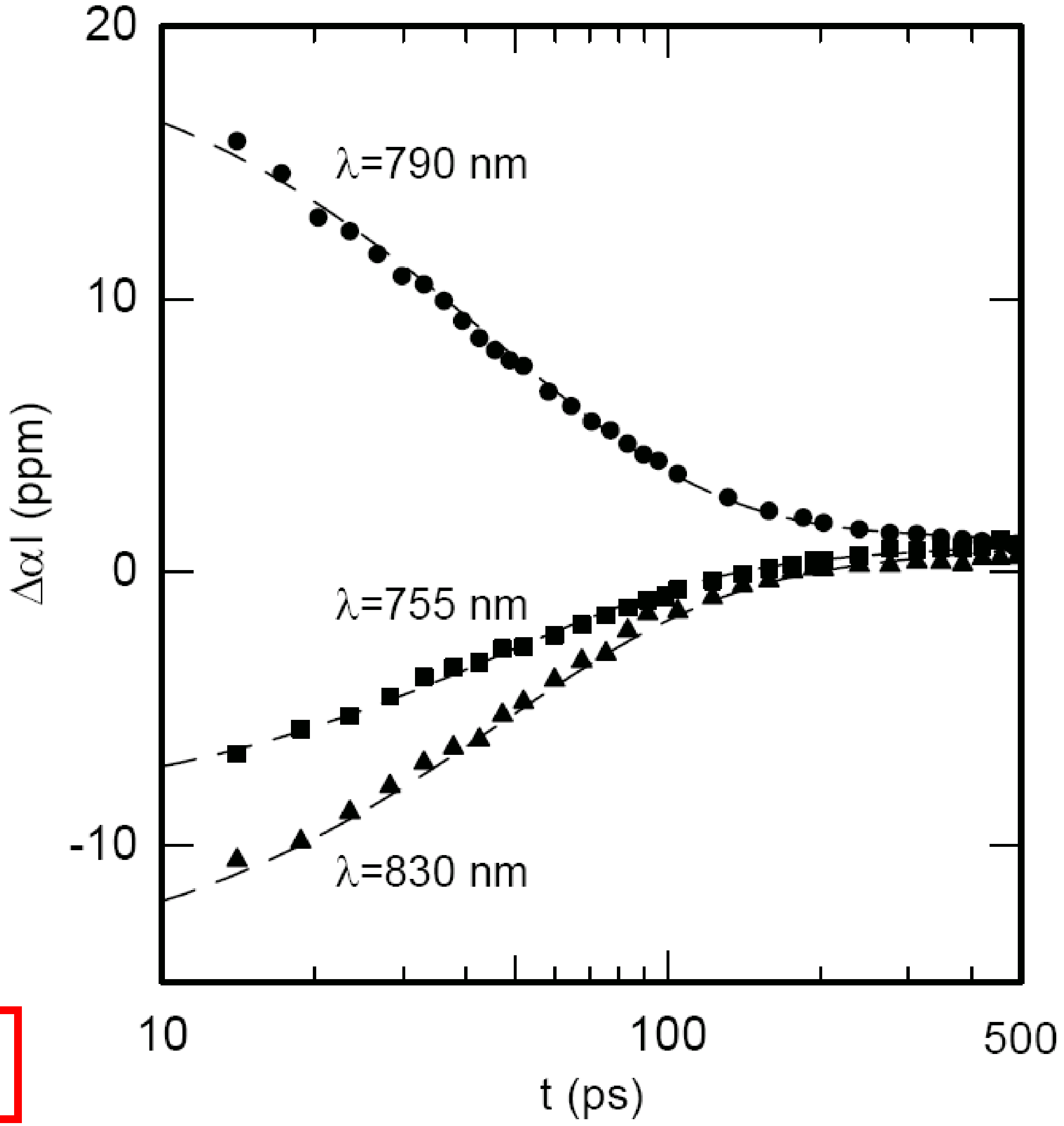


$$G = \frac{1}{Ak_BT^2}\int_0^\infty \langle q(t)q(0)\rangle\, dt$$

Nanotubes in surfactant in water

- Optical absorption depends on temperature of the nanotube
- Assume heat capacity is comparable to graphite
- Cooling rate (RC time constant) gives interface conductance

$G = 12 \text{ MW m}^{-2} \text{ K}^{-1}$



Critical aspect ratio of a fiber composite

- Isotropic fiber composite with high conductivity fibers (and infinite interface conductance)

$$\Lambda_c = \frac{1}{3} v_f \Lambda_{NT}$$

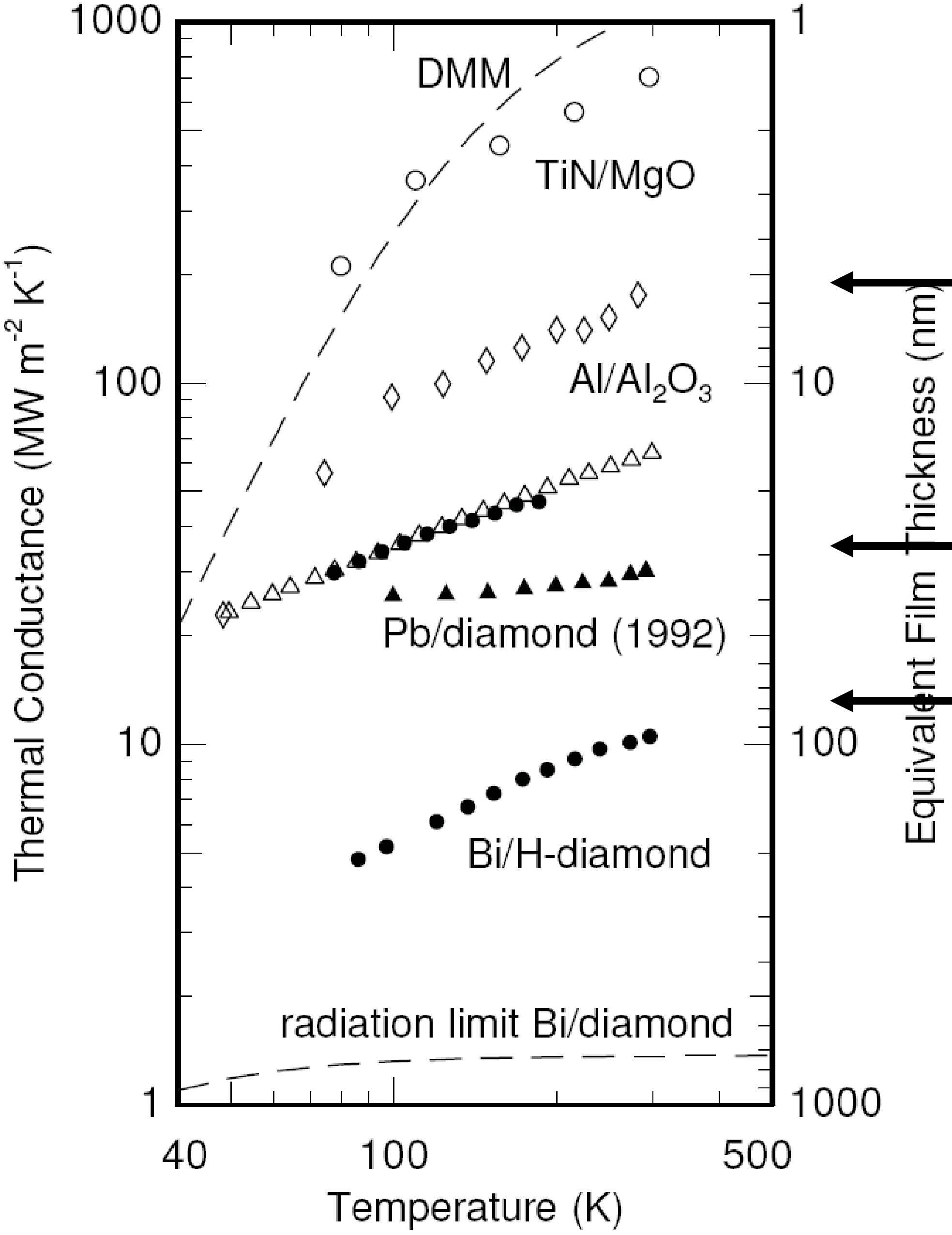
- But this conductivity if obtained only if the aspect ratio of the fiber is high

$$3\left(\frac{\Lambda_{NT}}{rG}\right)^{1/2} \approx 2000$$

- Troubling question: Did we measure the relevant value of the conductance?

"heat capacity G" vs. "heat conduction G"

Interface thermal conductance: Factor of 60 range at room temperature



W/Al_2O_3

Au/water

$PMMA/Al_2O_3$

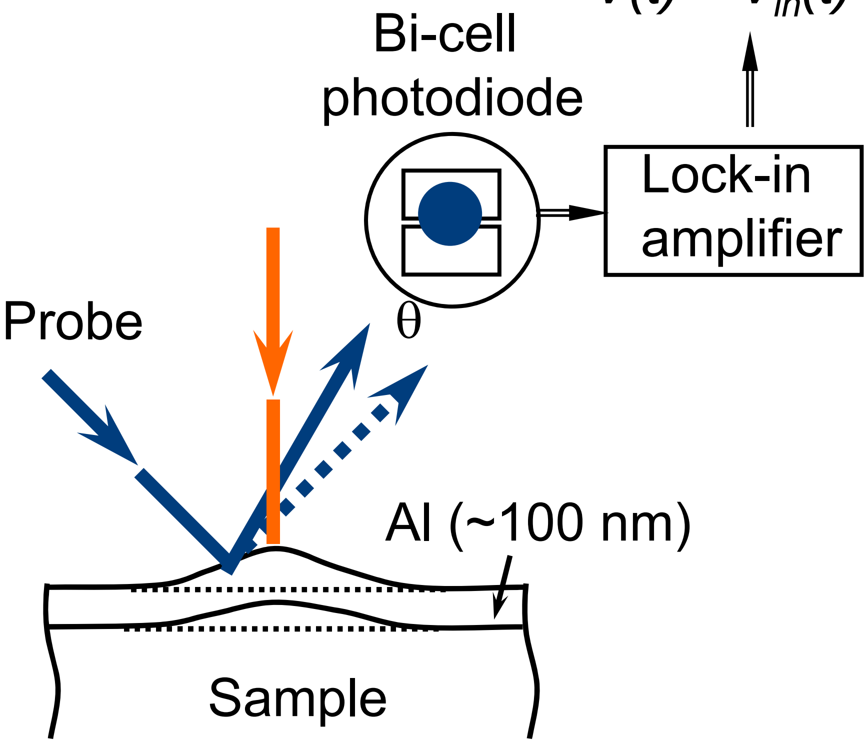
nanotube/alkane

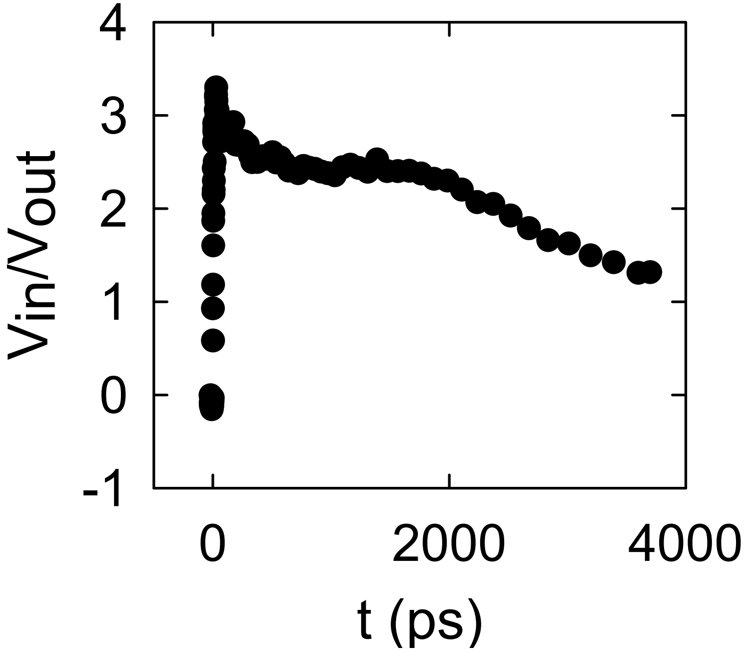
$L = \Lambda/G$

$\Lambda = 1\ W\ m^{-1}\ K^{-1}$

Time-domain probe beam deflection (TD-PBD) for CTE measurements

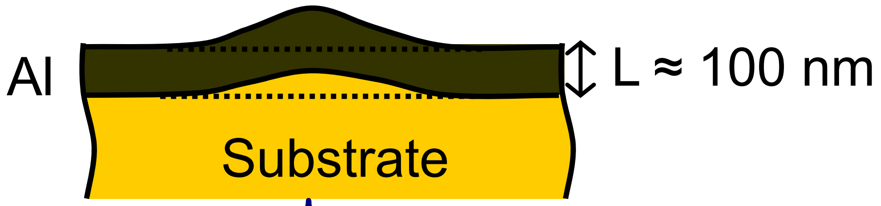
$$V(t) = V_{in}(t) + iV_{out}(t)$$



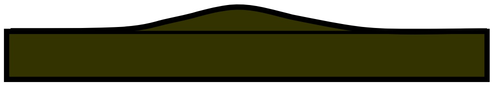


spatial resolution ~ laser spot size (~ 4 μm)

Model for the beam deflection

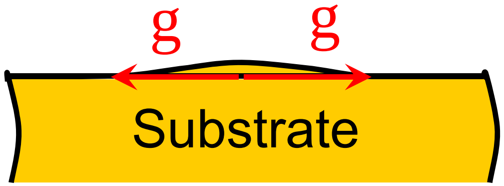


Thermal expansion of Al film



$$Z_1 = \int_0^L \frac{1+\upsilon_{Al}}{1-\upsilon_{Al}} \alpha_{T,Al} T_{Al}\, dz$$

Bi-axial stress of Al film



$$g = \int_0^L B_{Al} \alpha_{T,Al} T_{Al}\, dz$$

$$\frac{Y}{2(1+\upsilon)} \nabla^2 U + \frac{Y}{2(1+\upsilon)(1-2\upsilon)} \nabla(\nabla \cdot U) = \rho \ddot{U}$$

Z_2

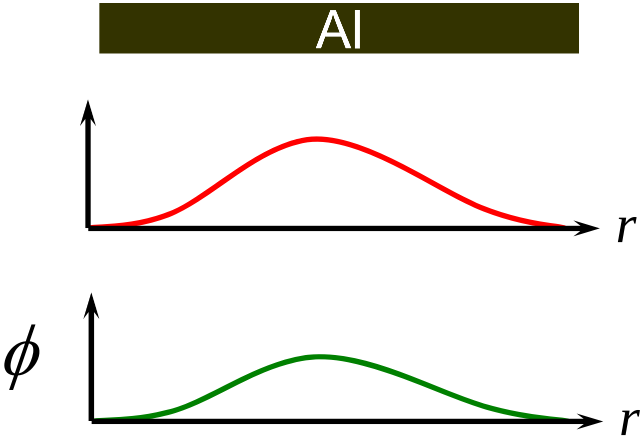
Thermal expansion and bi-axial stress of substrate

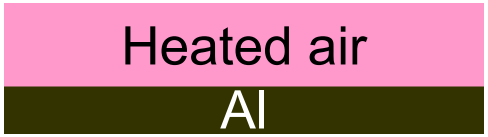
$$\frac{Y}{2(1+\upsilon)} \nabla^2 U + \frac{Y}{2(1+\upsilon)(1-2\upsilon)} \nabla(\nabla \cdot U) = \nabla T + \rho \ddot{U}$$

Z_3

Model for the beam defection (continued)

Temperature gradient on surface




Heated air

Al

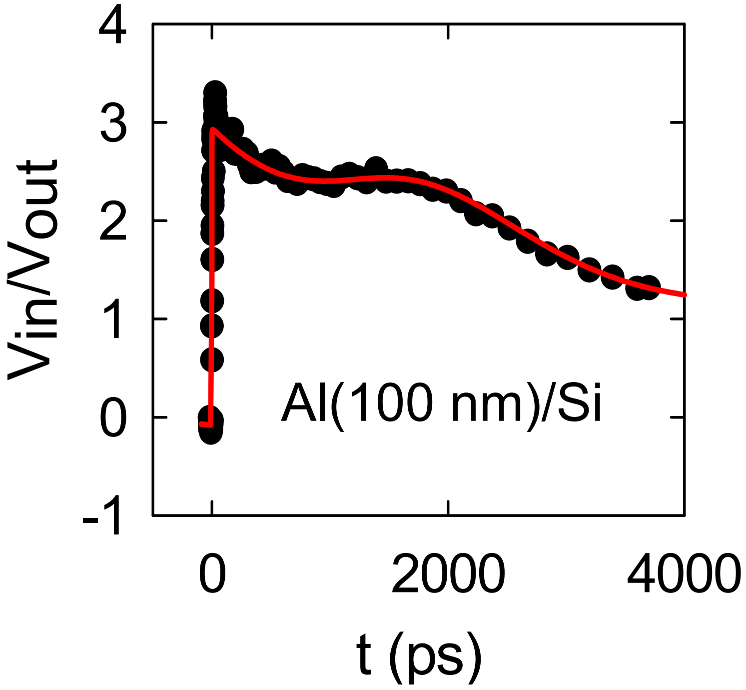
$$Z_{air} = 2\int_{-\infty}^{0} \frac{dn}{dT} T_{air}\, dz$$

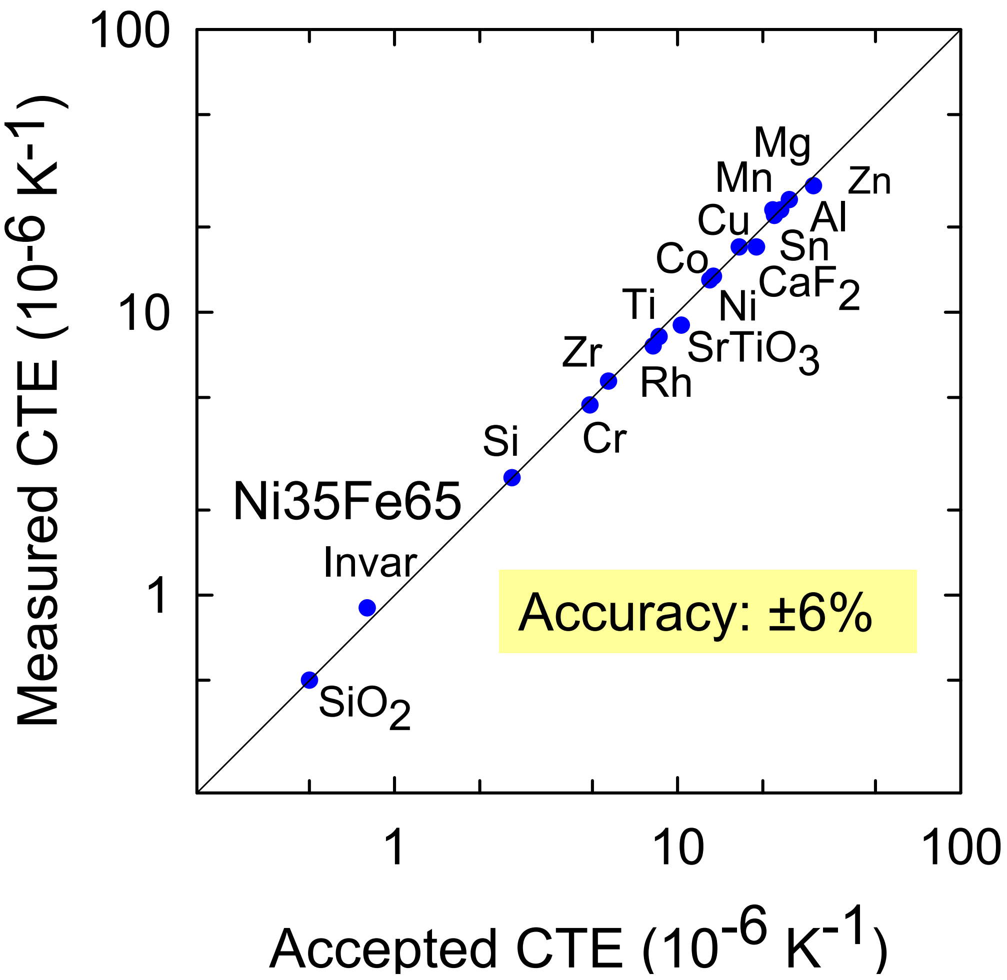
$$r = \frac{n+ik-1}{n+ik+1} = r_0 \exp(i\phi)$$

$$Z_{SurfaceTemp} = T_{surface} \frac{d\phi}{dT} \frac{\lambda}{4\pi}$$

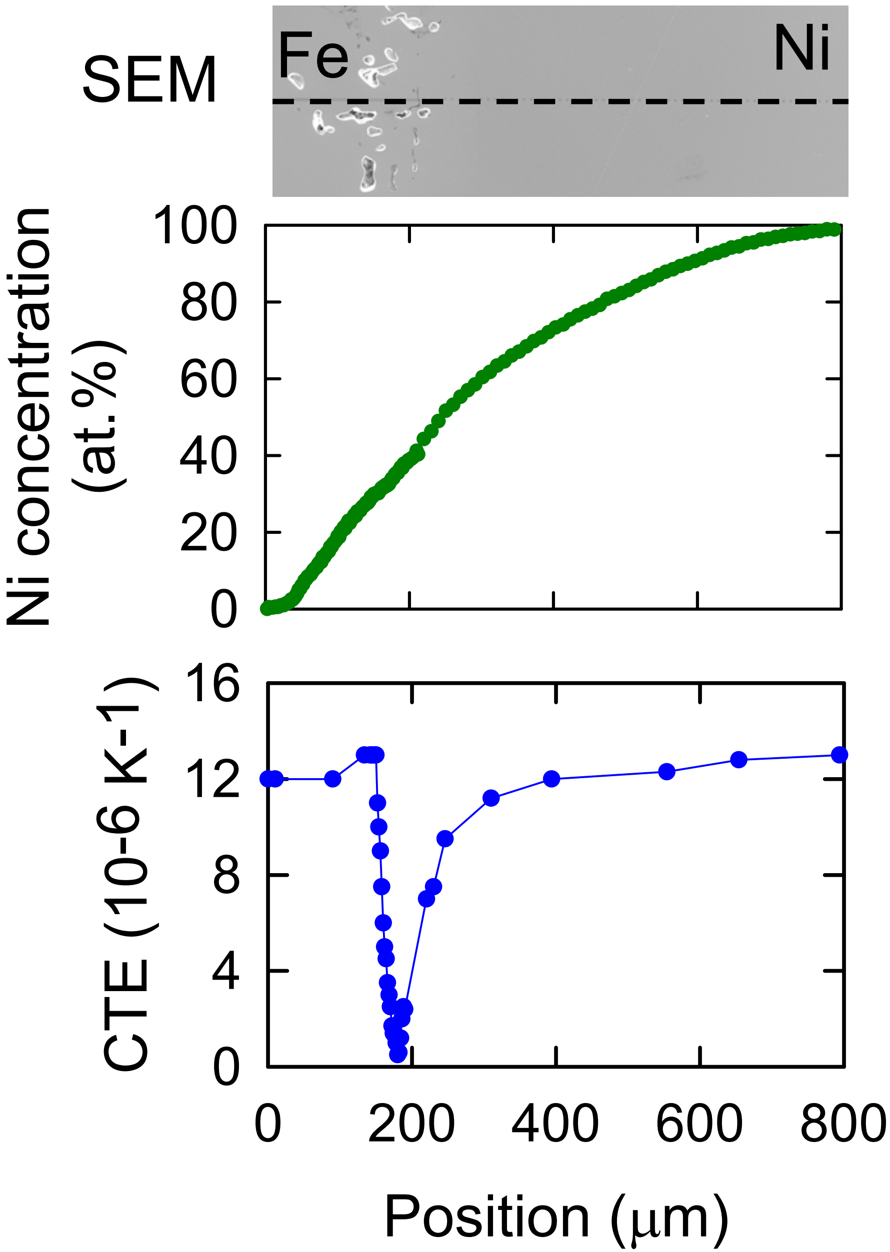
$$Z = Z_1 + Z_2 + Z_3 + Z_{SurfTemp} + Z_{air}$$

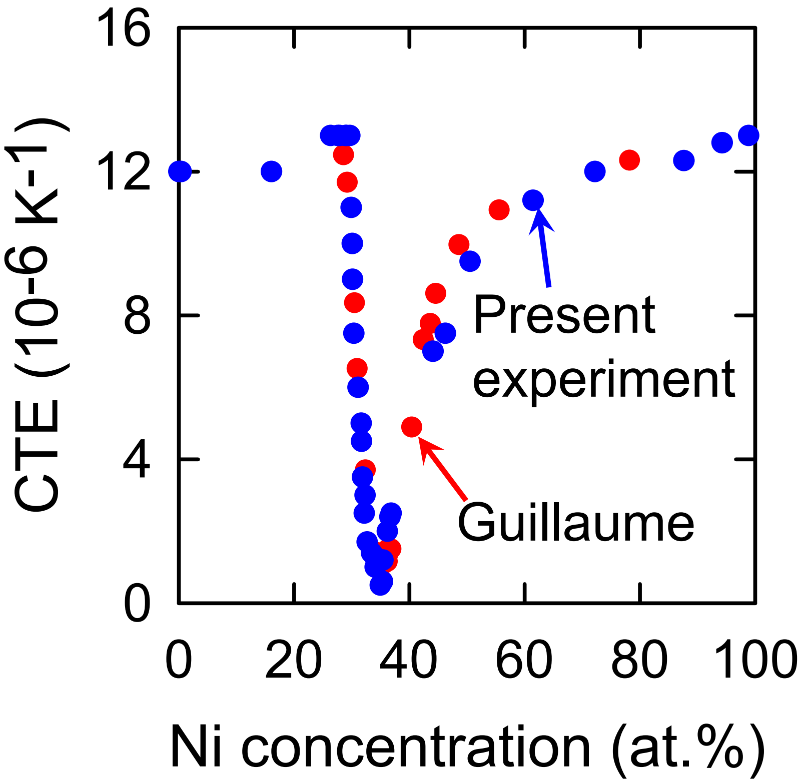
Validation of CTE measurements



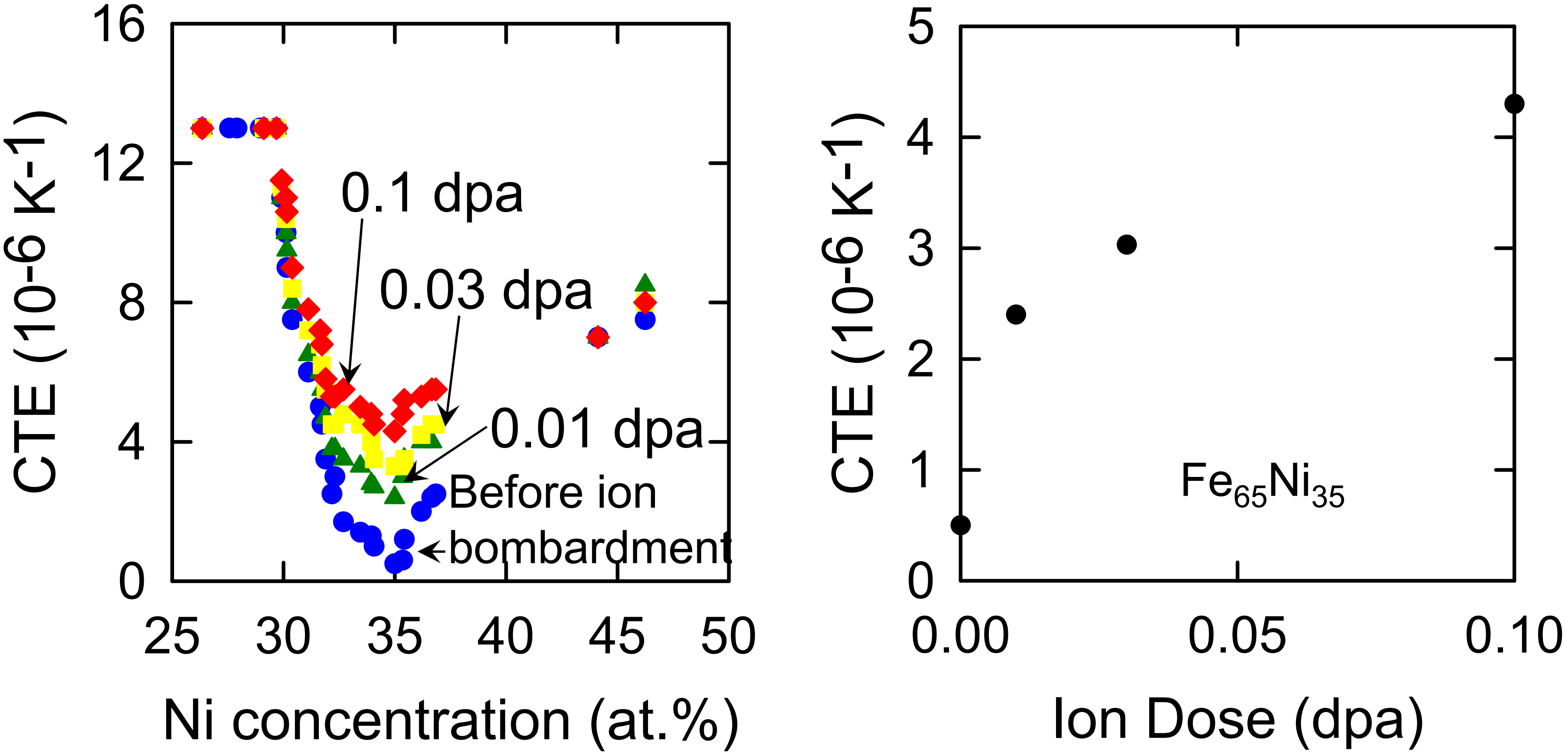

Accuracy: ±6%

CTE of Fe-Ni diffusion couple

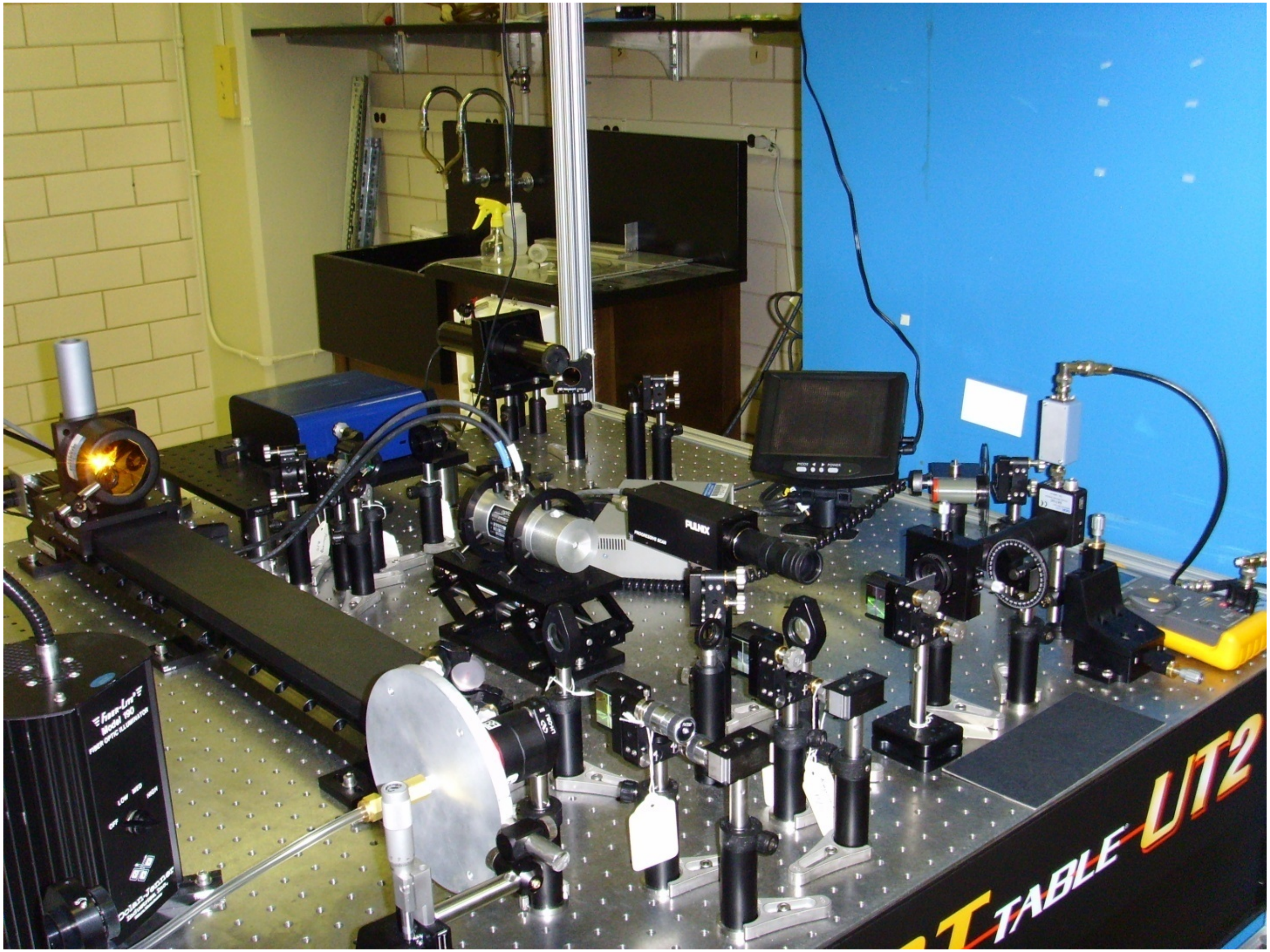




Use ion bombardment to reduce atomic short range order

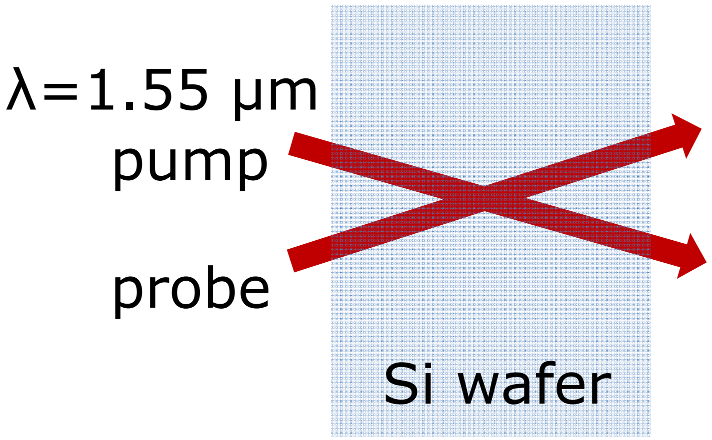


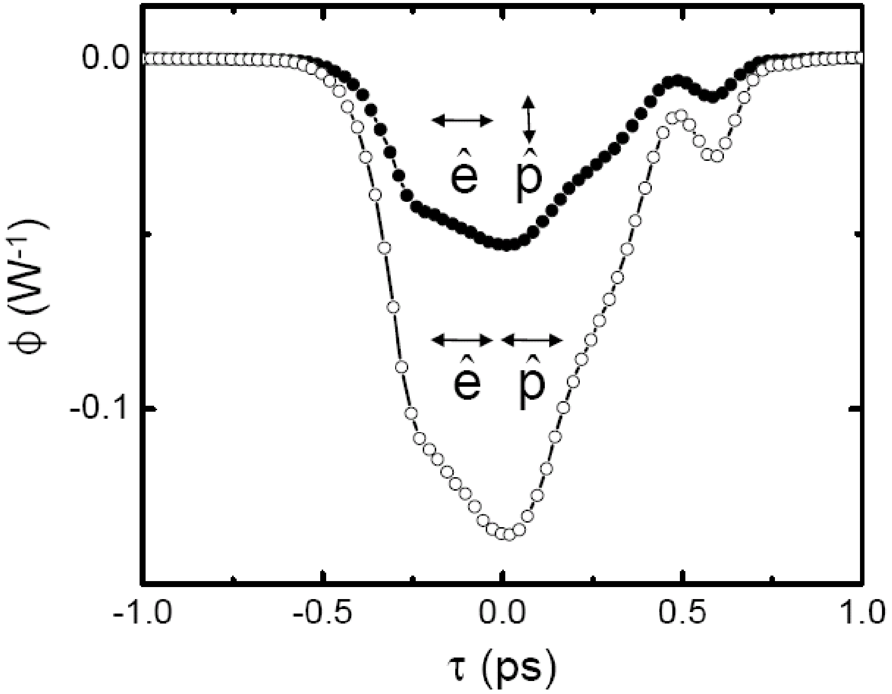
Er:fiber-laser pump-probe system at UIUC

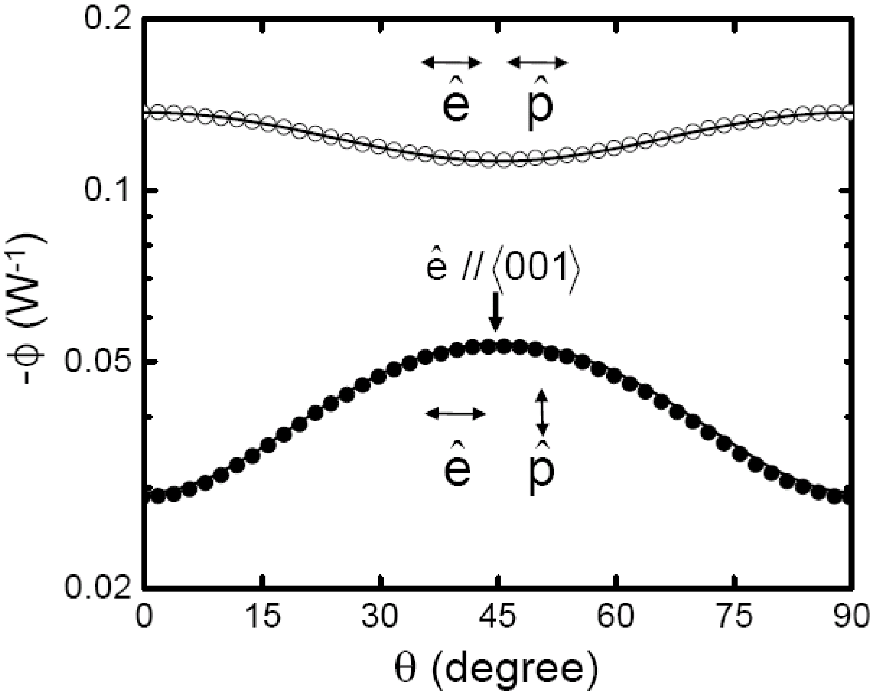


Transient absorption below the band gap of Si

- Pump-probe measurements of two-photon absorption
- Φ is the normalized absorption

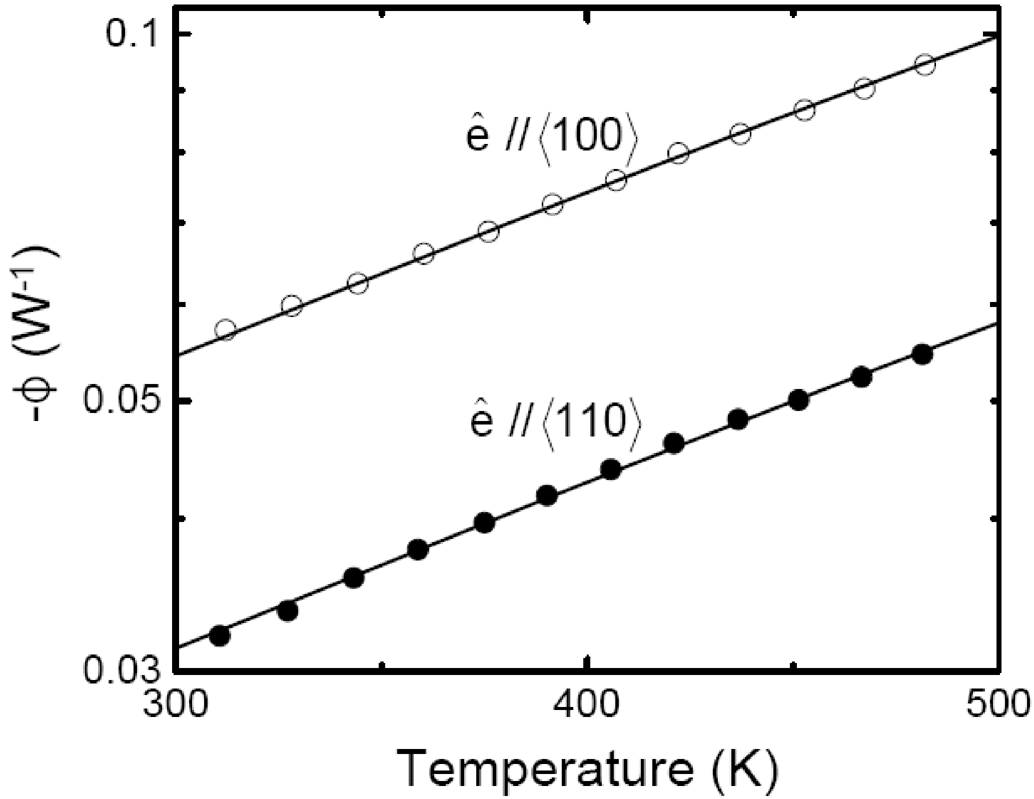






Two-photon (+one phonon) absorption is strongly temperature dependent

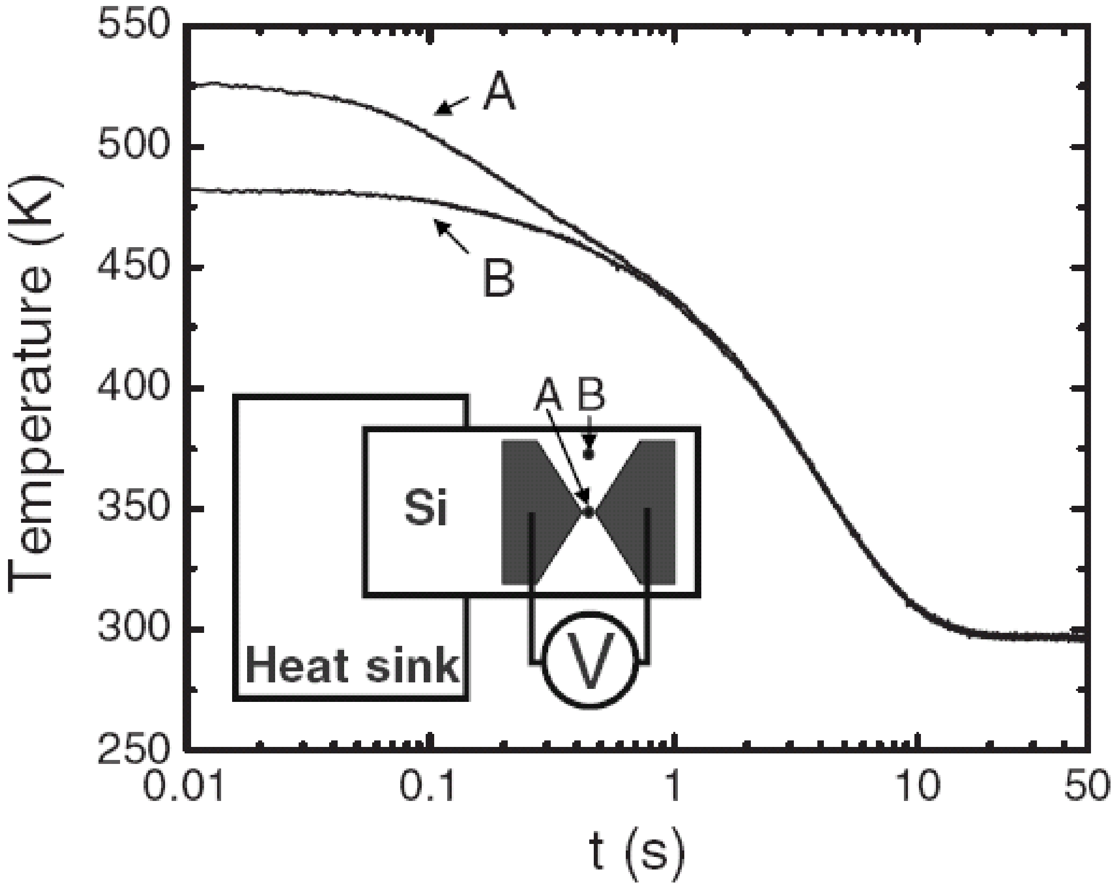
- 470 cm^{-1} (670 K) phonon population controls the optical absorption
- Simple calibration based on well-known physics



$$A(h\nu,T)=D\sum_{i=1}^{4}P_i\left[\frac{\left(h\nu-E_g(T)+\varepsilon_i\right)^2}{\exp\left(\varepsilon_i/kT\right)-1}+\frac{\left(h\nu-E_g(T)-\varepsilon_i\right)^2}{1-\exp\left(-\varepsilon_i/kT\right)}\right].$$

Thermometry by two-photon absorption

- Volume probed is 6×6×50 μm^3
- Sensitivity is < 1 K in a 1 kHz bandwidth



Conclusions

- Time domain thermoreflectance (TDTR) is now a robust and routine method for measuring the thermal conductivity of almost anything (that has a smooth surface).
- Difficult to take advantage of superlative properties of carbon nanotubes because of "thermally weak" interfaces.
- Time domain probe beam deflection (TD-PBD) provides micron-scale measurements of coefficient of thermal expansion
- Transient absorption by below band-gap (1.56 μm) two-photon absorption provides a fast, spatially resolved thermometer in Si