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Measurements of thermal transport at the nanoscale using kHz and ultrafast thermal waves

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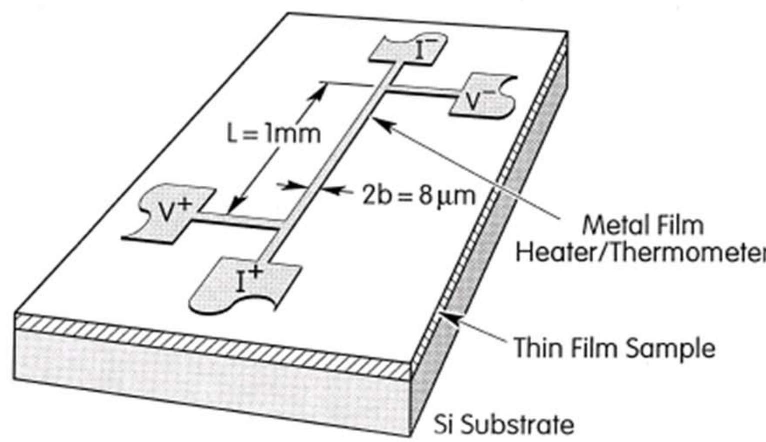
Outline

- 3ω method
- Time-domain thermoreflectance (TDTR)
- The heatl diffusion equation in not completely valid at high frequencies: frequency dependent thermal conductivity and ballistic phonon transport.

3ω Method

Uses single metal film for heater/thermometer (Birge, 1987); (Cahill, 1990).



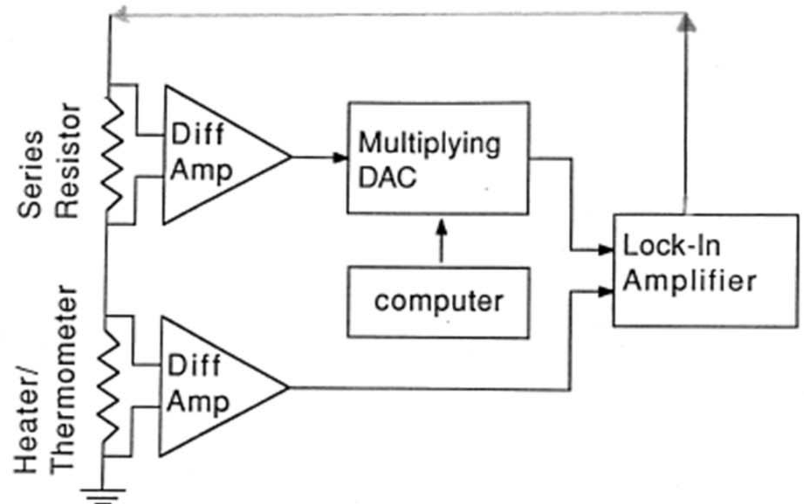
$$I \sim \exp(i\omega t)$$

$$P \sim \exp(i2\omega t) \;; \qquad \Delta T \sim \exp(i2\omega t)$$

$$\Delta R \sim \exp(i2\omega t)$$

$$\Delta V = I \Delta R \sim \exp(i3\omega t)$$

Measure ΔV with a bridge circuit and digital lockin amplifier.



Heat flow in a radial coordinate r.

$$\nabla^2 T - q^2 T = 0 \qquad q^2 = \frac{i\omega}{D}; \quad D = \frac{\Lambda}{C}$$

$$\nabla^2 = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial}{\partial r}\right).$$

Solution for an infinite half-space

$$\Delta T(r) = \frac{P}{l\pi\Lambda} K_0(qr)$$

K_0 is the zeroth order modified Bessel function
Think of this as the circular thermal wave

Take the Fourier transform of this frequency domain solution

$$\Delta T(k) = \int_0^\infty \Delta T(x)\cos(kx)\,dx,$$

$$\Delta T(k) = (P/2l\Lambda)[1/(k^2+q^2)^{1/2}].$$

For a low thermal conductivity thin film
on a high thermal conductivity substrate

$$\Delta T(\omega) = \Delta T_s + \Delta T_f$$

$$\Delta T_s = \frac{P}{l\pi\Lambda_s}\int_0^\infty \frac{\sin^2(kb)}{(kb)^2(k^2+q^2)^{1/2}}\,dk$$

$$\Delta T_f = \frac{Pd}{2Lb\Lambda_f}$$

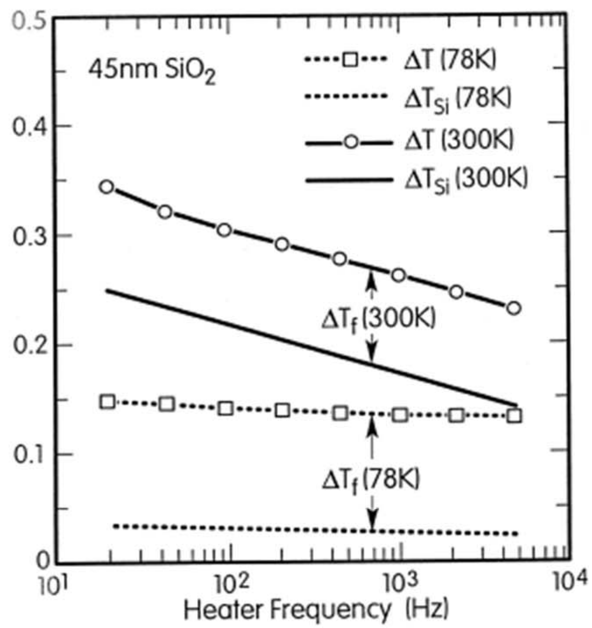
$$q^2 = \frac{2i\omega}{D_s}$$

(Factor of 2 because current is at frequency ω)

When $|q|b < 0.3$

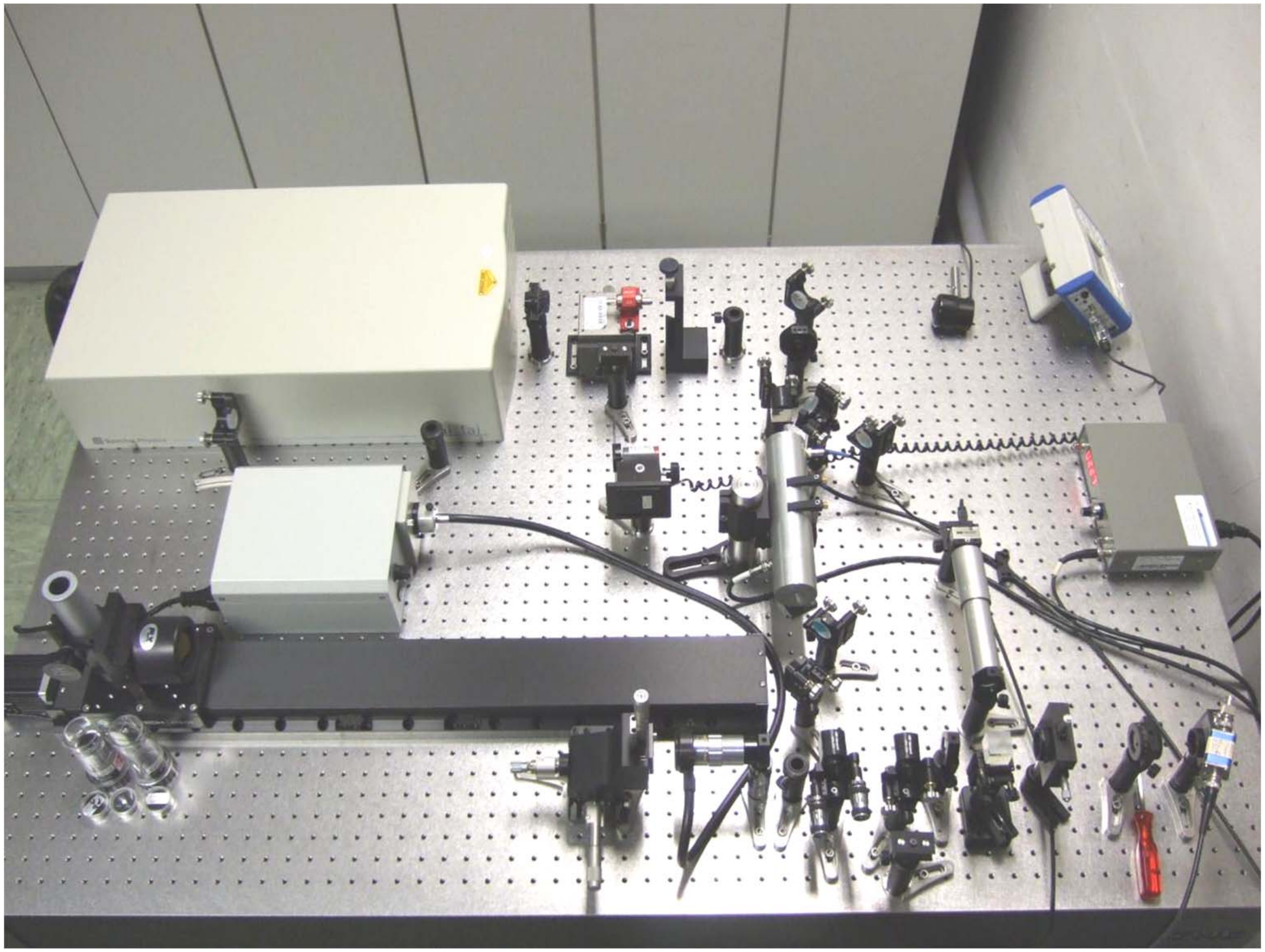
$$\Delta T_s(\omega) = \frac{P}{l\pi\Lambda_s}\left[\frac{1}{2}\ln\left(\frac{D_s}{b^2}\right) + \eta - \frac{1}{2}\ln(2\omega) - \frac{i\pi}{4}\right]$$

numerically $|q|b \ll 1$, gives $\eta = 0.92$. Empirically, $\eta \simeq 1.05$.



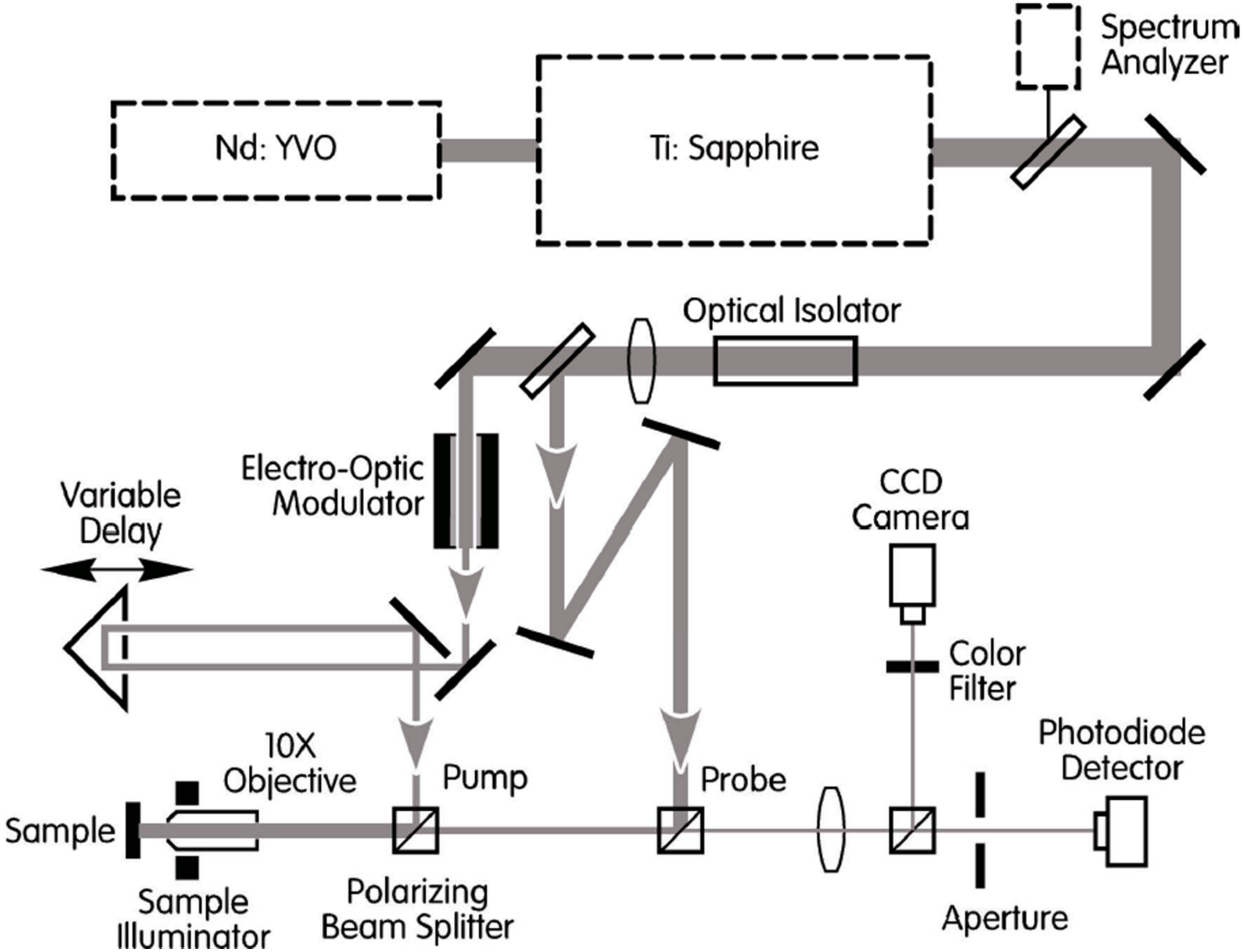
In-phase ΔT for a 45 nm SiO_2 film deposited on a Si wafer (Lee, 1996). Curves ΔT_{Si} are the calculated response of the substrate.

Time-domain thermoreflectance



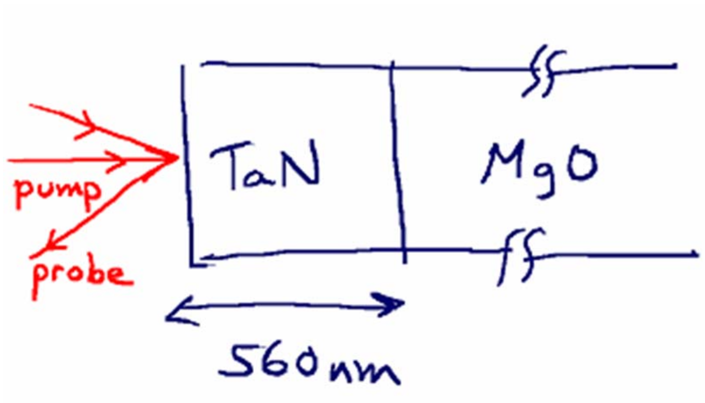
Clone built at Fraunhofer Institute for Physical Measurement, Jan. 7-8 2008

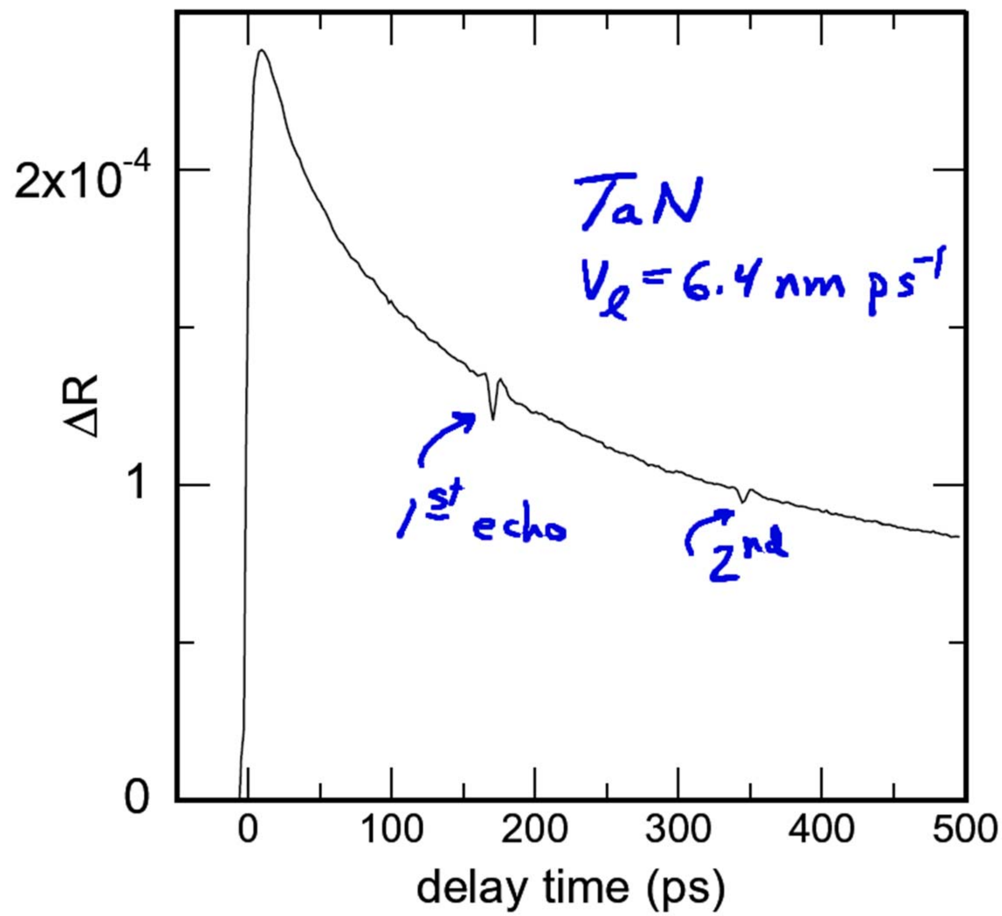
Time-domain thermoreflectance



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

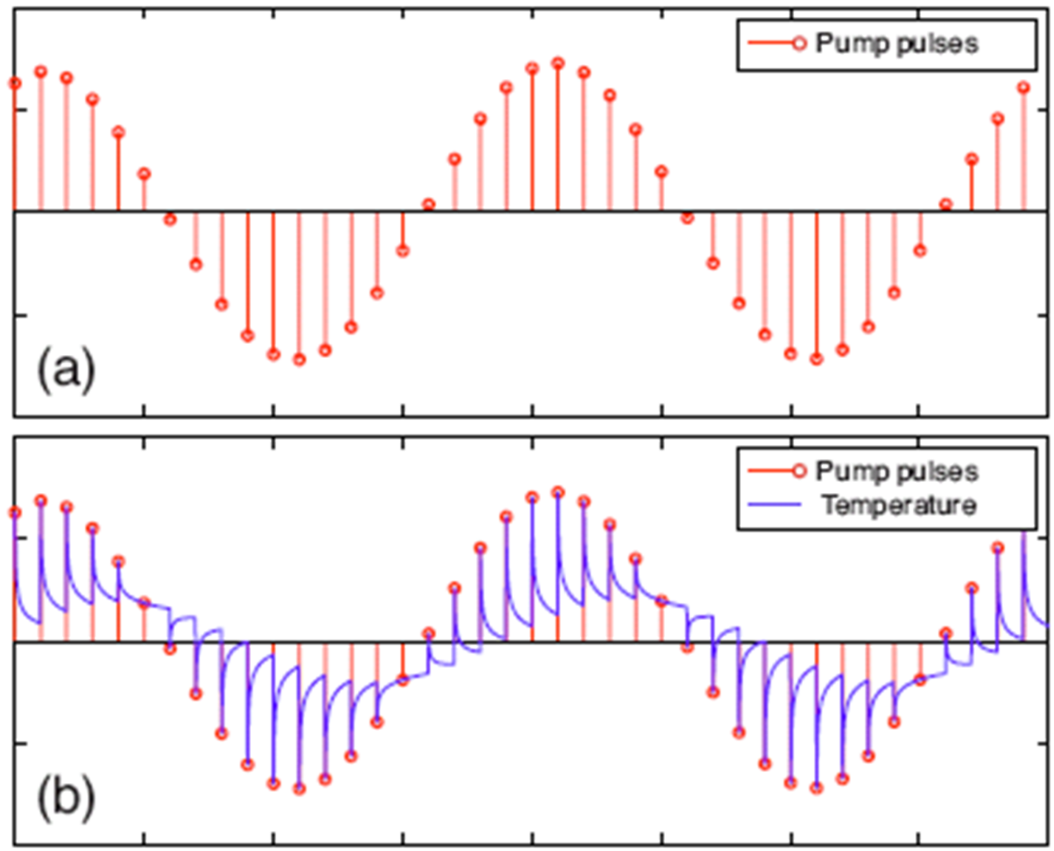




Schmidt et al., RSI 2008

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

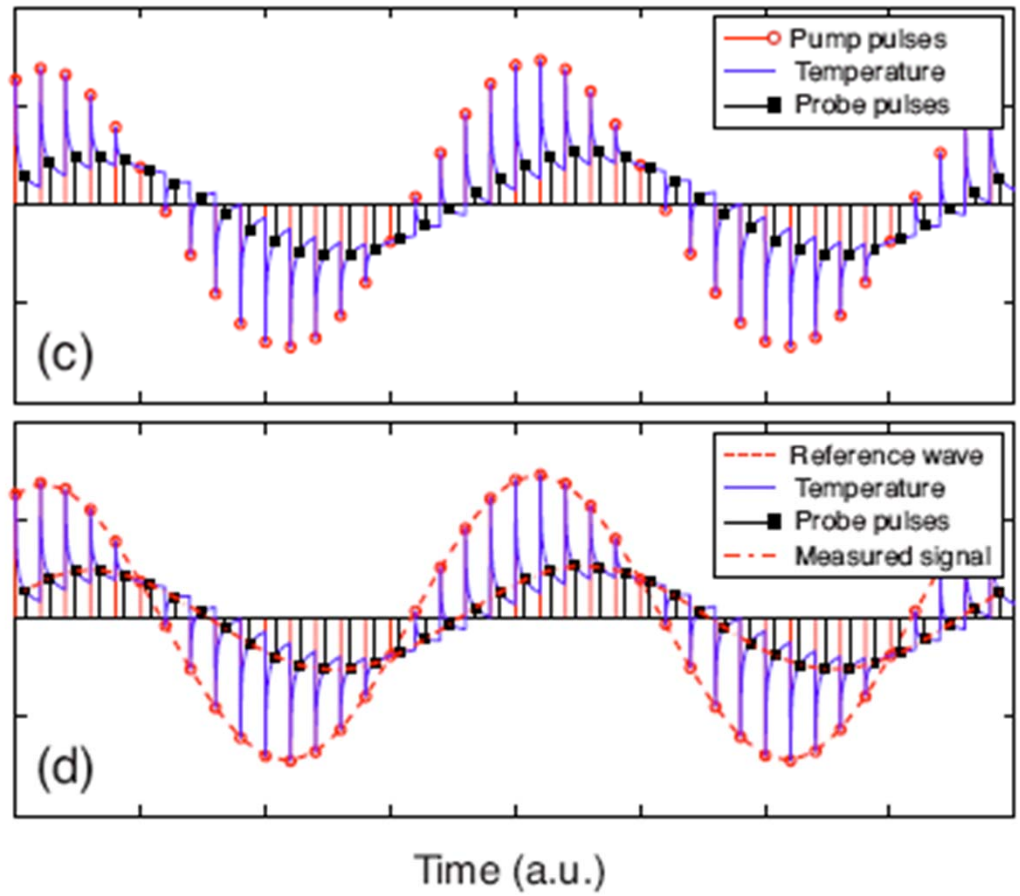
- Evolution of surface temperature



time

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

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

$$\times \begin{pmatrix} \gamma_n + \gamma_{n+1} & \gamma_n - \gamma_{n+1} \\ \gamma_n - \gamma_{n+1} & \gamma_n + \gamma_{n+1} \end{pmatrix} \begin{pmatrix} B^+ \\ B^- \end{pmatrix}_{n+1}$$

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

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

Signal analysis for the rf lock-in

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

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

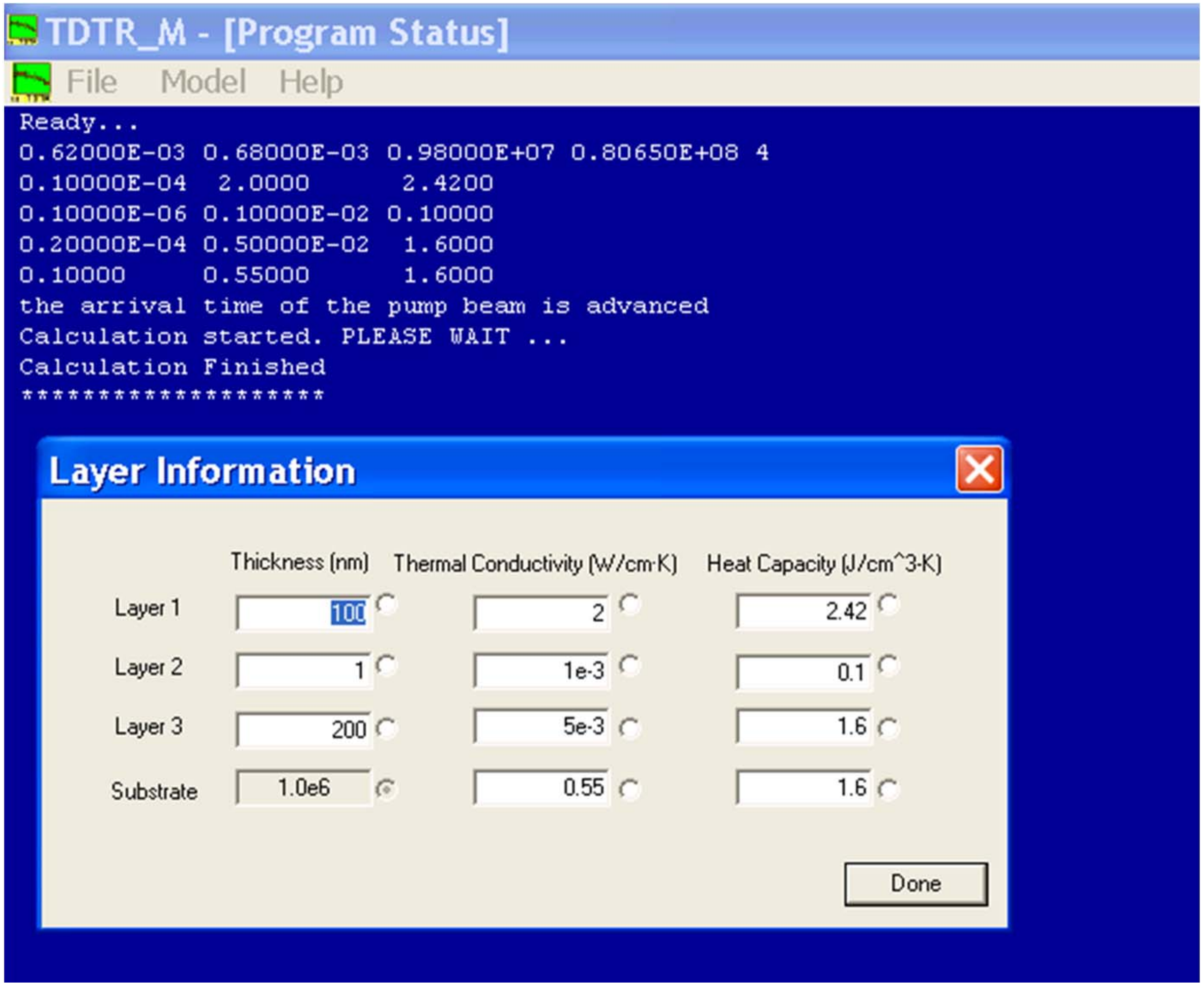
$$\mathrm{Im}\left[\Delta R_M(t)\right] = -i\frac{dR}{dT}\sum_{m=-M}^{M}\left(\Delta\mathcal{T}(m/\tau+f)-\Delta\mathcal{T}(m/\tau-f)\right)\exp(i2\pi mt/\tau)$$

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

Windows software

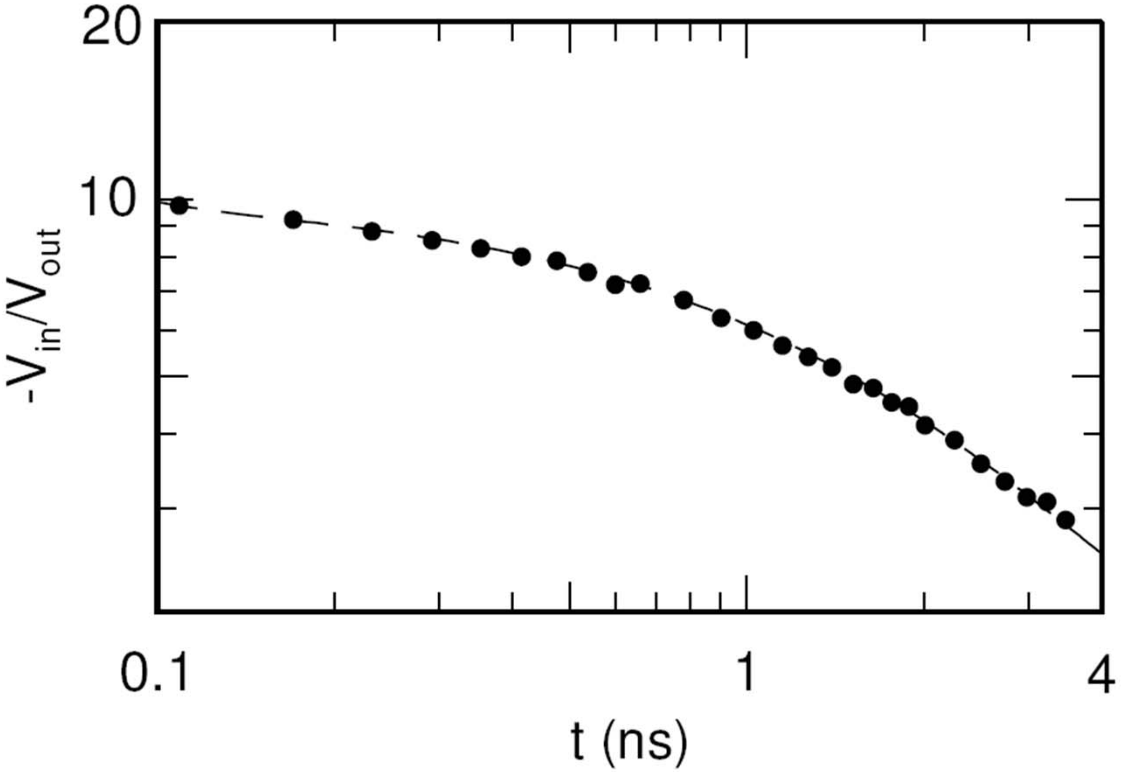
author: Catalin Chiritescu,

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

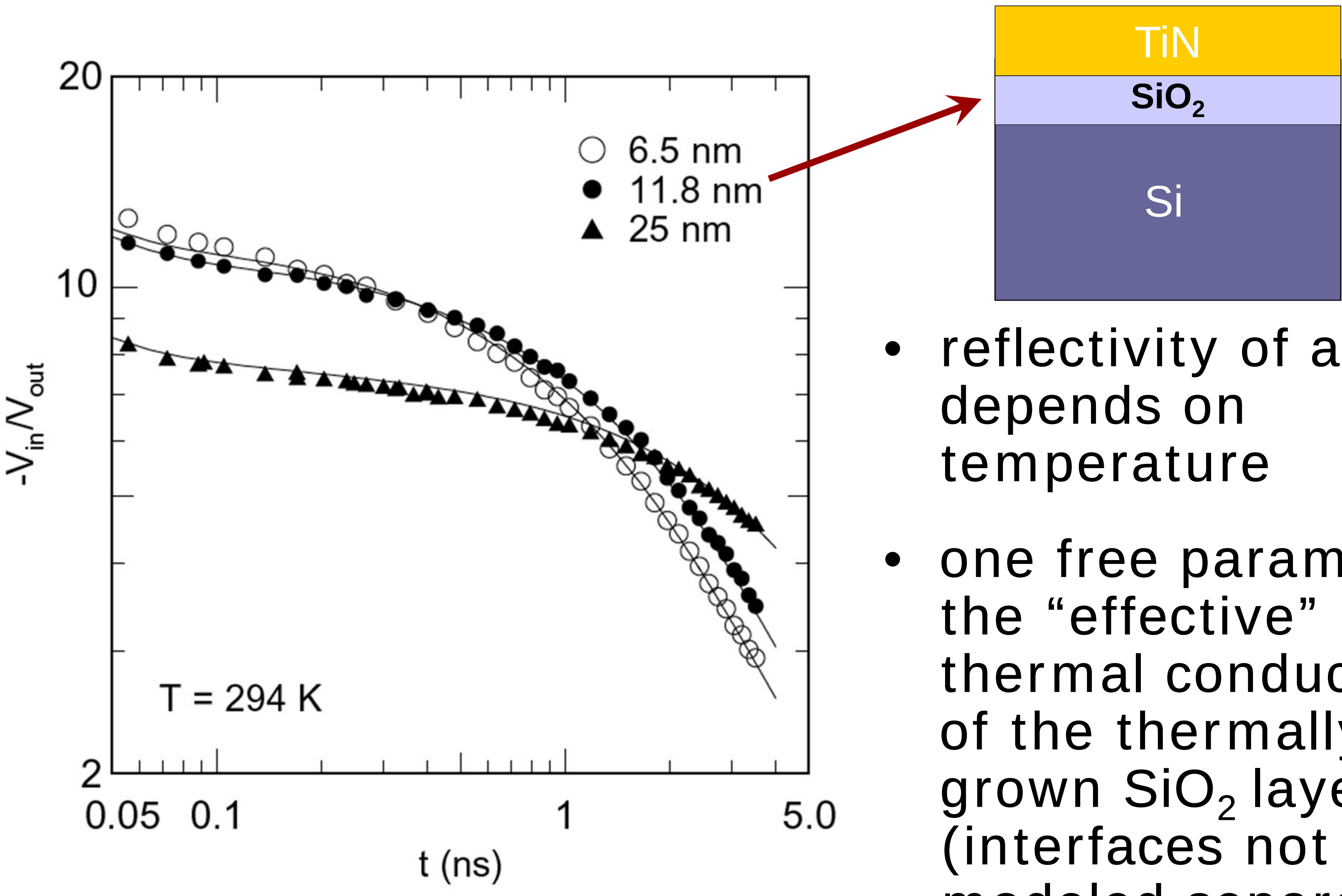


Thermoreflectance data for isotopically pure Si

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

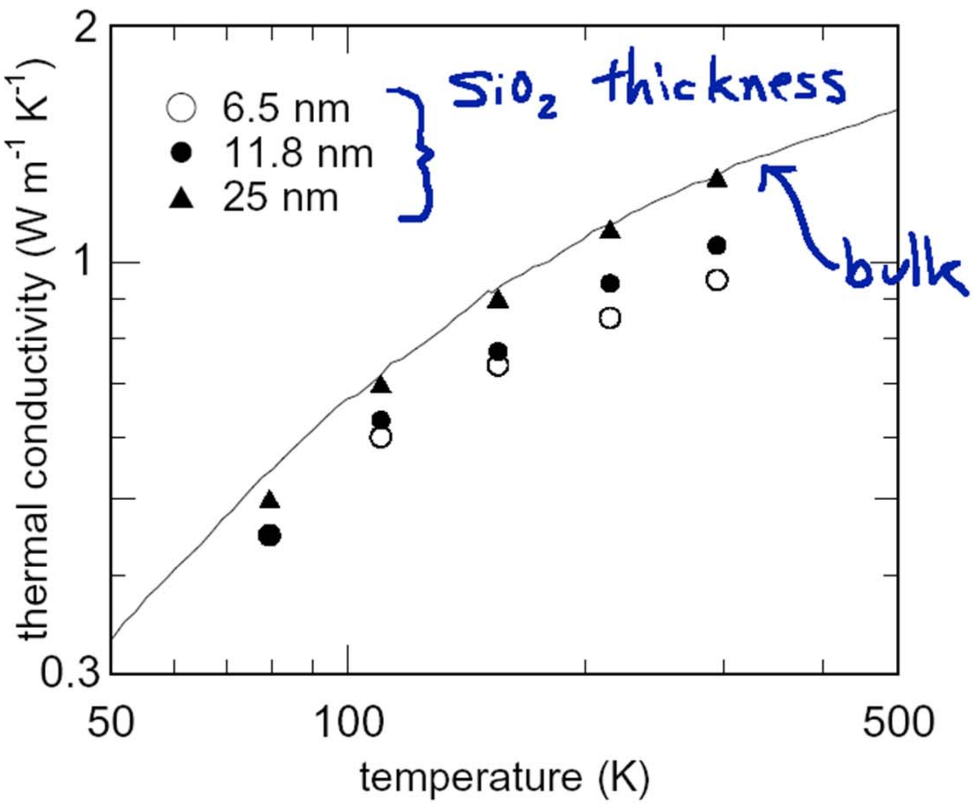


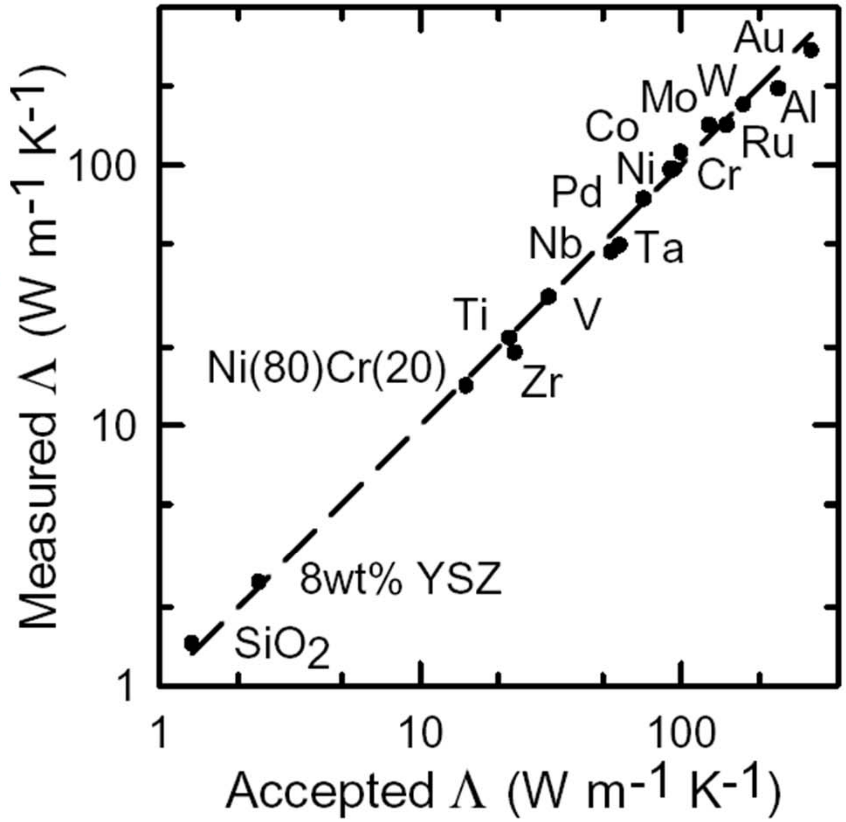
Time-domain Thermoreflectance (TDTR) data for TiN/SiO_2/Si



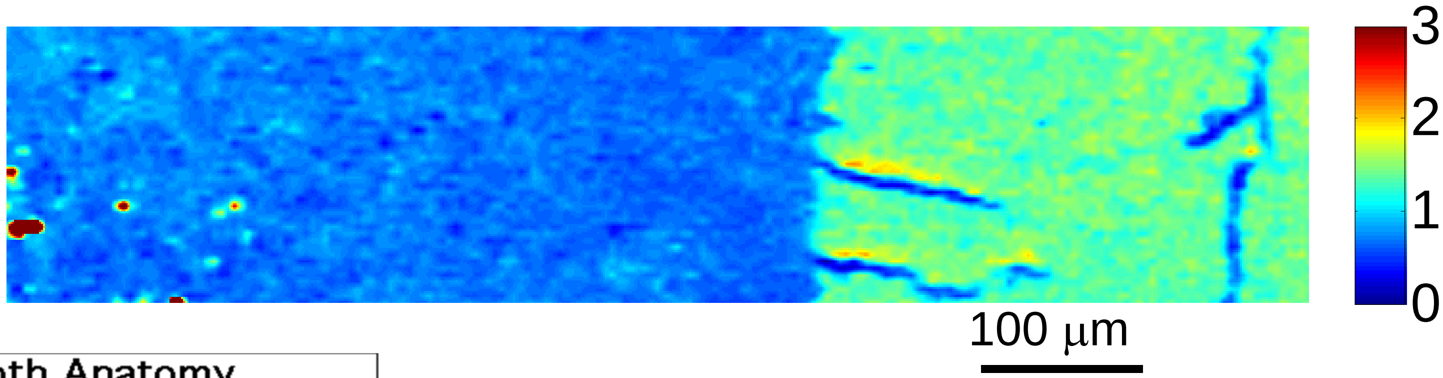
- reflectivity of a metal depends on temperature
- one free parameter: the “effective” thermal conductivity of the thermally grown SiO_2 layer (interfaces not modeled separately)

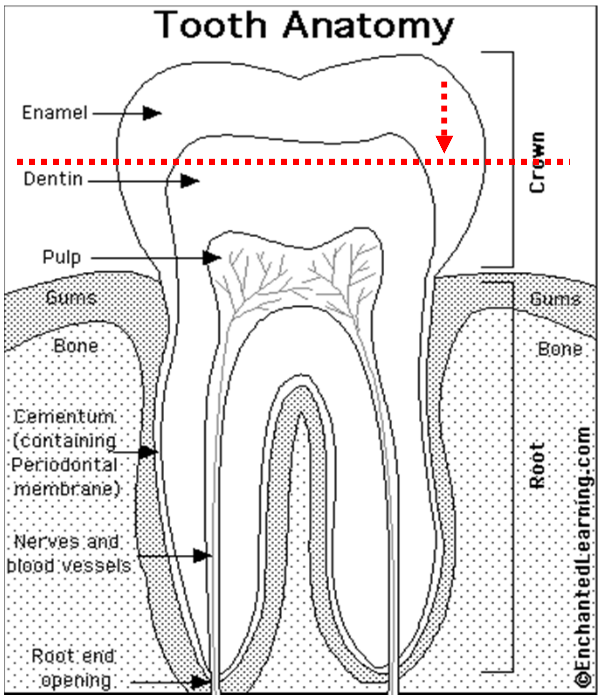
TDTR: early validation experiments



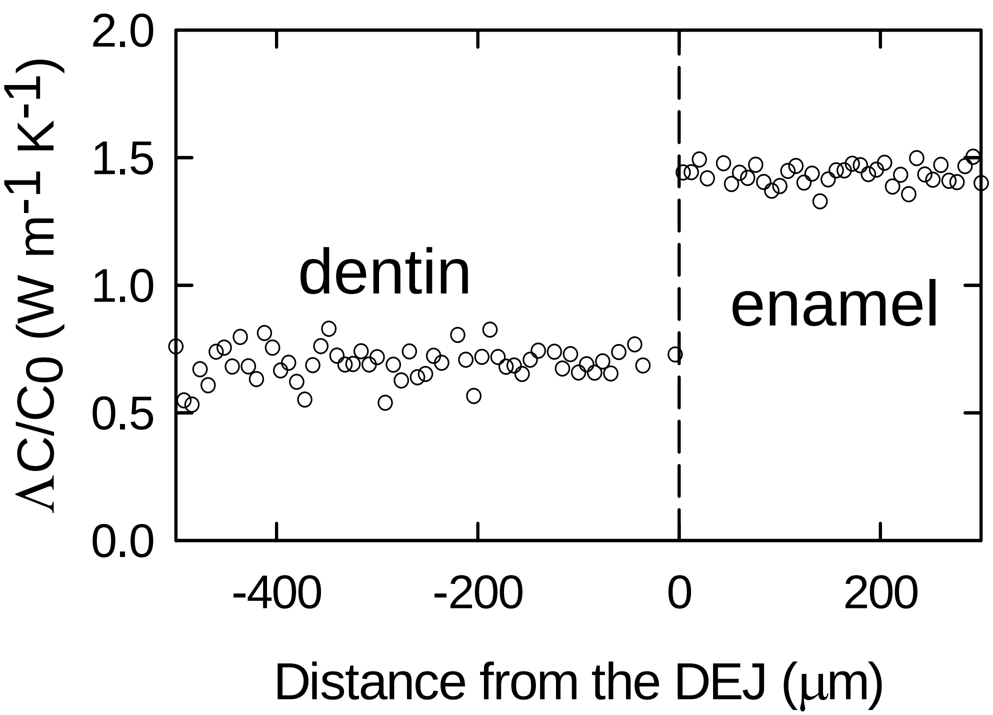


Thermal conductivity map of a human tooth





www.enchantedlearning.com/



Zheng *et al.*, JAP (2008)

The thermal penetration depth and phonon mean-free-path

- First, make the (incorrect) assumption that the mean-free-path of all phonons is the same.

$$\Lambda = \frac{1}{3} C v^2 \tau \qquad l = v\tau$$

$$d = \sqrt{\frac{\Lambda}{\pi f C}} \Rightarrow d = \frac{v}{3}\sqrt{\frac{\tau}{f}} \Rightarrow \frac{d}{l} = \frac{1}{3\sqrt{f\tau}} \gg 1$$

- $f\tau \ll 1$ so the penetration depth is large compared to the mean-free-path

In reality, heat is carried by phonons with a broad distribution of mean-free-paths

- Simplest case of thermal conductivity where resistive scattering dominates.

$$\Lambda = \frac{1}{3}\int_0^{\omega_c} c(\omega) v_g^2(\omega)\tau(\omega)d\omega$$

$c(\omega)$ = heat capacity of phonon mode

$v_g(\omega)$ = phonon group velocity

$\tau(\omega)$ = scattering time

ω_c = cut-off frequency

Make a "Klemens-like" calculation

- Assume linear dispersion for $\omega<\omega_c$ and $\tau^{-1} \propto \omega^2 T$

$$\Lambda = \frac{A}{T}\int_0^{\omega_c} d\omega = \frac{A}{T}\omega_c$$

- Convert to an integral over mean-free-path $l = \frac{B}{\omega^2 T}$

$$\Lambda = \frac{A\sqrt{B}}{2T^{3/2}}\int_{l_c}^{\infty} \frac{1}{l^{3/2}} dl$$

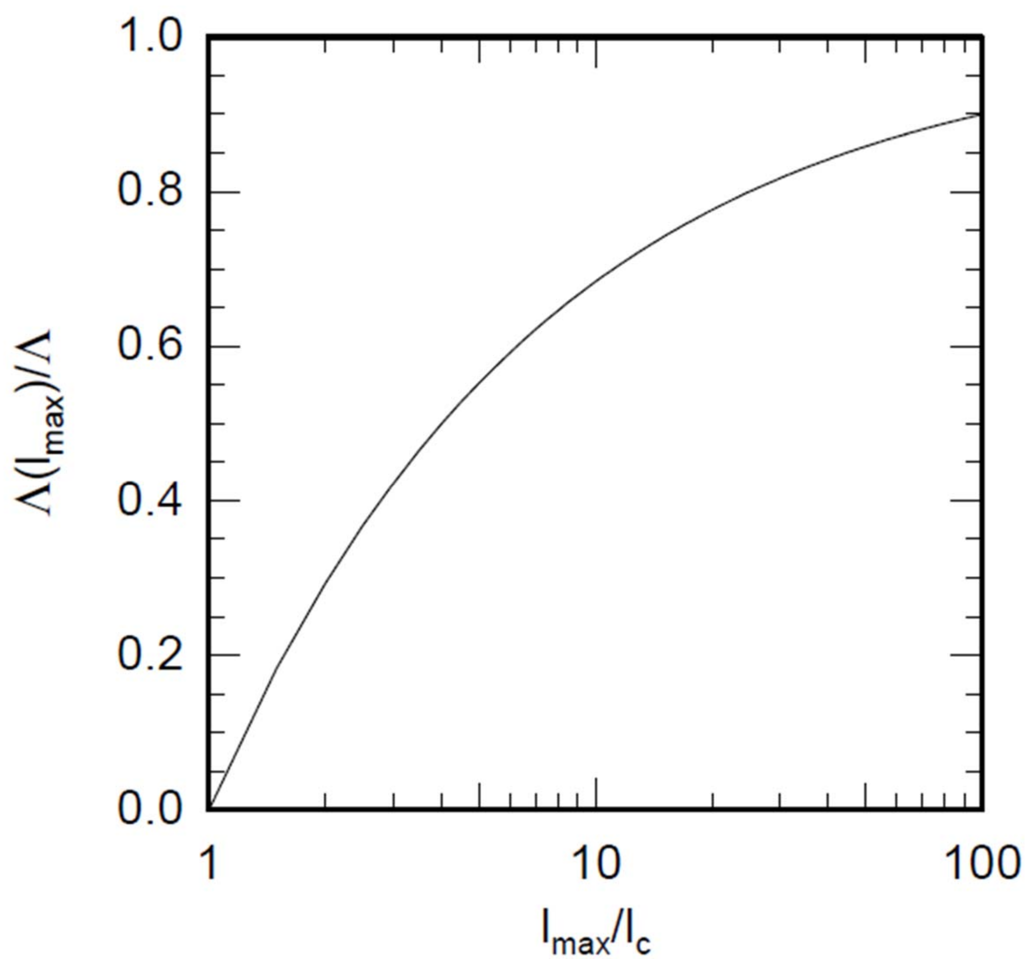
$$\frac{\Lambda(l_{\max})}{\Lambda} = 1 - \left(\frac{l_c}{l_{\max}}\right)^{1/2}$$

ℓ_c is the mean-free-path at the cut-off frequency

ℓ_{max} is the maximum mean-free-path that contributes to Λ

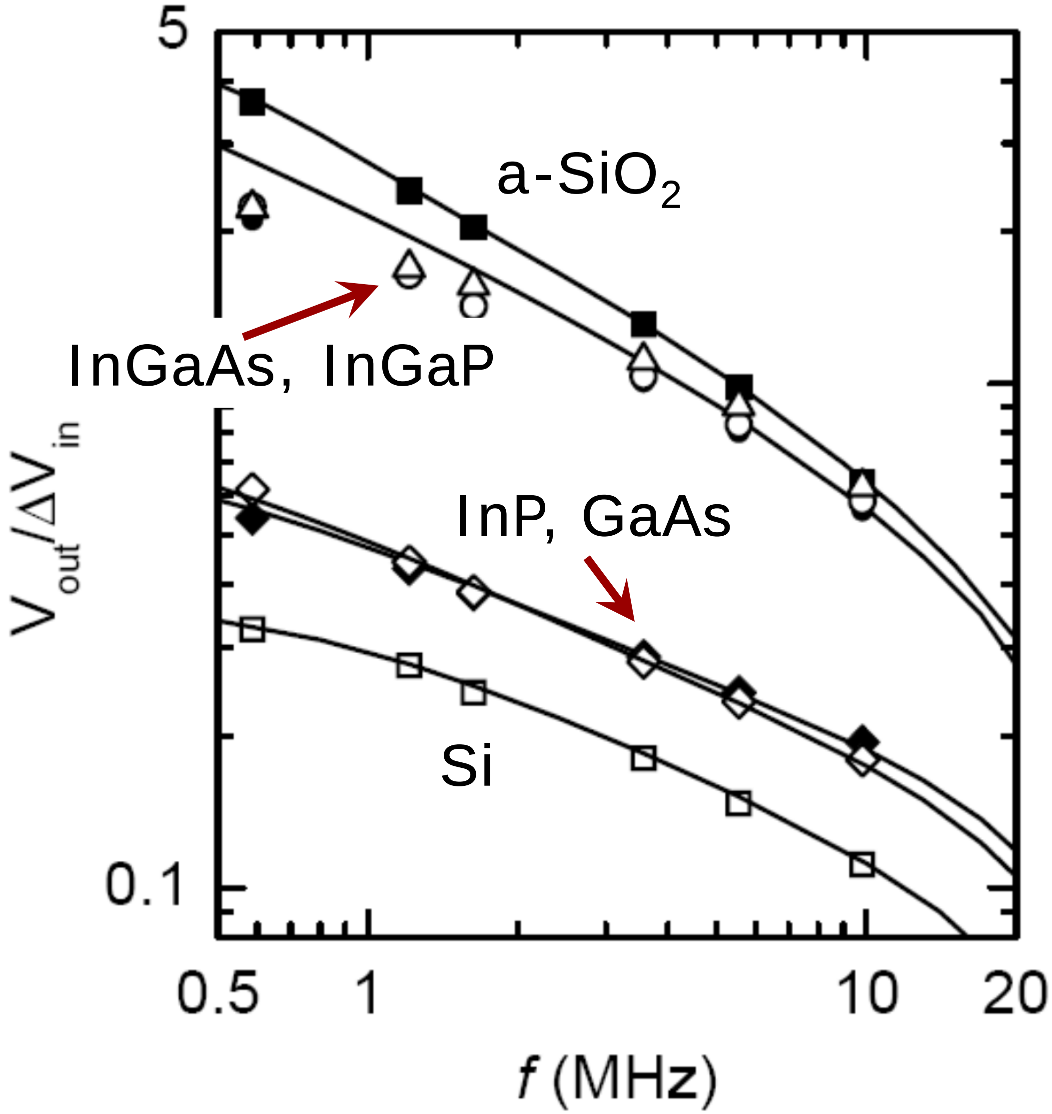
Heat is carried by phonons with a broad distribution of mean-free-paths

- Phonon scattering by charge carriers or boundaries will narrow the distribution.
- Alloying and point defects will broaden the distribution.
- Relaxational damping will eventually be a limiting factor.
- Details are probably important (scattering rates, normal processes, dispersion...)



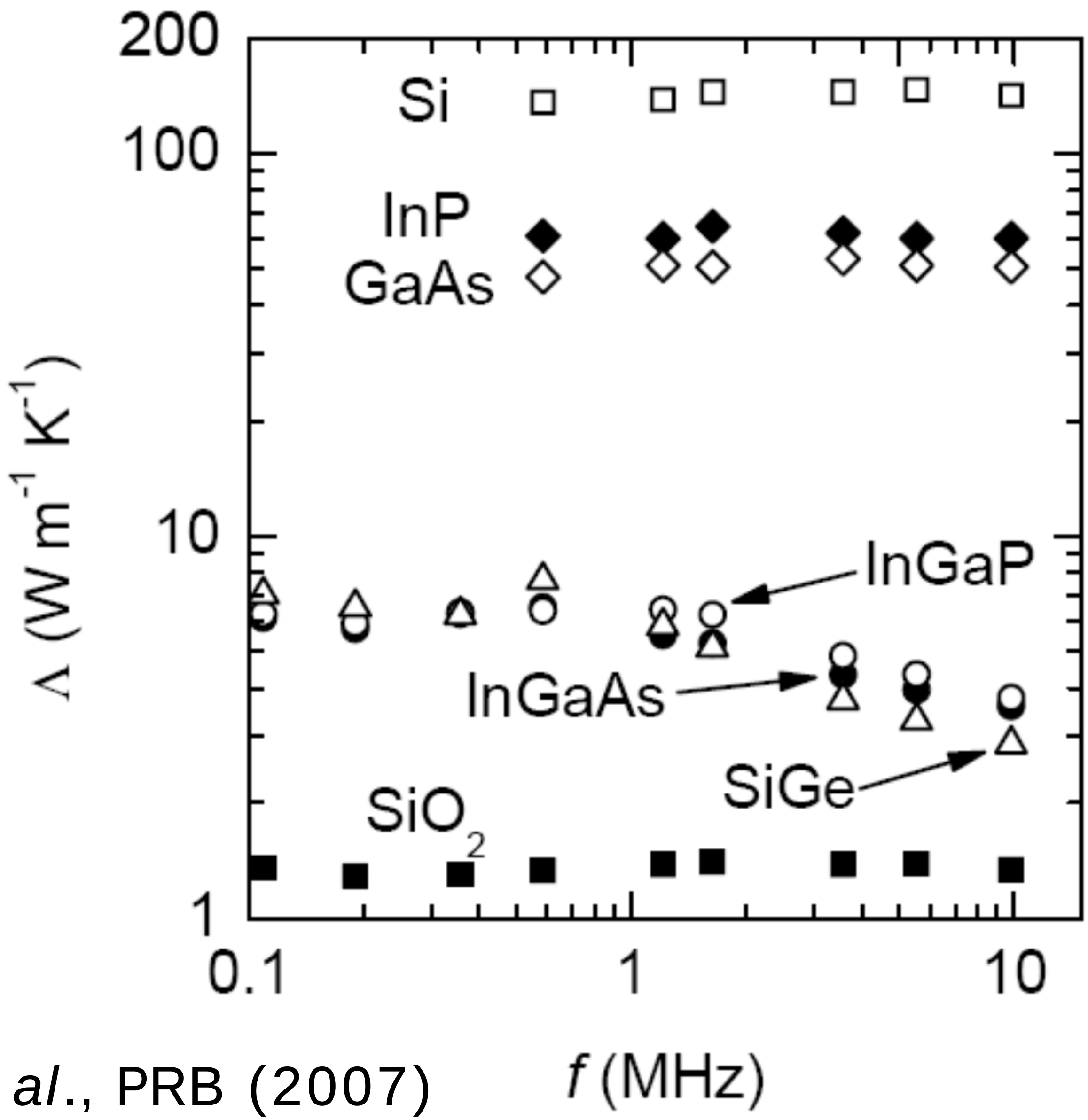
Thermoreflectance raw data at t=100 ps

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



Koh *et al*., PRB (2007)

Same data but allow Λ to vary with frequency f



Koh *et al*., 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 ~100 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}.

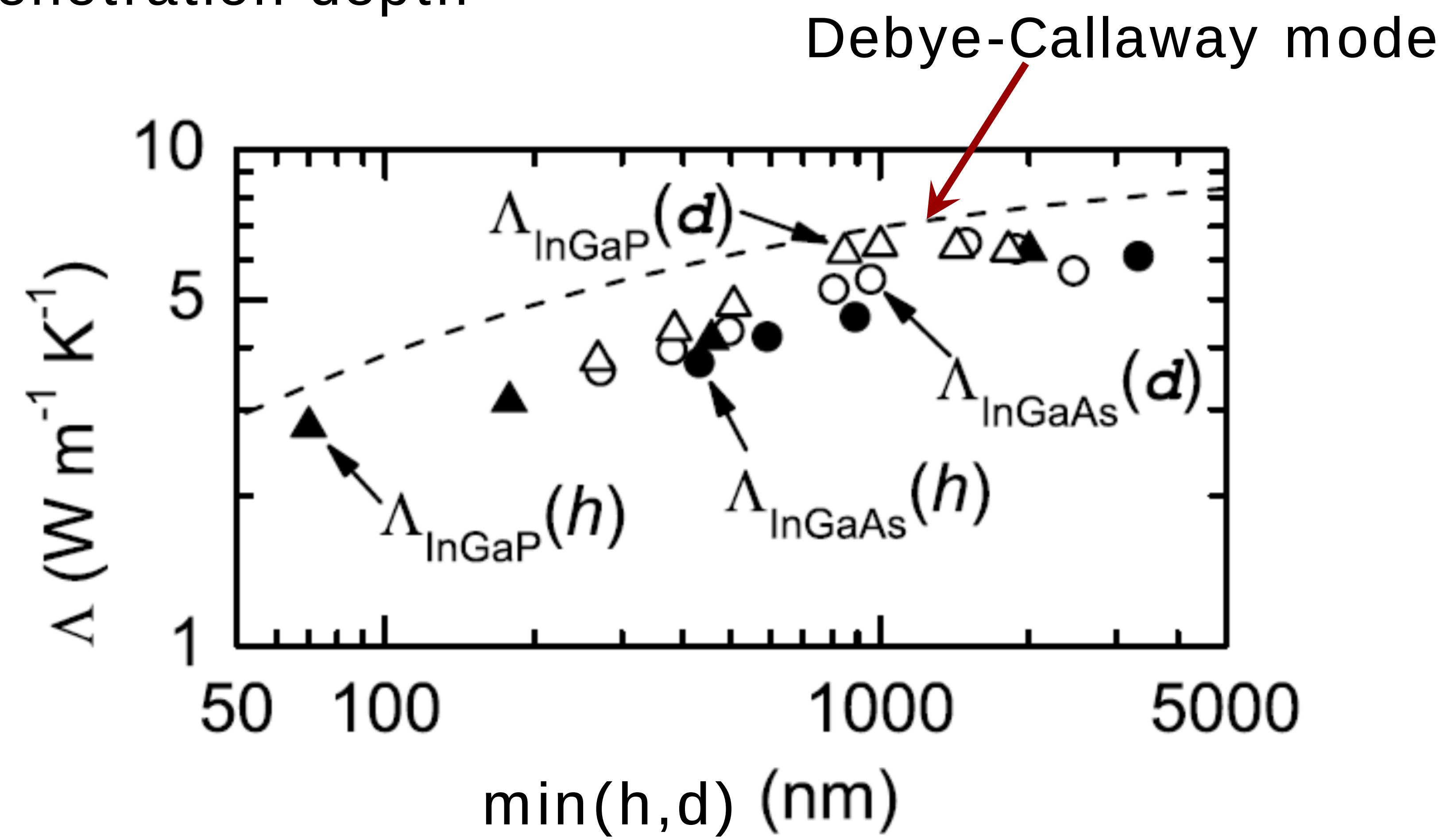
$$d = \sqrt{\Lambda / \pi C f}$$

- 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, ℓ<<d because $f\tau << 1$.

Frequency and thickness dependence for InGaP and InGaAs

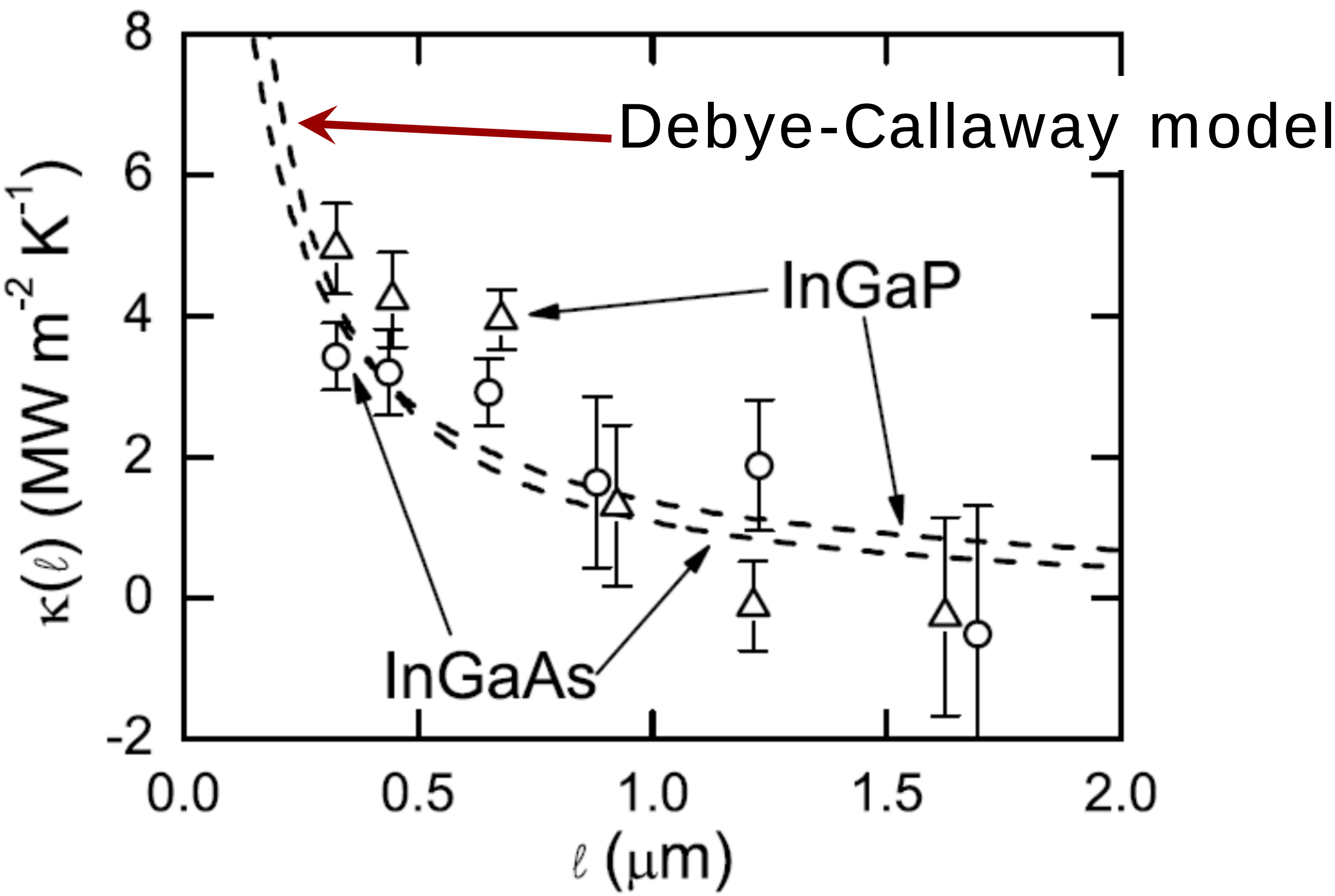
- h=film thickness; d = thermal penetration depth

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



Differentiate to get a distribution function

- Define $\kappa(\ell) = \dfrac{d\Lambda}{d\ell} \approx \dfrac{\Lambda_{n+1} - \Lambda_n}{\boldsymbol{d}_{n+1} - \boldsymbol{d}_n}$ $\qquad \boldsymbol{d} = \sqrt{\Lambda / \pi C f}$



Current open question: why don't we see frequency dependence in pure crystals?

$$\Lambda = \frac{1}{3}\int_0^{\omega_c} c(\omega) v_g^2(\omega)\tau(\omega) d\omega$$

$c(\omega) \propto \omega^2$

$v_g(\omega)$ = phonon group velocity

$\tau(\omega)$ = $(A\omega^2 T)^{-1}$

ω_c = cut-off frequency for heat carrying acoustic modes

ω_D = Debye frequency

- Fraction of thermal conductivity from phonons with mean-free-path smaller than the thermal penetration depth

$$\phi = 1 - \left(\frac{\omega_D^3 f \tau_c}{\omega_c^3}\right)^{1/4}$$

Current open question: why don't we see frequency dependence in pure crystals?

- Make an order-of-magnitude estimate

$$\left(\frac{\omega_D}{\omega_c}\right)^3 \sim 10; \;\; f \approx 10 \text{ MHz}; \quad \tau_c \sim 10 \text{ ps}; \quad f\tau_c \sim 10^{-4}$$

$$\Longrightarrow \quad \phi = 0.82$$

- This reduction is not observed.
- Why are pure crystals different than the alloy?
- Do we need to replace the diffusion equation as the basis for the analysis of TDTR experiments? (see also experiments by Minnich and Chen)
- Boundary conditions at the metal/sample interface are complicated. Too many unknowns (?). What experiment and theory can provide more constraints?