

# Time Domain Thermoreflectance

Tips, tricks, and a few extensions

# USNA Nanoscale Electronic and Thermal Characterization



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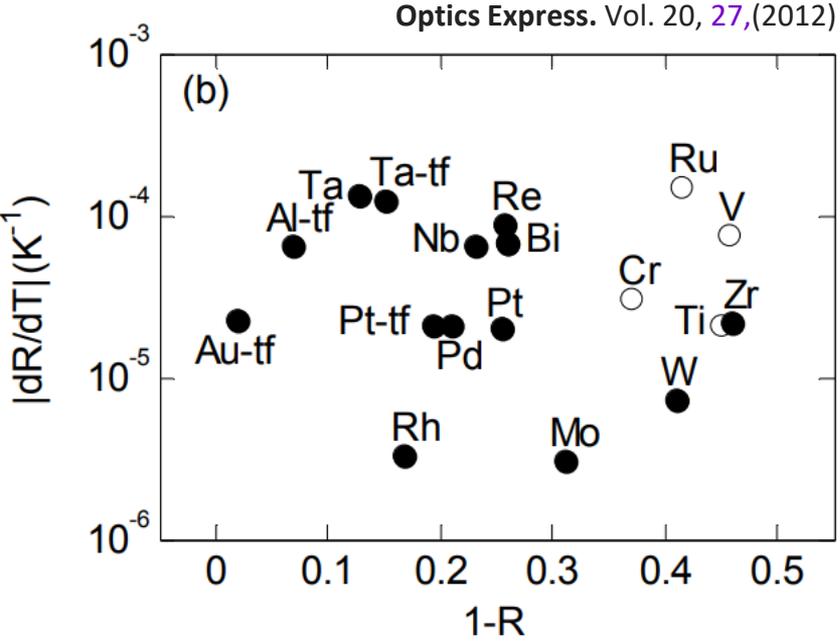
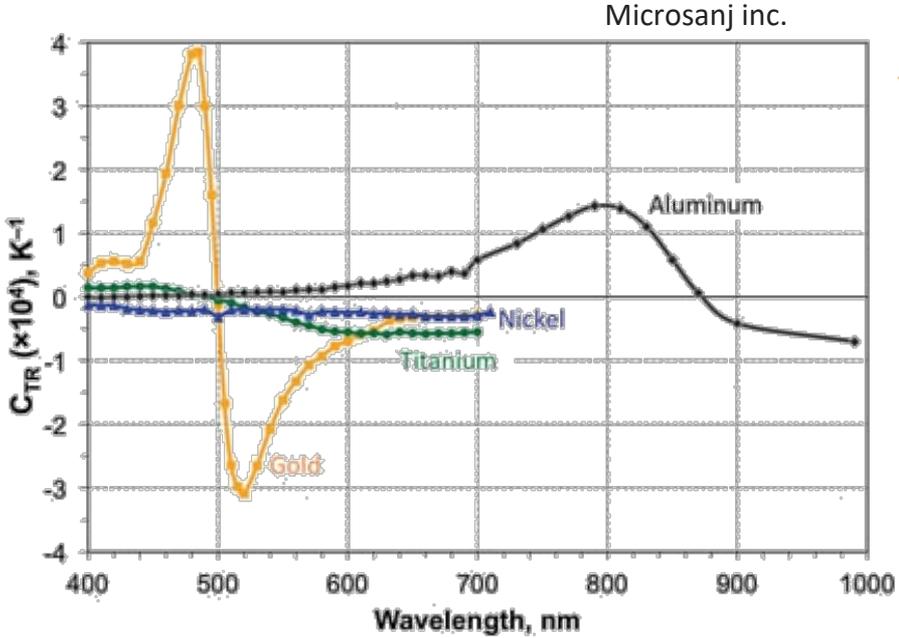
Assoc. Prof Michelle Jamer  
USNA Physics



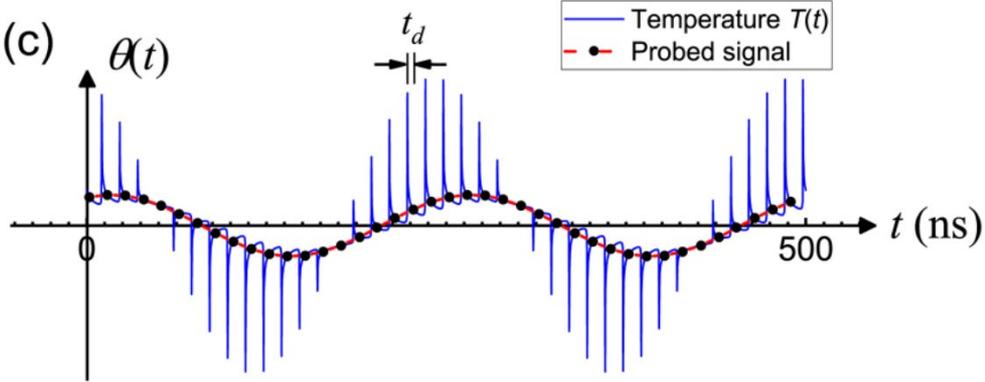
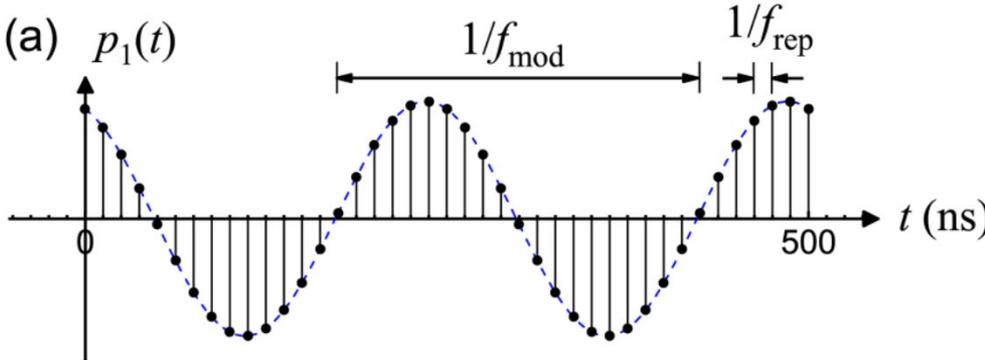
**You can determine lots of interesting thermal properties with Time Domain Thermoreflectance.**



# Thermoreflectance



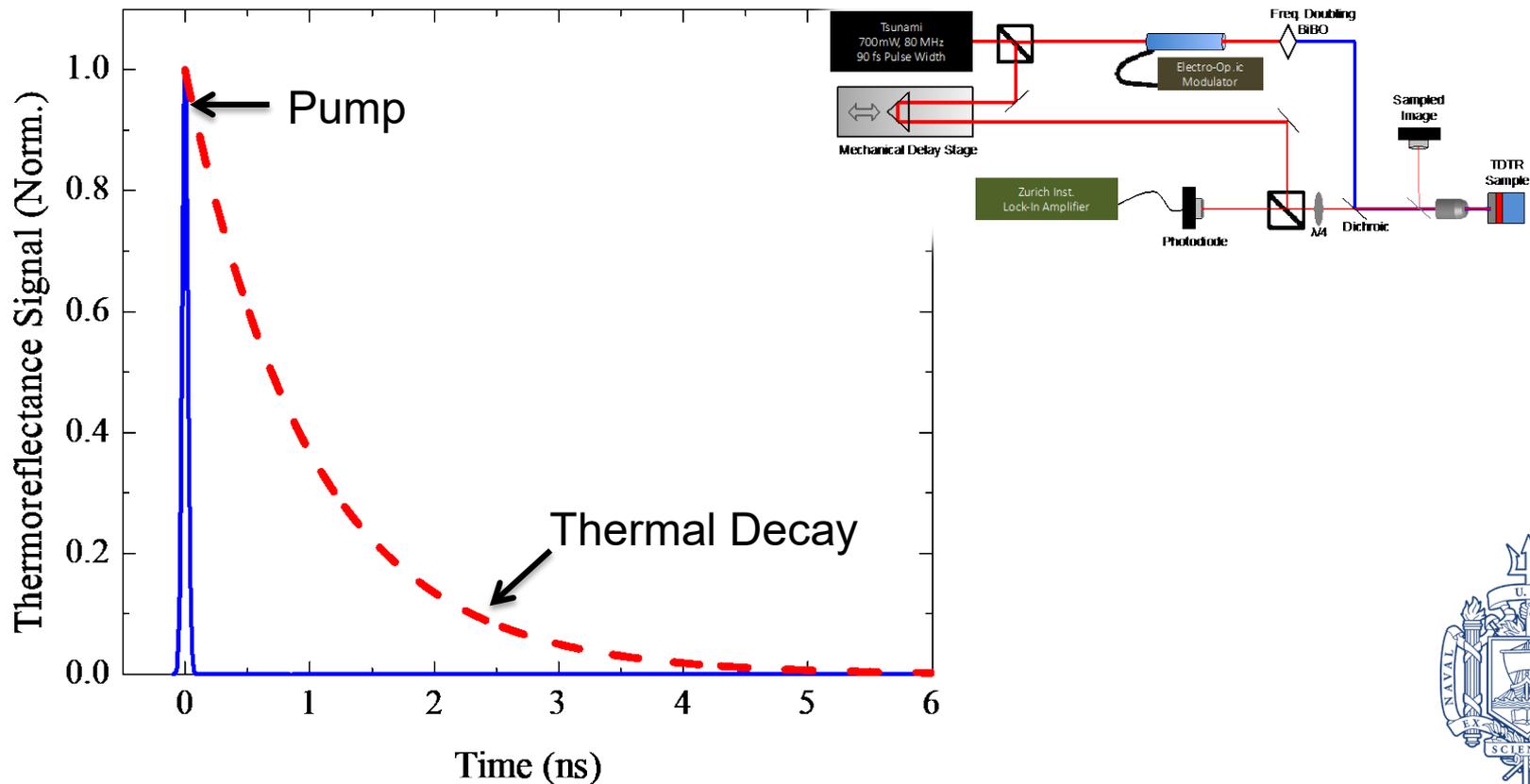
# Time Domain Thermoreflectance (TDTR)



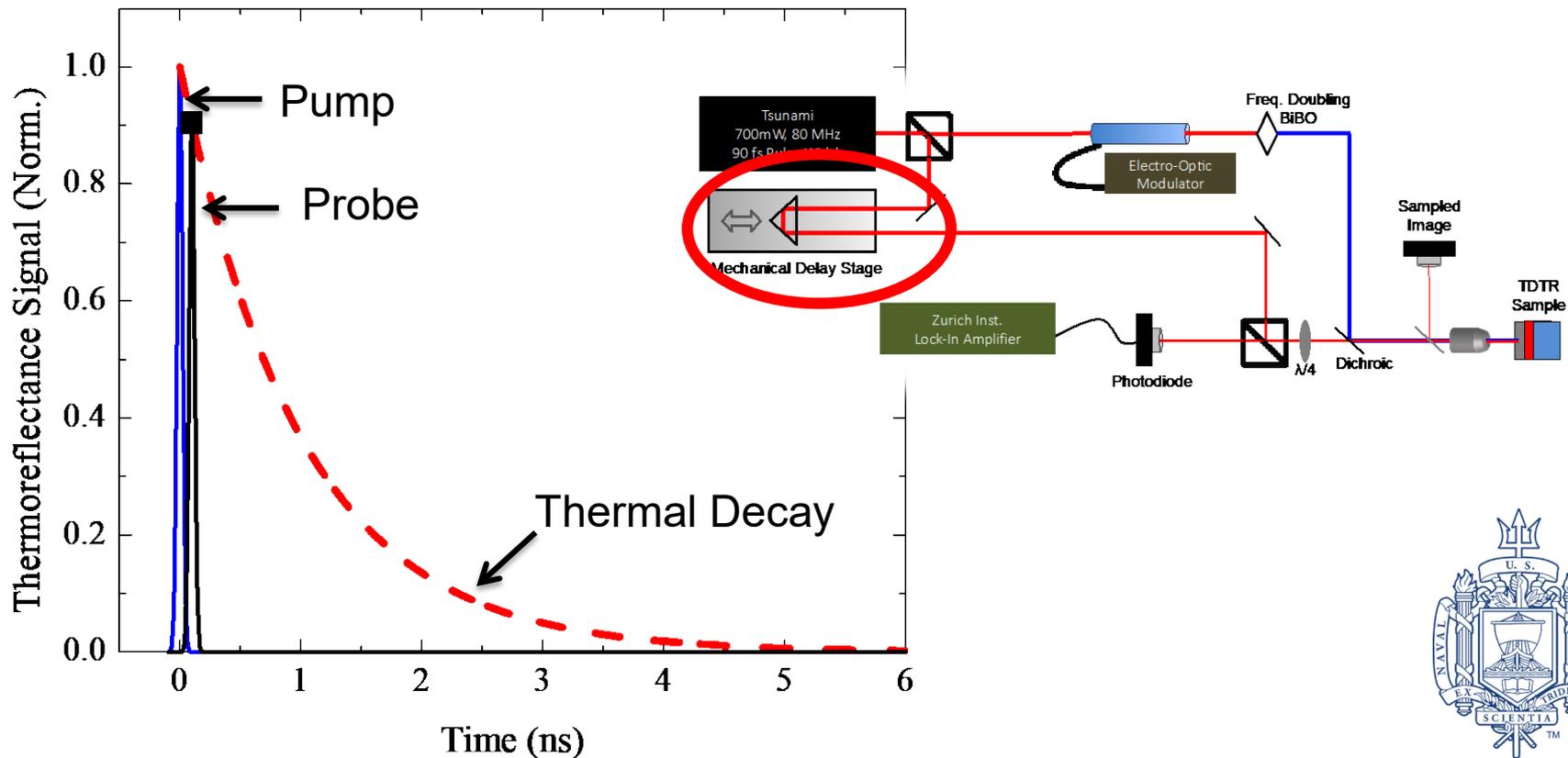
- Thin Film Thermal Conductivity
- Thermal Boundary Conductance
- Electron-phonon coupling
- Nanoscale Thermal Physics
- Hot Spot Generations
- ...



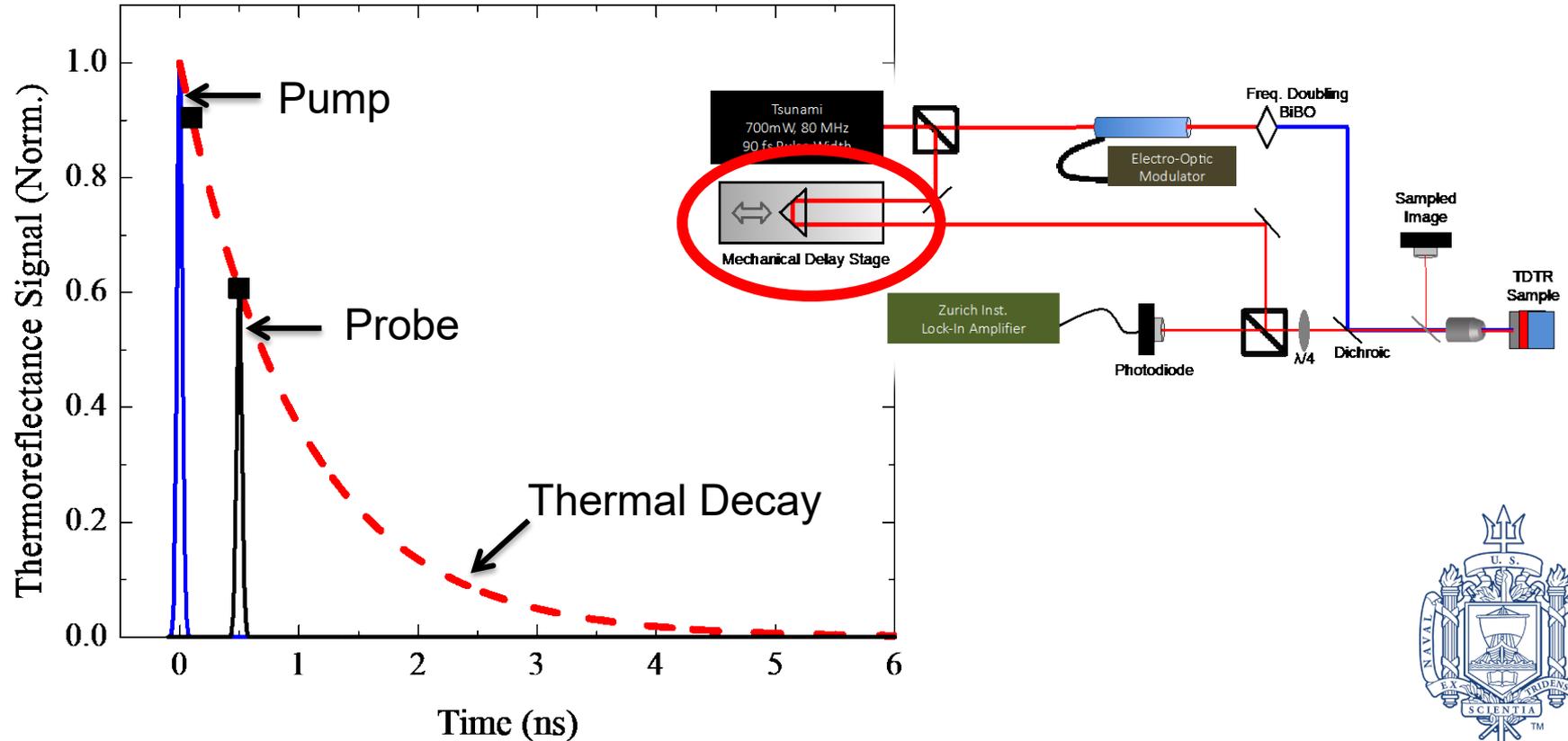
# Optical Pump-Probe Thermal Lifetime



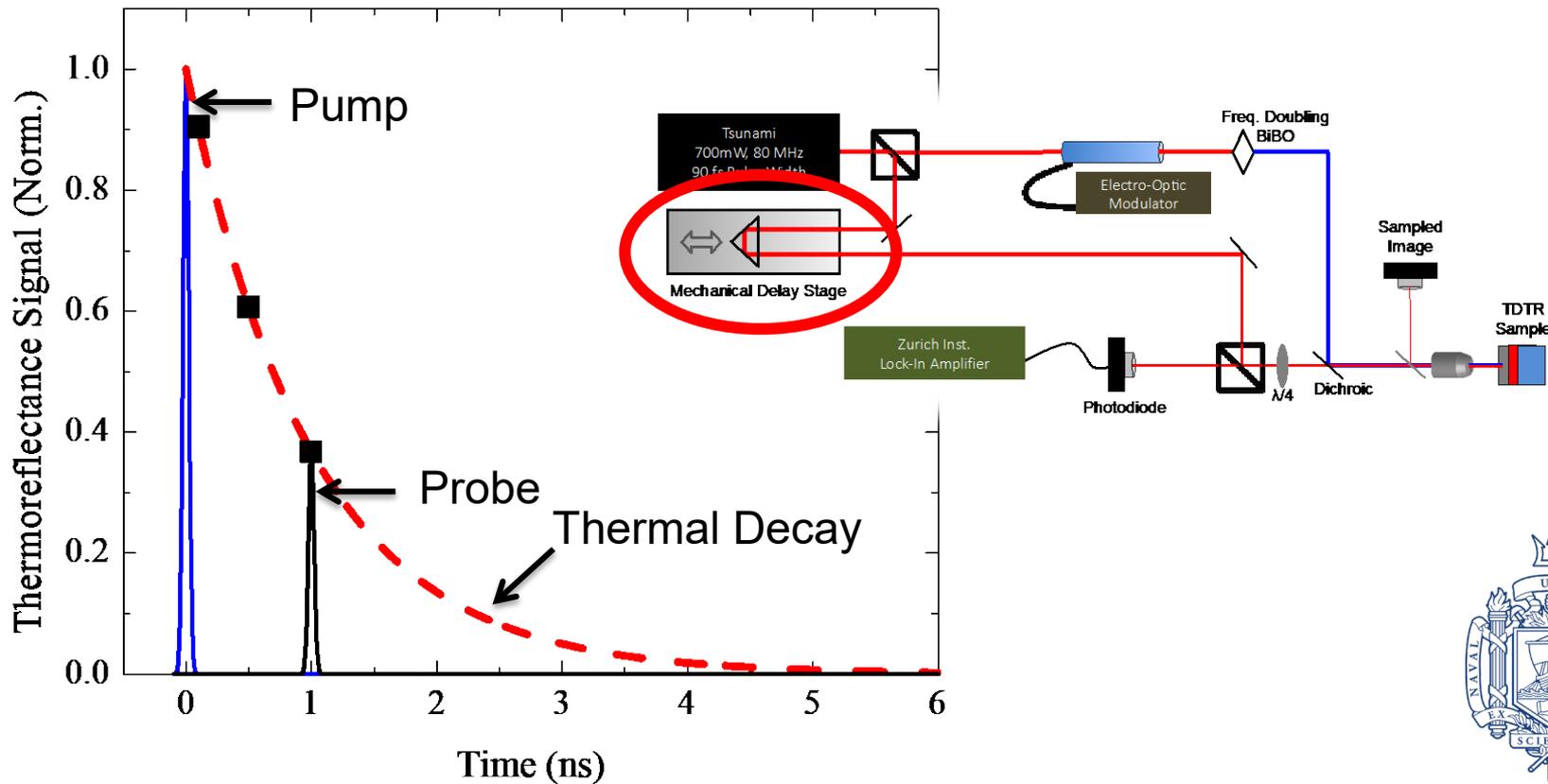
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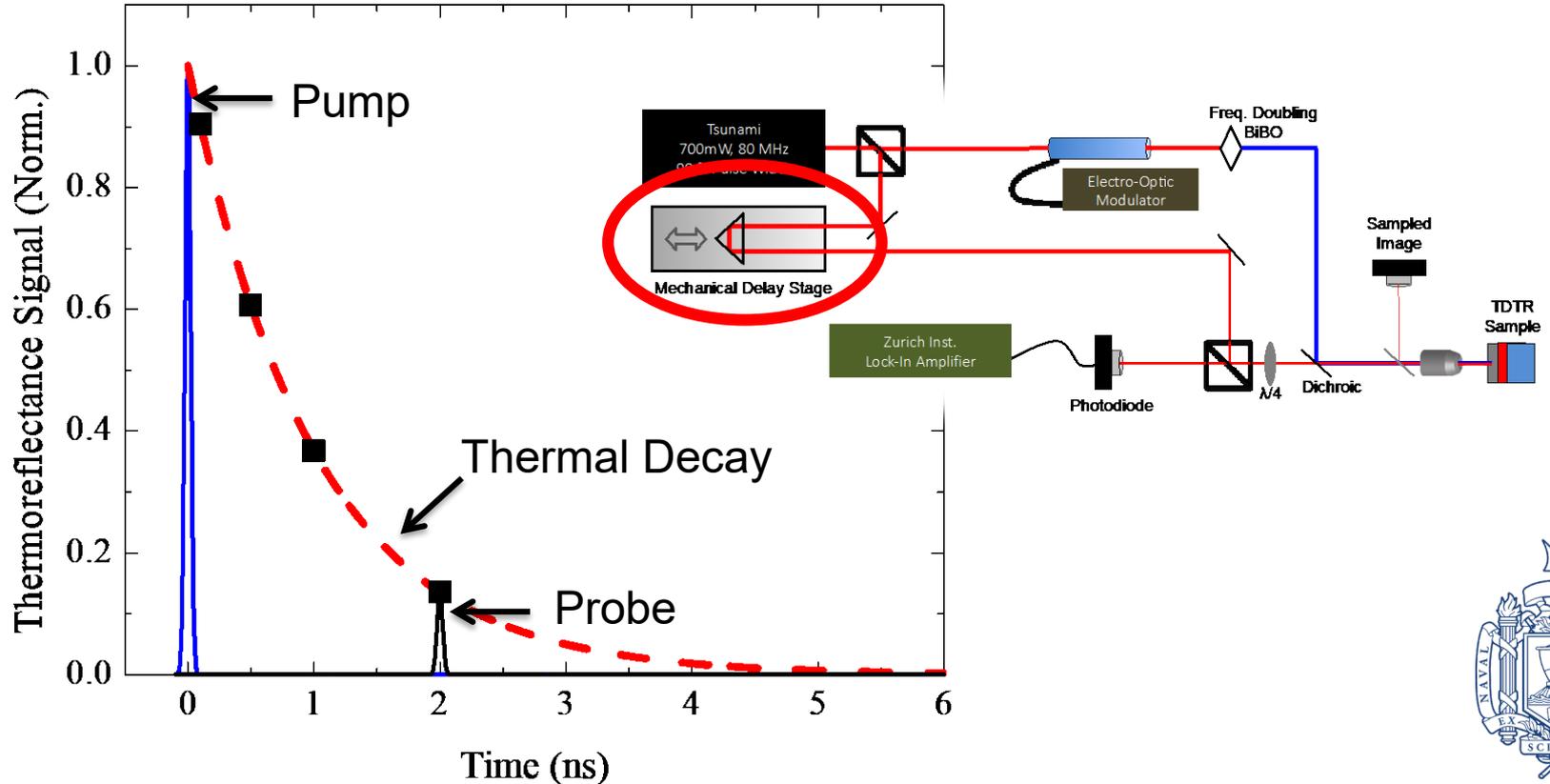
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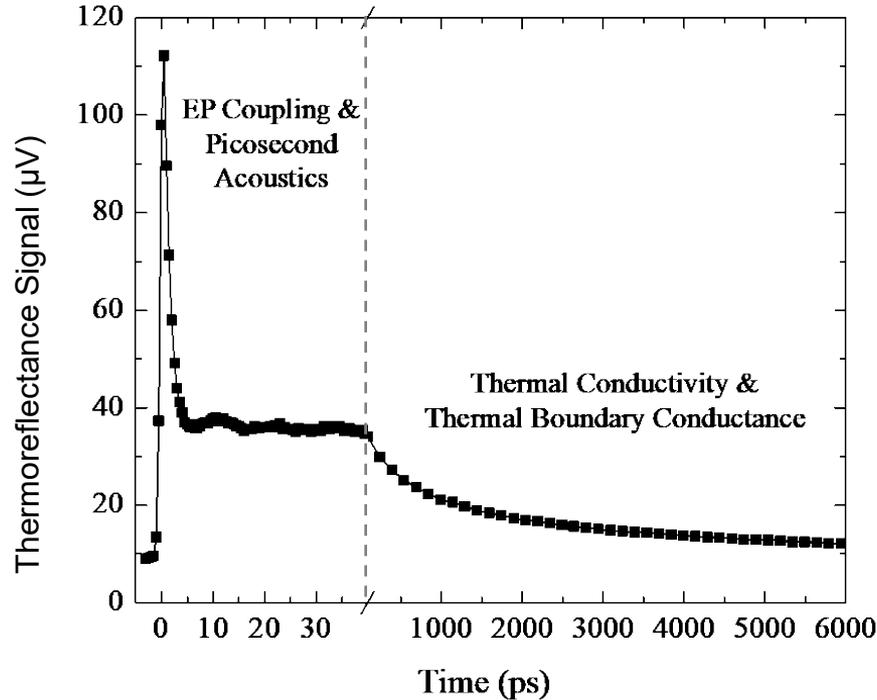
# Optical Pump-Probe Thermal Lifetime



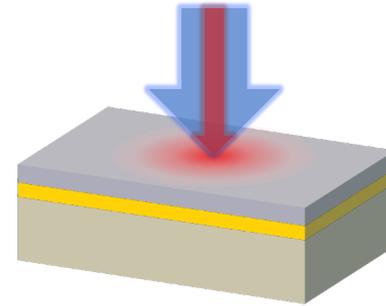
# Optical Pump-Probe Thermal Lifetime



# Optical Pump-Probe Thermal Lifetime Measurement



Thermoreflectance decay fit to model for multilayer thermal transport to determine thermophysical properties



# Multilayer Laser Heating

$$C_v \frac{\partial \theta}{\partial t} = \frac{\kappa_r}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \theta}{\partial r} \right) + \kappa_z \frac{\partial^2 \theta}{\partial z^2}$$

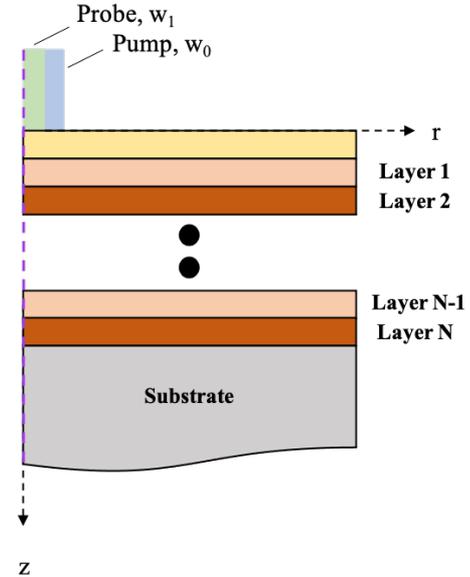
$$\kappa_z \frac{\partial^2 \theta(\omega, k, z)}{\partial z^2} = (\kappa_r k^2 + C_v i \omega) \theta(\omega, k, z)$$

$$\begin{bmatrix} \theta_{n,b} \\ f_{n,b} \end{bmatrix} = \begin{bmatrix} \cosh(qd) & -\frac{1}{\kappa_z q} \sinh(qd) \\ -\kappa_z q \sinh(qd) & \cosh(qd) \end{bmatrix} \begin{bmatrix} \theta_{n,t} \\ f_{n,t} \end{bmatrix} \quad \begin{bmatrix} \theta_{n+1,t} \\ f_{n+1,t} \end{bmatrix} = \begin{bmatrix} 1 & -G^{-1} \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \theta_{n,b} \\ f_{n,b} \end{bmatrix}$$

$$\begin{bmatrix} \theta_b \\ f_b \end{bmatrix} = M_n M_{n-1} \cdots M_2 M_1 = \begin{bmatrix} A & B \\ C & D \end{bmatrix} \begin{bmatrix} \theta_t \\ f_t \end{bmatrix}$$

$$H(\omega) = \frac{A_0}{2\pi} \int_0^\infty k \left( -\frac{D}{C} \right) \exp\left( -\frac{k^2(w_0^2 + w_1^2)}{8} \right) dk$$

$$\phi_{LI} = \tan^{-1} \frac{\Im(H(\omega))}{\Re(H(\omega))} + \phi_{ext}$$



**You can determine lots of interesting thermal properties with Time Domain Thermoreflectance....**

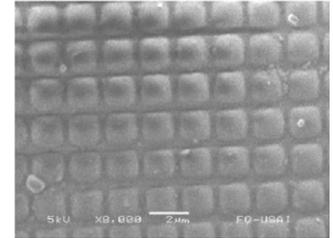
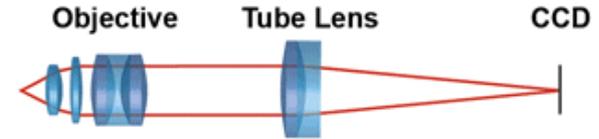
**But it can be hard.**



# TDTR

## Tips & Tricks

- Understand your camera & imaging
- Calibrate a LOT
- Fiber-based pulsed lasers
- Workhorse + Exploratory separate systems
- Electronics & Detection
  - BPD, tidy wire yokes, amplifiers, bandpass filters
- FIND AN EXPERT!



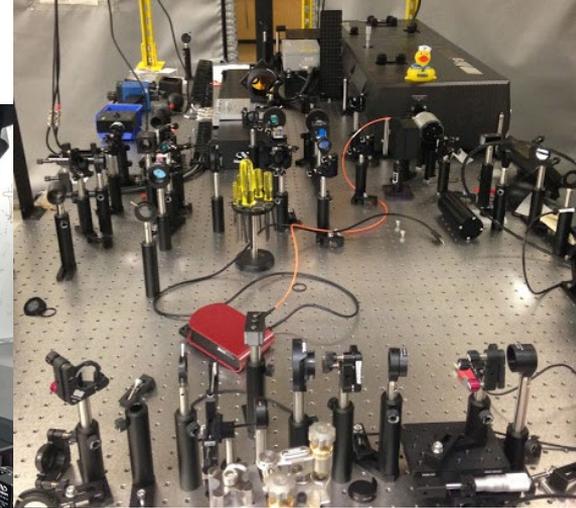
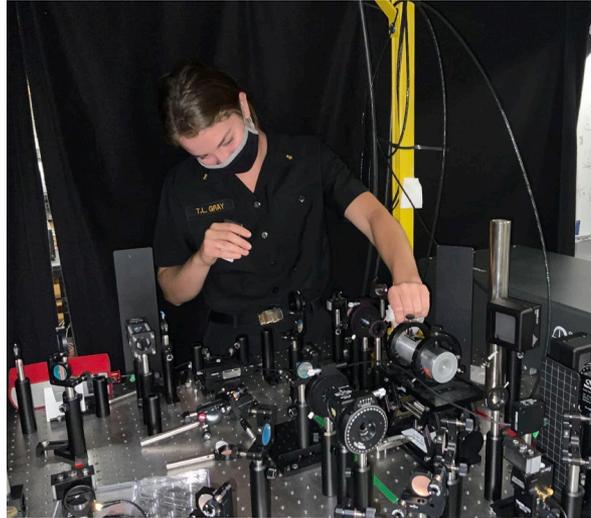
SEM of CMOS Camera pixels  
DOI:[10.13140/RG.2.1.1602.5689](https://doi.org/10.13140/RG.2.1.1602.5689)



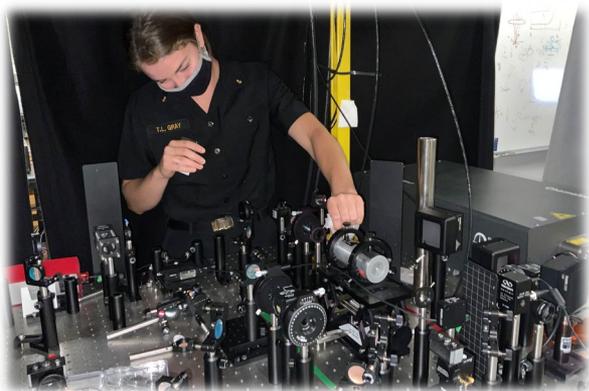
# TDTR

## Shortcomings

- Complexity
- Optical Alignment
- Transducers
- Popularity
- Temperature Dependence
- Sample Limitations
- ...



# Accessing Full Time Delay TDTR



Review of  
Scientific Instruments

ARTICLE

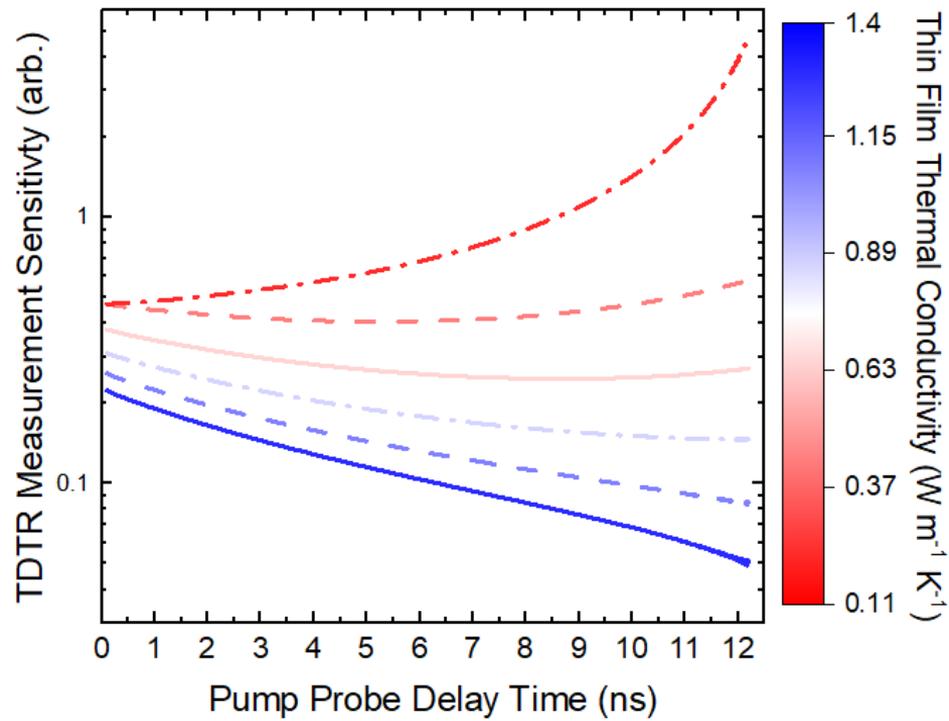
scitation.org/journal/rsi

## Unwrapping a full temporal cycle in time domain thermoreflectance for enhanced measurement sensitivity in thermally insulating materials

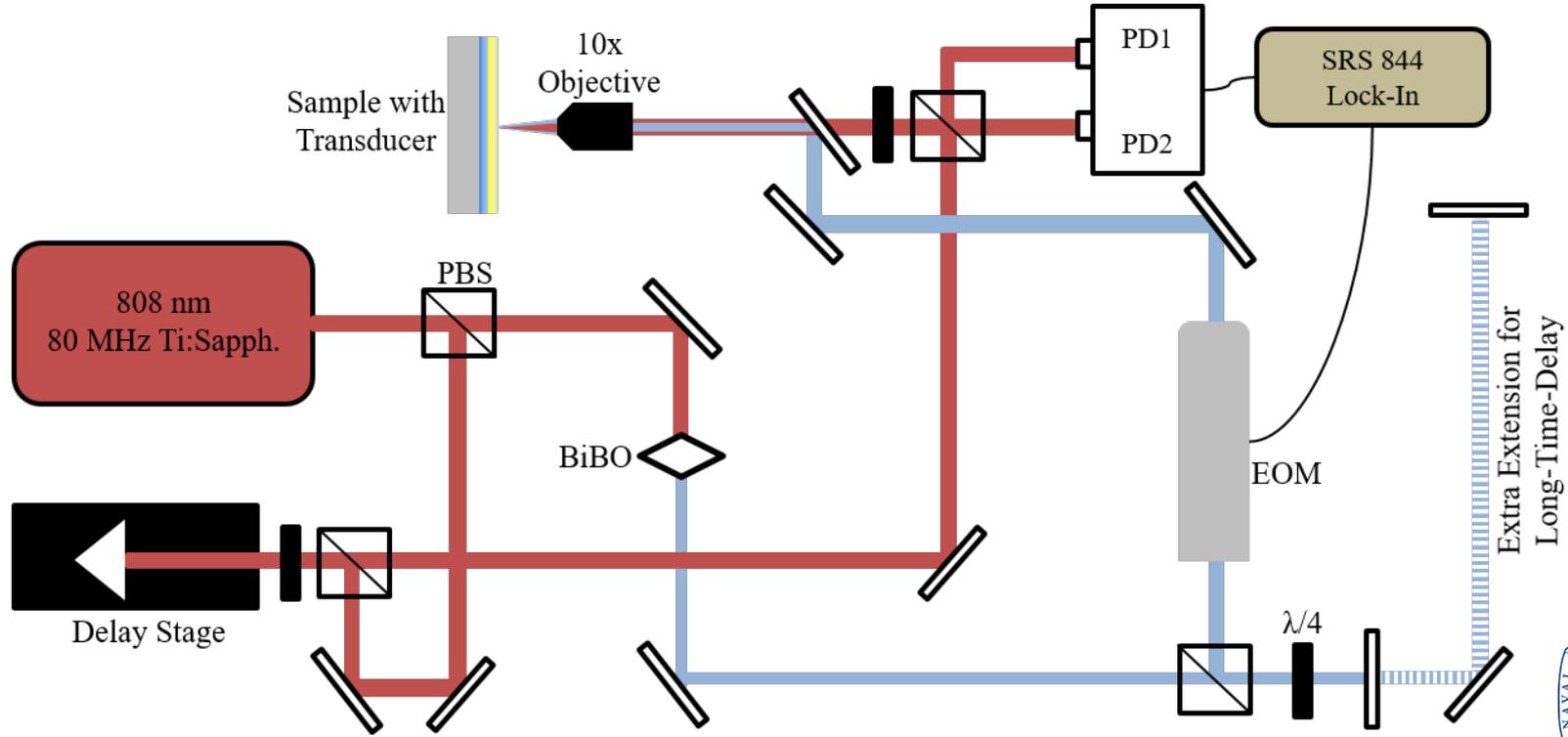
Cite as: Rev. Sci. Instrum. 93, 084904 (2022); doi: 10.1063/5.0089075  
Submitted: 22 February 2022 • Accepted: 14 July 2022 •  
Published Online: 19 August 2022



Brian F. Donovan,<sup>1(a)</sup> Taylor L. Gray,<sup>1</sup> Adam A. Wilson,<sup>2</sup> and Ronald J. Warzoha<sup>3</sup>

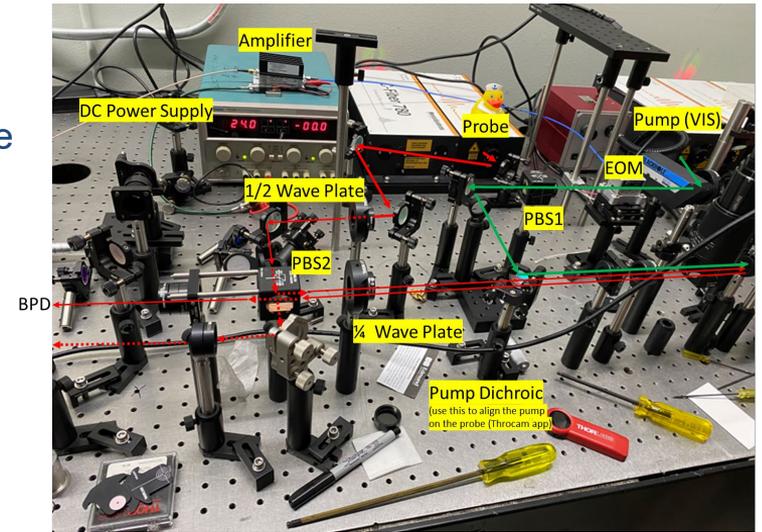
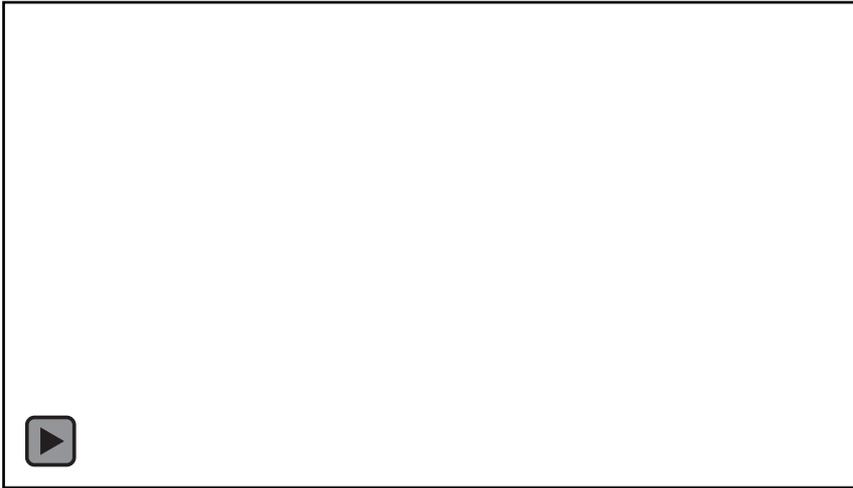


# Accessing Full Time Delay TDTR



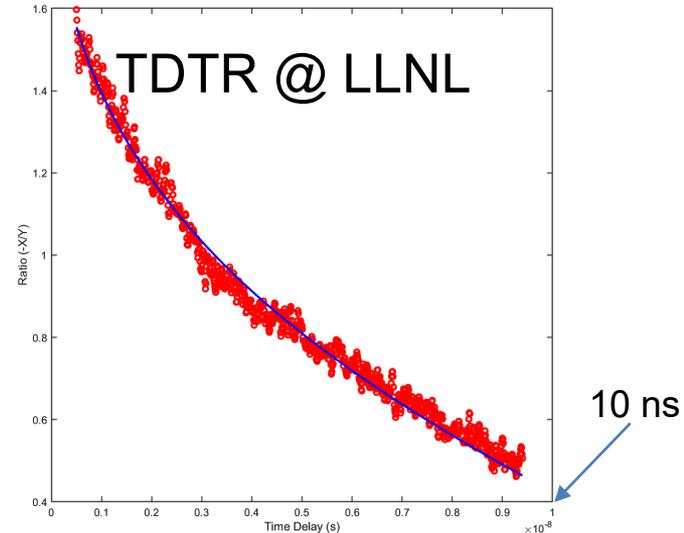
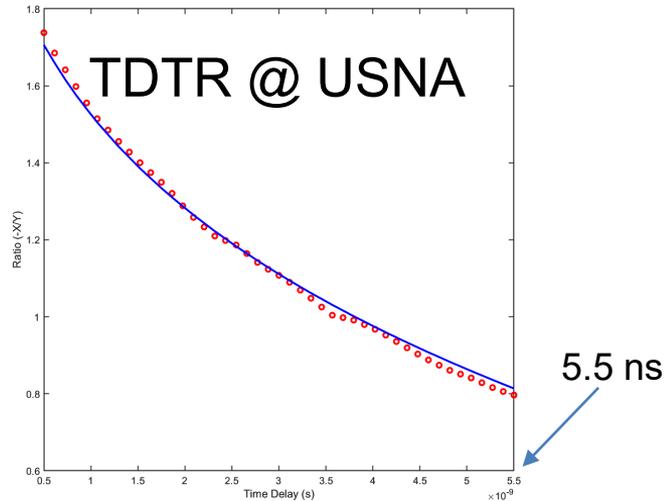
# Accessing Full Time Delay TDTR – ASOPS!

- Pump-Probe delay from “beat” signal
  - Slight rep-rate difference between pump and probe
- No Delay Stage!



# ASOPS TDTR

- Tons of data and fast scans (set by  $\Delta f$  and lock-in TC)
- Uniform time spacing (double edged sword!)
  - Hold sweeping in 1/10 windows in time (then you can set phase difference)
- Full Time delay



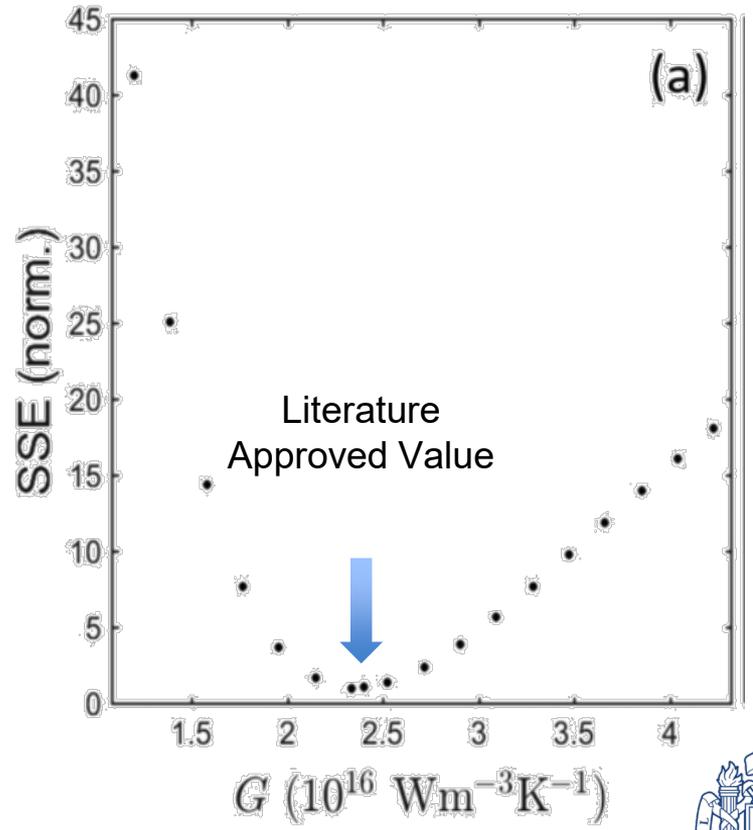
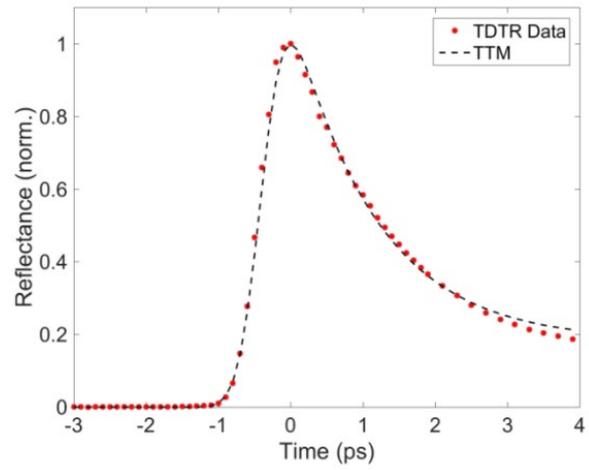
**You can determine lots of interesting  
thermal properties with Time Domain  
Thermoreflectance....**

**But it can be hard.  
(you might need more time)**



# Sensitivity

Do we ignore fitting for the physics?

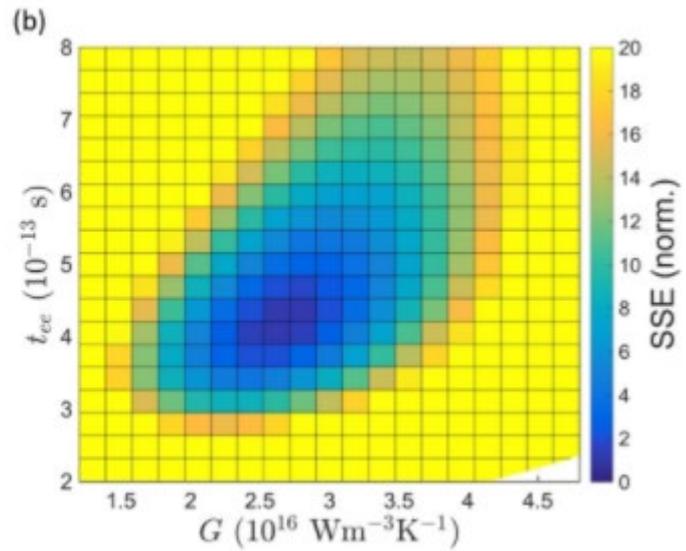
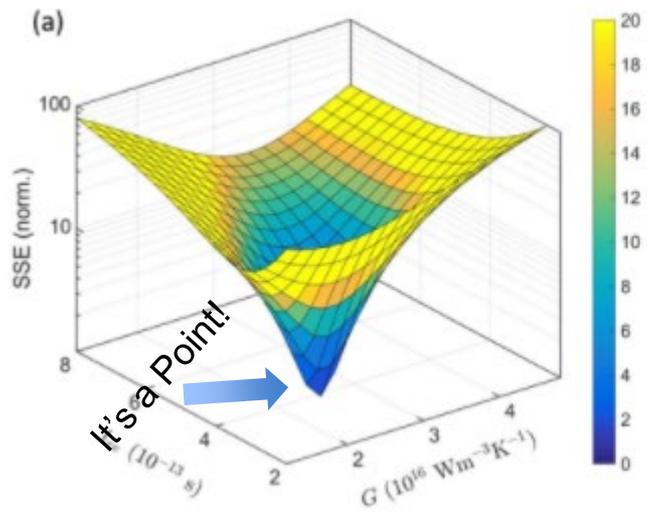


Naldo, Bernotas, Donovan. *J.A.P.* 2020.



# Sensitivity

What can we fit confidently?

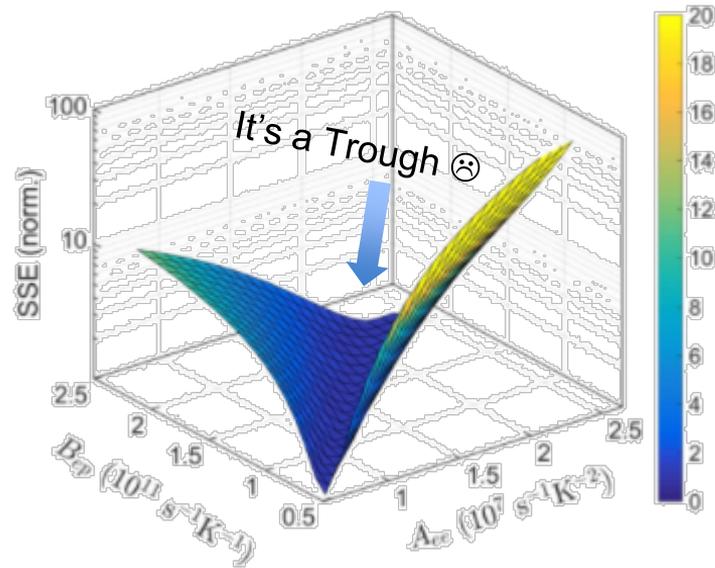
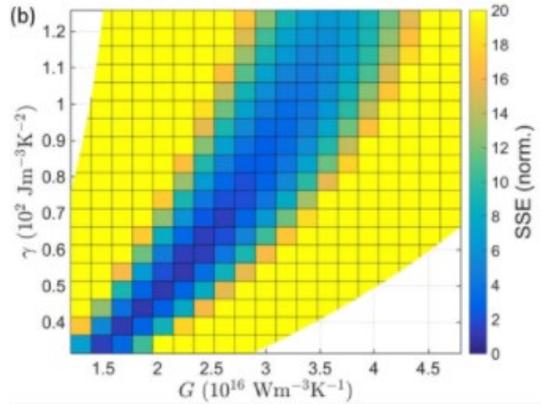
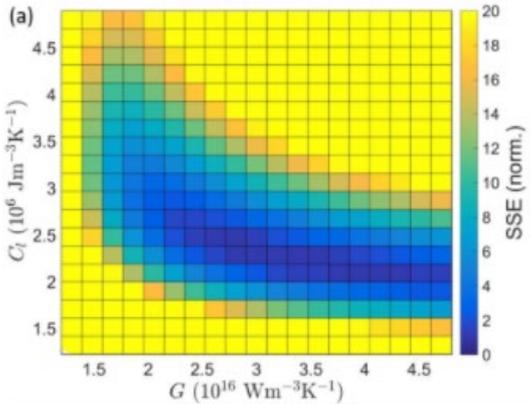


Electron relaxation time ( $t_{ee}$ ) and EP coupling ( $G$ ) can be fit for together.



# Sensitivity

What can we fit confidently?

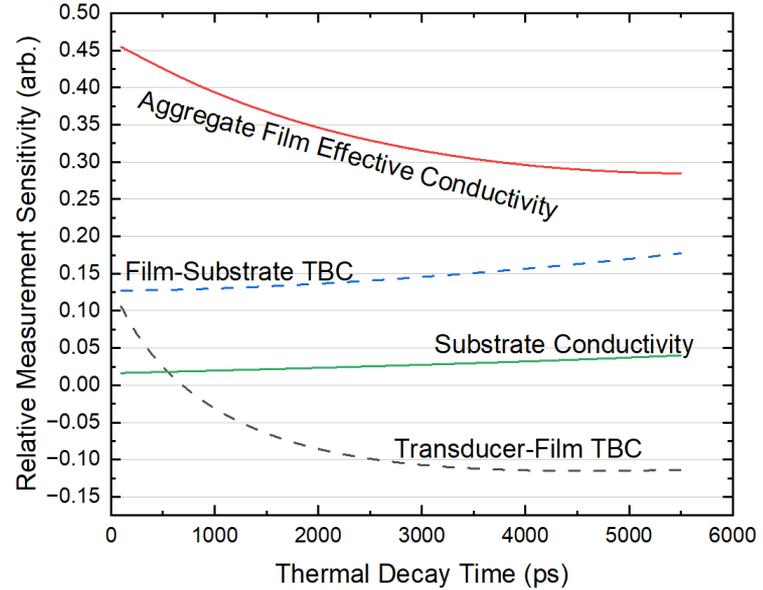
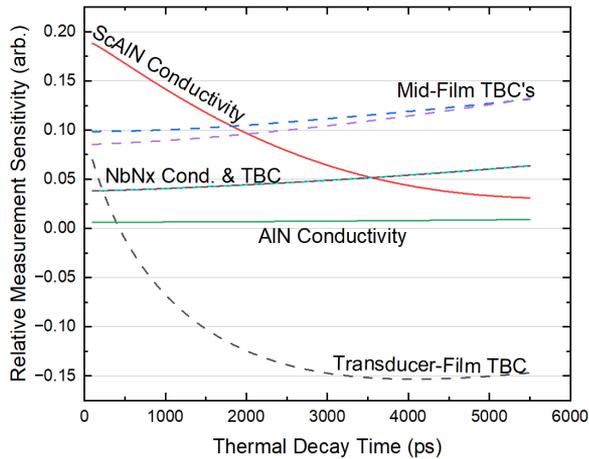


Certain parameters may lead to a false confidence in fitted values!



# Sensitivity

## What can we fit confidently?



Maybe if you aren't sensitive to it, it's not the thing that matters...

**You can determine lots of interesting thermal properties  
with Time Domain Thermoreflectance....**

**But it can be hard.  
(you might need more time)**

**And you should check your physical sensitivity  
and your fitting uncertainty**

Are those  
different???



# TDTR: Time Domain ThANKYOUermoreflectance



**NIST**



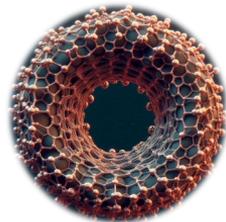
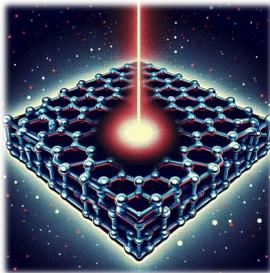
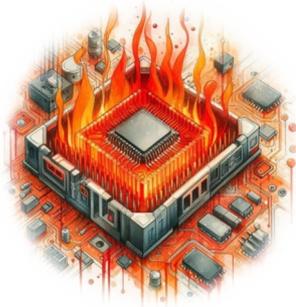
**LASER  
THERMAL**



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# Thanks!!!

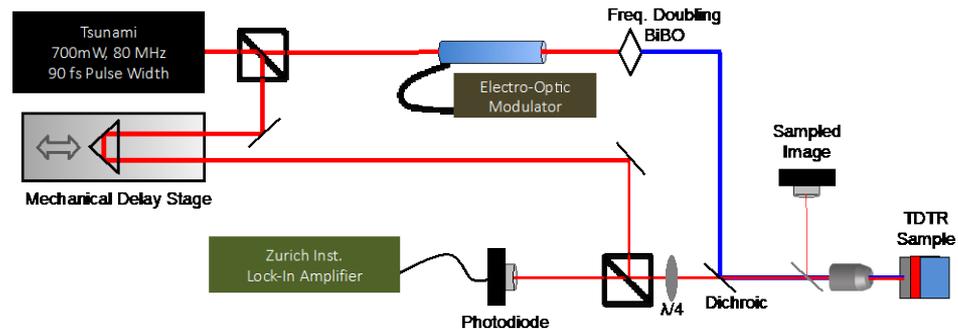
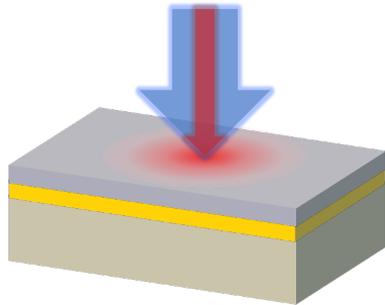
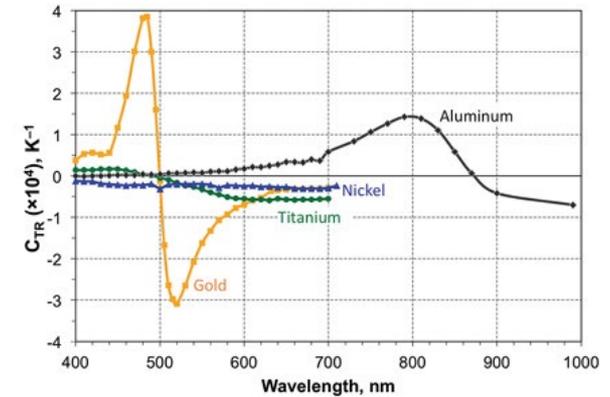


# NLST

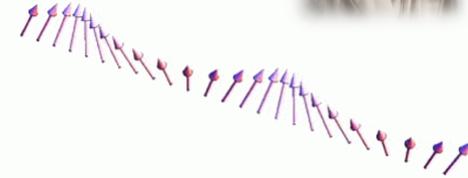
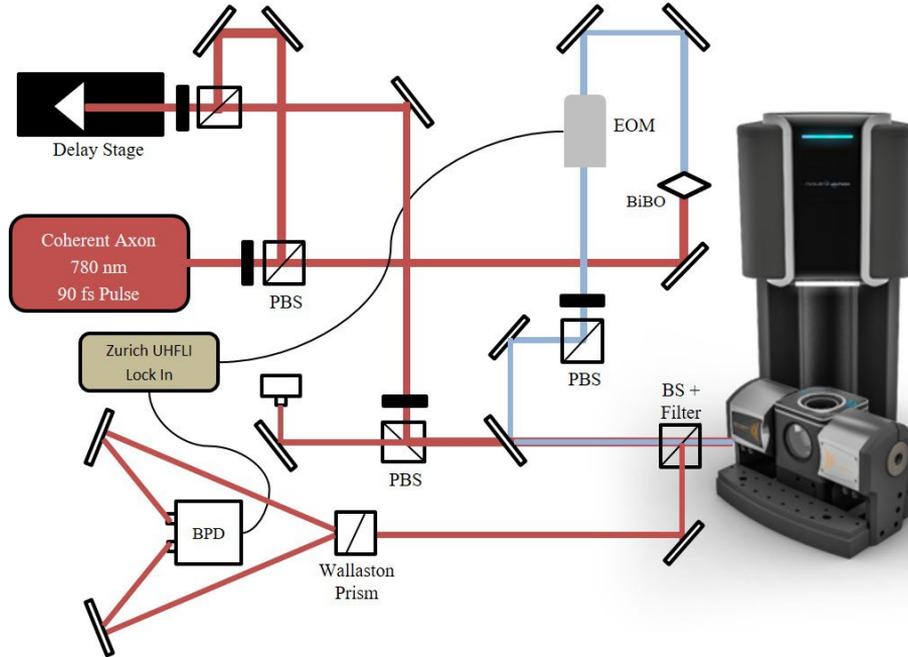


# Optical Modulated Heating & Sensing

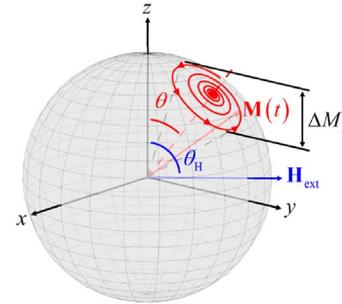
- Change in reflectance with temperature ( $\sim 10^{-5}$ )
- Modulated optical absorption
- Pump-Probe thermal measurement



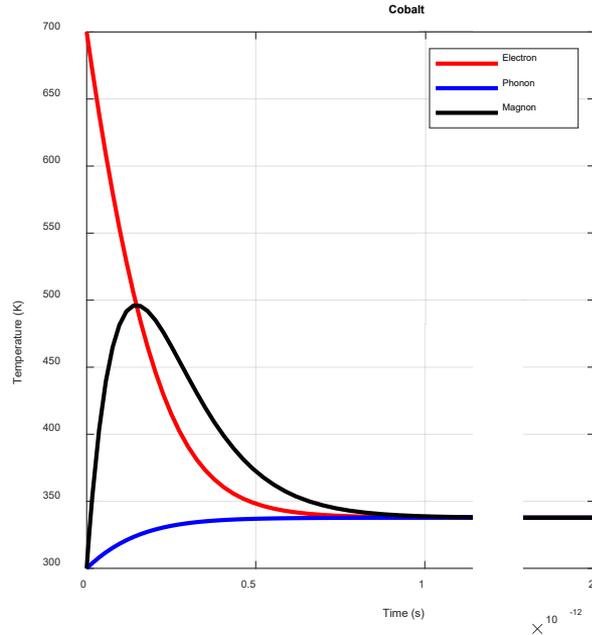
# Time Resolved Magneto Optic Kerr Effect



# Ultrafast Magneto-dynamics on Nanoparticles



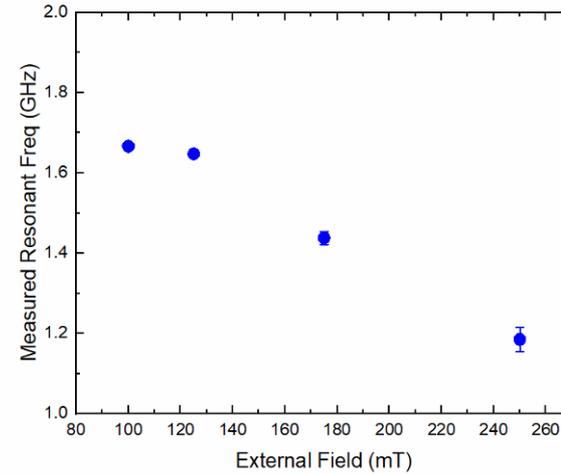
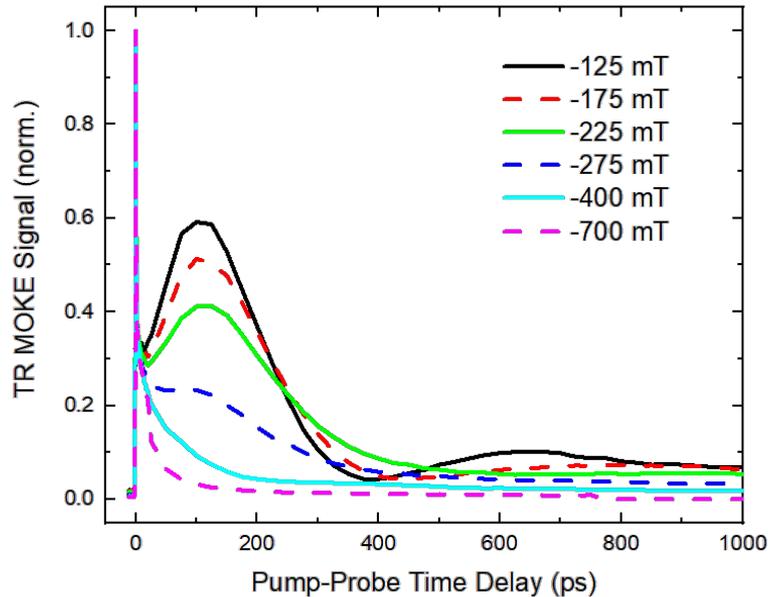
Lattery et al., APL 113 (2018)



$$A + \exp\left(-\frac{t}{t_{ph}}\right) + B * \exp\left(-\frac{t}{t_m}\right) * \cos(2\pi ft)$$



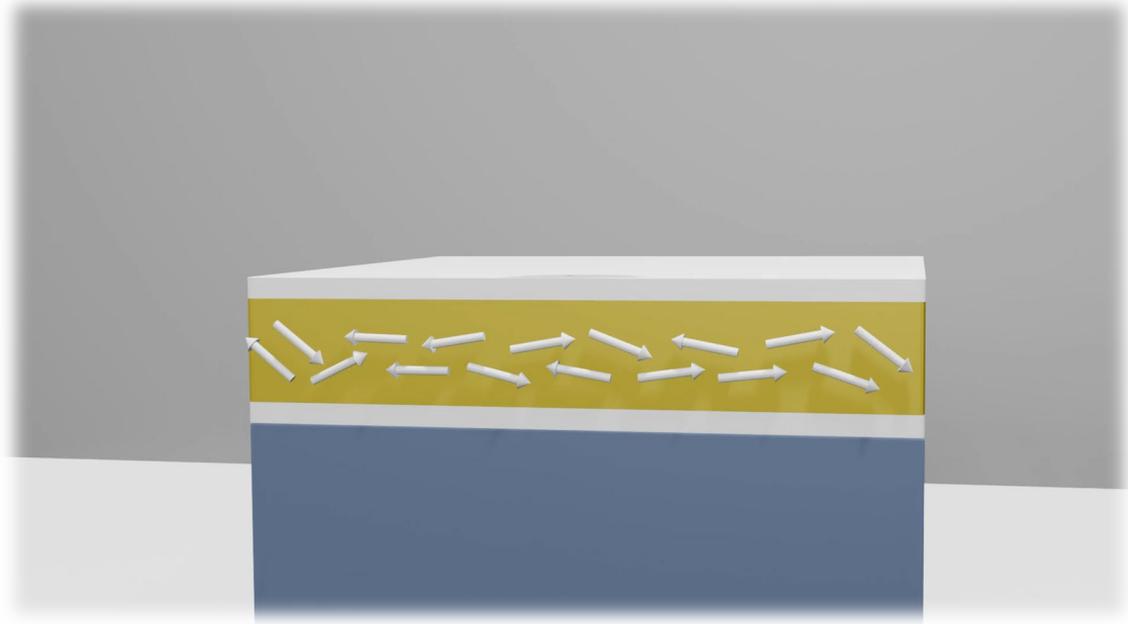
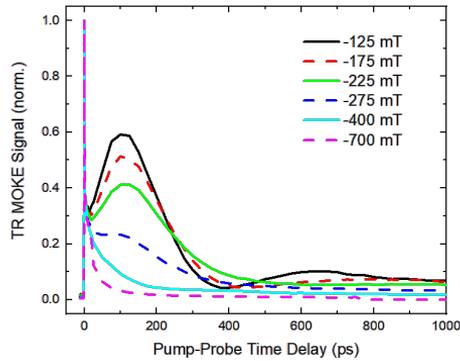
# Ultrafast Magneto-dynamics on Nanoparticles



Magnetic resonance measurements coupled with energy transport



# Ultrafast Magneto-dynamics on Nanoparticles



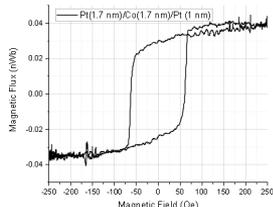
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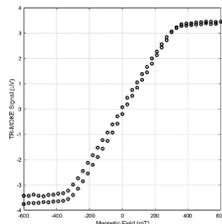
Pt/Co/Pt thin films (1.7 nm Co)

Magnetic anisotropy in-plane

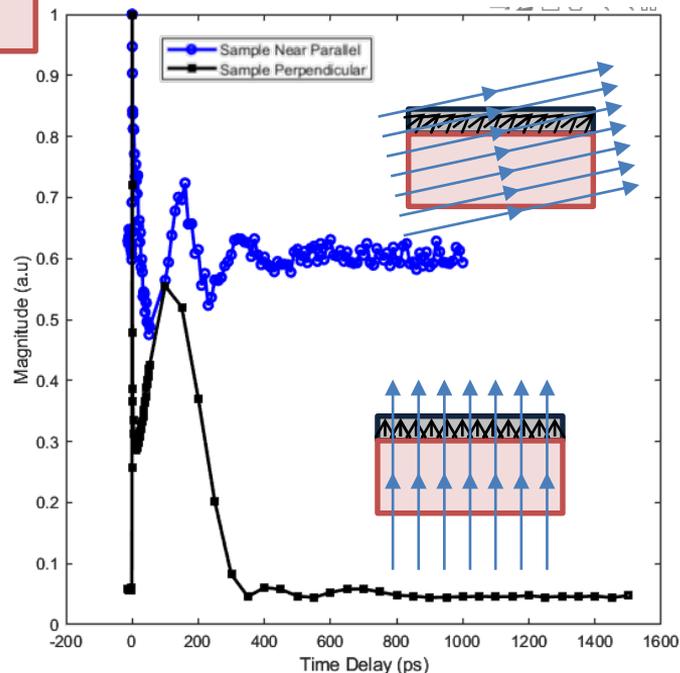
Clear difference in TR-MOKE signature depending on field direction



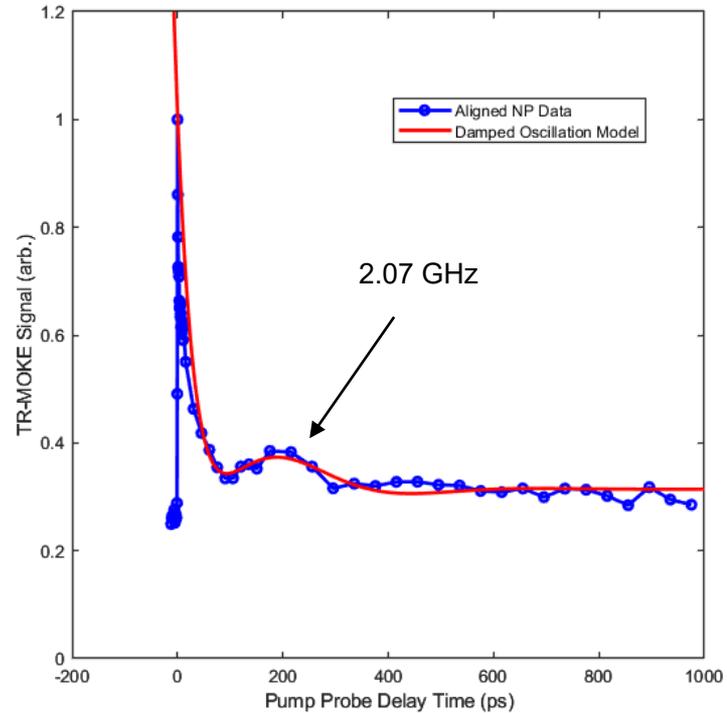
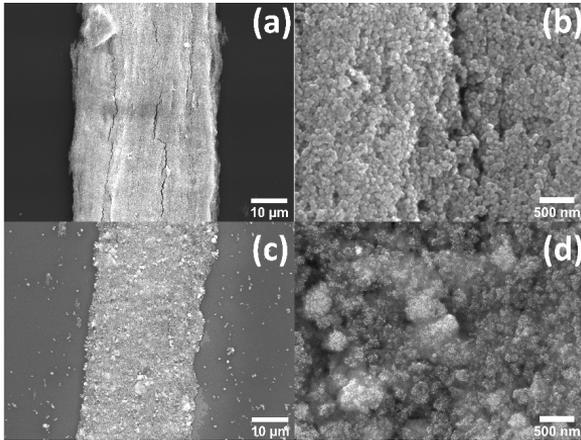
In Plane



Out of Plane



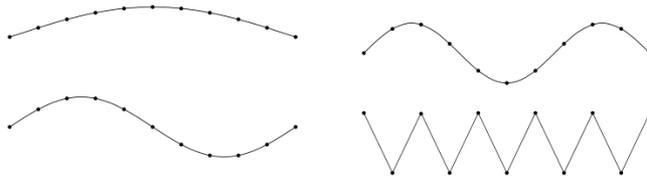
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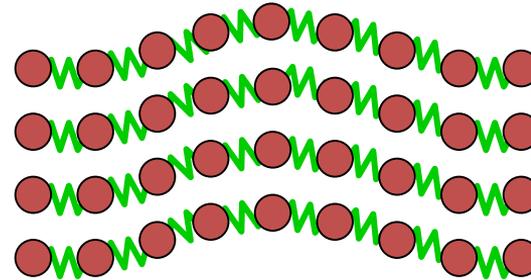
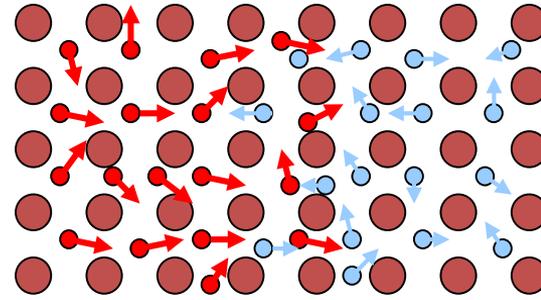
# Nanoscale Thermal Physics

$$\kappa_{tot} = \kappa_L + \kappa_e$$

- Electrons
  - Carrier of electronic current
  - Dominate in conductors
- Phonons
  - Quantization of lattice vibrations
  - Dominant in semiconductors & insulators



Diffusion of “hot” electrons →



Phonon propagation →

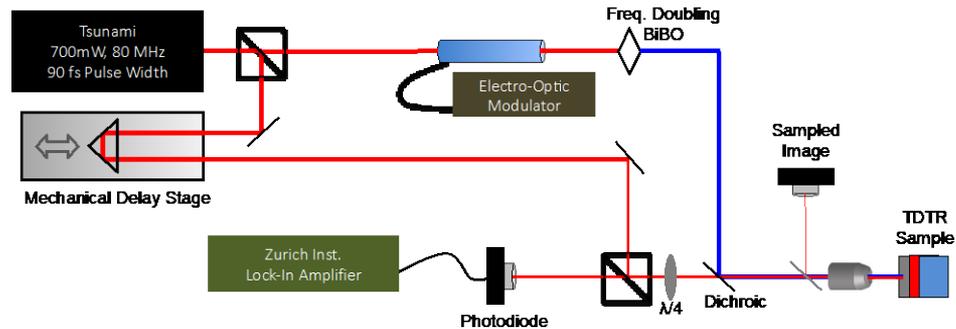
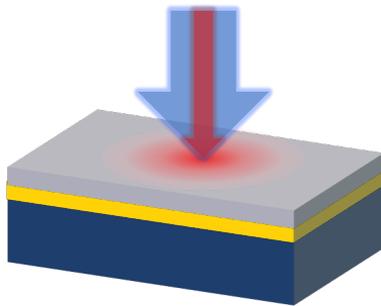
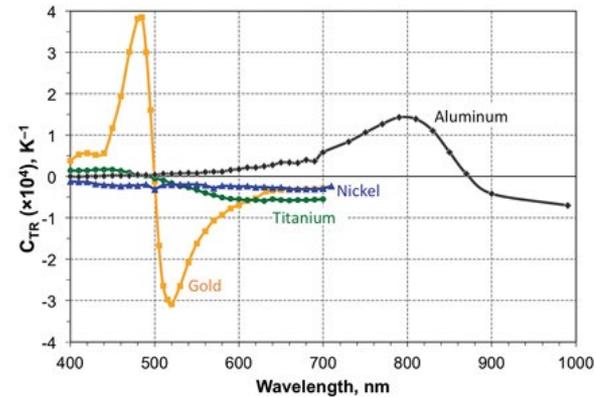
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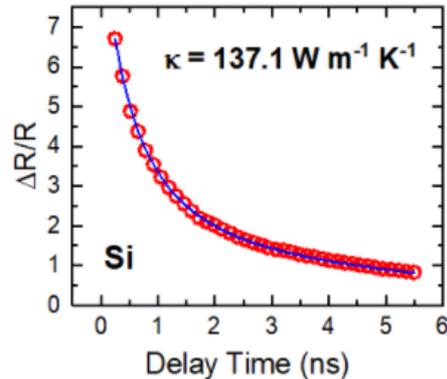
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# Data Fitting

- Data taken is Thermoreflectance vs. Time
- Fit for thermo-physical parameters we care about
  - Thermal conductivity, thermal conductance, heat capacity
  - Least squares nonlinear fitting (thanks Matlab!)



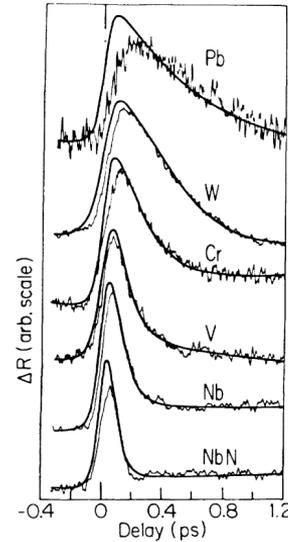
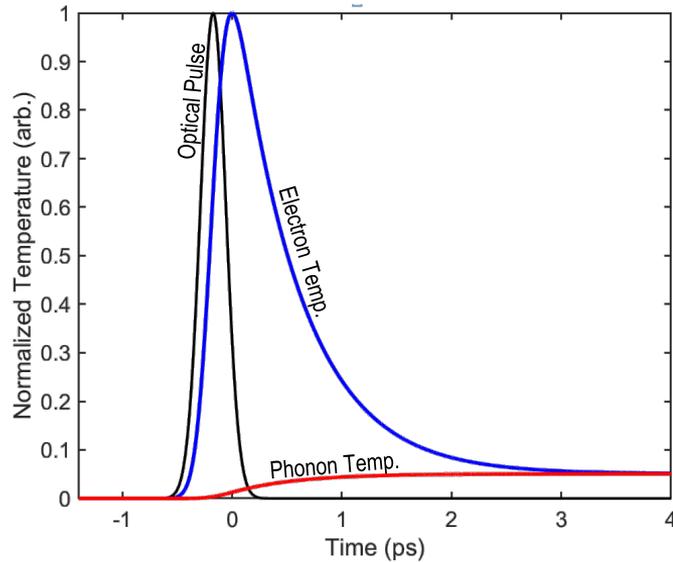
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# Electron-Phonon Coupling

Optically excited electrons scatter with atomic nuclei resulting in increased phonon activity.



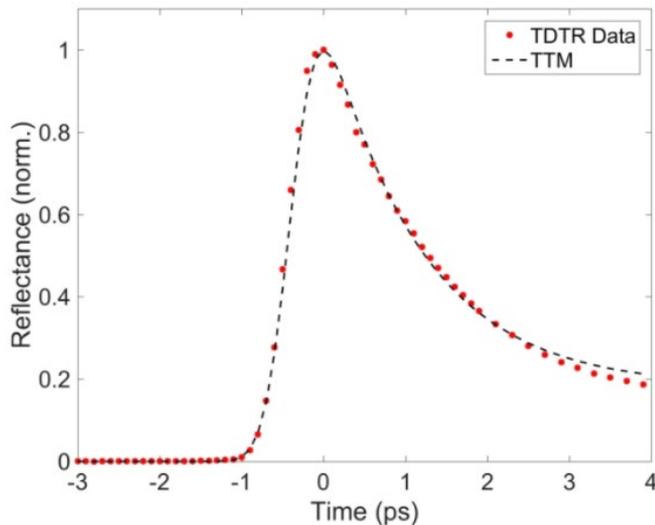
Brorson *et al.* PRL 1990

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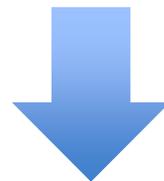
# The Two Temperature Model



$$\gamma T_e \frac{\partial T_e}{\partial t} = G(T_p - T_e) + S(t)$$

$$C_p \frac{\partial T_p}{\partial t} = G(T_e - T_p)$$

$$S(t) = \frac{0.94F_A}{d(t_p + t_{ee})} \times \exp \left[ -2.77 \left( \frac{t - 2(t_p + t_{ee})}{t_p + t_{ee}} \right)^2 \right]$$



Need to Understand  
Fitting Sensitivities!

$$SSE = \sum_{i=1}^n (y_i - F(\{b_1, b_2, \dots, b_m\}, x_i))^2$$



Naldo, Bernotas, Donovan. *J.A.P.* 2020.

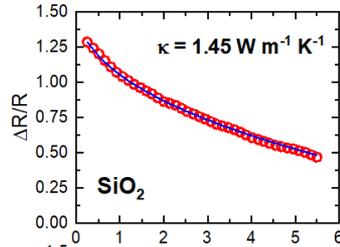
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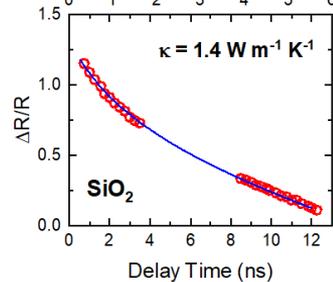


# Sensitivity – Thermal Parameters

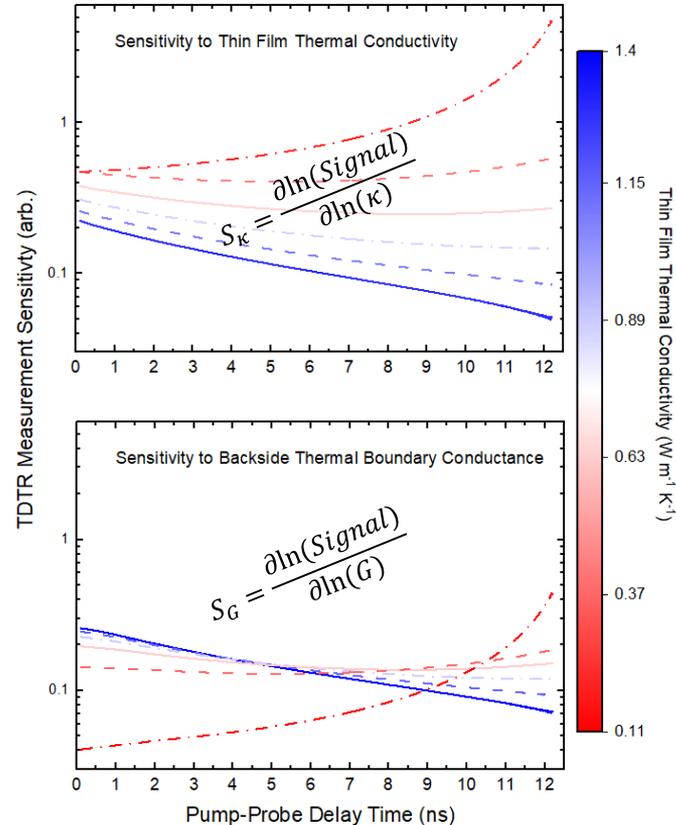
Another approach from our lab → More Thermal Diffusion Time



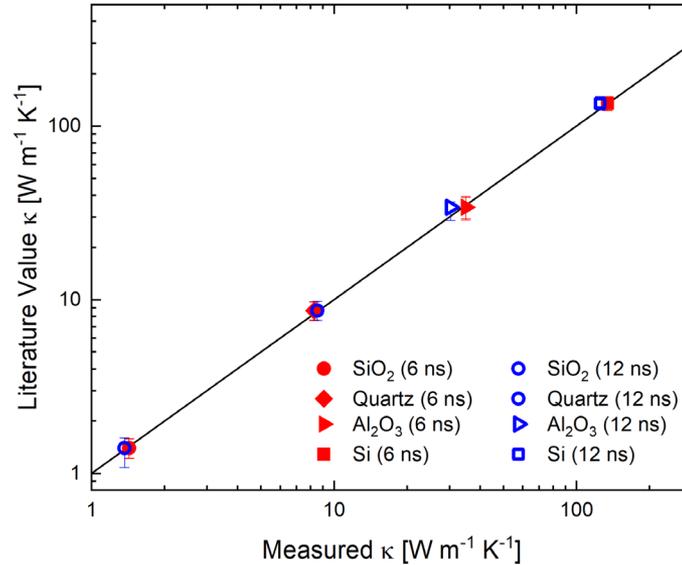
Typical Scan



Long-time Scan



# Longtime Cals



# EP Coupling Optics

$$R = \frac{(n_1 - 1)^2 + n_2^2}{(n_1 + 1)^2 + n_2^2}, \quad (5)$$

where  $n_1$  and  $n_2$  are the real and imaginary parts of the complex index of refraction. These are given by

$$n_1 = \frac{1}{\sqrt{2}} [(\epsilon_1^2 + \epsilon_2^2)^{1/2} + \epsilon_1]^{1/2}, \quad (6)$$

$$n_2 = \frac{1}{\sqrt{2}} [(\epsilon_1^2 + \epsilon_2^2)^{1/2} - \epsilon_1]^{1/2}, \quad (7)$$

where  $\epsilon_1$  and  $\epsilon_2$  are the real and imaginary parts of the optical dielectric function,  $\hat{\epsilon}$ , respectively. This is where the temperature dependence of the reflectivity comes in, particularly for a system that is out of equilibrium with different temperatures with respect to the electron and phonon populations,  $T_e$  and  $T_l$ , respectively,

$$\hat{\epsilon} = 1 - \frac{\omega_p^2}{\omega^2 + i\omega(A_{ee}T_e^2 + B_{ep}T_l)}. \quad (8)$$

In this equation,  $\omega$  is the frequency of the absorbed light and  $\omega_p$  is the plasma frequency.  $A_{ee}$  and  $B_{ep}$  are scattering coefficients

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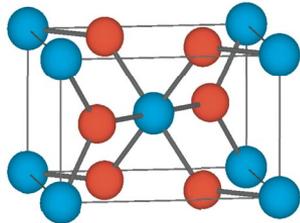
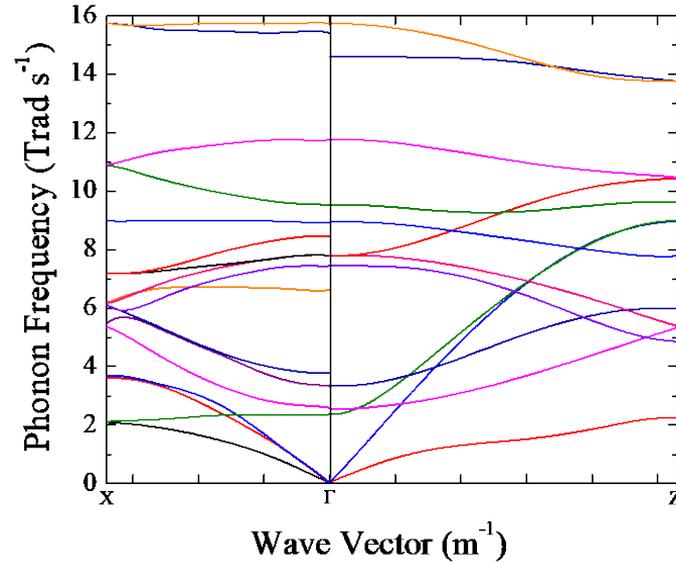


# Anisotropic Phonon Thermal Conductivity

44

Donovan *et al.* Under Review

- Isotropic crystal
  - Model Brillouin zone as a sphere
  - $\kappa_{xx} = \kappa_{yy} = \kappa_{zz}$
- Tetragonal Crystal
  - Model Brillouin zone as a cylinder
  - Need  $\Gamma \rightarrow X$  and  $\Gamma \rightarrow Z$
  - $\kappa_{xx} = \kappa_{yy} \neq \kappa_{zz}$



$$K = \begin{bmatrix} K_{xx} & K_{xy} & K_{xz} \\ K_{yx} & K_{yy} & K_{yz} \\ K_{zx} & K_{zy} & K_{zz} \end{bmatrix}$$

