Comparison of the 3ω method and time-domain thermoreflectance

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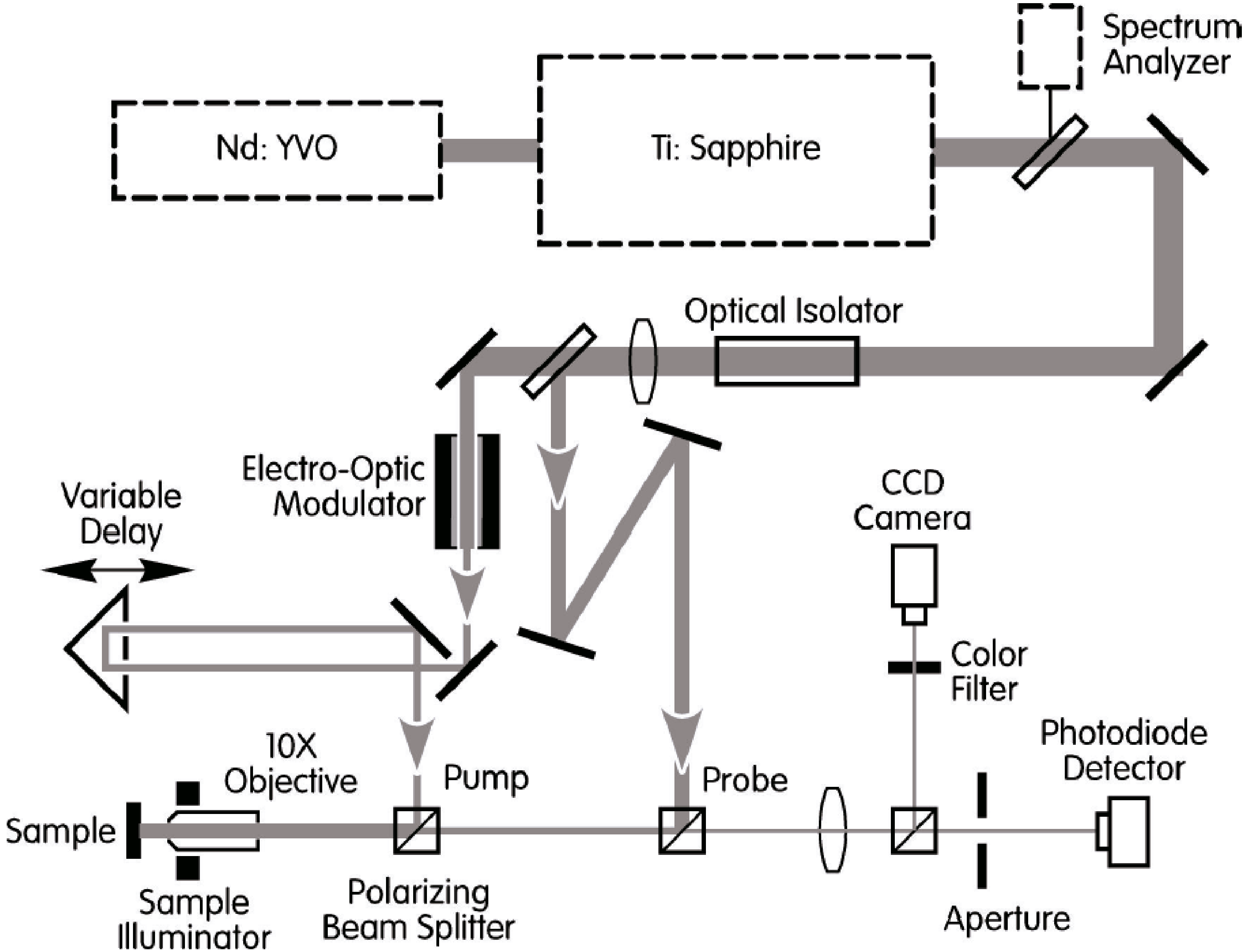
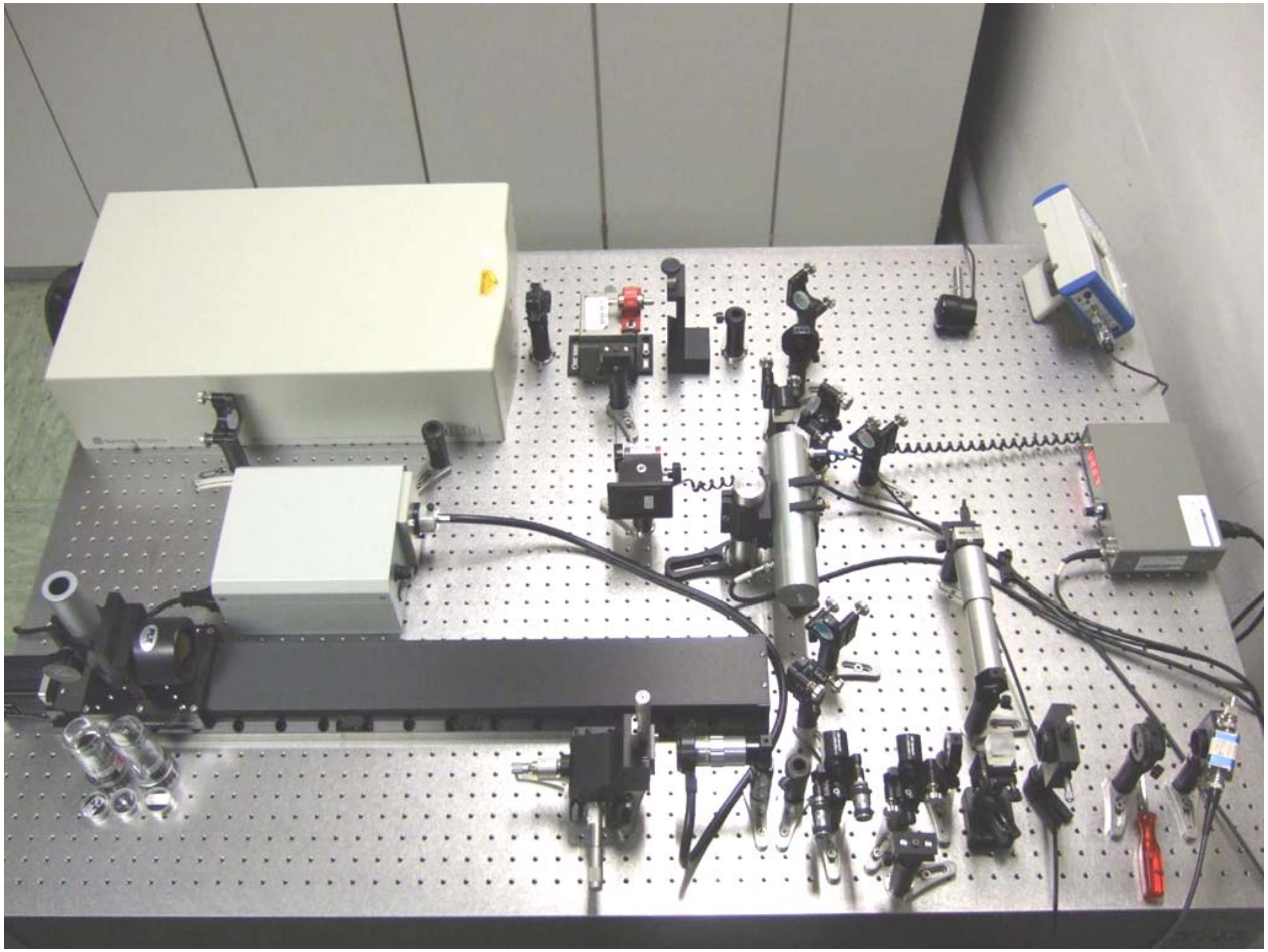
Outline

- Introduction to time-domain thermoreflectance (TDTR)
- Pros and cons: 3ω versus TDTR
- Digression: what limits 3ω accuracy and precision?
- TDTR advantages for high thermal conductivity thin layers, spatial resolution, and semiconductors.
- Additional issues: Frequency dependent thermal conductivity of semiconductor alloys.

Time-domain thermoreflectance



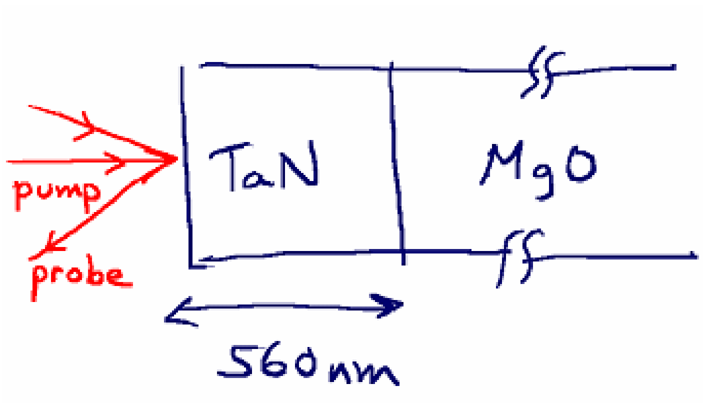
Time-domain thermoreflectance

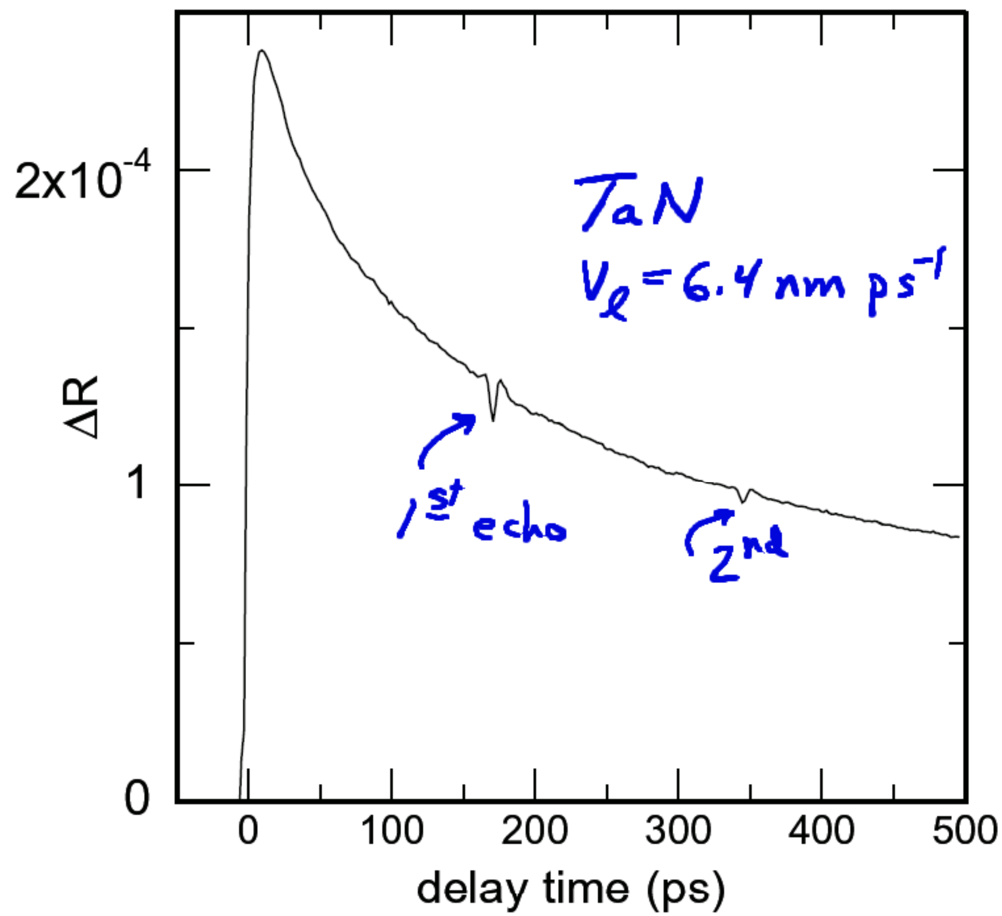


Clone built at Fraunhofer Institute for
Physical Measurement, Jan. 7-8 2008

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

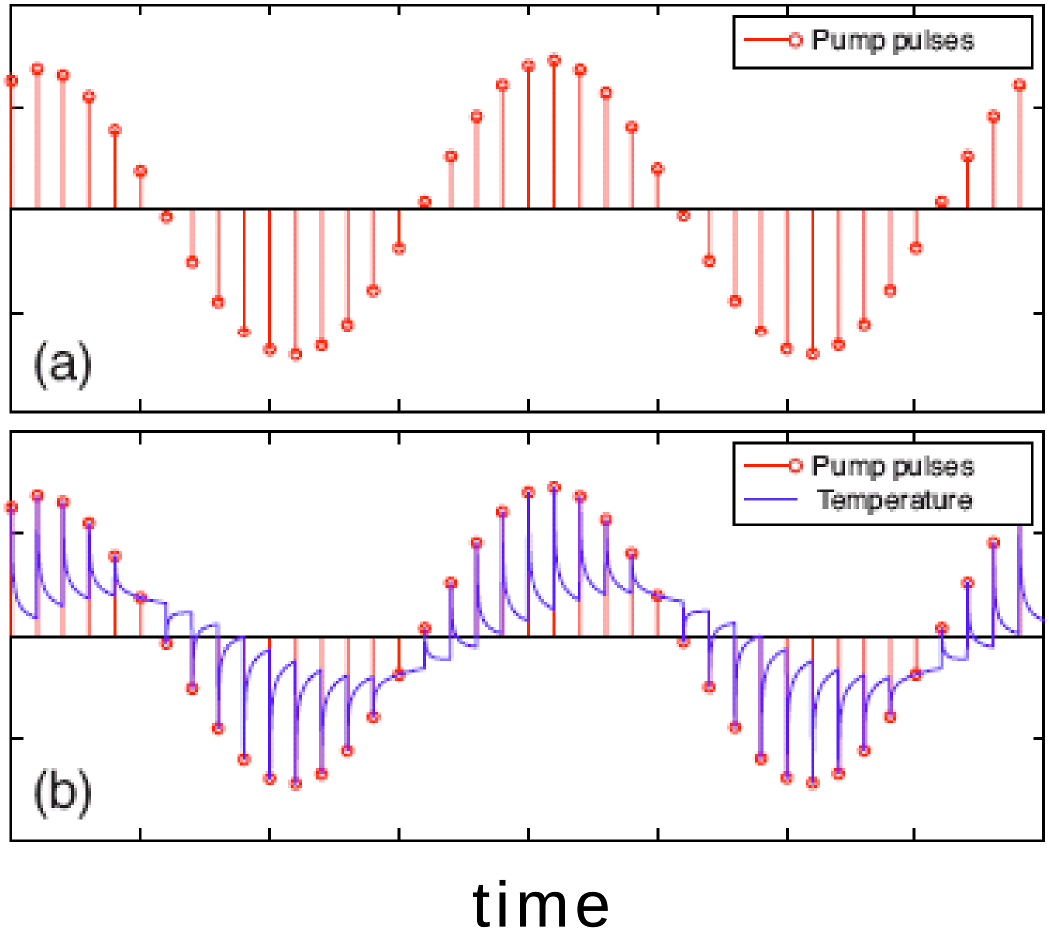




Schmidt et al., RSI 2008

- Heat supplied by modulated pump beam (fundamental Fourier component at frequency f)

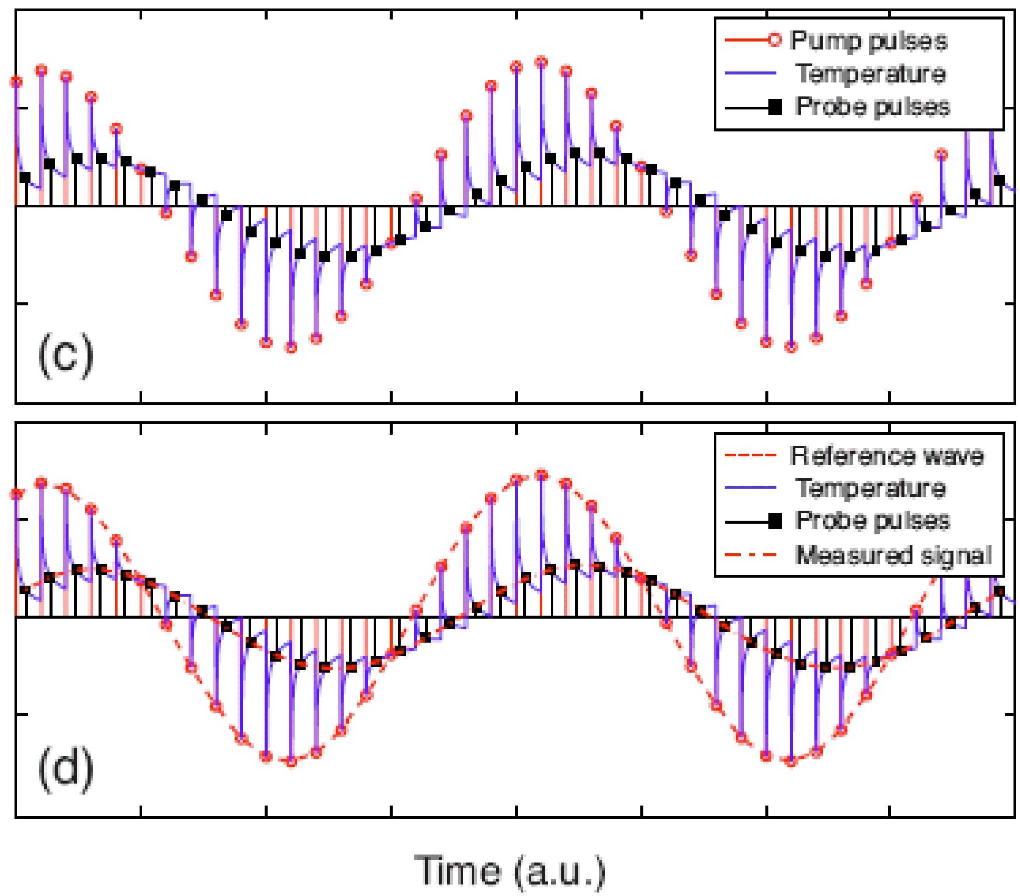
- Evolution of surface temperature



Schmidt et al., RSI 2008

- Instantaneous temperatures measured by time-delayed probe

- Probe signal as measured by rf lock-in amplifier



Analytical solution to 3D heat flow in an infinite half-space, Cahill, RSI (2004)

- spherical thermal wave

$$g(r) = \frac{\exp(-qr)}{2\pi\Lambda r} \quad q^2 = (i\omega/D)$$

- Hankel transform of surface temperature

$$G(k) = \frac{1}{\Lambda(4\pi^2k^2 + q^2)^{1/2}}$$

- Multiply by transform of Gaussian heat source and take inverse transform

$$P(k) = A\exp(-\pi^2k^2w_0^2/2)$$

$$\theta(r) = 2\pi\int_0^\infty P(k)G(k)J_0(2\pi kr)\ k\ dk$$

- Gaussian-weighted surface temperature

$$\Delta T = 2\pi A\int_0^\infty G(k)\exp\left(-\pi^2k^2\left(w_0^2 + w_1^2\right)/2\right)\ k\ dk$$

Iterative solution for layered geometries

$$\begin{pmatrix} B^+ \\ B^- \end{pmatrix}_n = \frac{1}{2\gamma_n} \begin{pmatrix} \exp(-u_n L_n) & 0 \\ 0 & \exp(u_n L_n) \end{pmatrix} \times \begin{pmatrix} \gamma_n + \gamma_{n+1} & \gamma_n - \gamma_{n+1} \\ \gamma_n - \gamma_{n+1} & \gamma_n + \gamma_{n+1} \end{pmatrix} \begin{pmatrix} B^+ \\ B^- \end{pmatrix}_{n+1}$$

$$u_n = \left(4\pi^2 k^2 + q_n^2\right)^{1/2} \qquad q_n^2 = \frac{i\omega}{D_n} \qquad \gamma_n = \Lambda_n u_n$$

$$G(k) = \left(\frac{B_1^+ + B_1^-}{B_1^- - B_1^+}\right) \frac{1}{\gamma_1}$$

Frequency domain solution for 3ω and TDTR are essentially the same

3ω

- “rectangular” heat source and temperature averaging.
- One-dimensional Fourier transform.
- “known” quantities in the analysis are Joule heating and dR/dT calibration.

TDTR

- Gaussian heat source and temperature averaging.
- Radial symmetric Hankel transform.
- “known” quantity in the analysis is the heat capacity per unit area of the metal film transducer.

TDTR signal analysis for the lock-in signal as a function of delay time *t*

- In-phase and out-of-phase signals by series of sum and difference over sidebands

$$\mathrm{Re}\left[\Delta R_M(t)\right] = \frac{dR}{dT}\sum_{m=-M}^{M}\left(\Delta\mathcal{T}(m/\tau+f)+\Delta\mathcal{T}(m/\tau-f)\right)\exp(i2\pi mt/\tau)$$

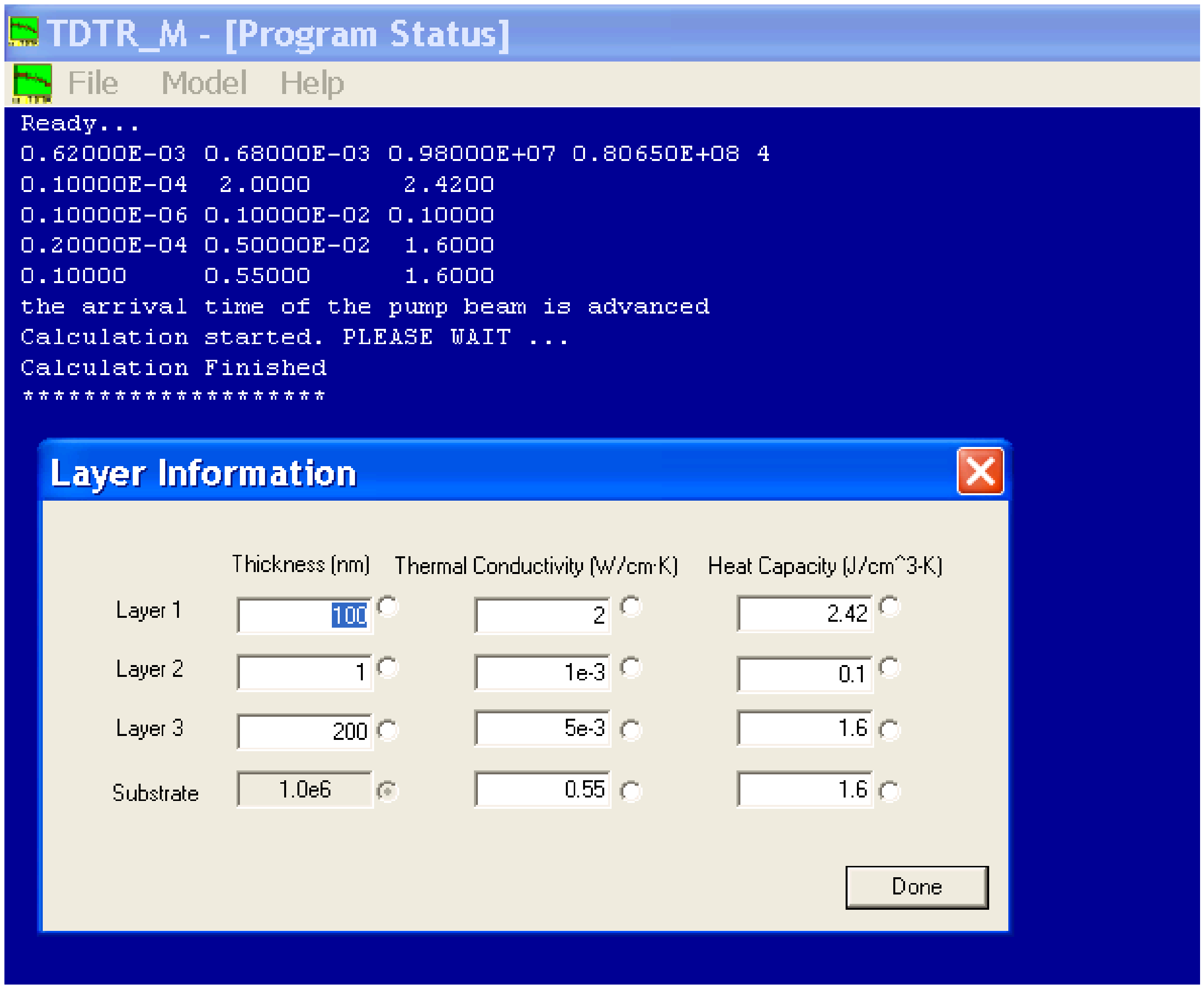
$$\mathrm{Im}\left[\Delta R_M(t)\right] = -i\frac{dR}{dT}\sum_{m=-M}^{M}\left(\Delta\mathcal{T}(m/\tau+f)-\Delta\mathcal{T}(m/\tau-f)\right)\exp(i2\pi mt/\tau)$$

- out-of-phase signal is dominated by the $m=0$ term (frequency response at modulation frequency *f*)

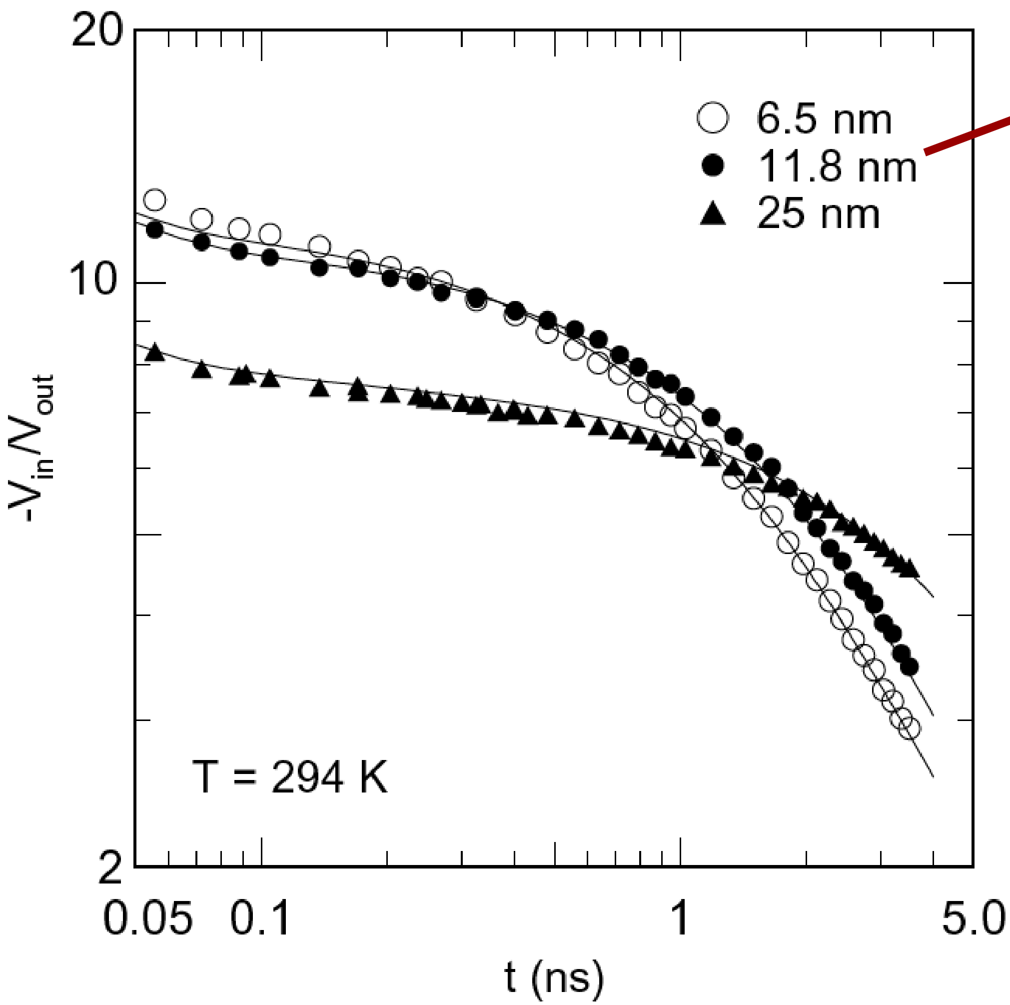
Windows software

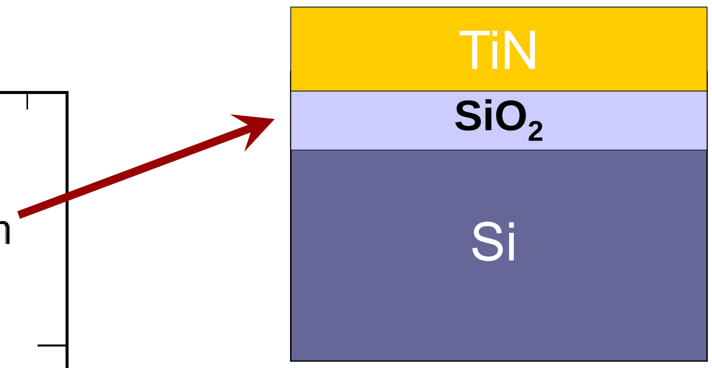
author: Catalin Chiritescu,

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



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

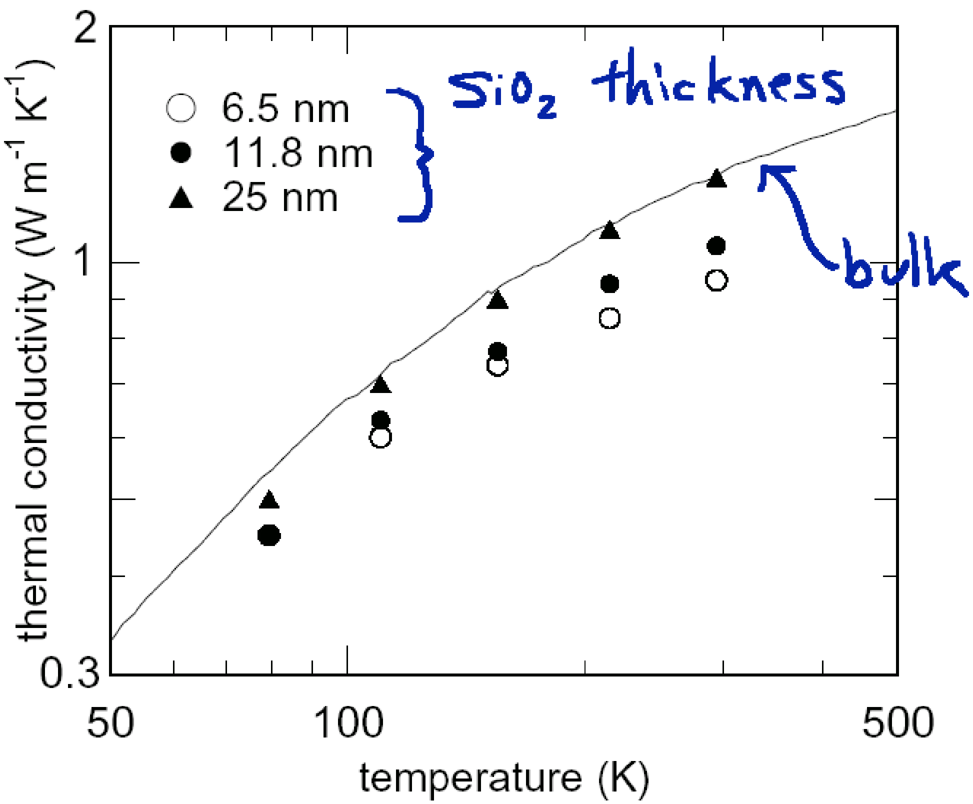


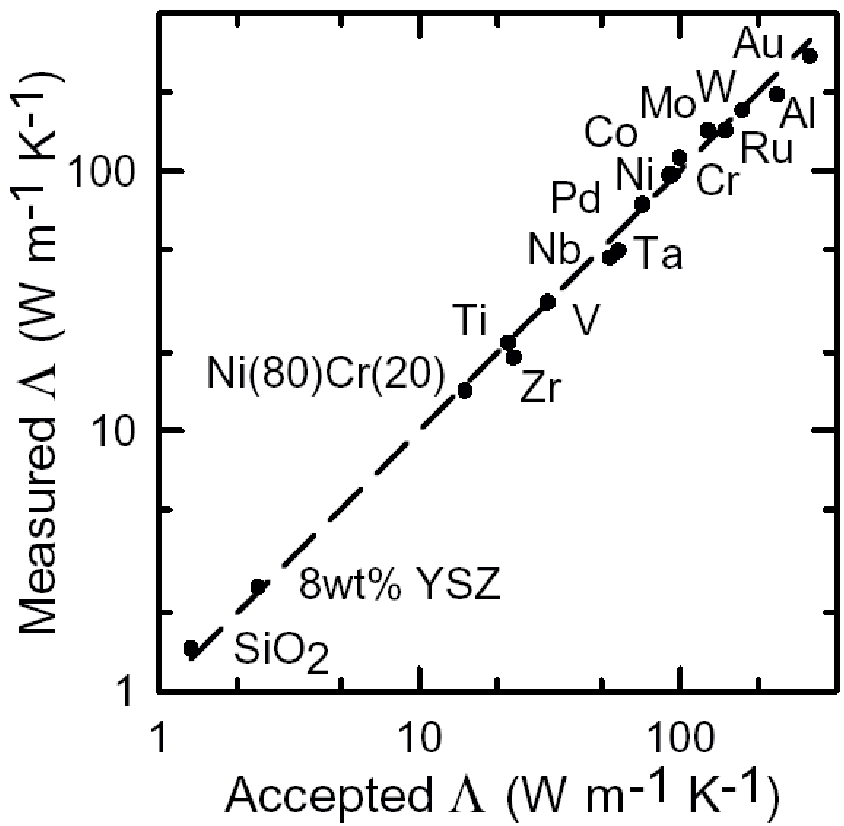


- reflectivity of a metal depends on temperature
- one free parameter: the "effective" thermal conductivity of the thermally grown SiO_2 layer (interfaces not modeled separately)

Costescu *et al*., PRB (2003)

TDTR: early validation experiments





Costescu *et al*., PRB (2003)

Zhao *et al*., Materials Today (2005)

Each have advantages and disadvantages

<u>3ω</u>

- High accuracy, particularly for bulk materials and low thermal conductivity dielectric films
- Accuracy is reduced for semiconducting thin films and high thermal conductivity layers
  - Need electrical insulation: introduces an additional thermal resistance.
  - Cannot separate the metal/film interface thermal conductance from the thermal conductivity
- Wide temperature range ($30 < T < 1000$ K)
  - But very high temperatures are not usually accessible for semiconductors

Each have advantages and disadvantages

TDTR

- Accuracy is typically limited to several percent due to uncertainties in the many experimental parameters
  - Metal film thickness
  - Heat capacity of the sample if film is thick
- But many experimental advantages
  - No need for electrical insulation
  - Can separate the metal/film interface thermal conductance from the thermal conductivity
  - High spatial resolution
  - Only need optical access: high pressures, high magnetic fields, high temperatures

Digression: what limits the accuracy of 3ω data?

- 1990's: approximations made for low thermal conductivity film on high thermal conductivity substrate and film thickness<heater-width
  - No need for those approximations now. Feldman and co-workers (1999), and others shortly after, pointed out that a transfer matrix approach for layered geometries is equally applicable for linear and radial heat flow.
  - DOS program: multi3w.exe available at users.mrl.uiuc.edu/cahill/tcdata.html
  - Anisotropy is easy to add

Digression: what limits the accuracy of 3ω data?

- Contributions from the heater line.
  - Not explicitly included in the heat flux boundary conditions of the solutions
  - Heat capacity matters at very high frequencies, see, for example, Tong *et al*. RSI (2006).
  - Lateral heat flow in heater line was considered recently by Gurrum *et al*., JAP (2008).

Digression: what limits the accuracy of 3ω data?

- In my experience, the dR/dT calibration is the biggest issue.
  - use physics to fix the calibration

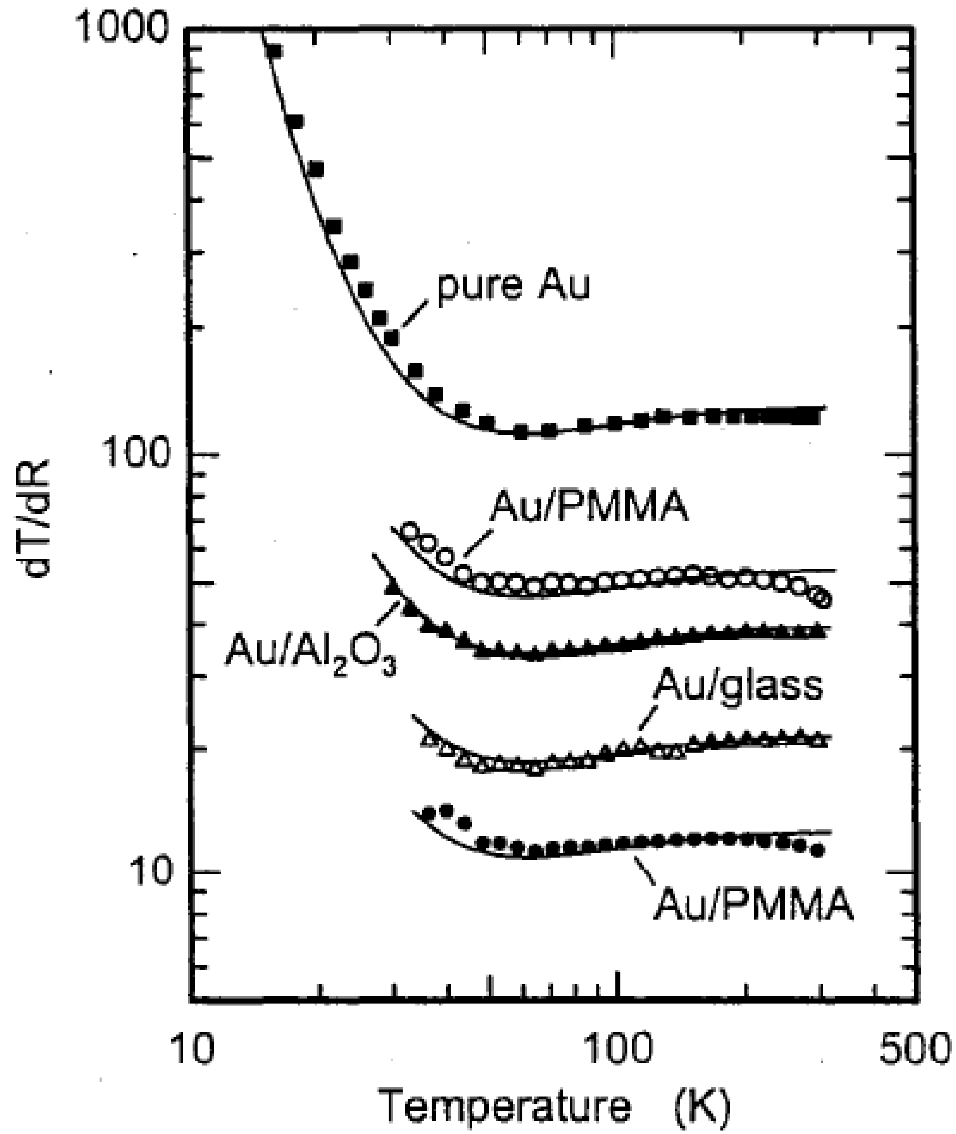
$$\mathrm{R}(T) = \frac{l}{A}\, \varrho_{\mathrm{BG}}(T) + \mathrm{R}_o,$$

Bloch-Grüneisen resistivity of a metal

$$\varrho_{\mathrm{BG}}(T) = C_{\mathrm{BG}} \left(\frac{T}{\theta_D}\right)^5 \int_0^{\theta_D/T} \frac{z^5}{(\exp(z)-1)(1-\exp(-z))}\, dz,$$

Calibration of Au thermometer line

- Materials with large coefficient of thermal expansion create an interesting problem
  - during calibration of R(T) substrate strain is homogeneous
  - but during 3ω measurement, ac strain field is complex so the determination of dR/dT is not really correct.

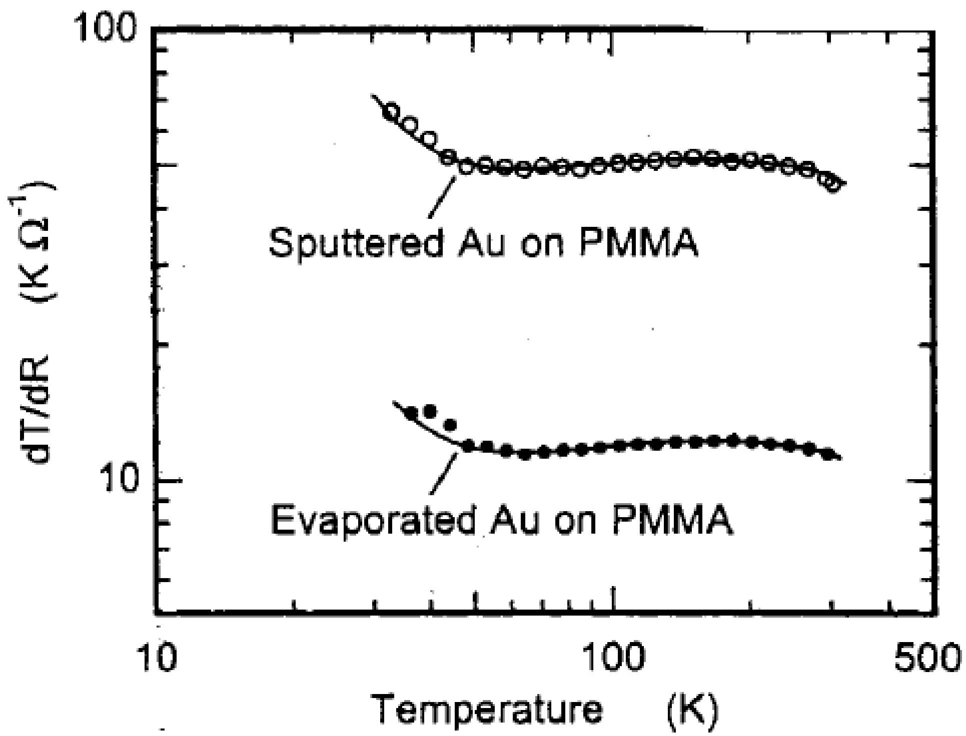


High thermal expansion coefficients

- Add terms to account for effect of strain on the Bloch-Grüneisen resistivity and the residual resistivity.

$$\mathrm{R}(T) = \frac{l}{A}\, \varrho_{\mathrm{BG}}(T)\, [1 + c_3\, 5.75\, (\alpha(T) - \alpha(T_o))]$$

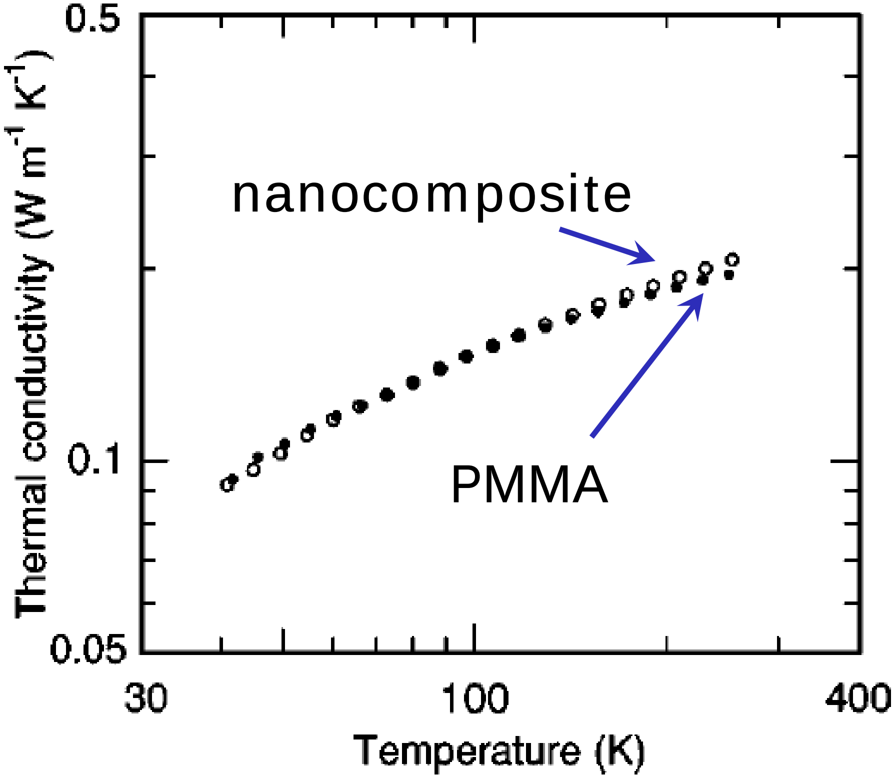
$$+ \mathrm{R}_o\, [1 + c_3\, 2.45\, (\alpha(T) - \alpha(T_o))]\,,$$

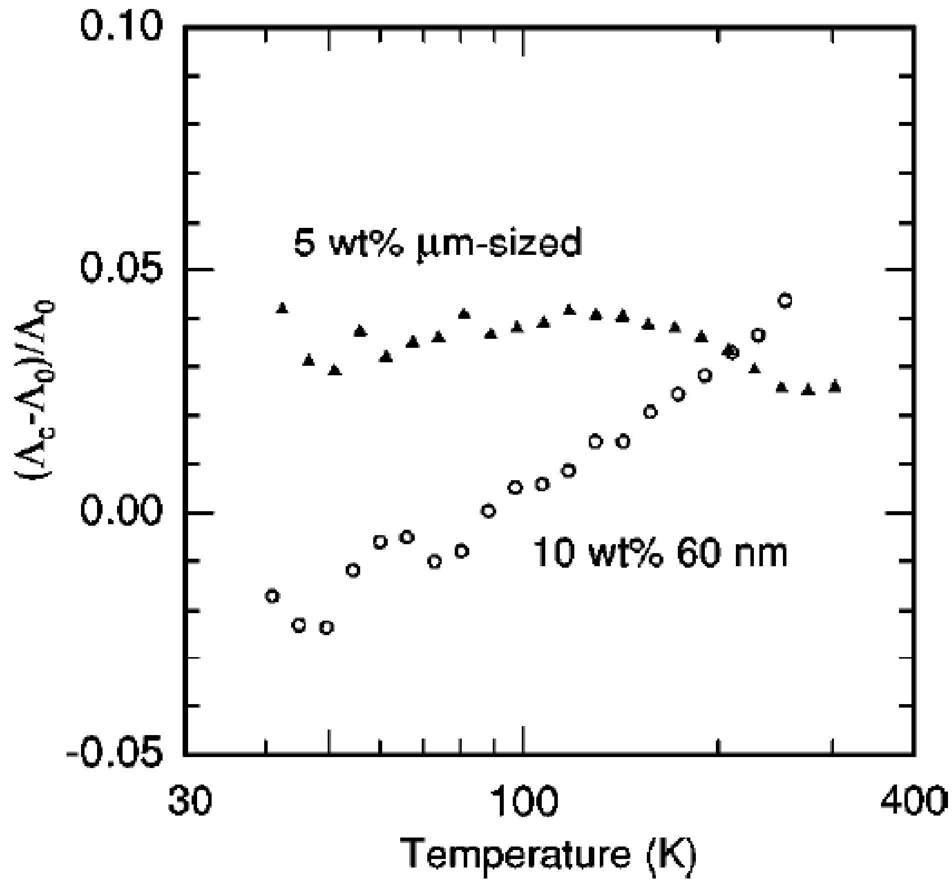


- CTE of PMMA is ≈50 ppm/K
- CTE of PbTe is ≈20 ppm/K

Highest precision measurements at Illinois using 3ω: polymer nanocomposites

- PMMA mixed with 60 nm γ-Al_2O_3 nanoparticles

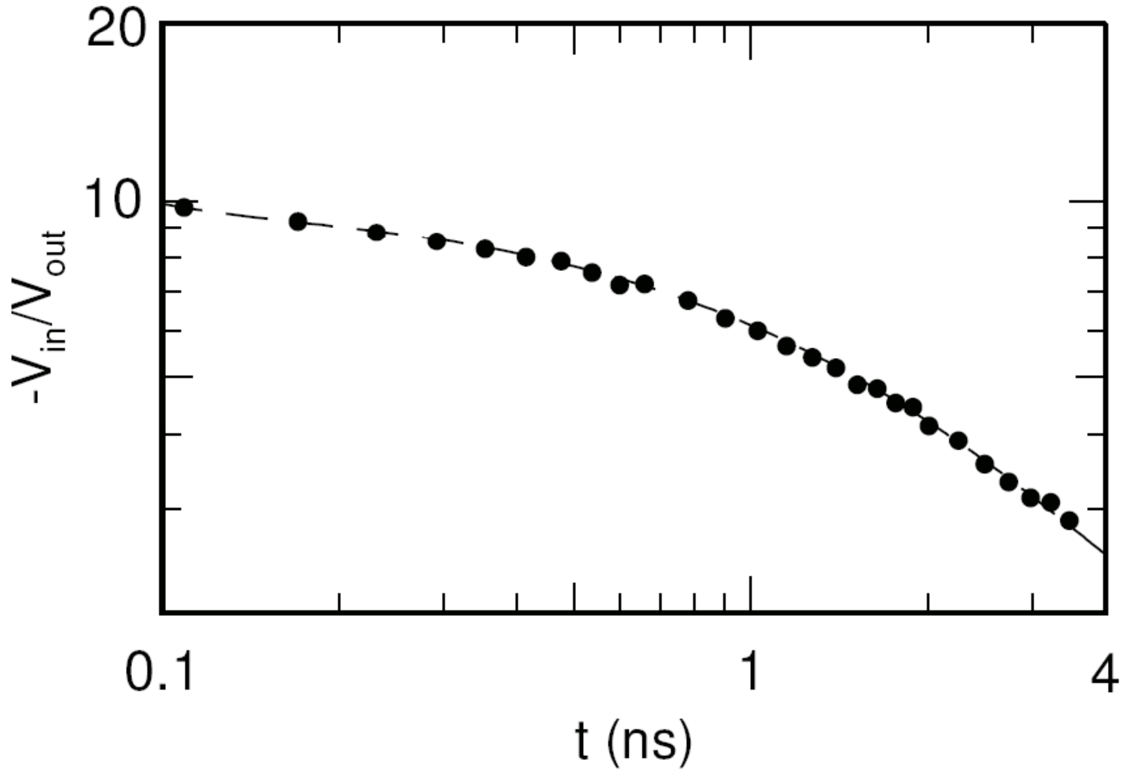




Putnam *et al*., JAP (2003)

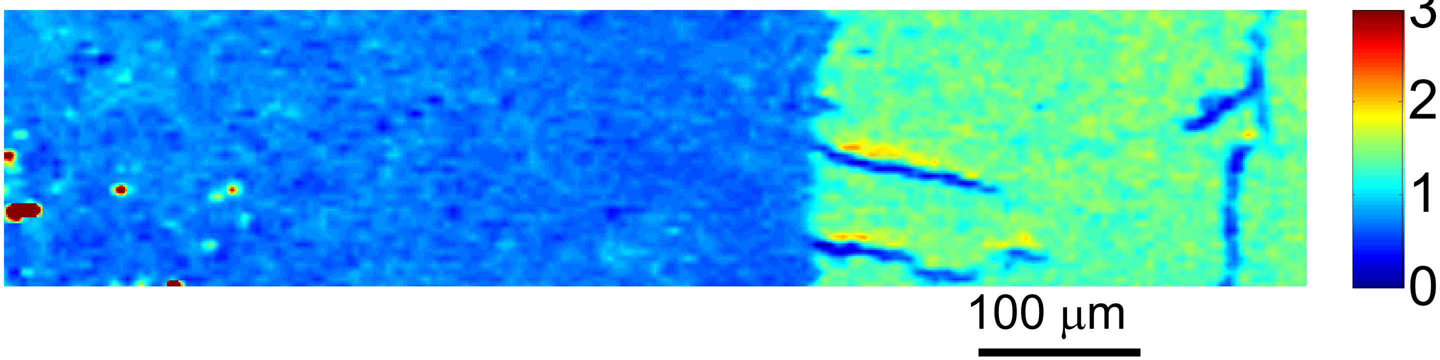
Something not possible with 3ω: TDTR data for isotopically pure Si epitaxial layer on Si

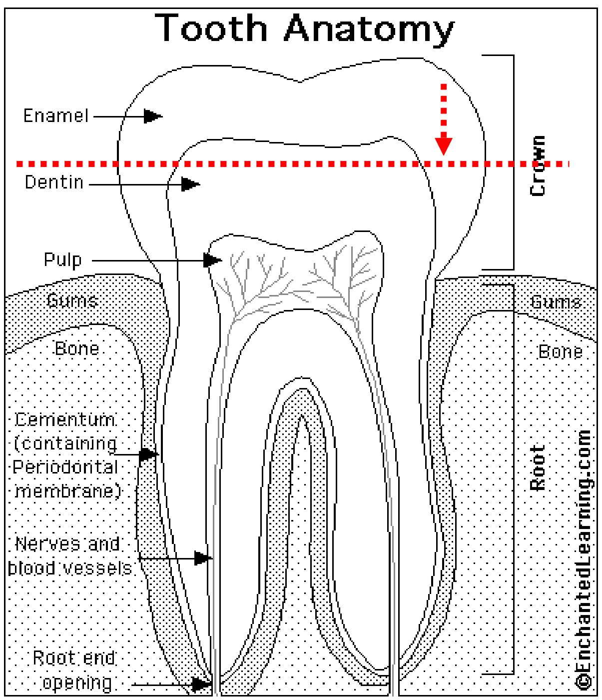
- Two free fitting parameters
  - thermal conductivity, 165 W/m-K
  - Al/Si interface conductance, 185 MW/m^2-K



Cahill *et al*., PRB (2004)

Thermal conductivity map of a human tooth





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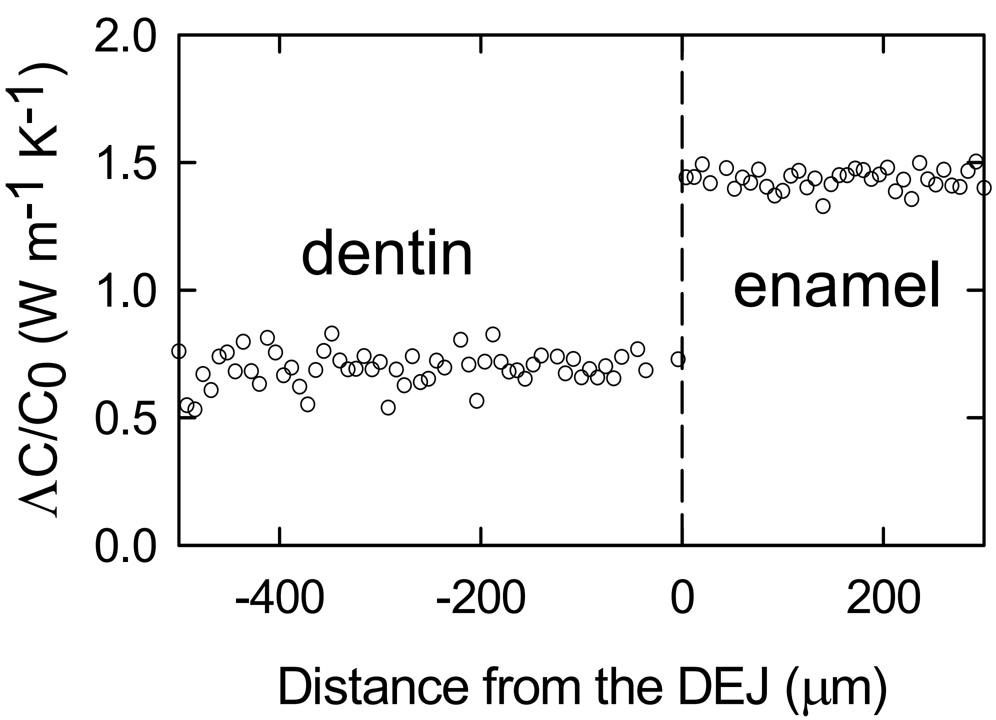
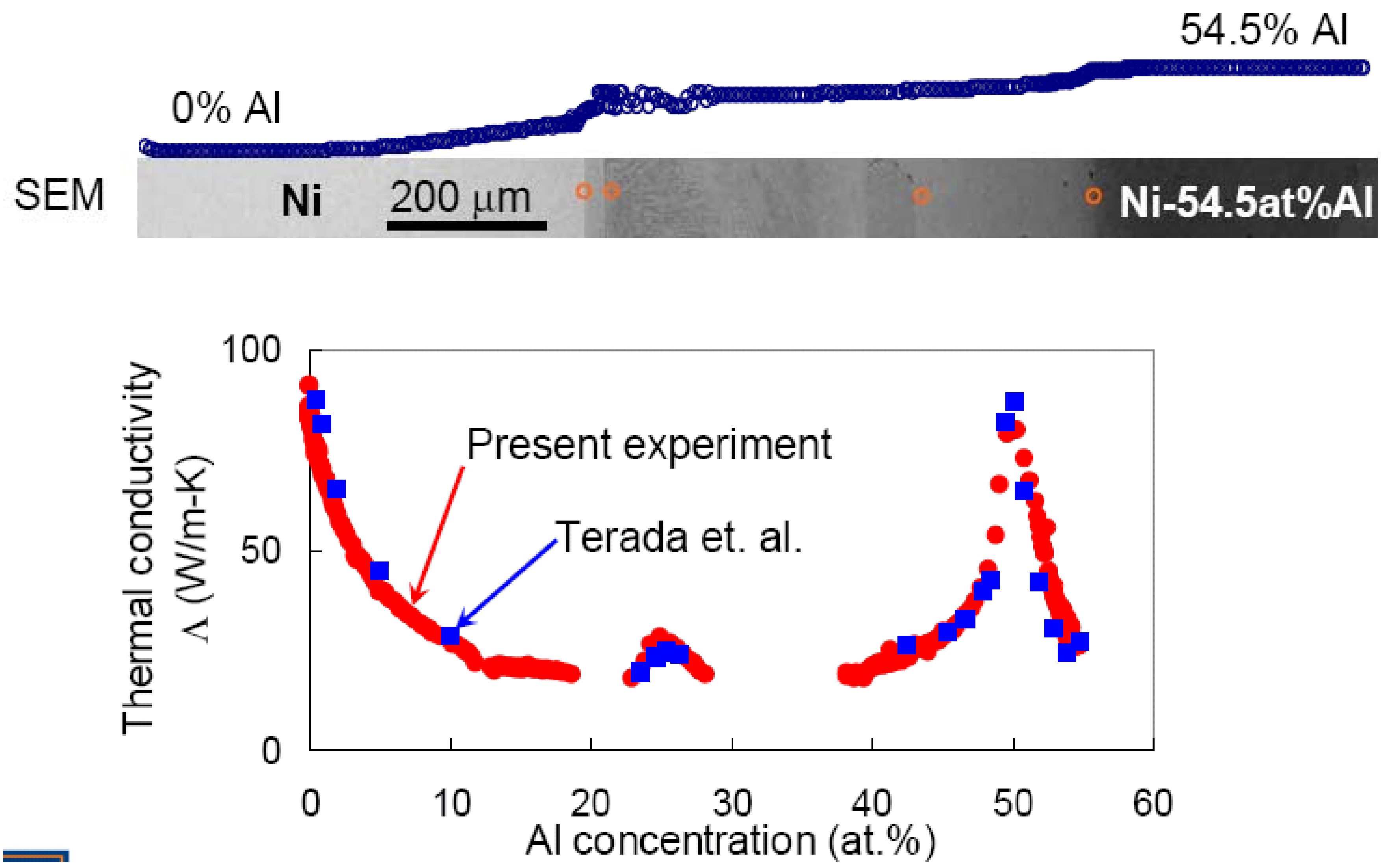
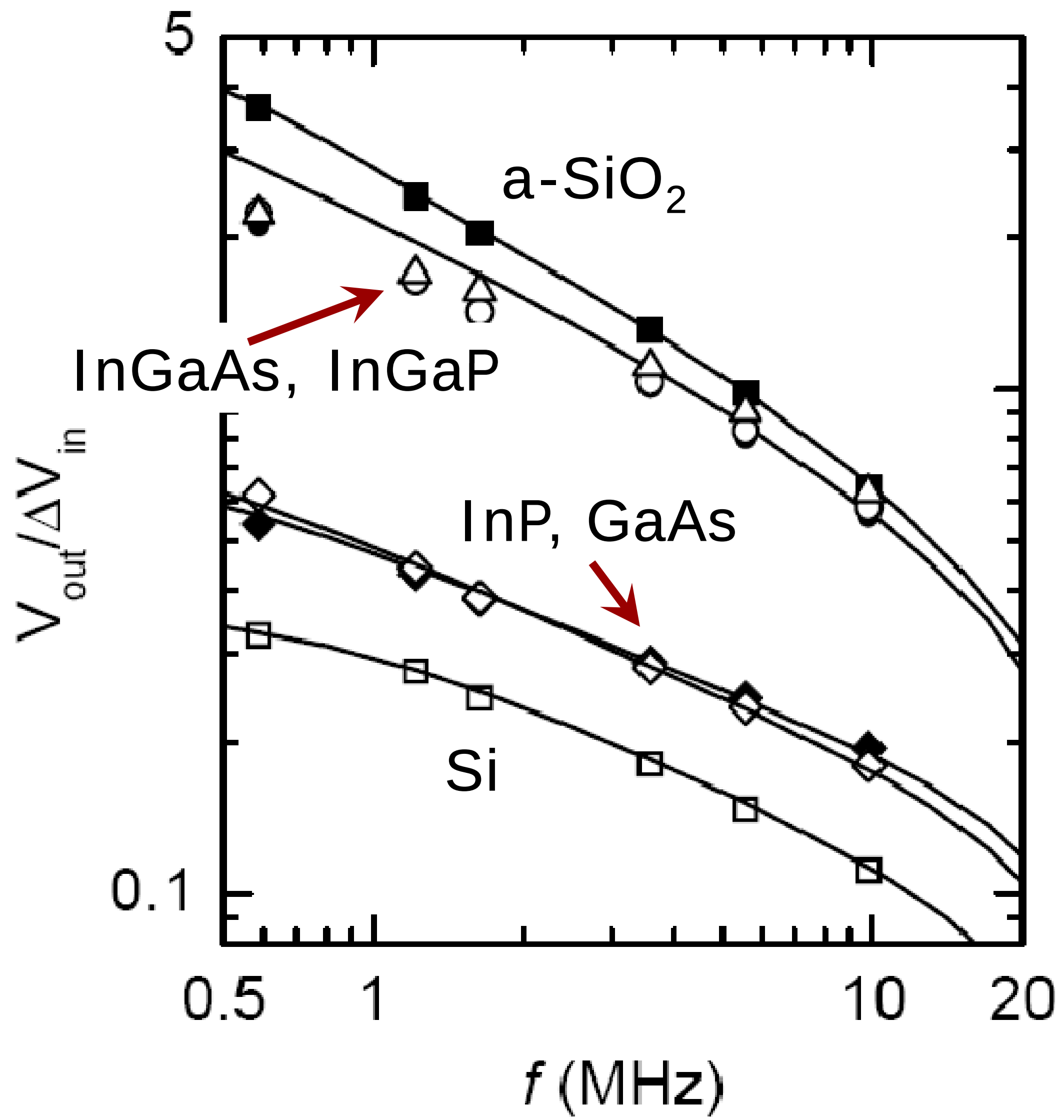


High throughput data using diffusion couples



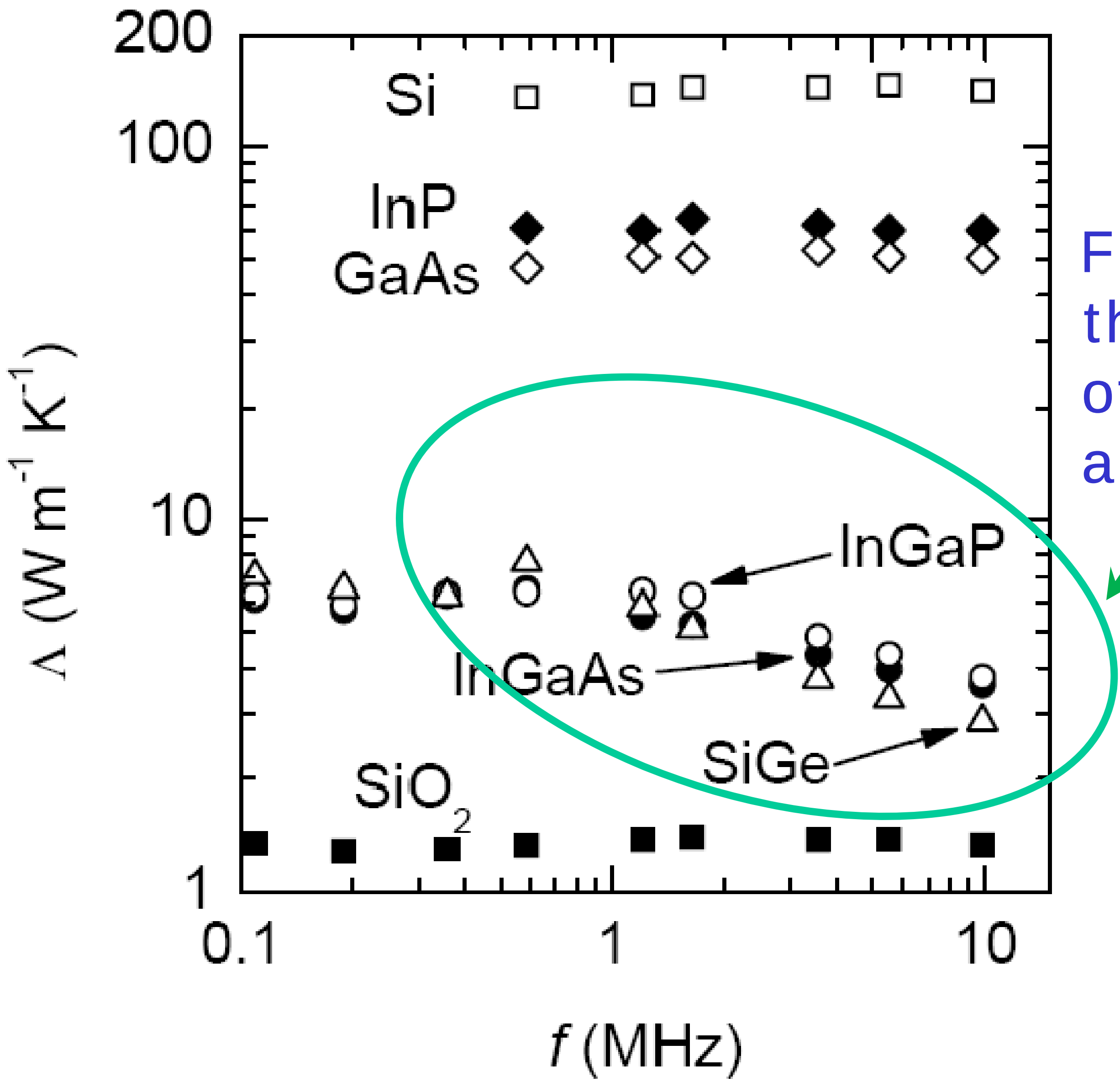
Thermoreflectance raw data at t=100 ps

- fix delay time and vary modulation frequency *f*.
- Change in V_{in} doesn't depend on *f*. V_{out} mostly depends on $(f\Lambda C)^{-1/2}$
- semiconductor alloys show deviation from fit using a single value of the thermal conductivity



Koh and Cahill PRB (2007)

Same data but fit Λ at each frequency *f*



Frequency dependent thermal conductivity of semiconductor alloys

Koh and Cahill PRB (2007)

How can thermal conductivity be frequency dependent at only a few MHz?

- $2\pi f\tau << 1$ for phonons that carry significant heat. For dominant phonons, τ ~50 ps, and $2\pi f\tau$ ~ 10^{-3}.
- But the thermal penetration depth d is not small compared to the dominant mean-free-path ℓ_{dom}.

$$d = \sqrt{\Lambda / \pi C f}$$

- Ansatz: phonons with $\ell(\omega) > d$ do not contribute to the heat transport in this experiment.
- True only if the “single-relaxation-time approximate” fails strongly. For single relaxation time τ, $\ell << d$ because $f\tau << 1$.

For non-equilibrium, add effusivity instead of conductivity

- Consider a "two-fluid" model with

$$\Lambda_1 \approx \Lambda_2$$

$$C_1 >> C_2$$

- Equilibrium,

$$(\Lambda C)^{1/2} = [(\Lambda_1 + \Lambda_2)(C_1 + C_2)]^{1/2}$$

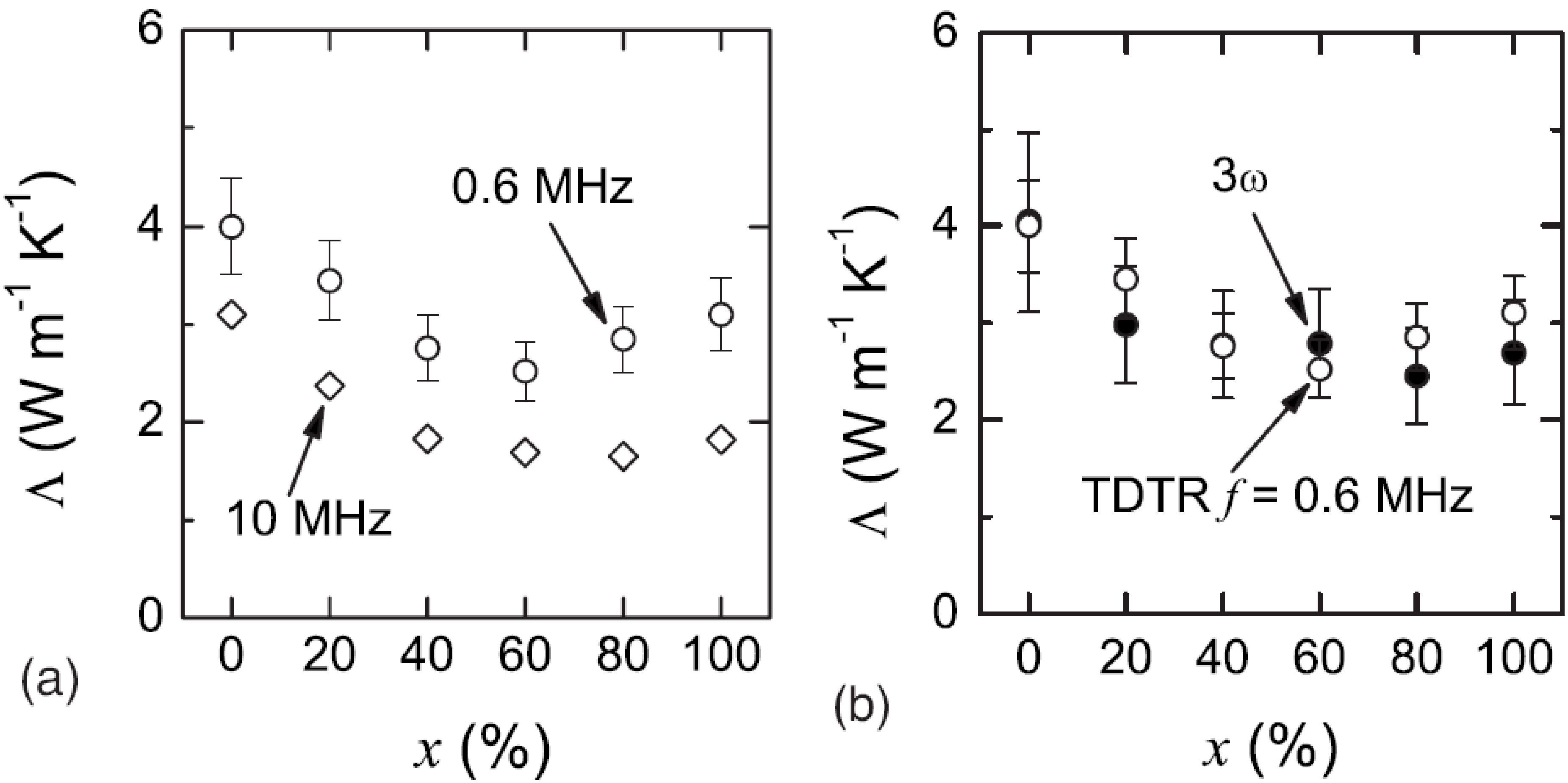
- Out-of-equilibrium,

$$(\Lambda C)^{1/2} = (\Lambda_1 C_1)^{1/2} + (\Lambda_2 C_2)^{1/2}$$

$$\approx (\Lambda_1 C_1)^{1/2}$$

f<1 MHz frequency TDTR agrees with 3ω

2 μm thick $(In_{0.52}Al_{0.48})_x(In_{0.53}Ga_{0.47})_{1-x}As$



Koh *et al*., JAP (2009)

Summary and Conclusions

- Usually, 3ω has higher accuracy because Joule heating and dR/dT calibration are electrical measurements and geometry is precisely known.
- For semiconducting thin films, because of extra thermal resistance of electrical isolation layers, accuracy of TDTR is comparable.
- TDTR has tremendous advantages in experimental convenience—once the high initial cost and set-up has been overcome.