



UNIVERSITÀ
DEGLI STUDI DI TRIESTE



Advances in time domain spectroscopies, a table-top approach complementary to FEL and SR sources



*Daniele Fausti,
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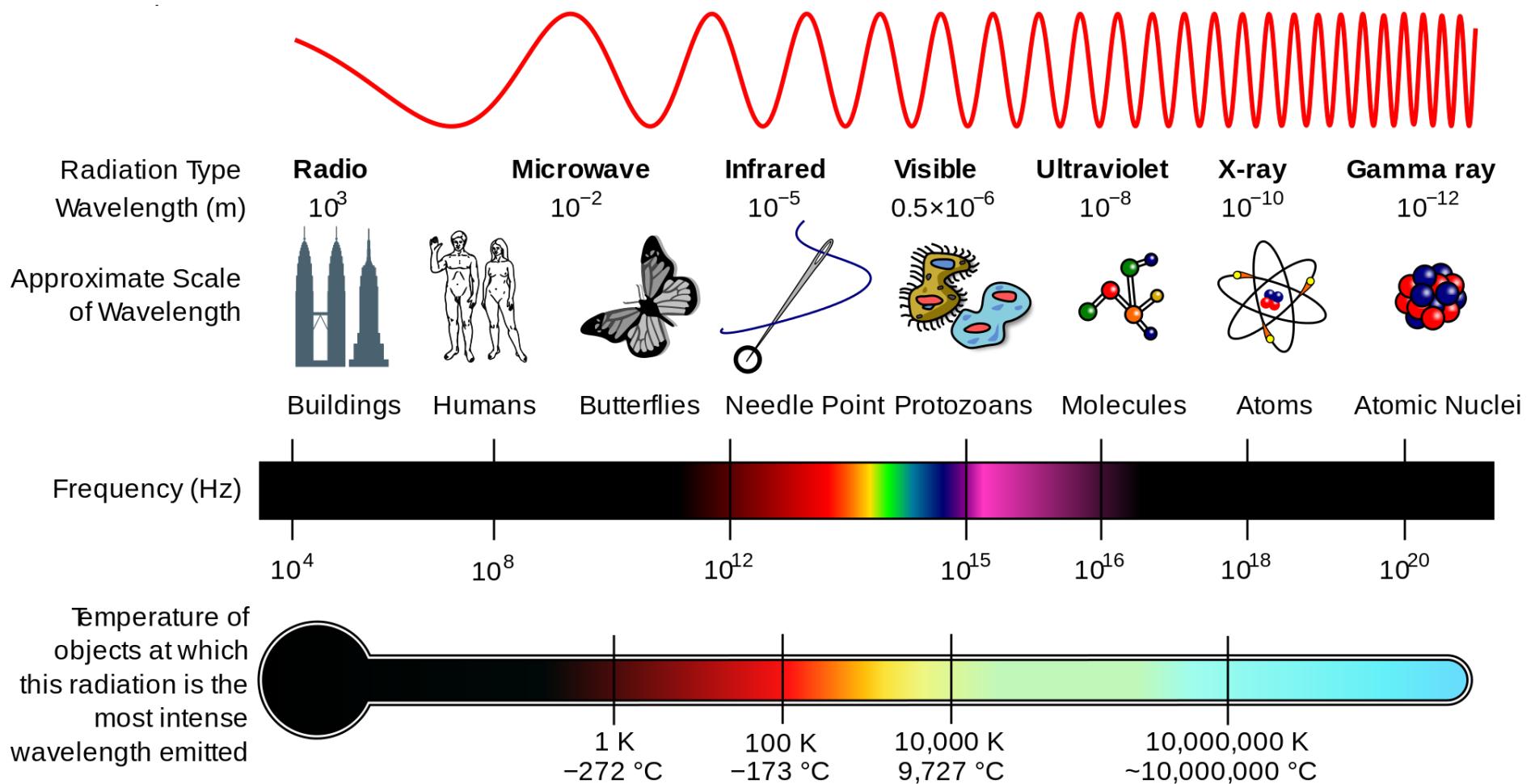


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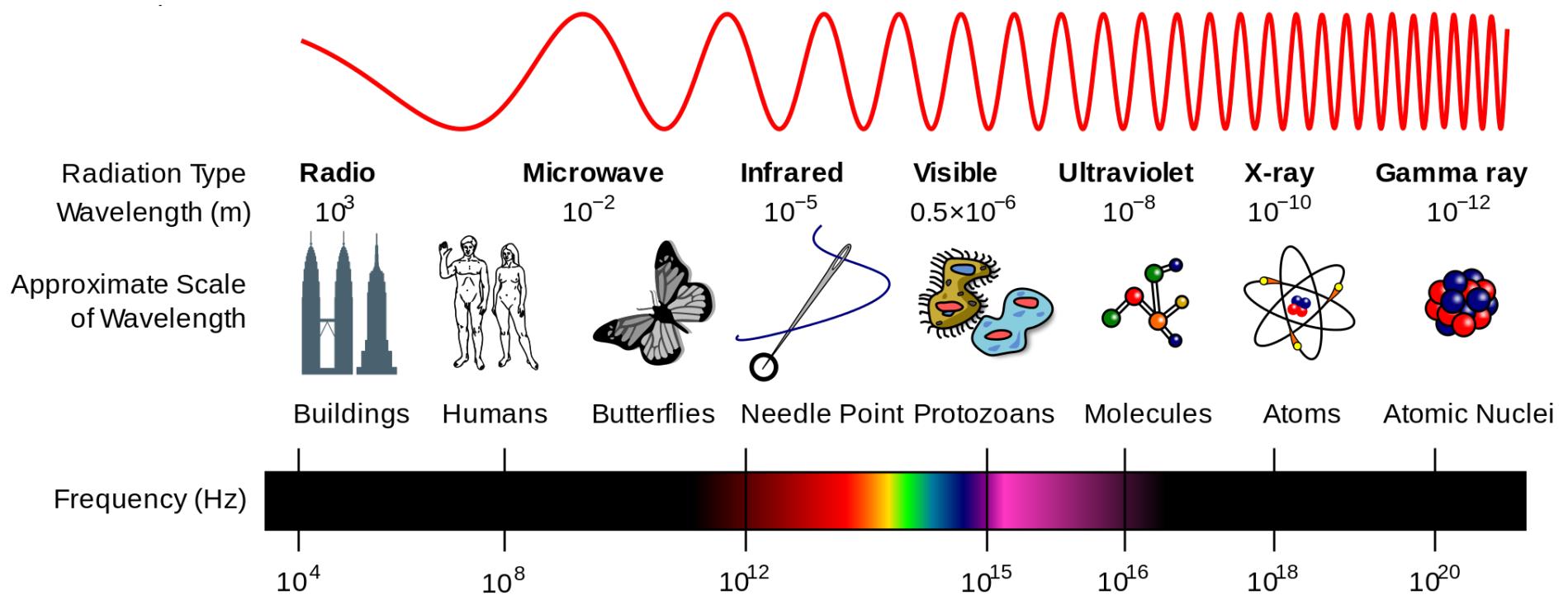


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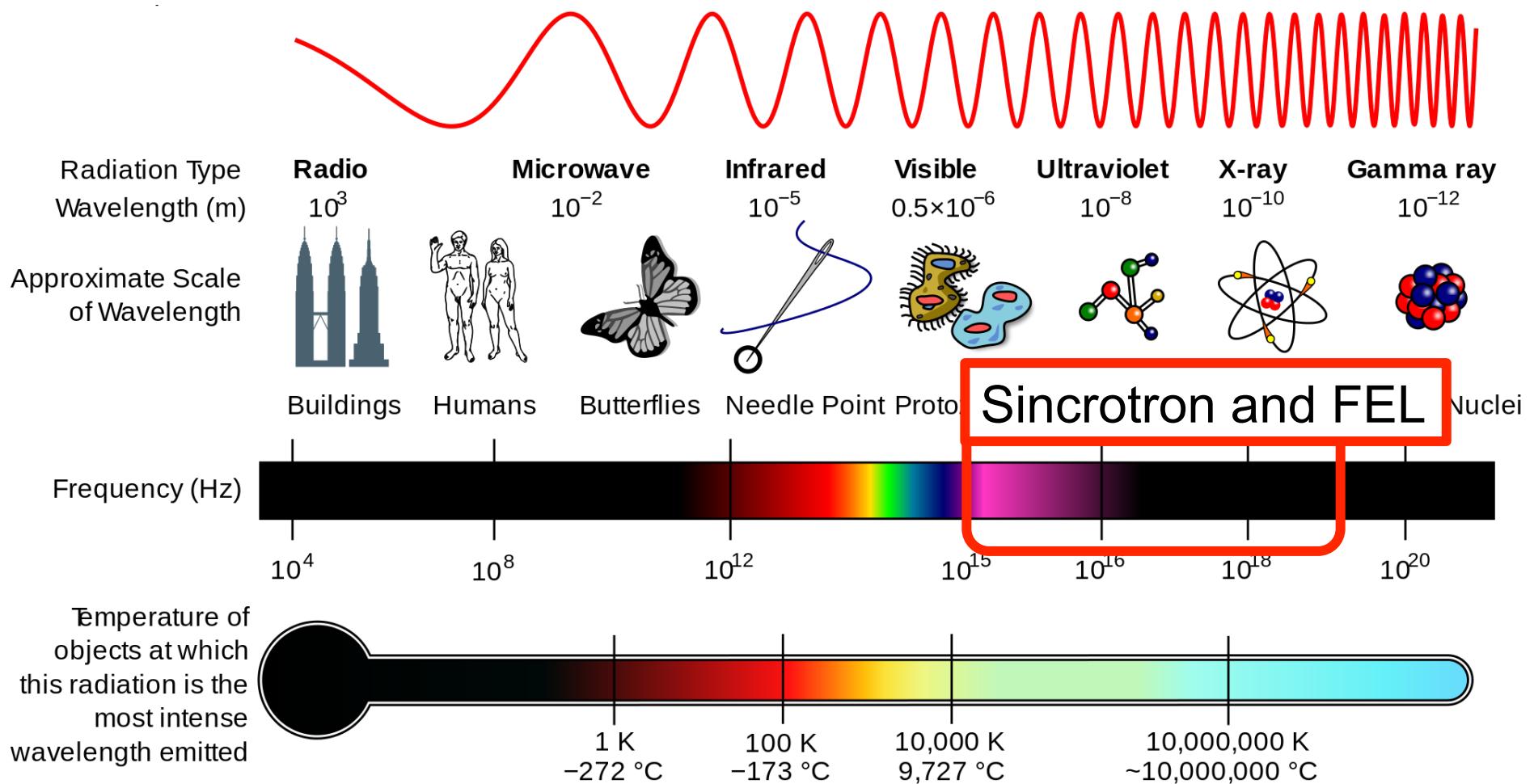
The energy scale of Table top



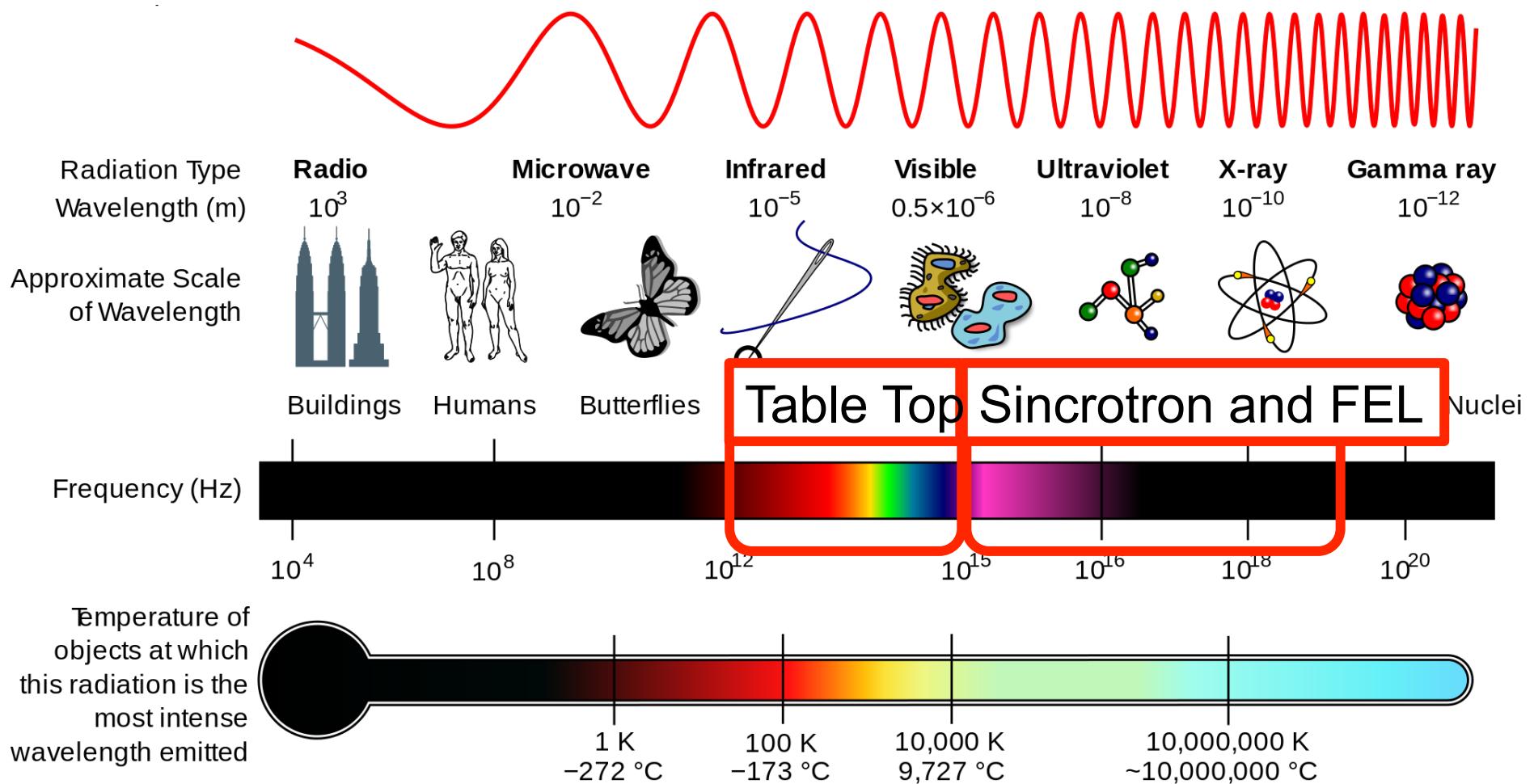
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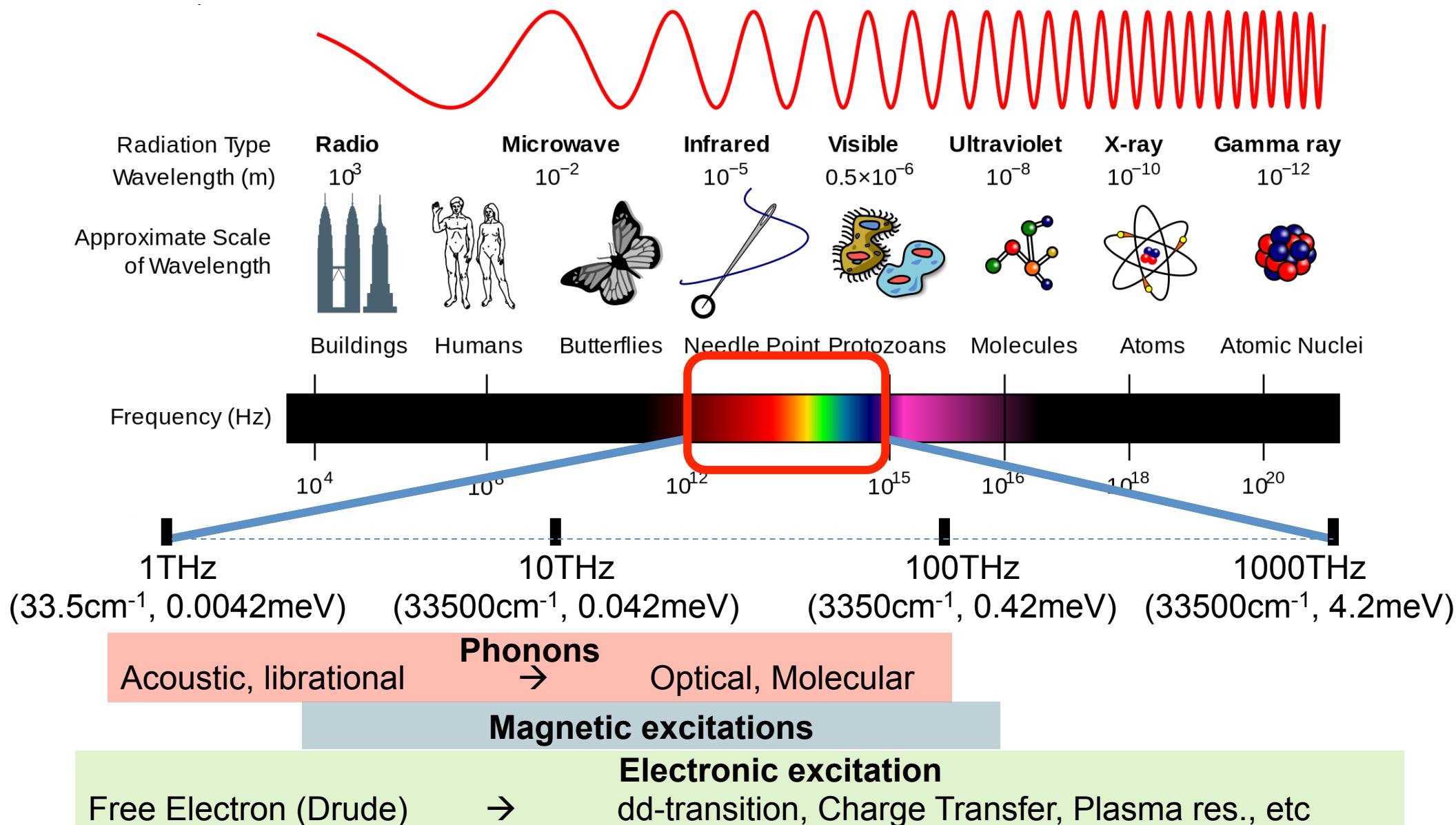
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Outline

✓ Equilibrium Optical Spectroscopy

- The optical conductivity in the Visible and Near-IR
- What do we learn from optical conductivity?
- An example, metal insulator transition in complex oxides

✓ Non-equilibrium optical (visible near-IR) spectroscopy

- Pump&probe the main idea
- «Single color» Pump and probe
- Broadband P&P spectroscopy

✓ Self referential examples:

- Revealing the excitonic nature of excitation (Hubbard Exciton)
- The electron-phonon (EP) interaction in with strong electronic correlation

✓ Non-equilibrium Infrared spectroscopy

- Optical pump and THz probe spectroscopy (technique and example)
- THz pump and optical probe spectroscopy
- Phonon pump optical probe spectroscopy (MidIR pulse generation)

✓ Perspectives

- Beyond classical spectroscopy
- Using the quantum state of light as a new spectroscopyc tool
- Table top Vs. FEL and sincrotrons

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Equilibrium optical conductivity

Characterization of the response of the material
to a field at a specific frequency

-Optical conductivity $\rightarrow \sigma_1(\omega)$ and $\sigma_2(\omega)$

*Links the current density to the electric field for
general frequencies $\rightarrow J(\omega) = \sigma(\omega)E(\omega)$*

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$$\tilde{\epsilon(\omega)} =$$

$$+ \sum_{i=1}^M \frac{\omega_{P,i}^2}{(\omega_i^2 - \omega^2) - i\Gamma_i\omega}$$

$$\omega_P^2 = \frac{4\pi Ne^2}{m}$$

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$\Gamma_0(\omega)$ frequency dependent to describe all scattering processes

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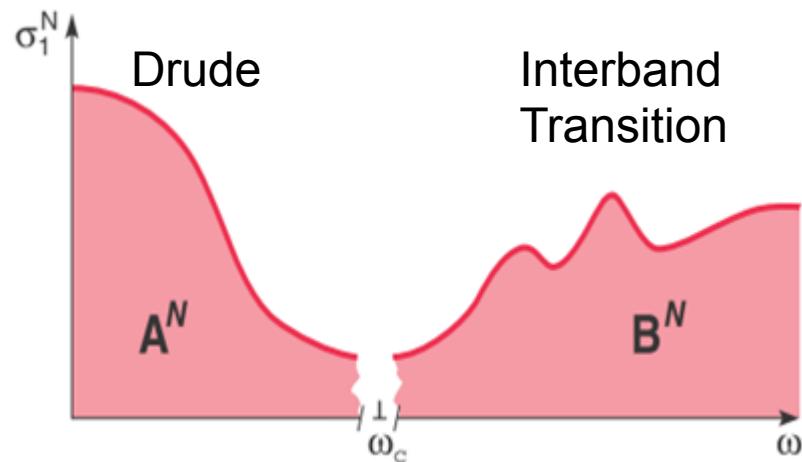
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Integral of sigma one is a conserved quantity (sum rule...)

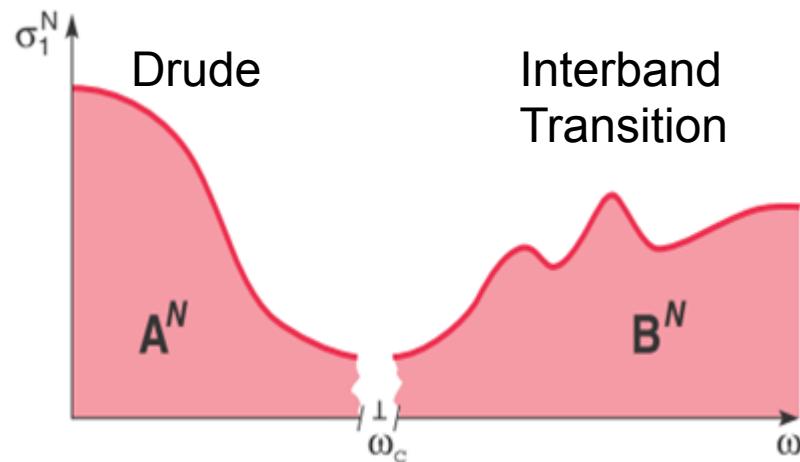
What do we learn from equilibrium optical conductivity?

Optical Conductivity of a Metal

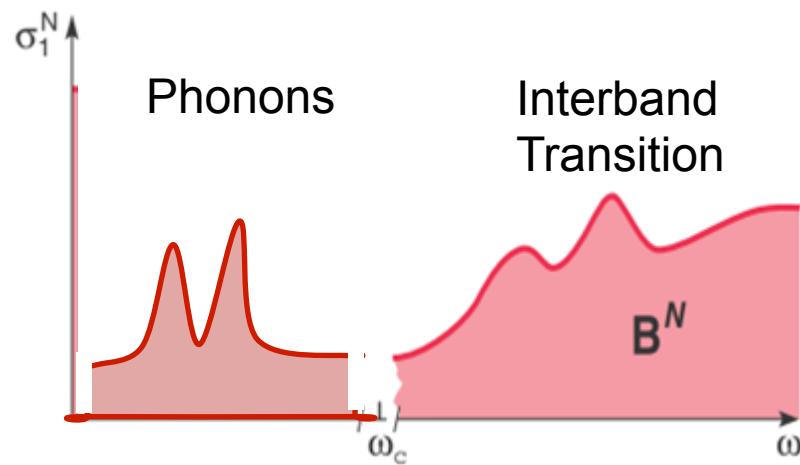


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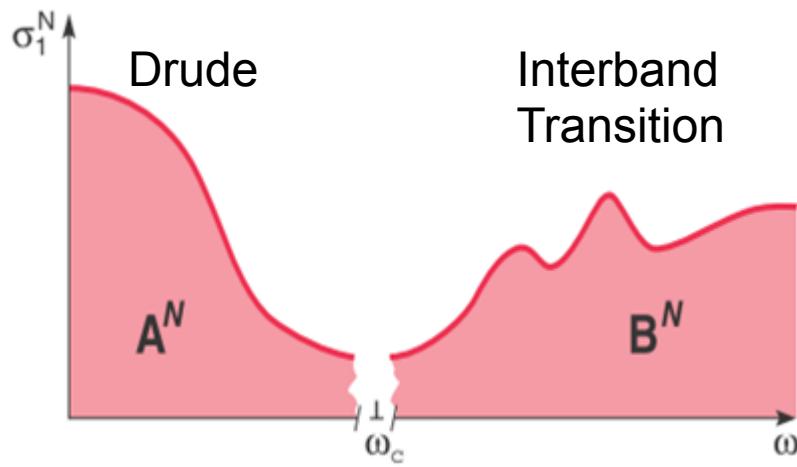


Optical Conductivity of an Insulator



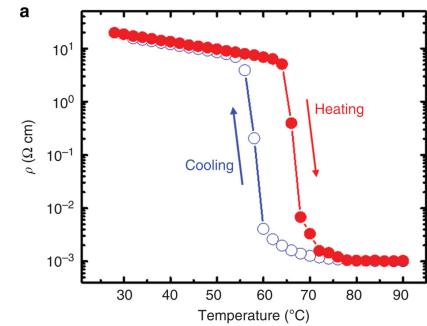
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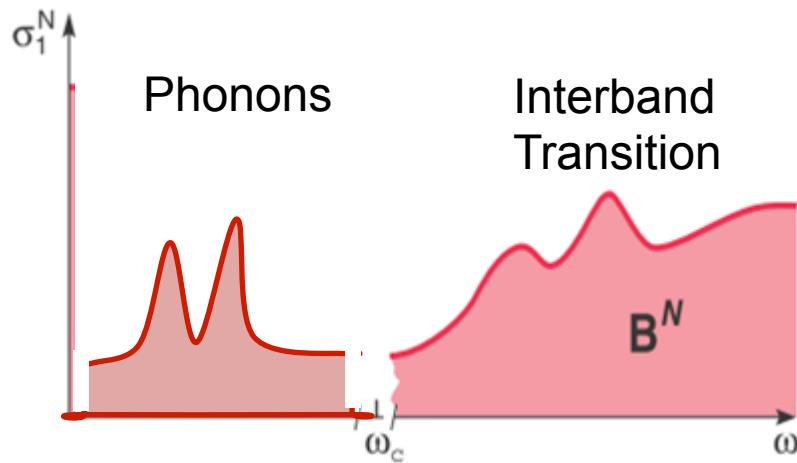


Metal insulator Transition in VO₂

Transition to a metallic state at 60 °C

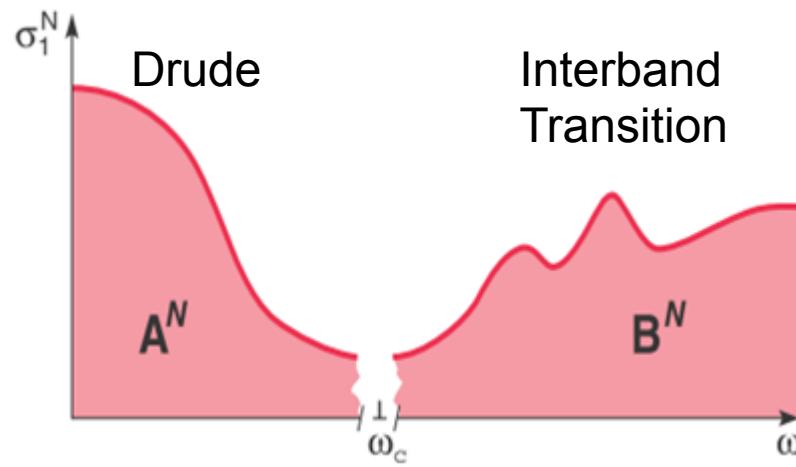


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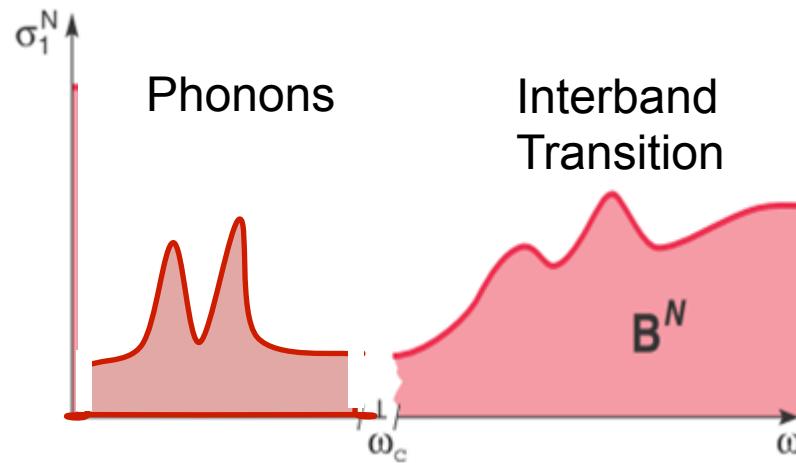


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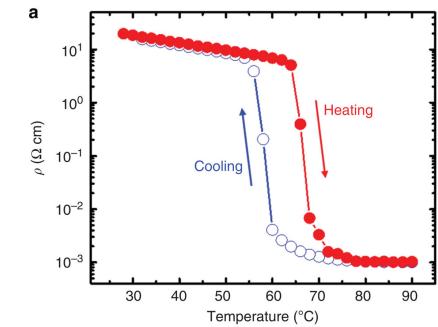


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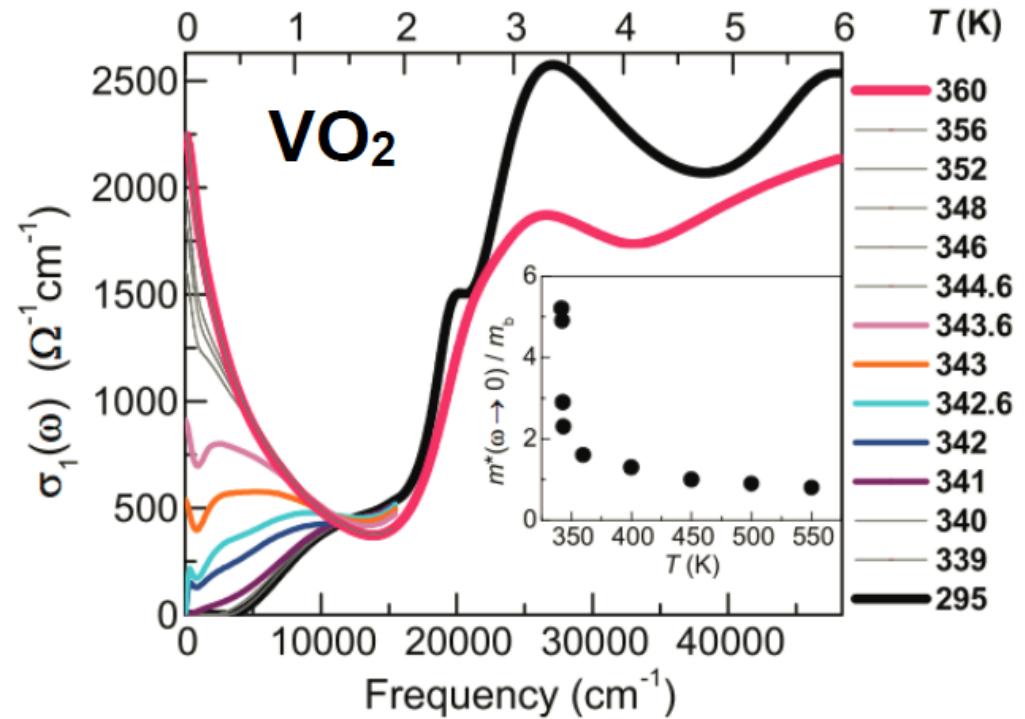


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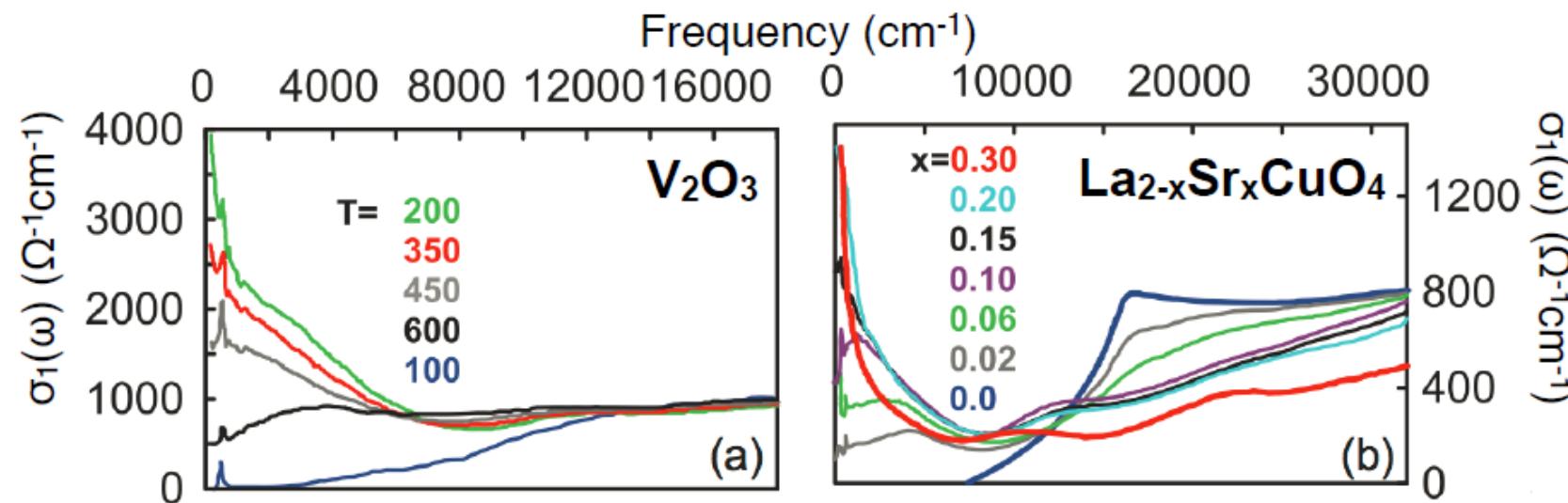
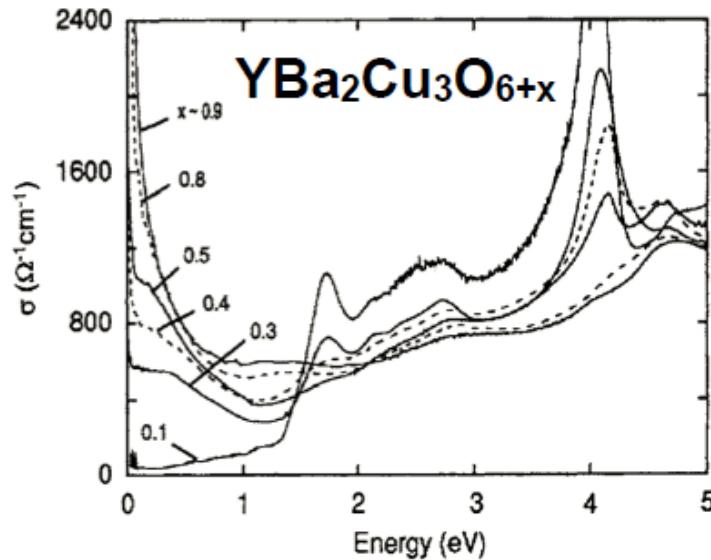


Photon energy (eV)



What do we learn from equilibrium optical conductivity?

Different examples of optical conductivity



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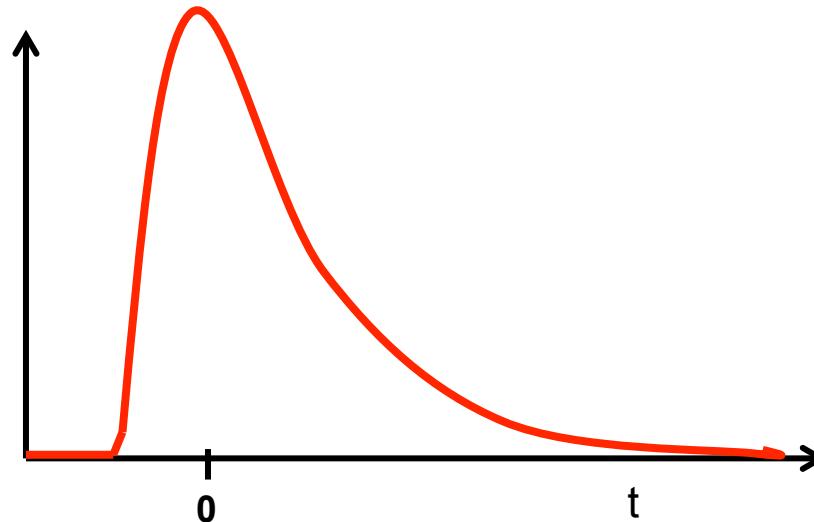
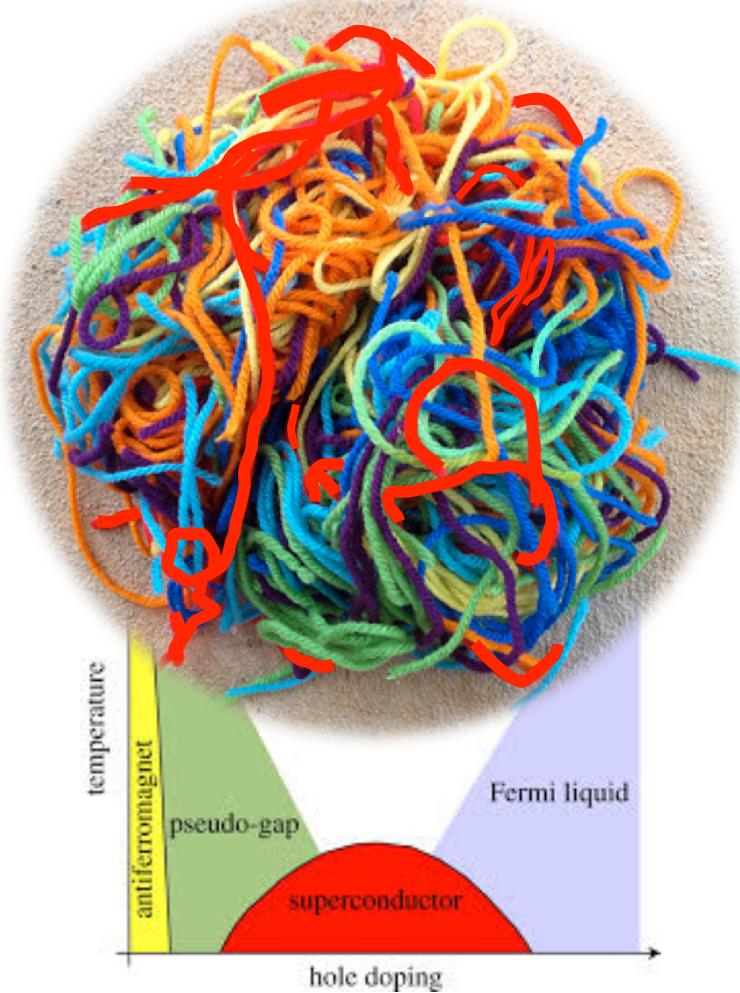
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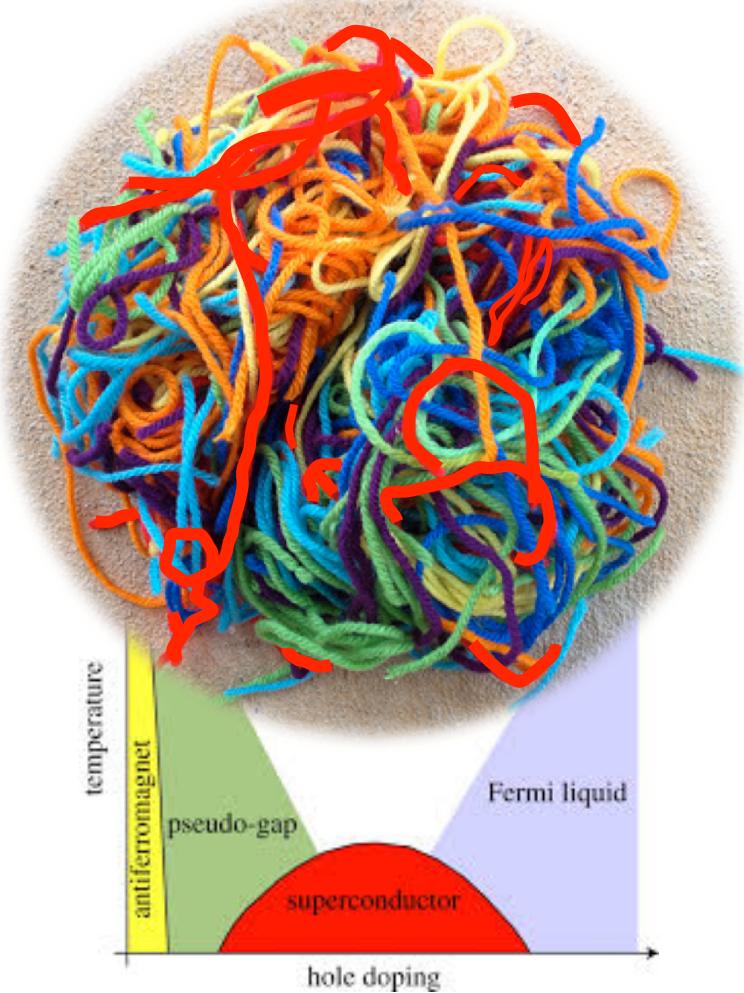
Why non-equilibrium physics?

Non-Equilibrium studies
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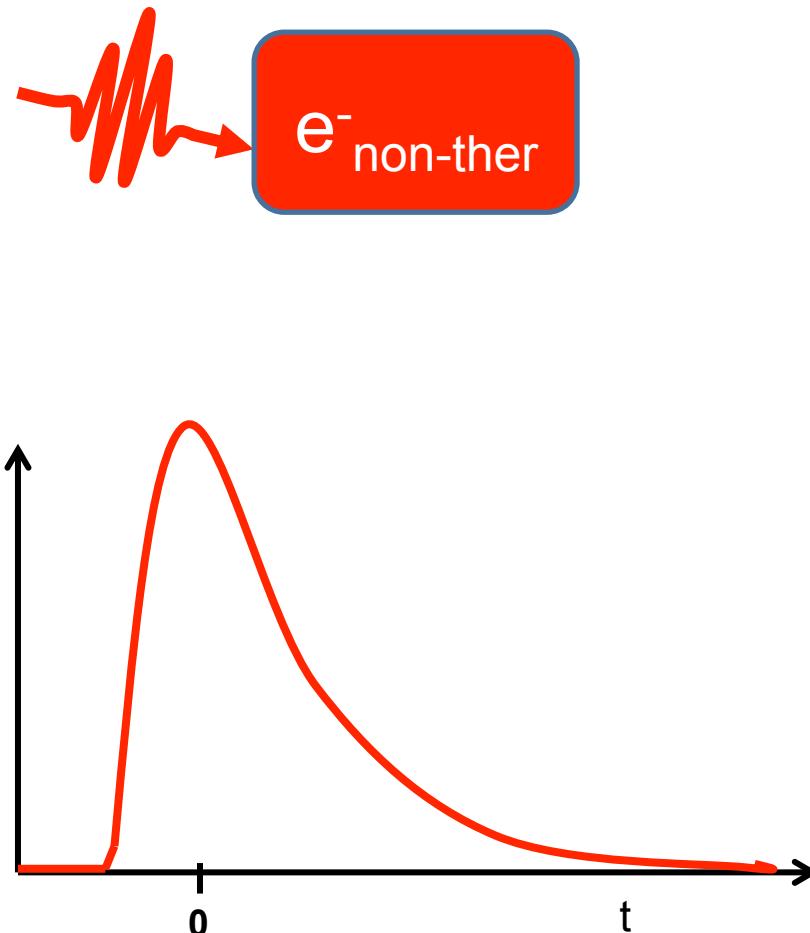


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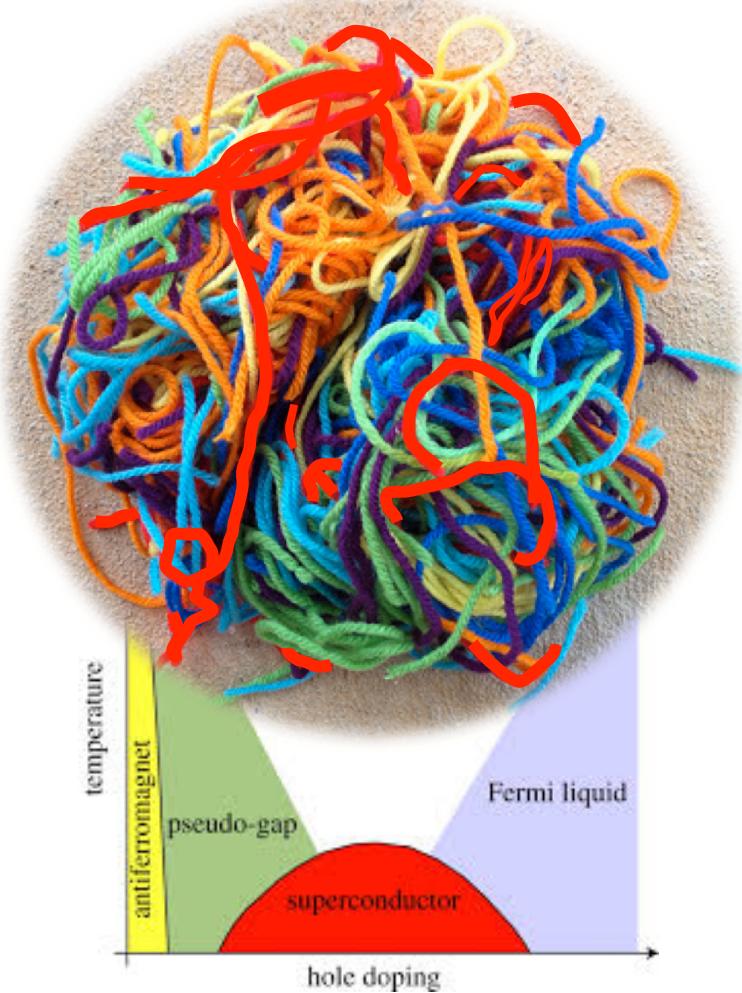
Dynamical response dominated by a
Higherarchy in the time scales



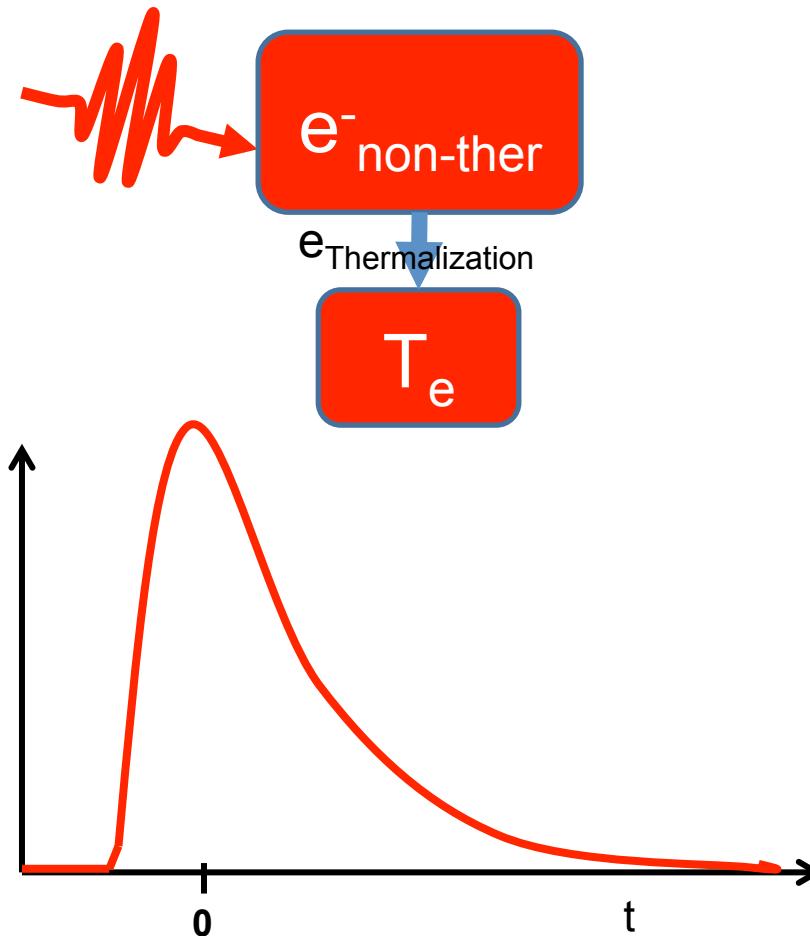
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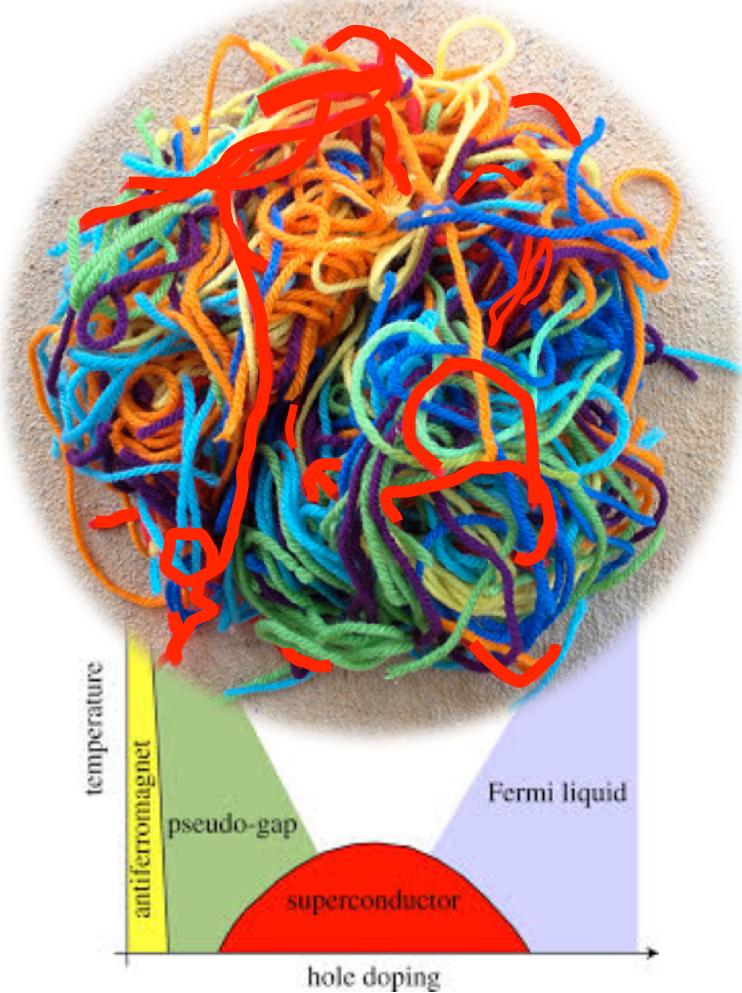
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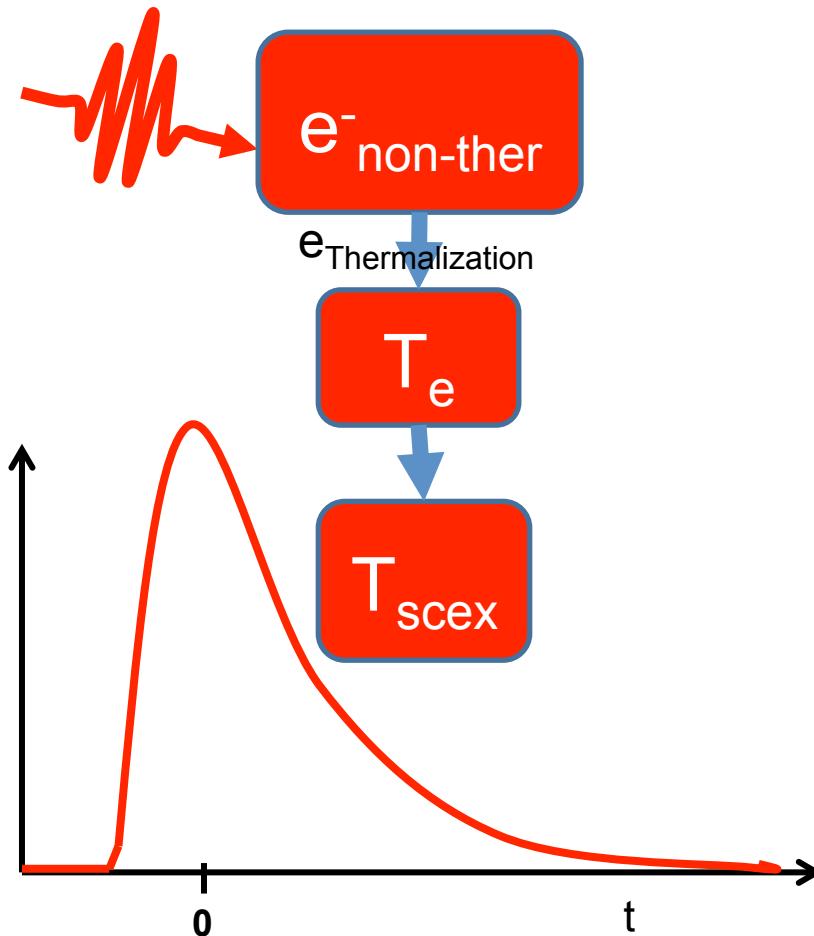
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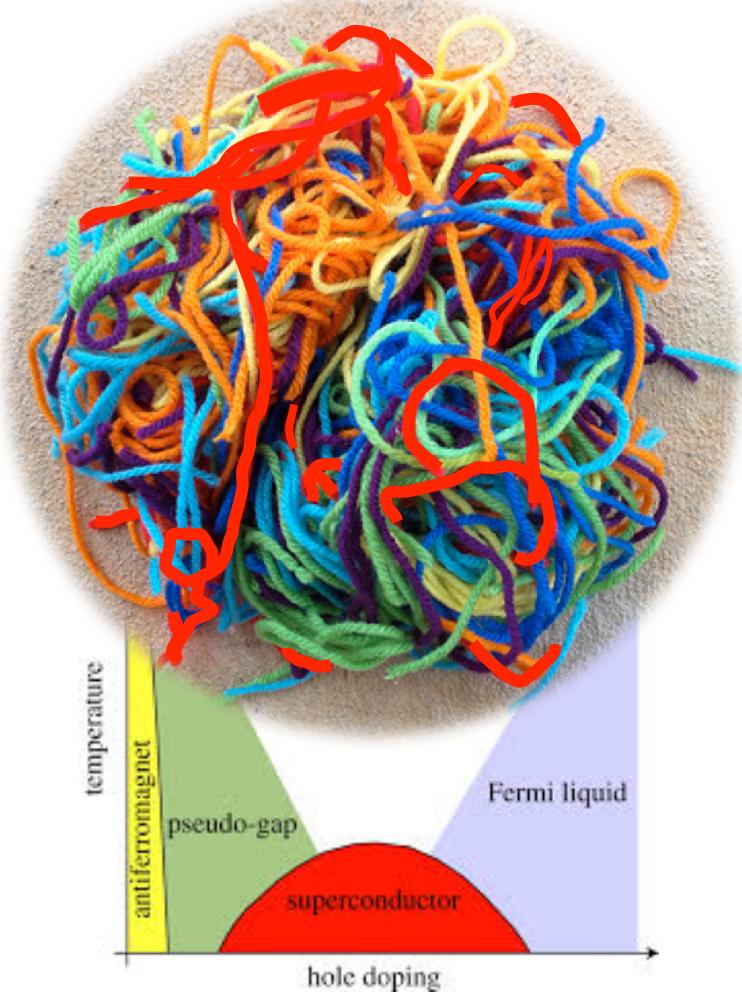
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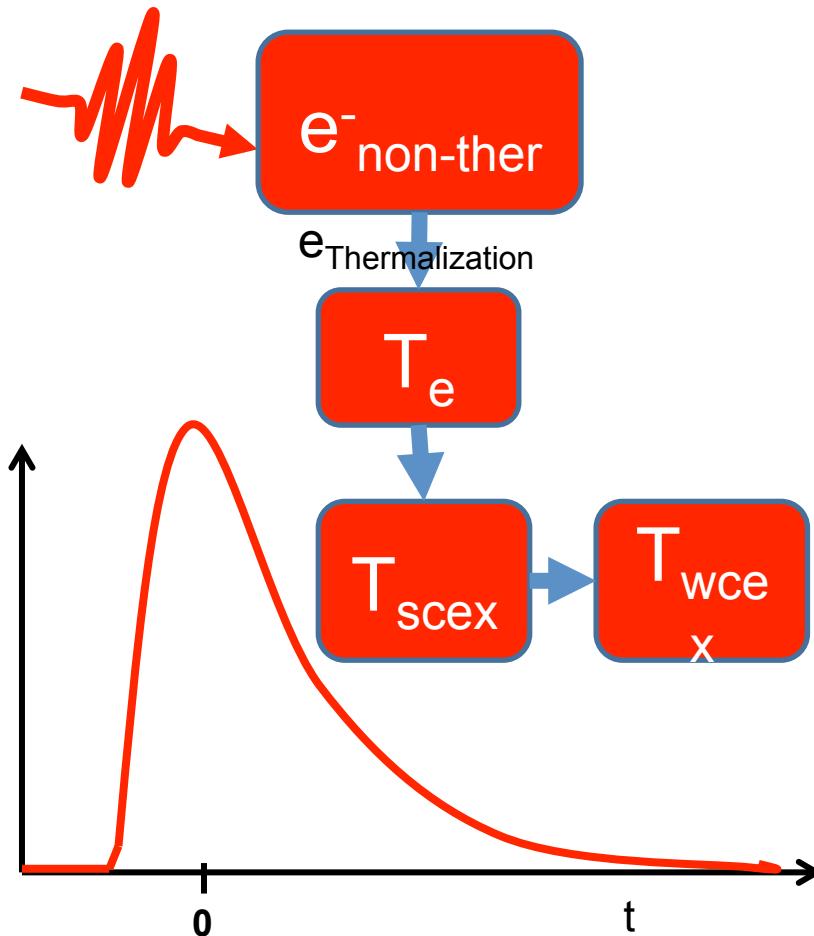
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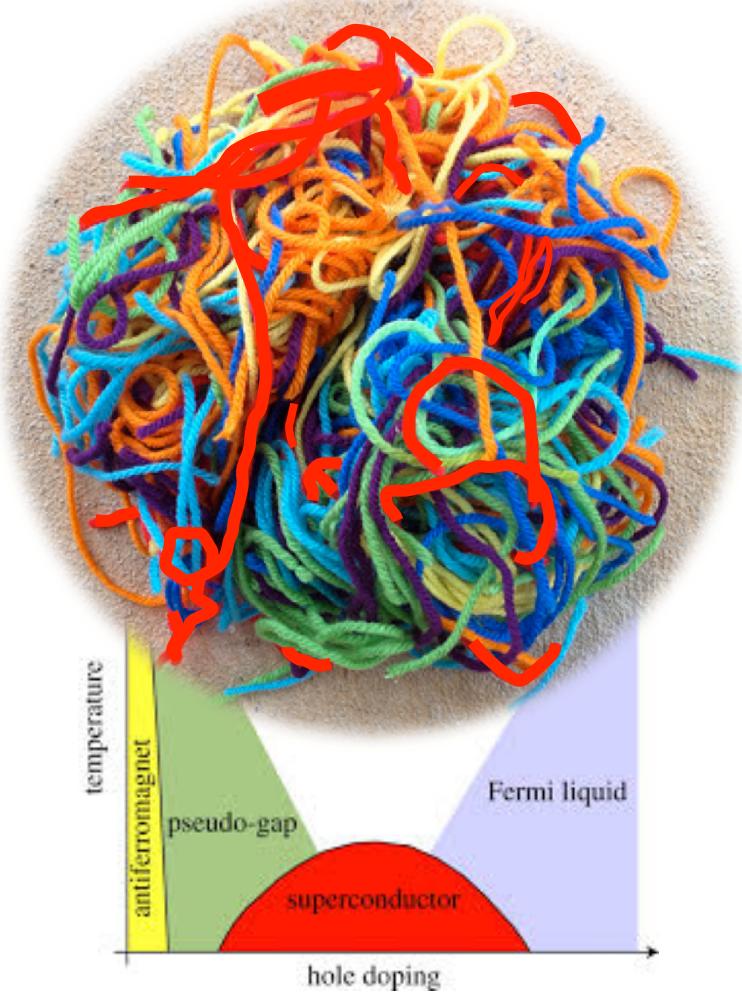
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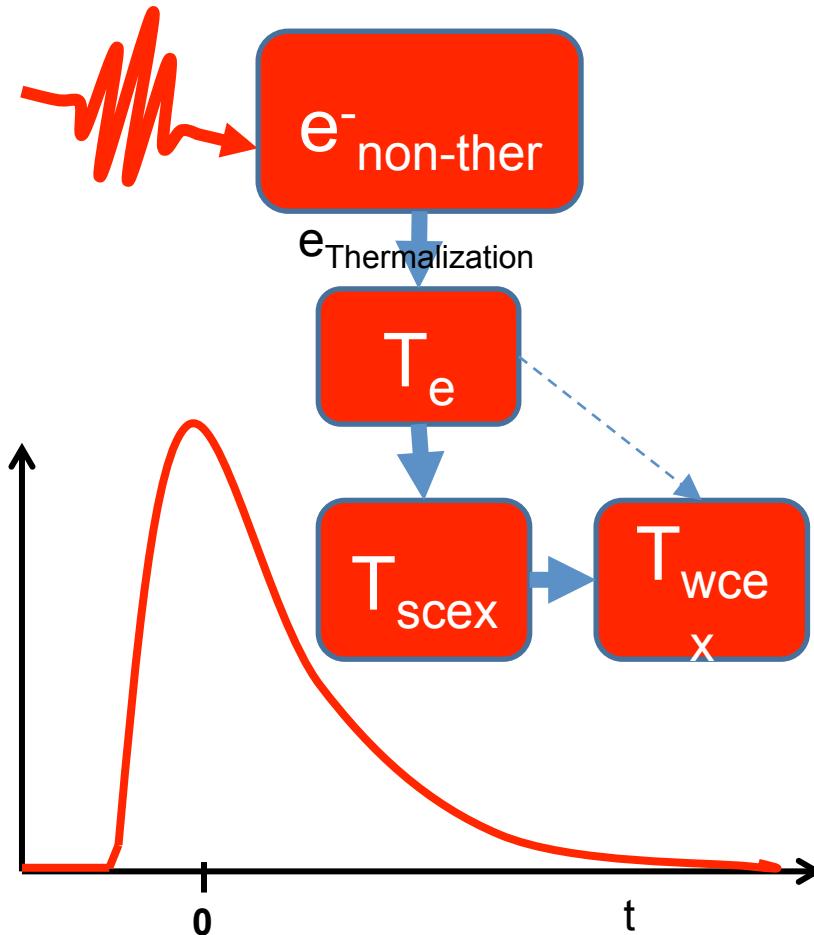
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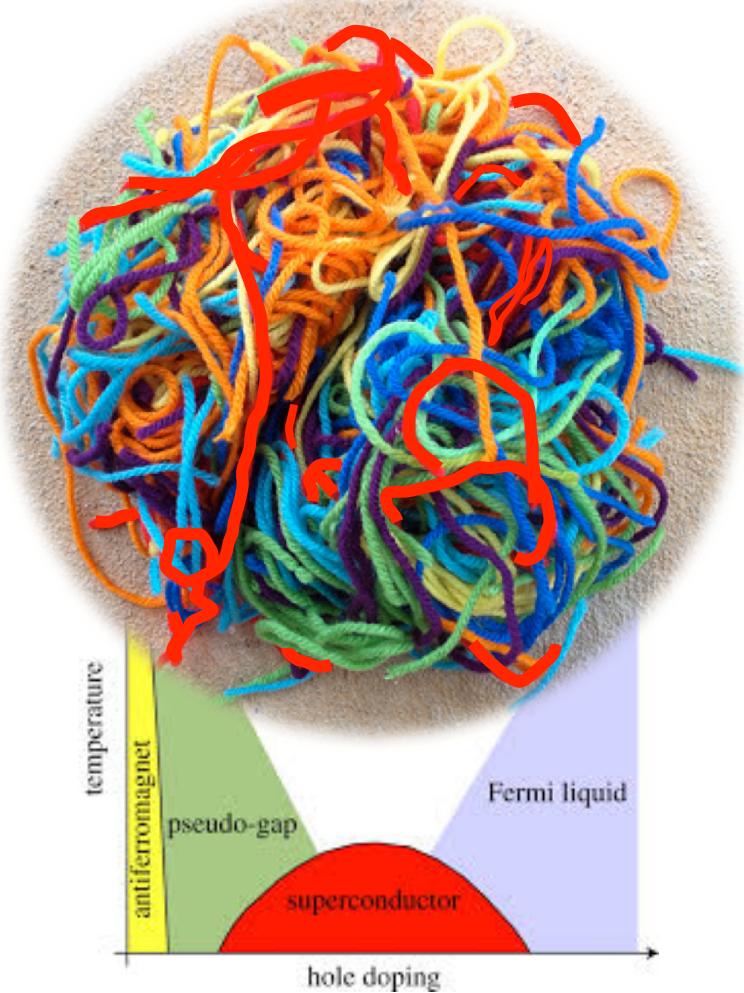
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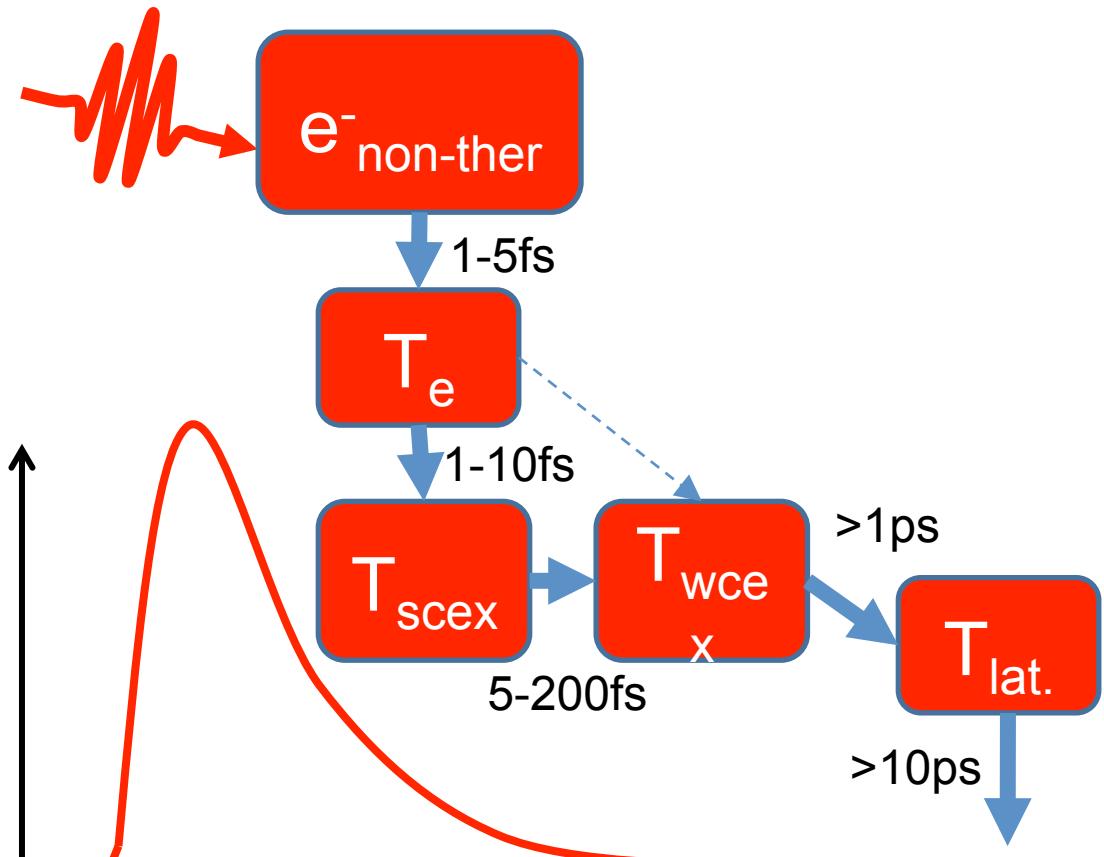
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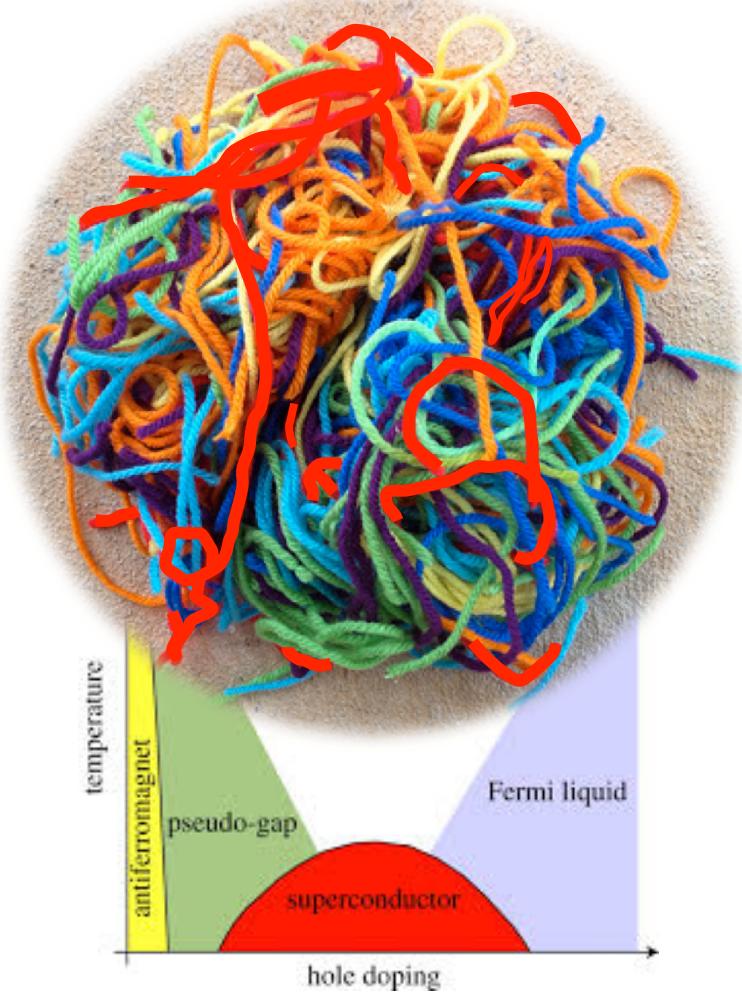
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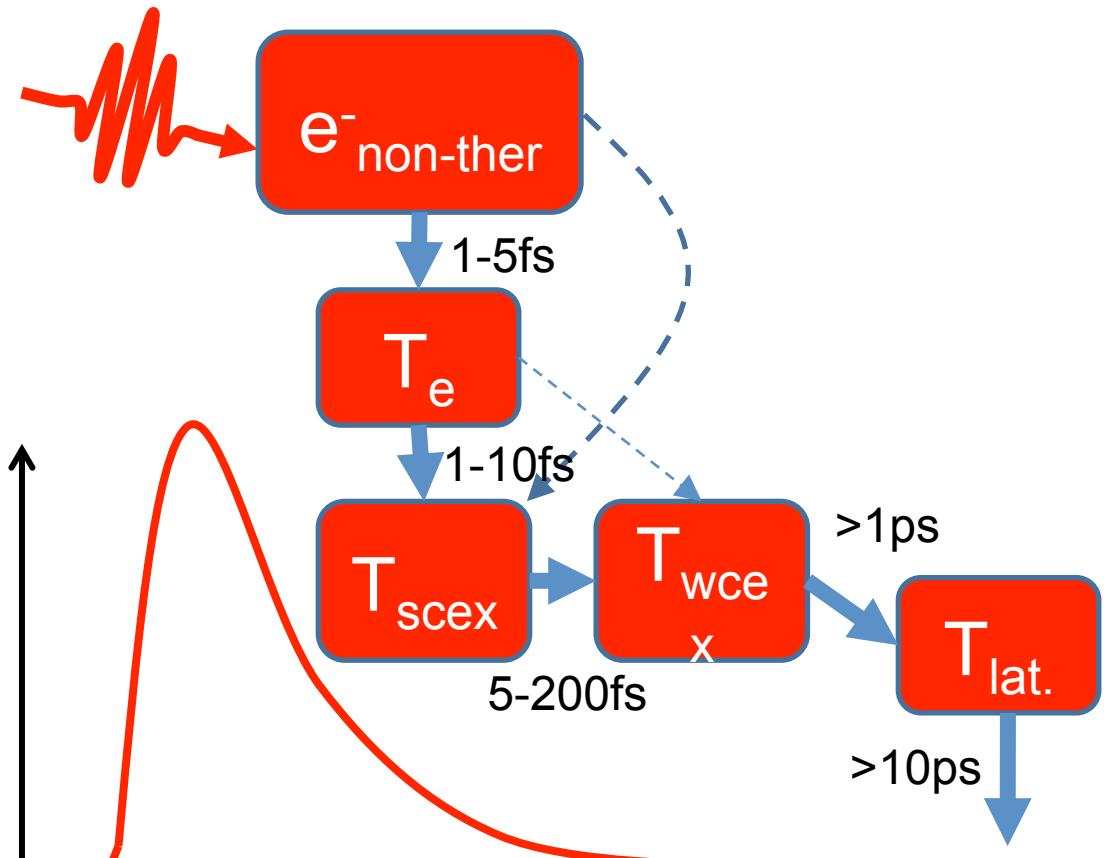
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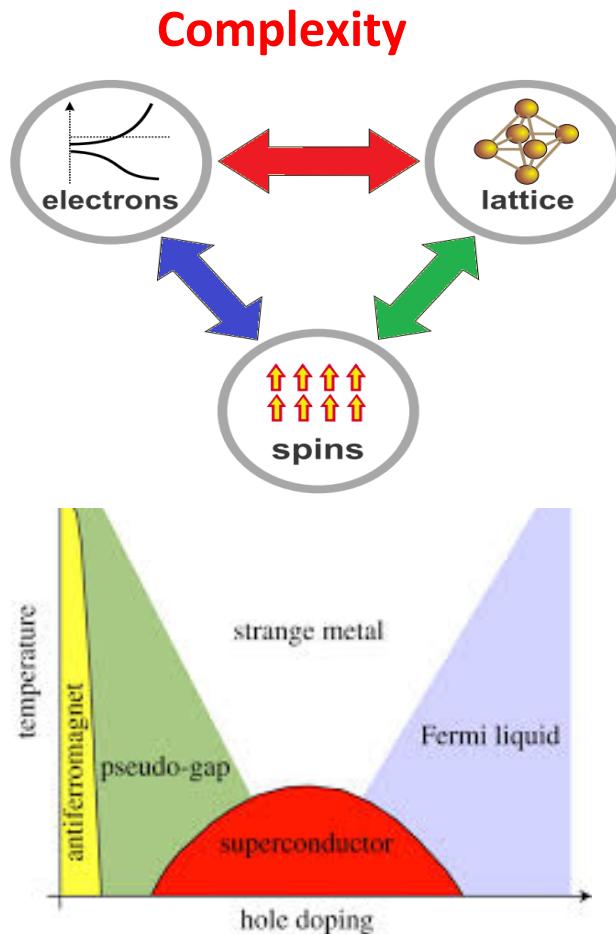
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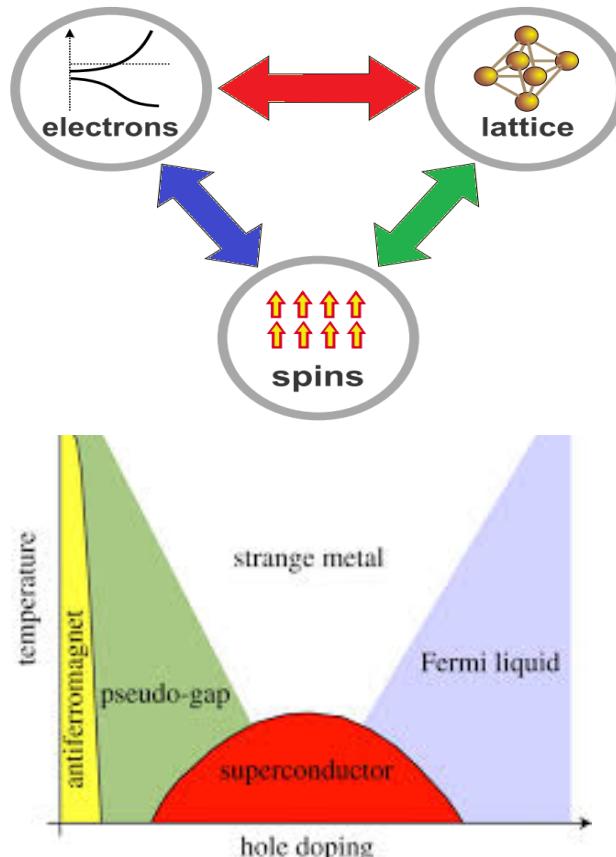
Nat. Comm. 5, 5112, 2014

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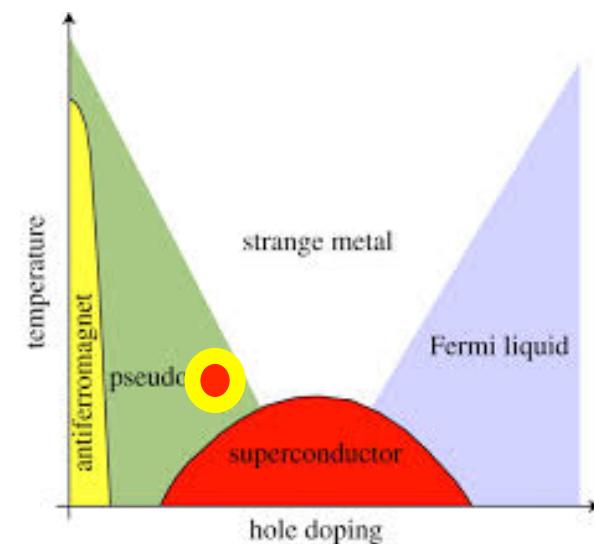
Non-Equilibrium studies
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Non-Equilibrium physics
for optical control

Complexity



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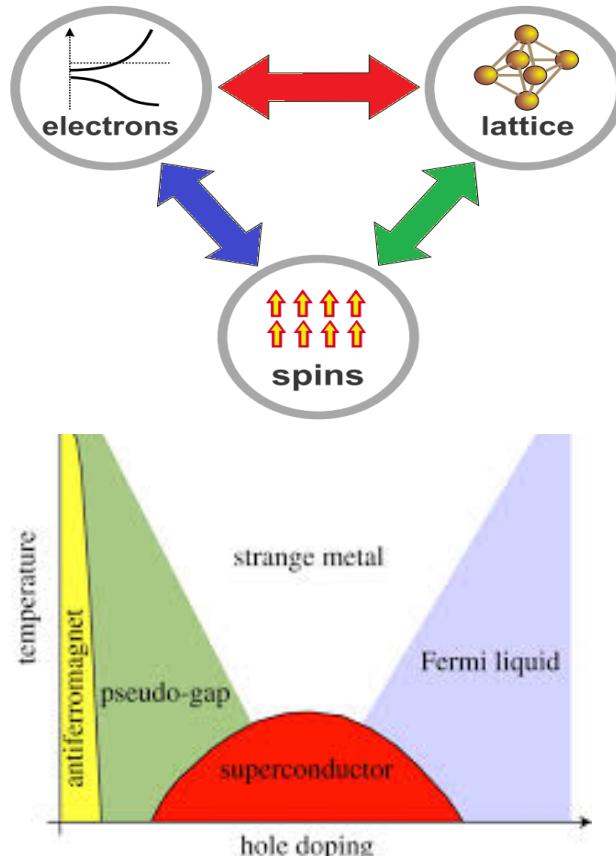


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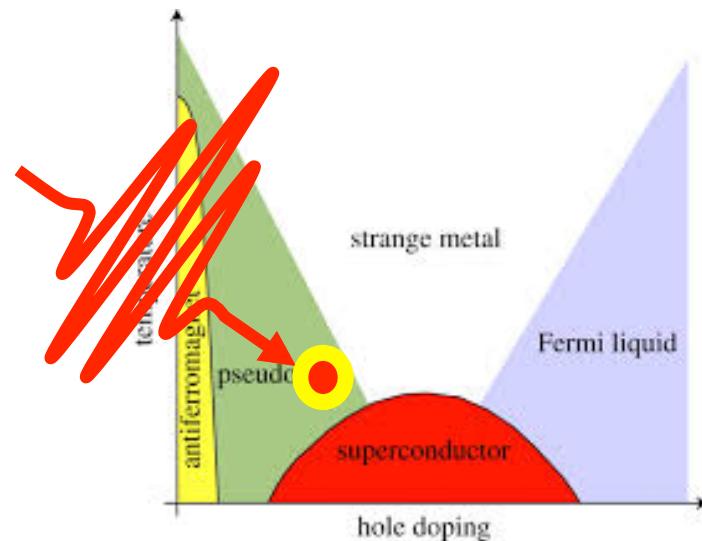
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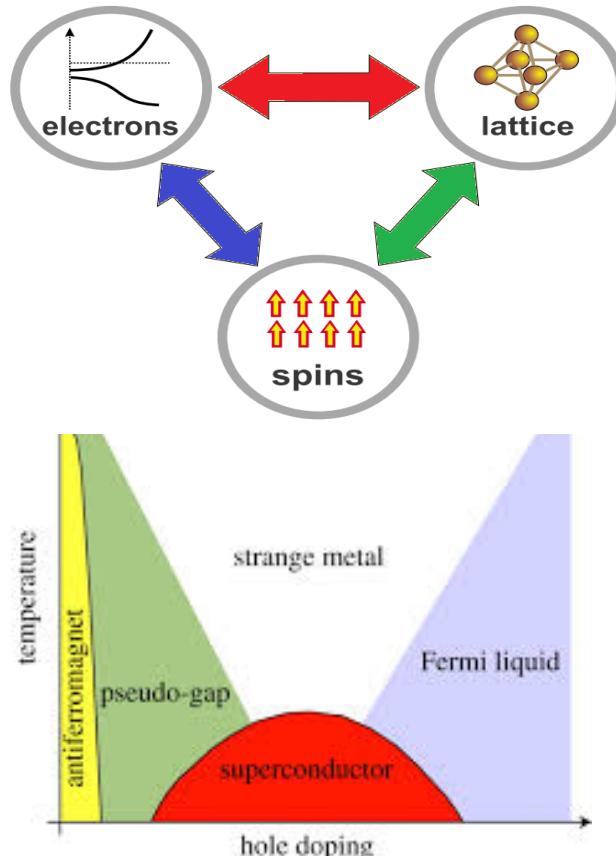


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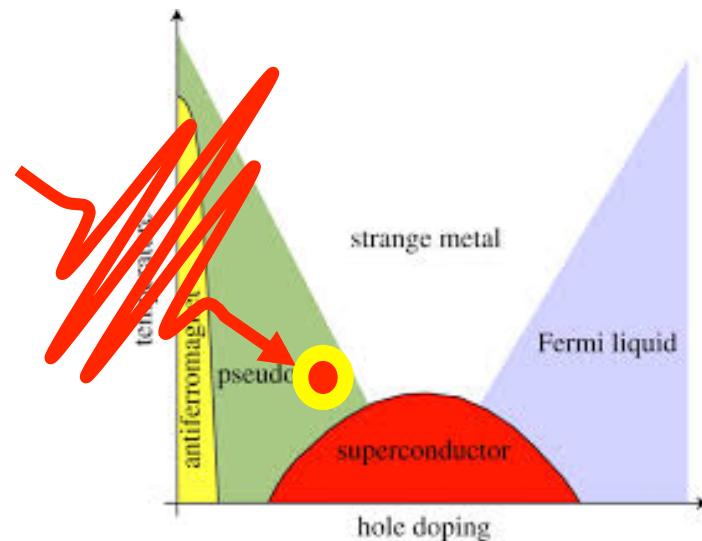
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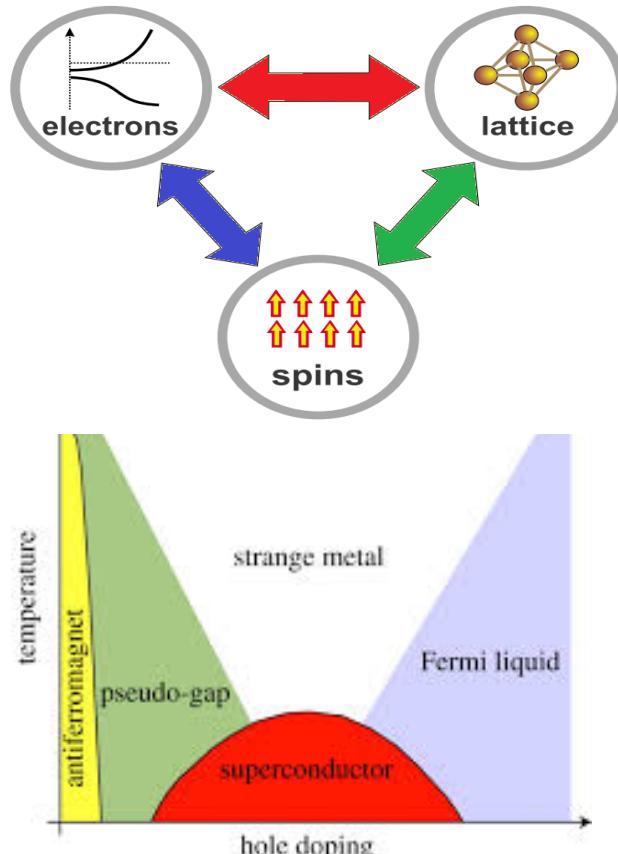


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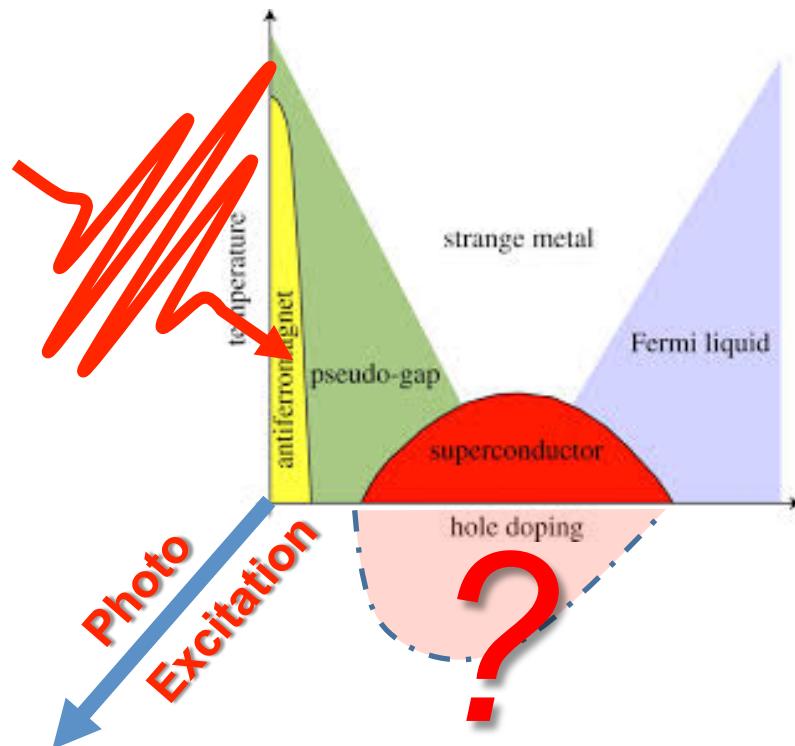
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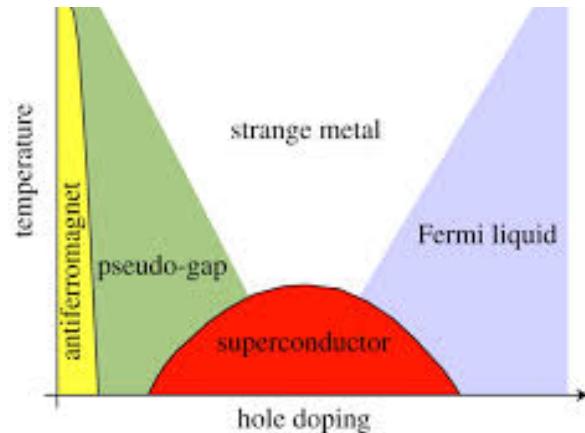
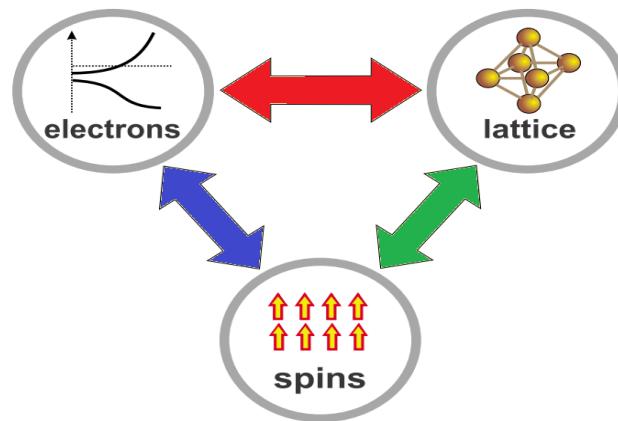
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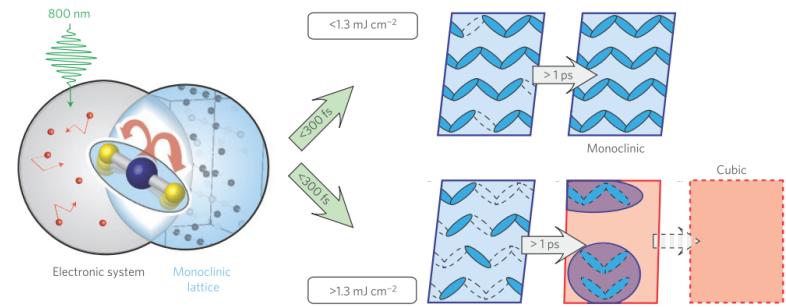
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Non-Equilibrium physics for optical control

Insulator-to-metal transition

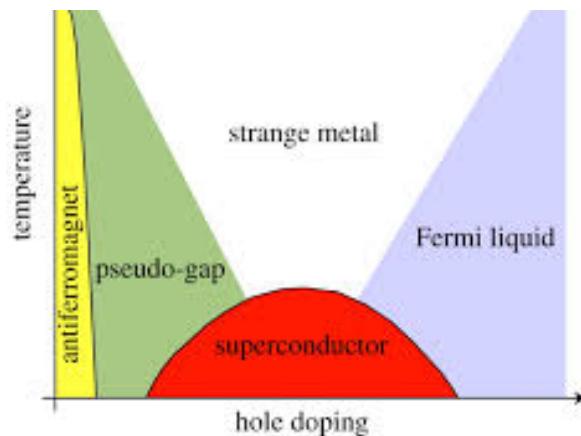
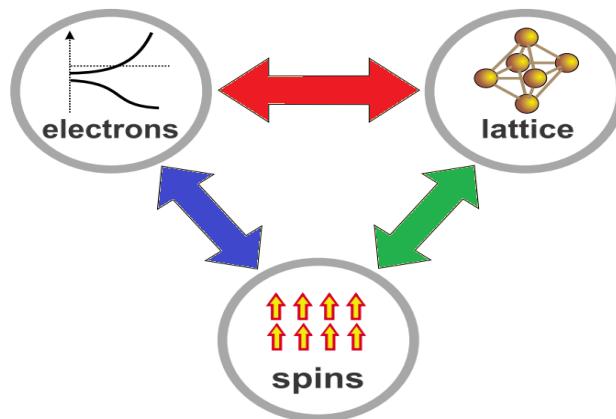


Nature Material 12, 882 (2013)
Phys. Rev. B 93, 054305 (2016)

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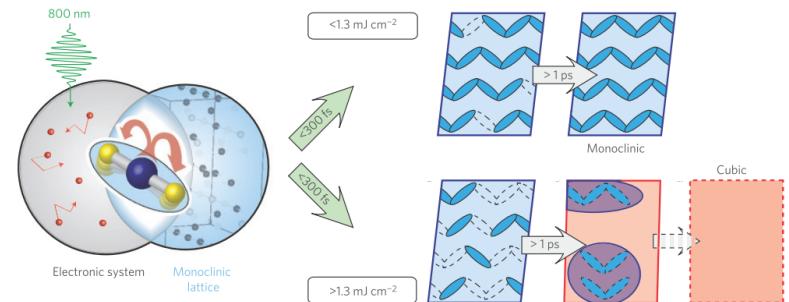
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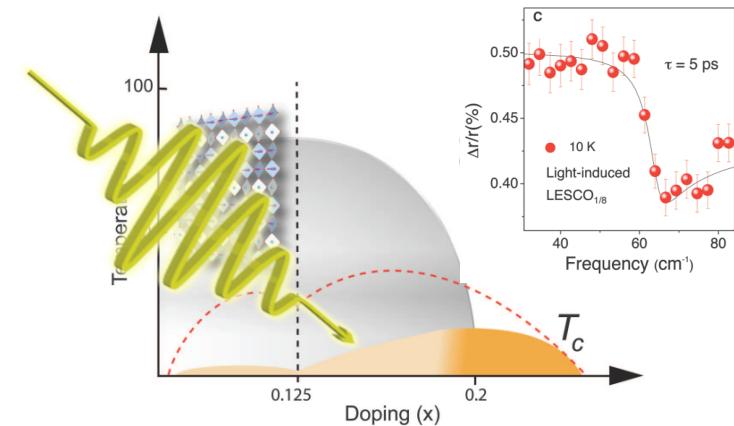
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Insulator-to-metal transition



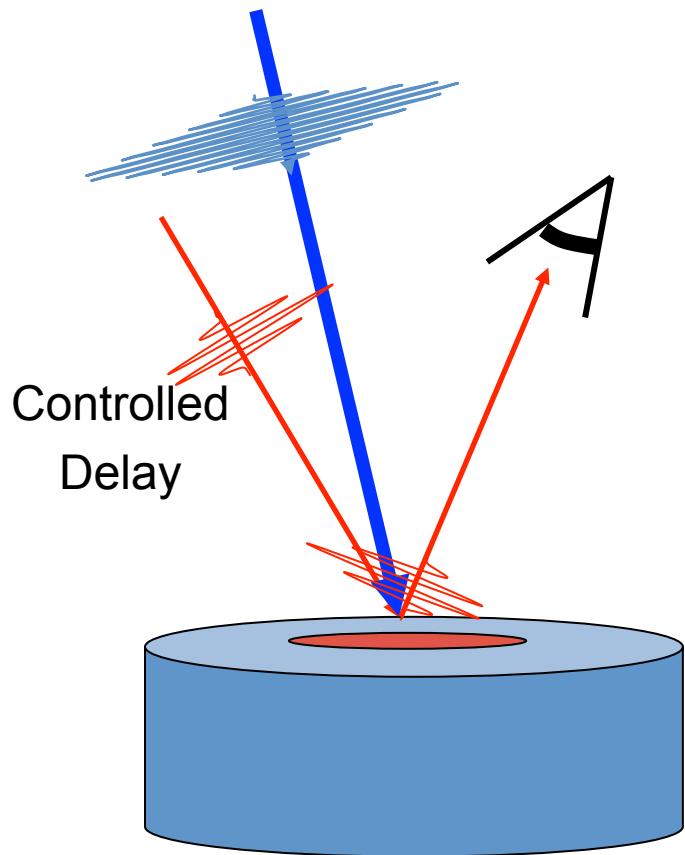
Nature Material 12, 882 (2013)
Phys. Rev. B 93, 054305 (2016)

Photo-induced Superconductivity



Science 331, 189 (2011)

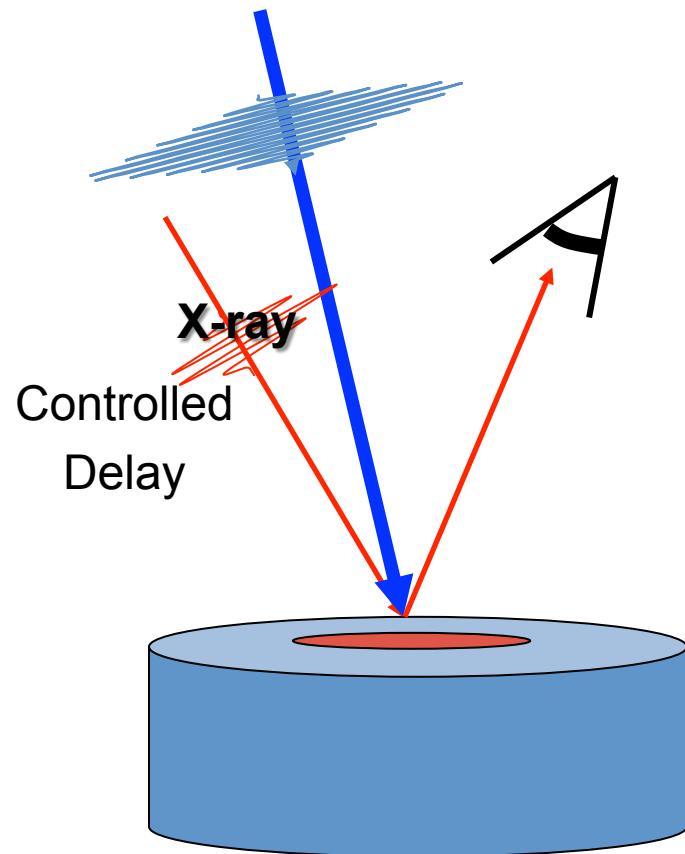
The prime tool for non-equilibrium physics: Pump and probe



-«All Optical» pump and probe spectroscopy

-Reflectivity, Transmission, Kerr, Moke...

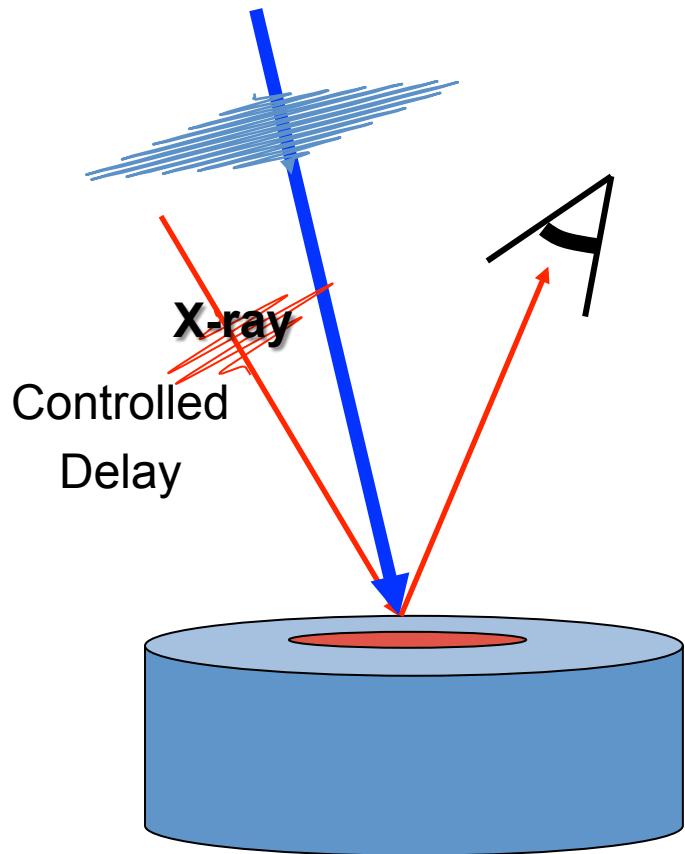
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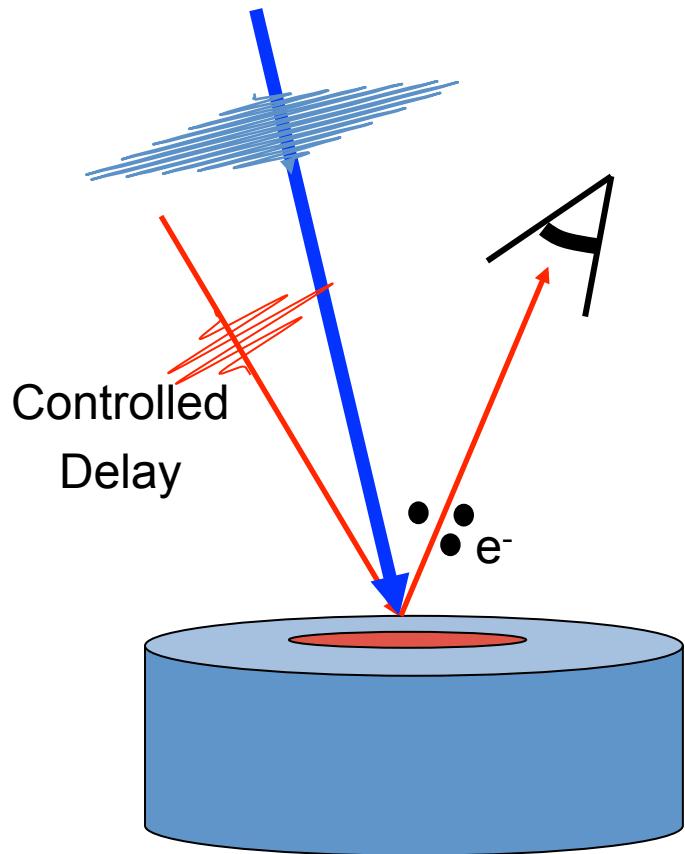


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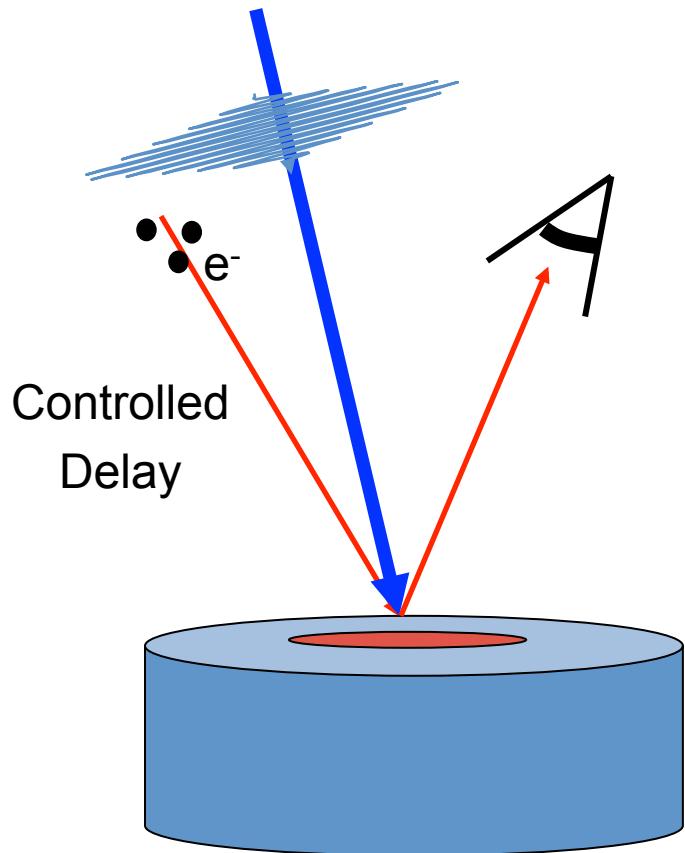
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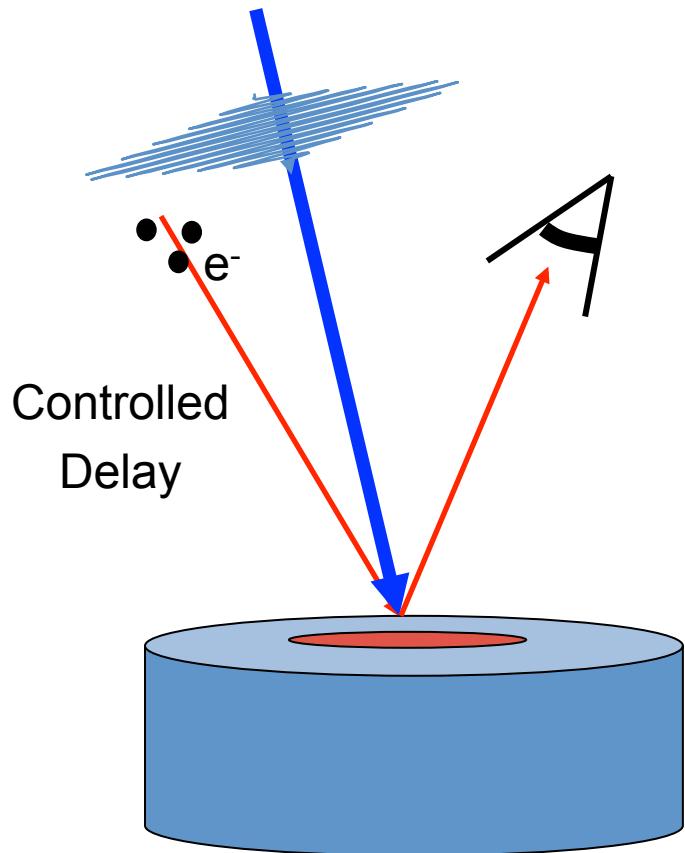
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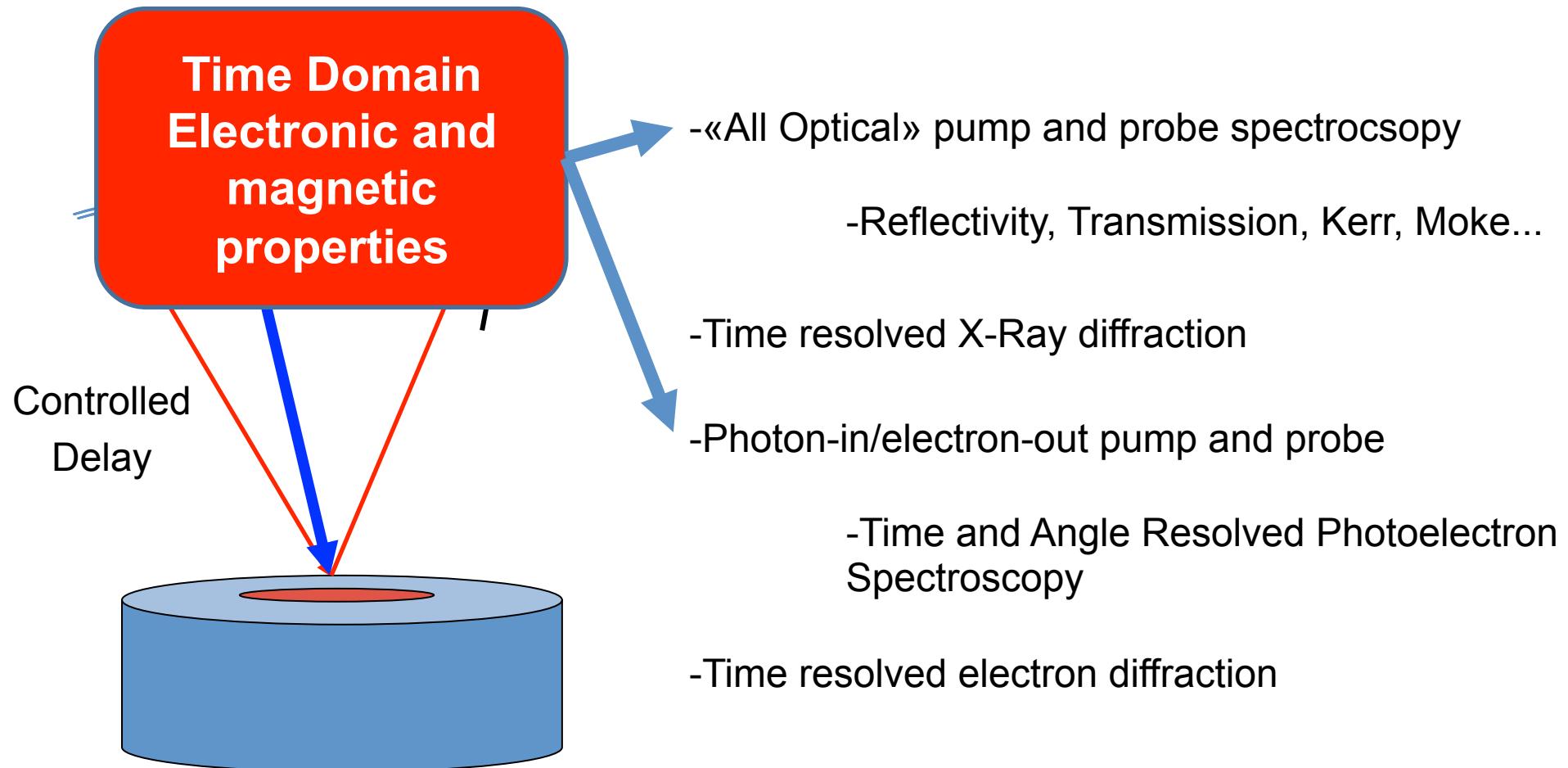
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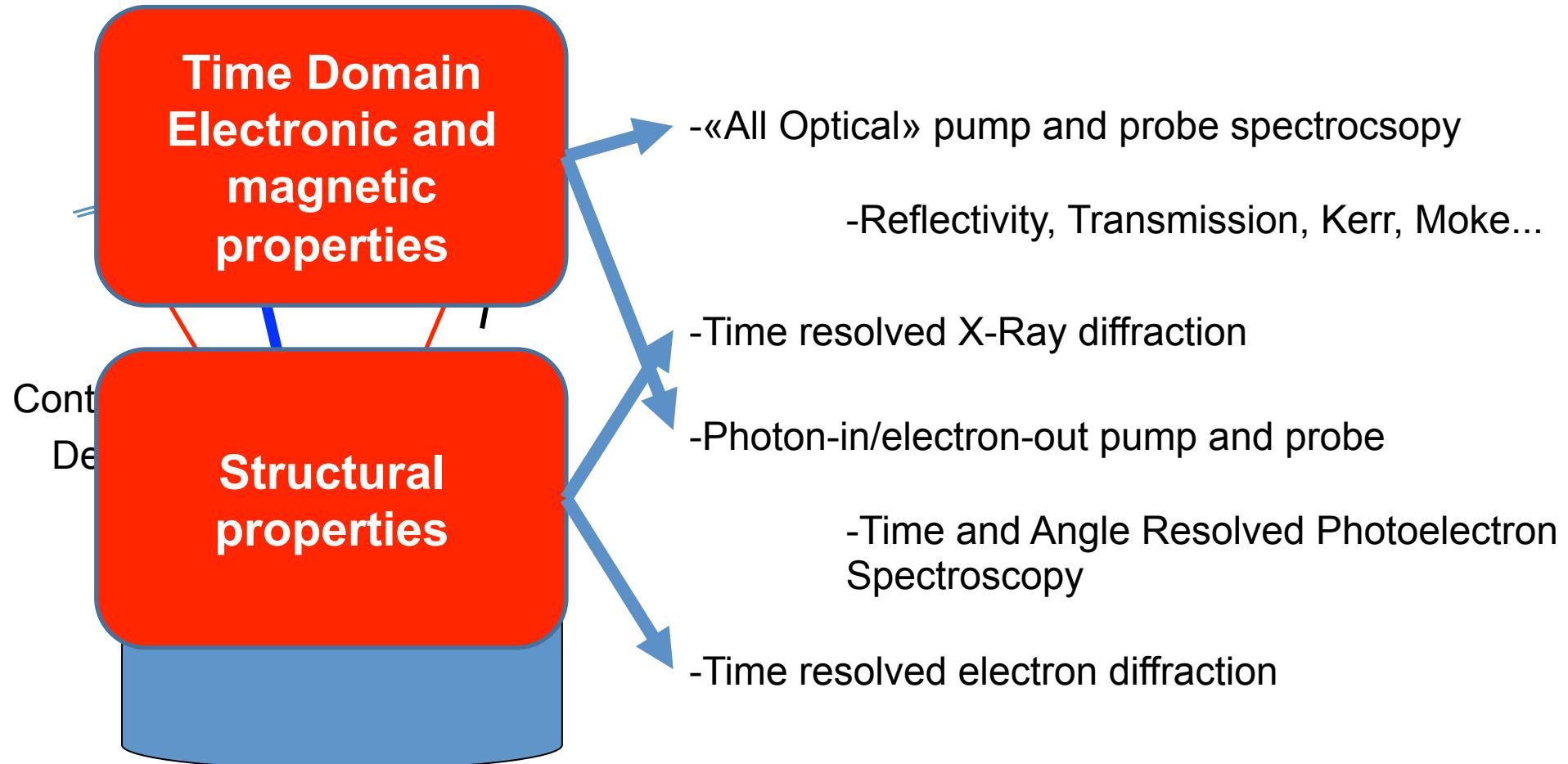


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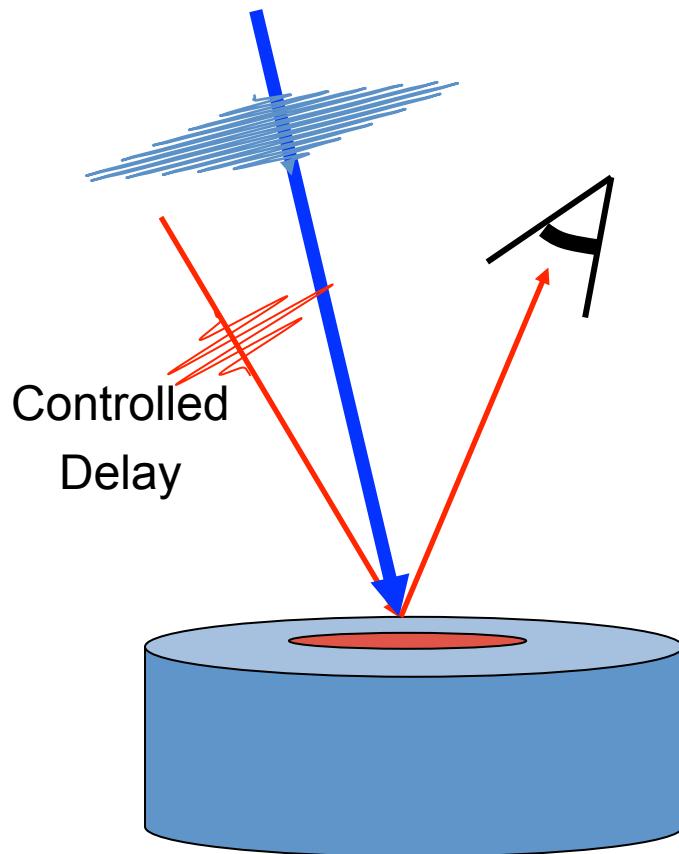
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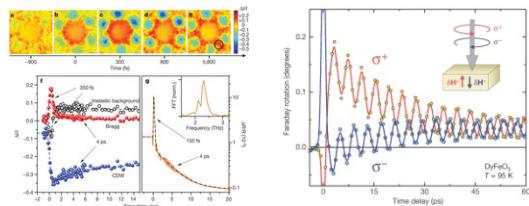
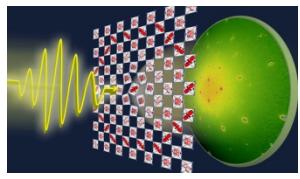


-What do i measure?

Non-equilibrium “Parameters Space”

-Pump & Probe

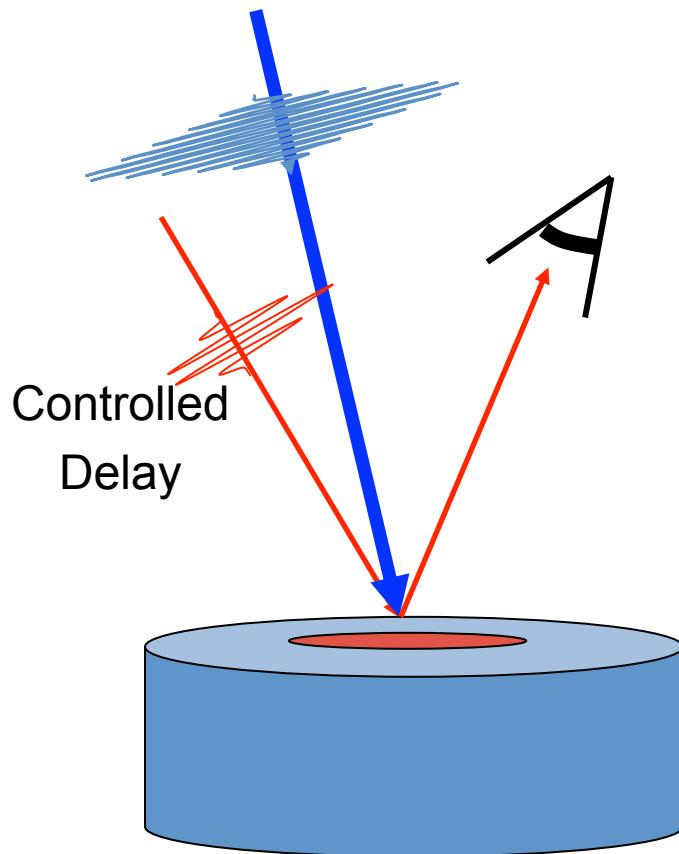
Time resolved spectroscopies



Phys. Rev. Lett., 106, 217401 (2011), Nature 468, 799–802, Nature 435, 655 (2005)

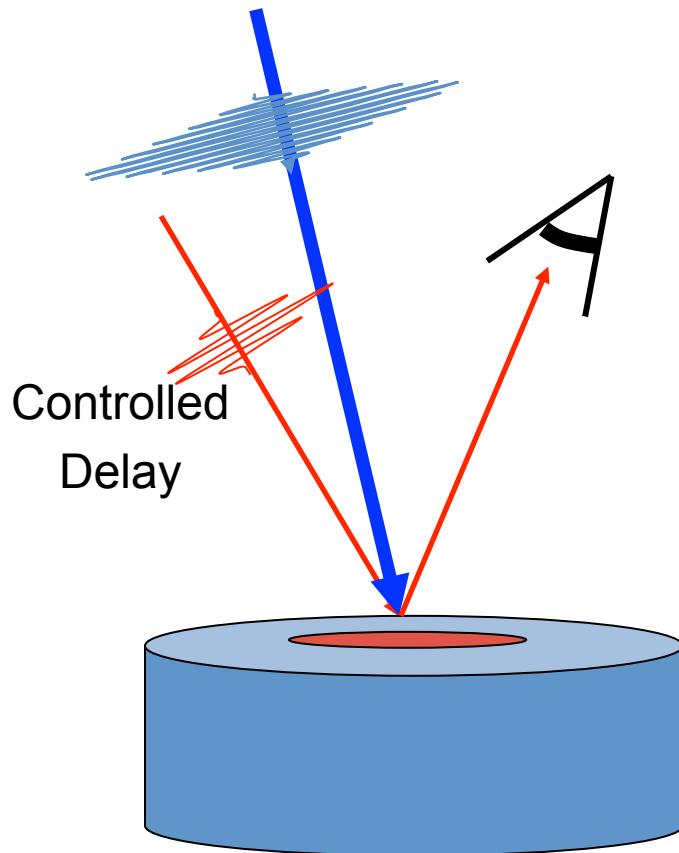
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- ✓ TR Kerr, moke and X-MCD, Arpes
- ✓ TR Spectroscopy, TRRaman

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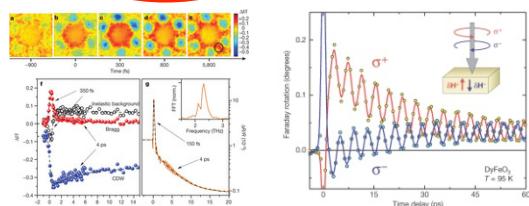
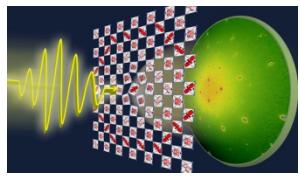
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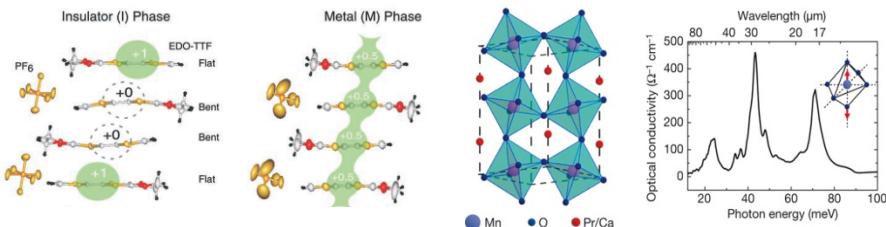


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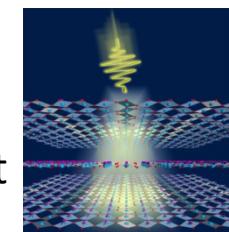
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Optical Control of Material



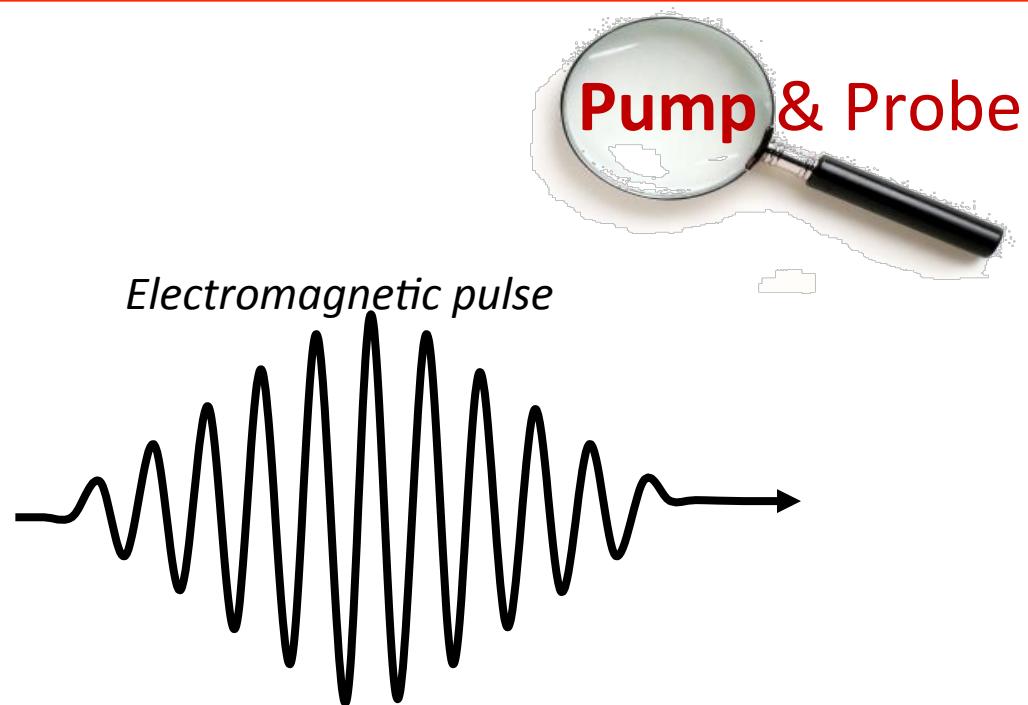
Science 307, 2005, Nature 449(2007)

- ✓ Photo-Induced phase transitions
- ✓ Coherent control (IR and THz)
- ✓ Light control of quantum coherent phases

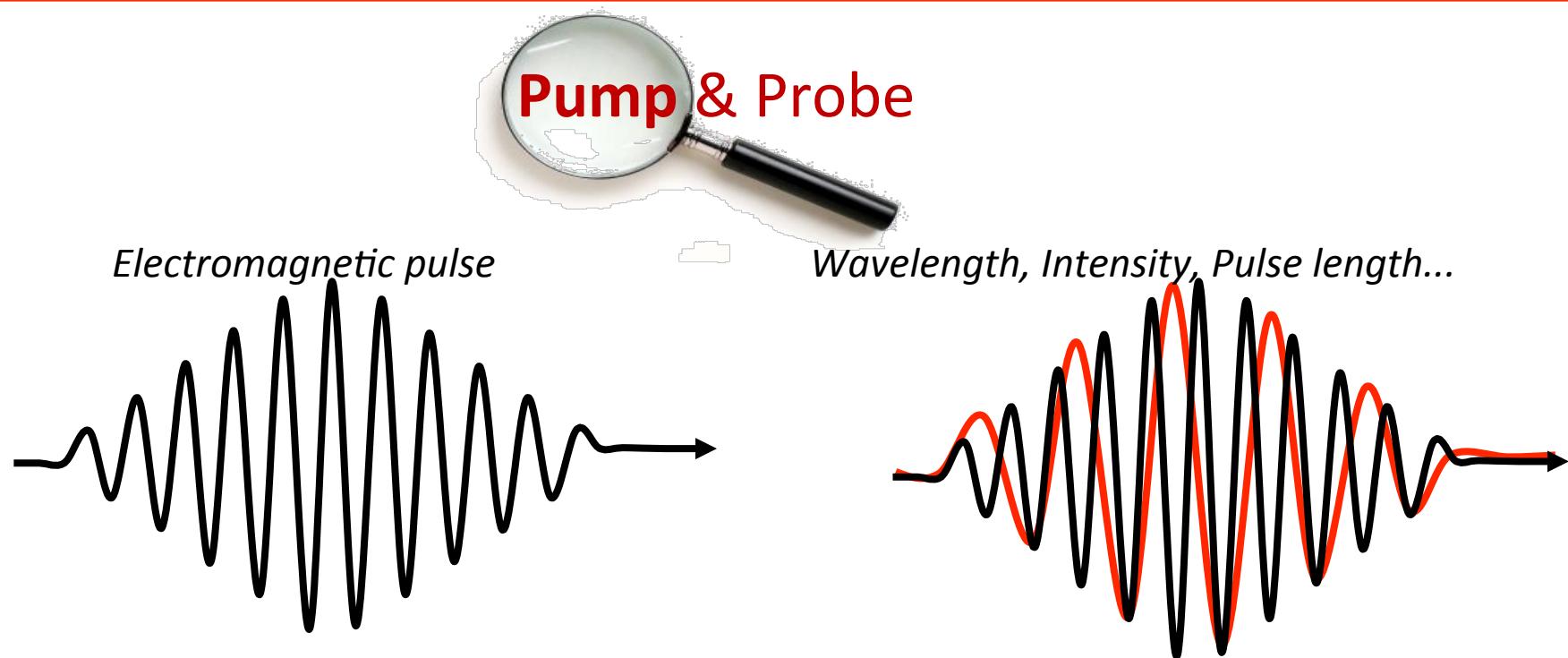


Science 331, 2011

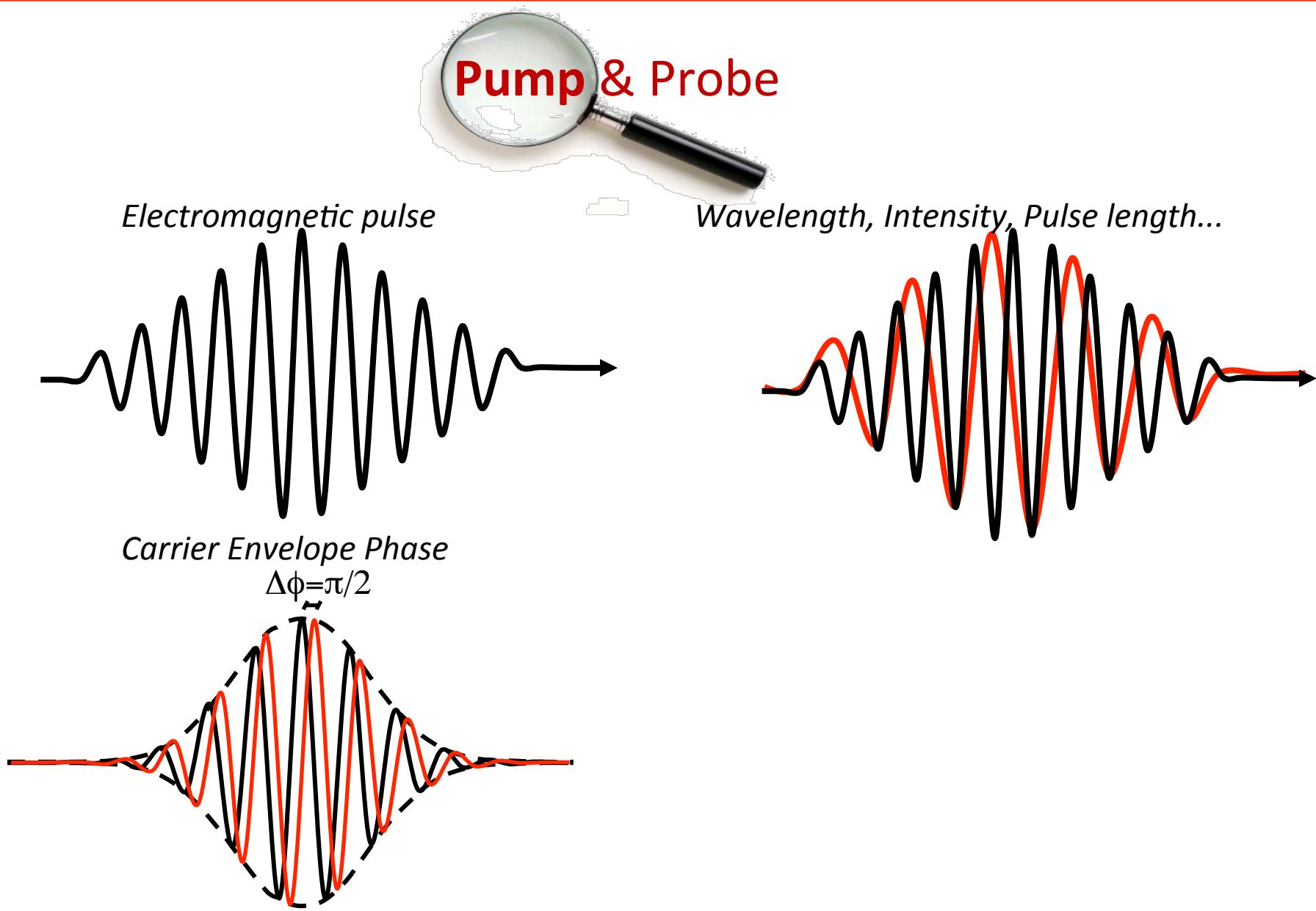
Non-equilibrium “Parameters Space”



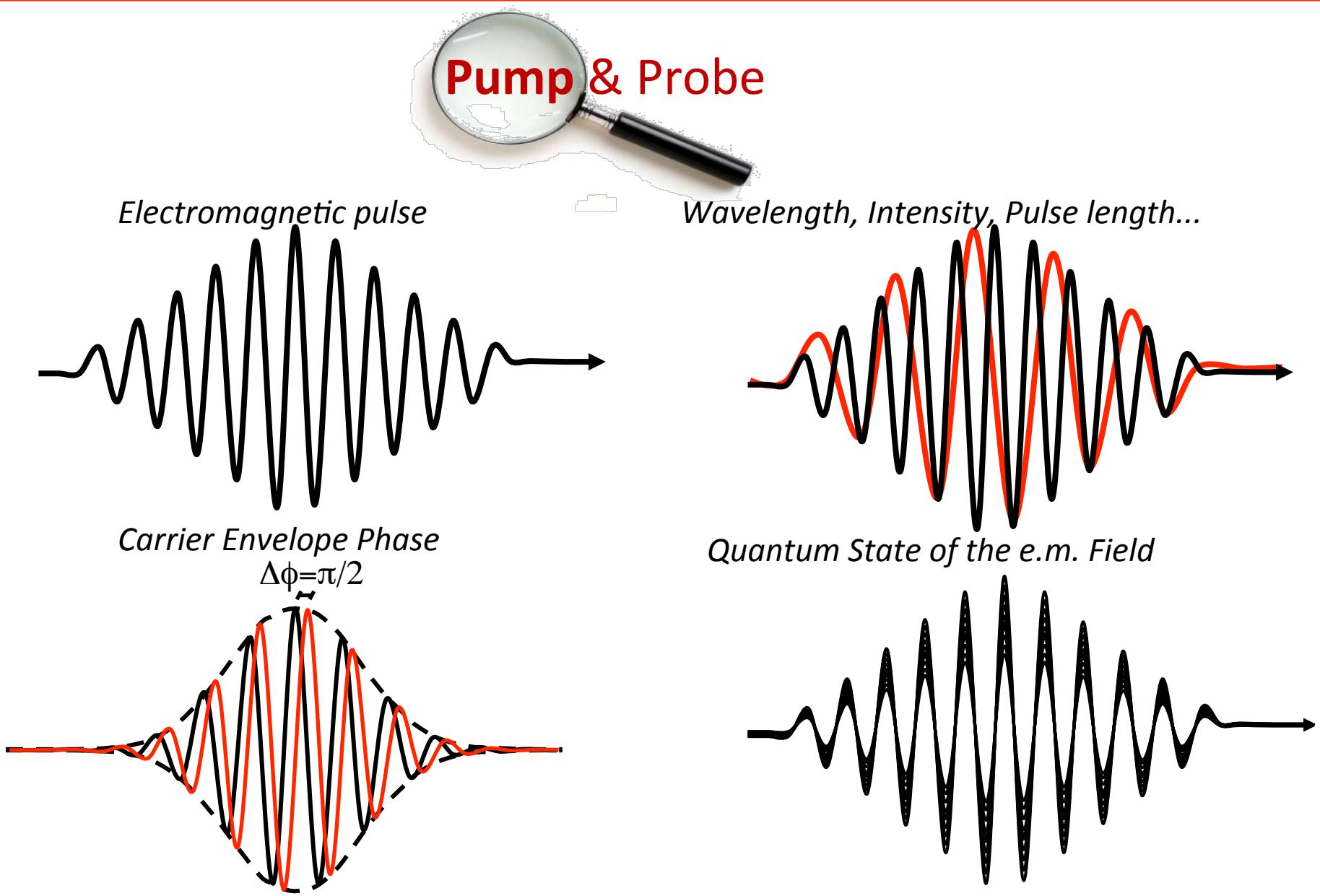
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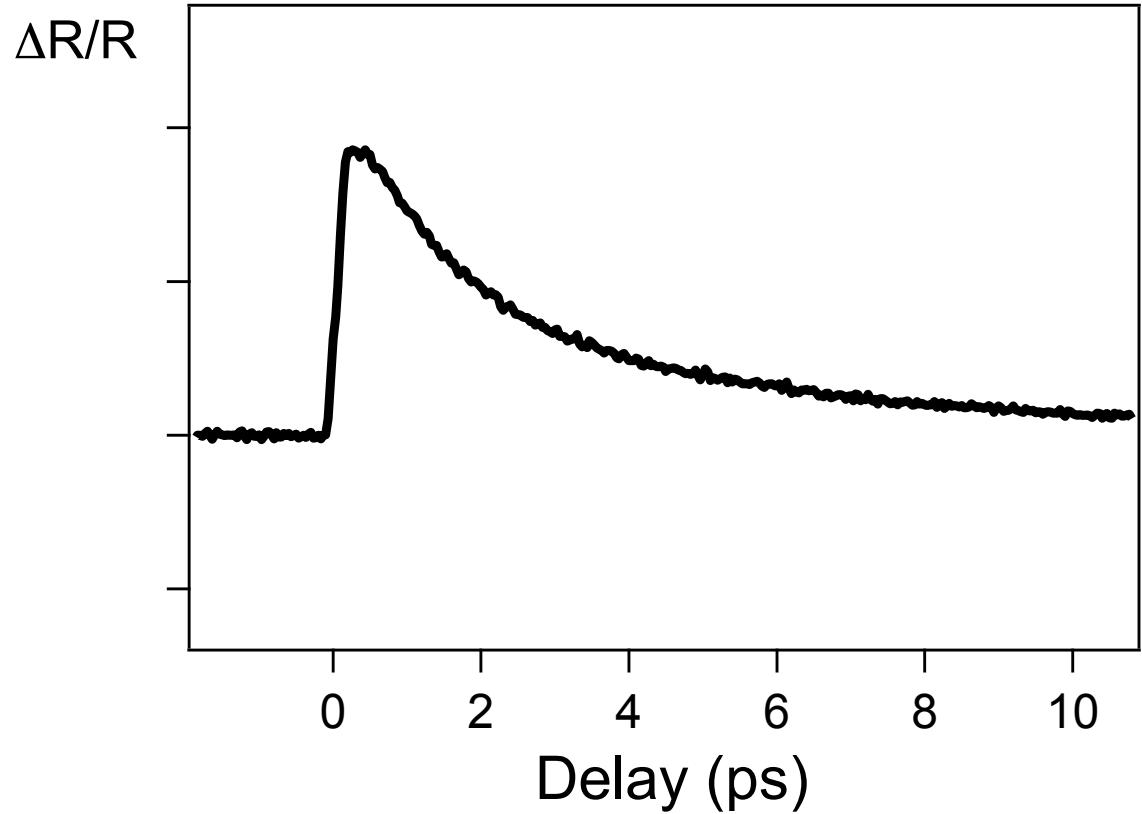
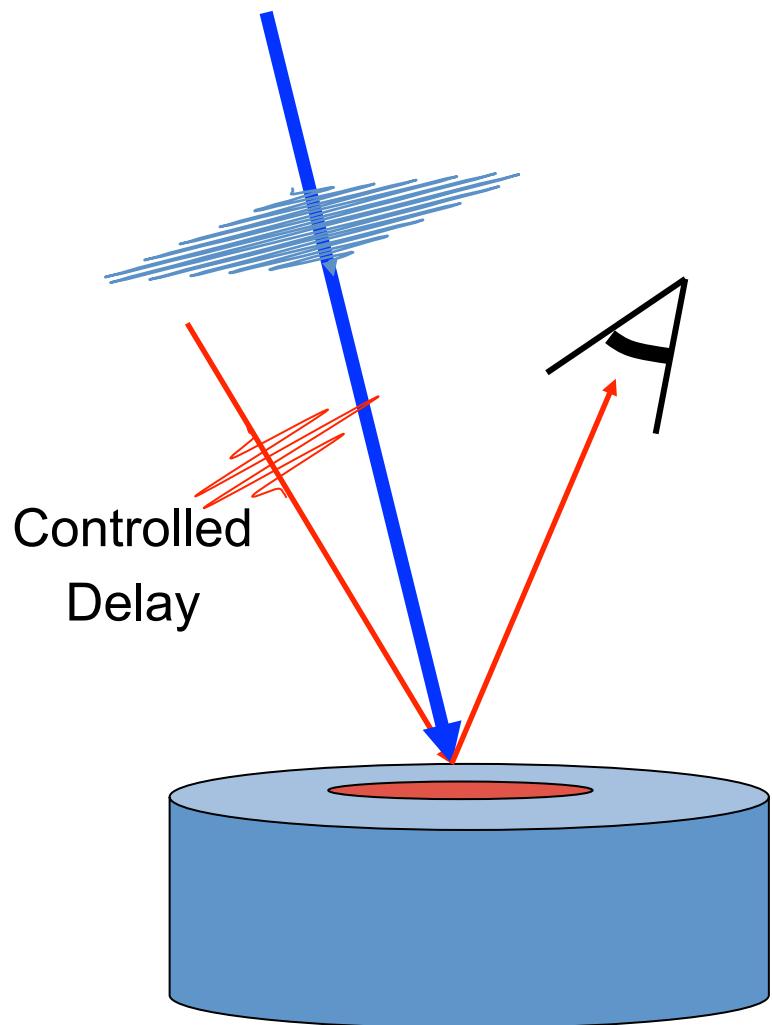
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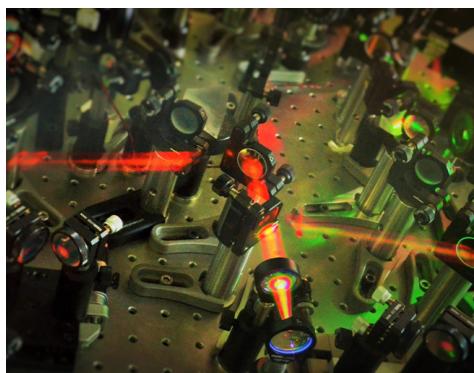
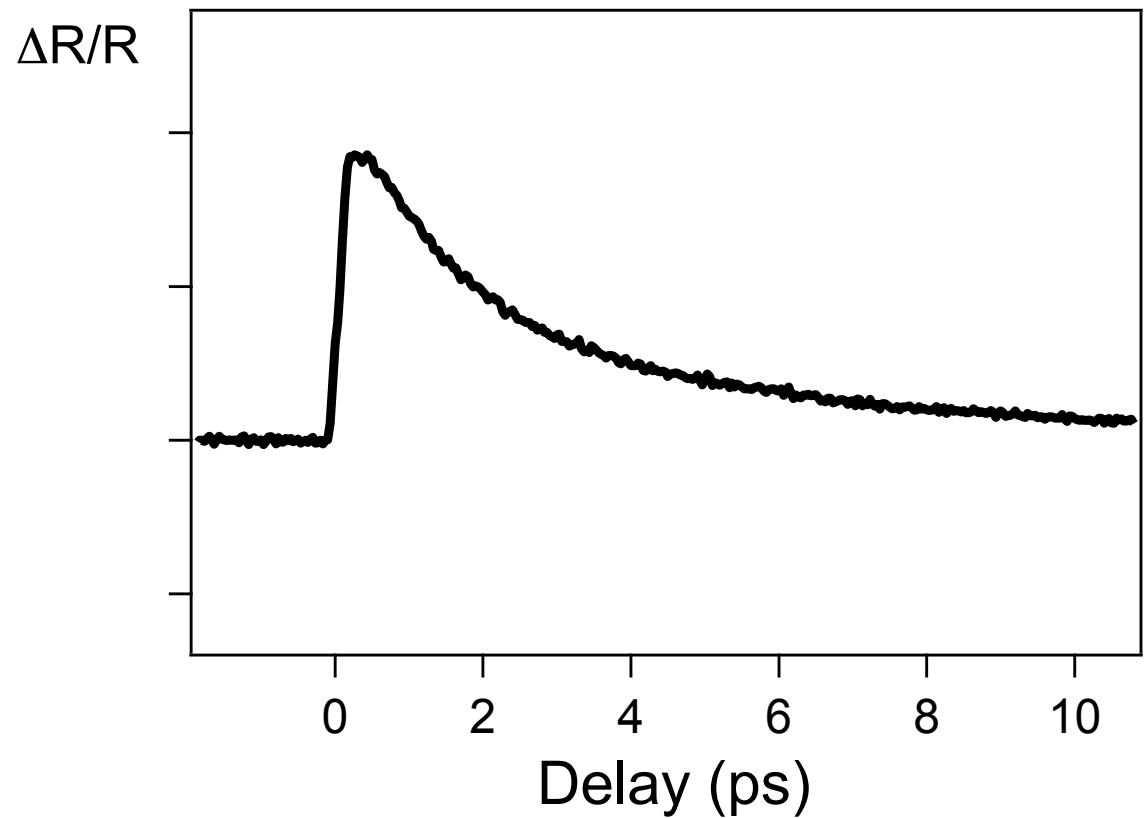
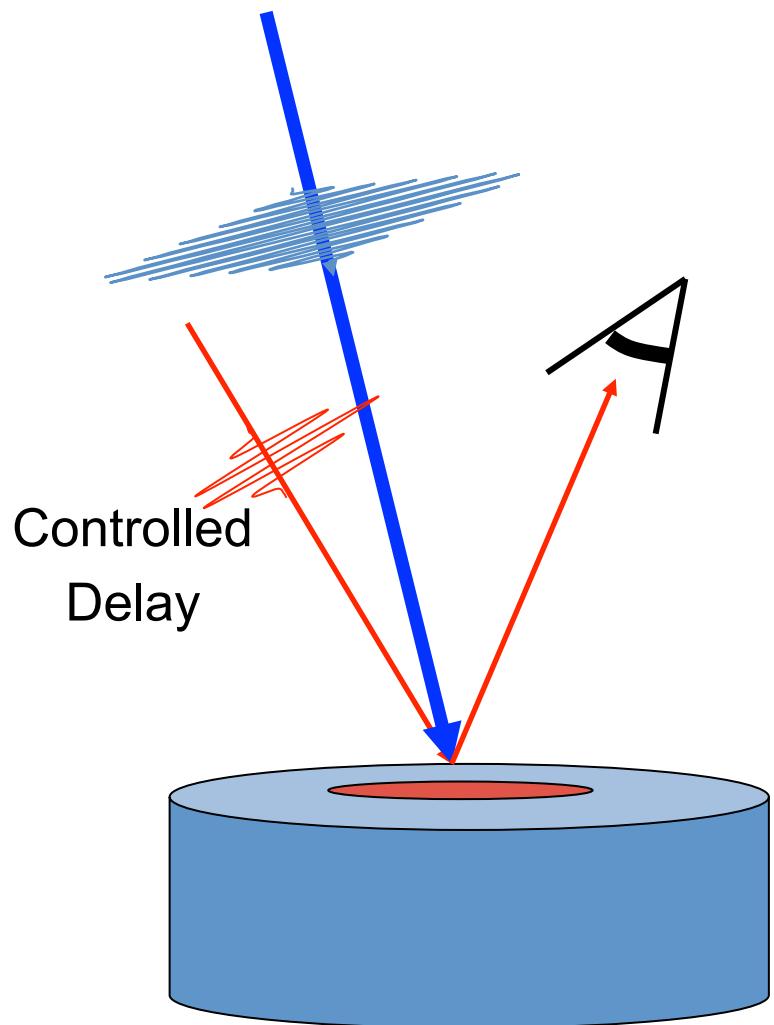
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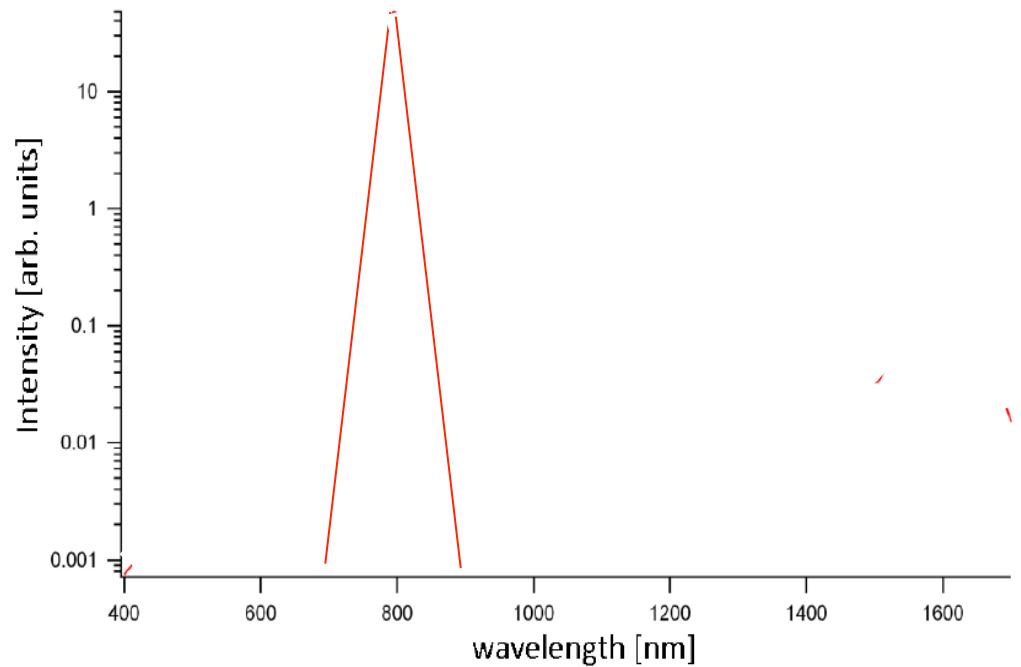
From P&P measurements to P&P spectroscopy



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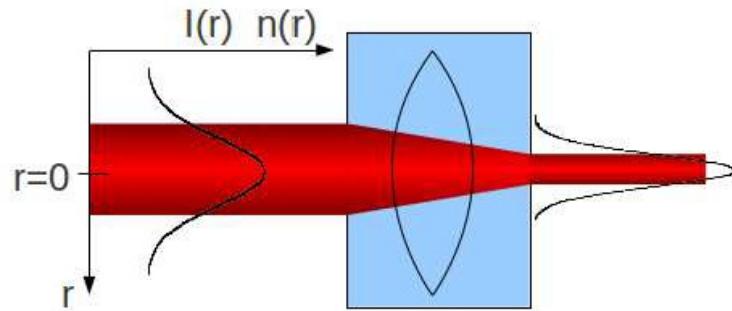


Non-Linear Optical Processes: White light generation

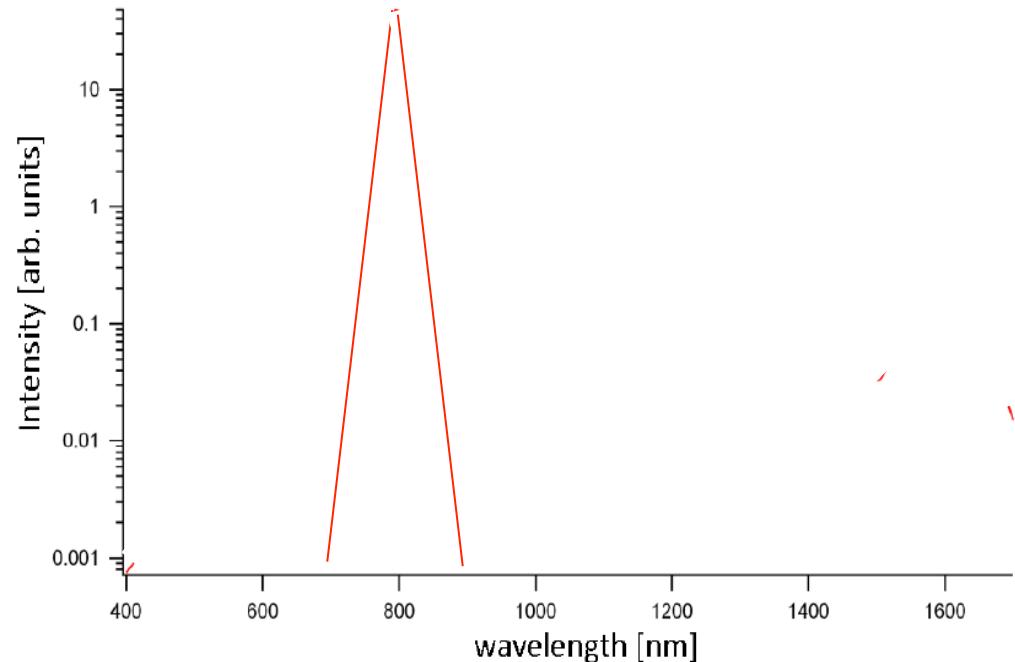
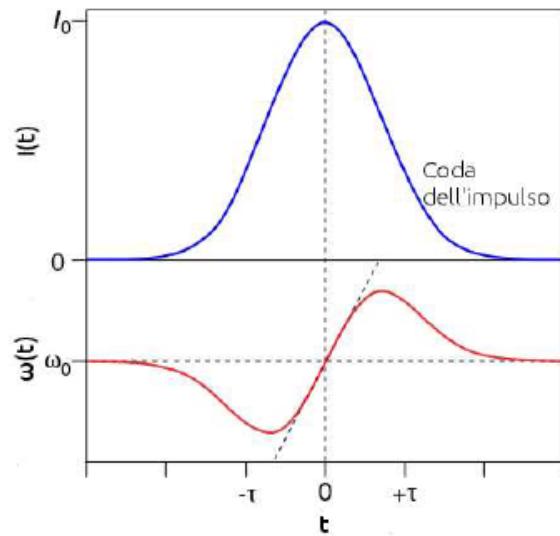


Non-Linear Optical Processes: White light generation

Self Focusing

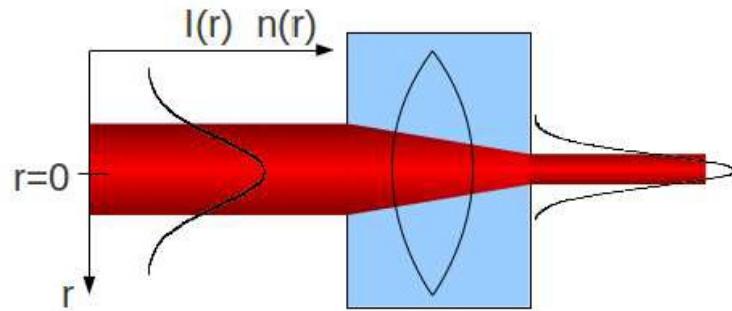


Self Phase modulation

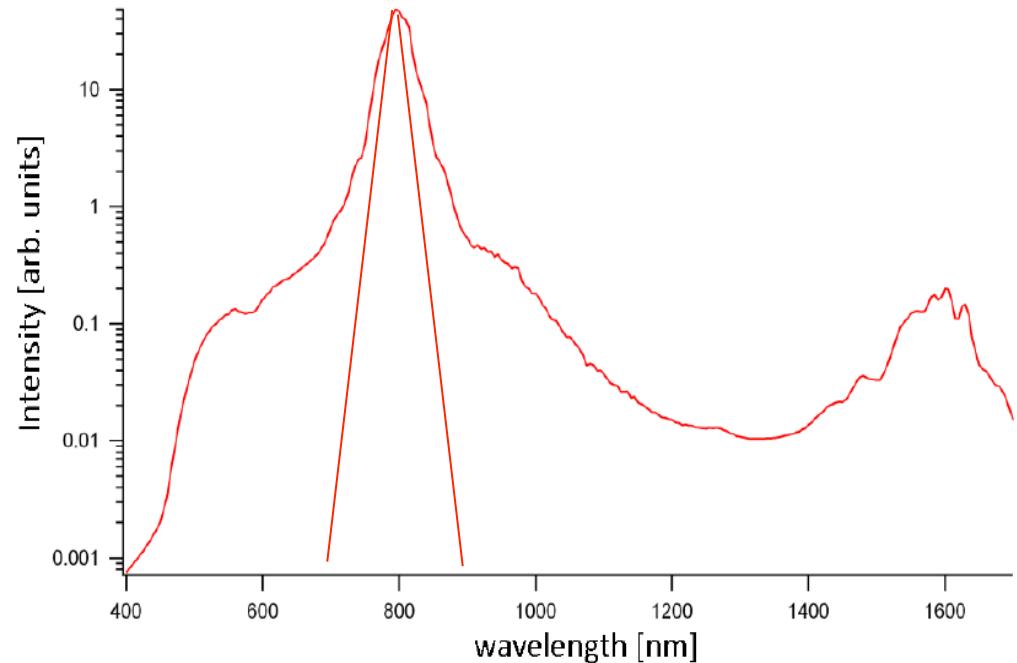
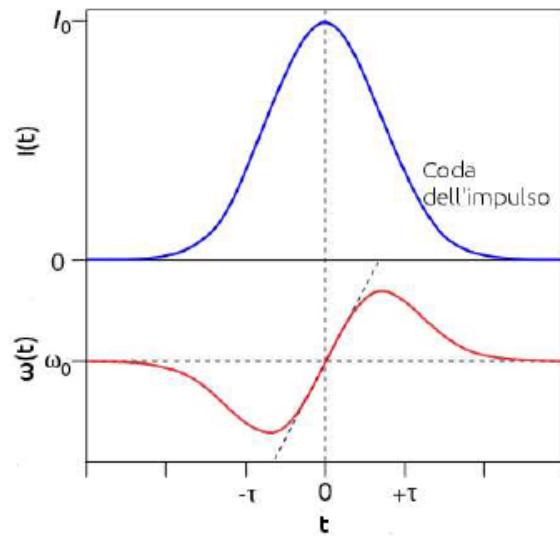


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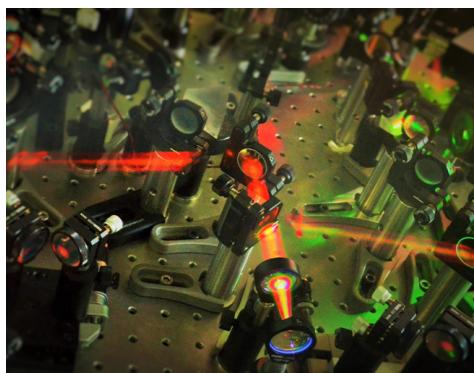
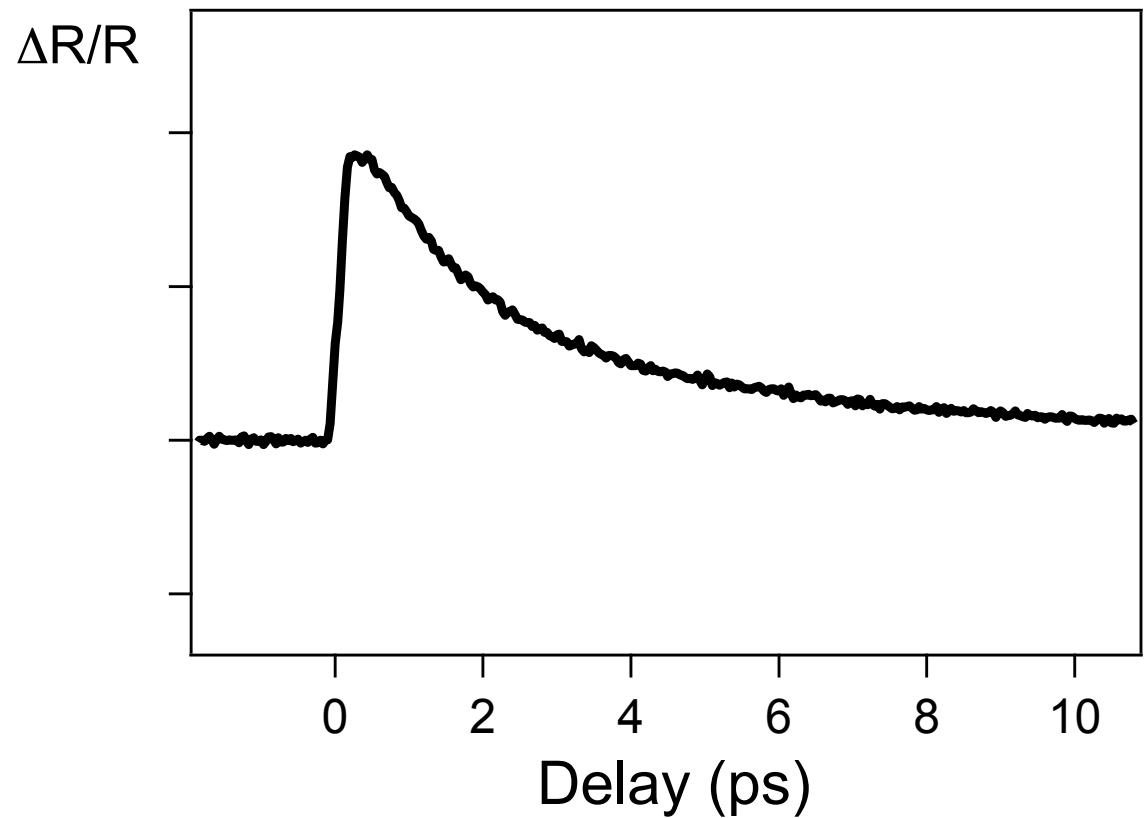
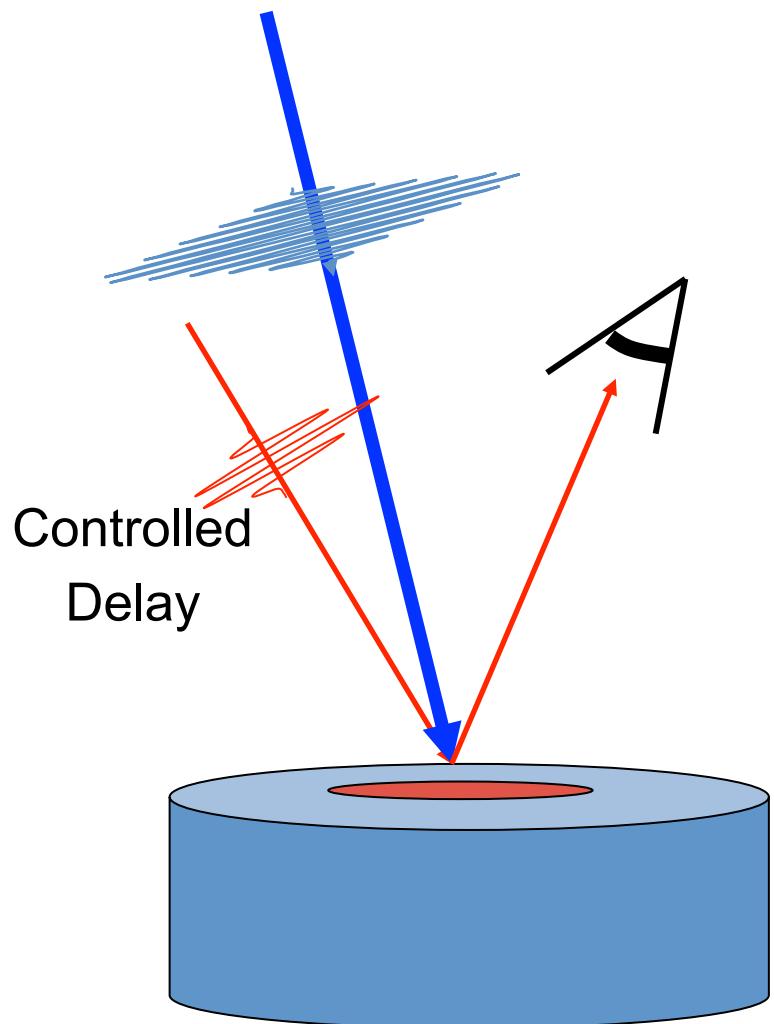
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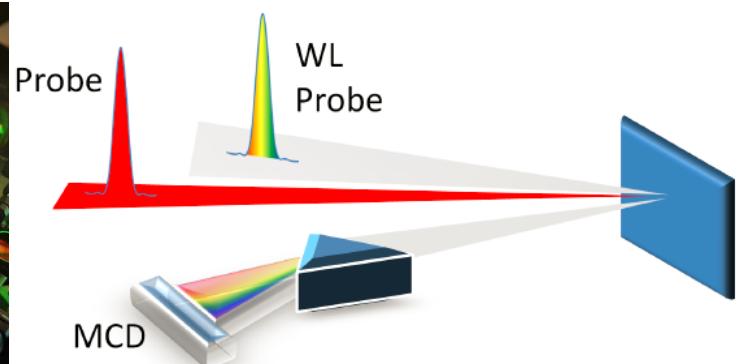
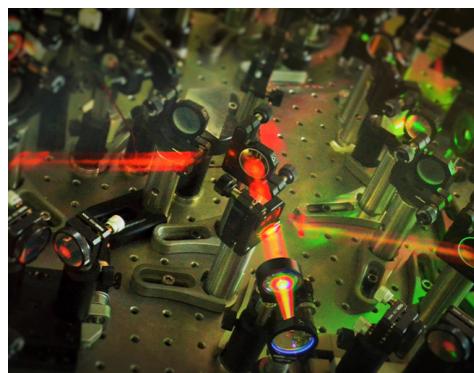
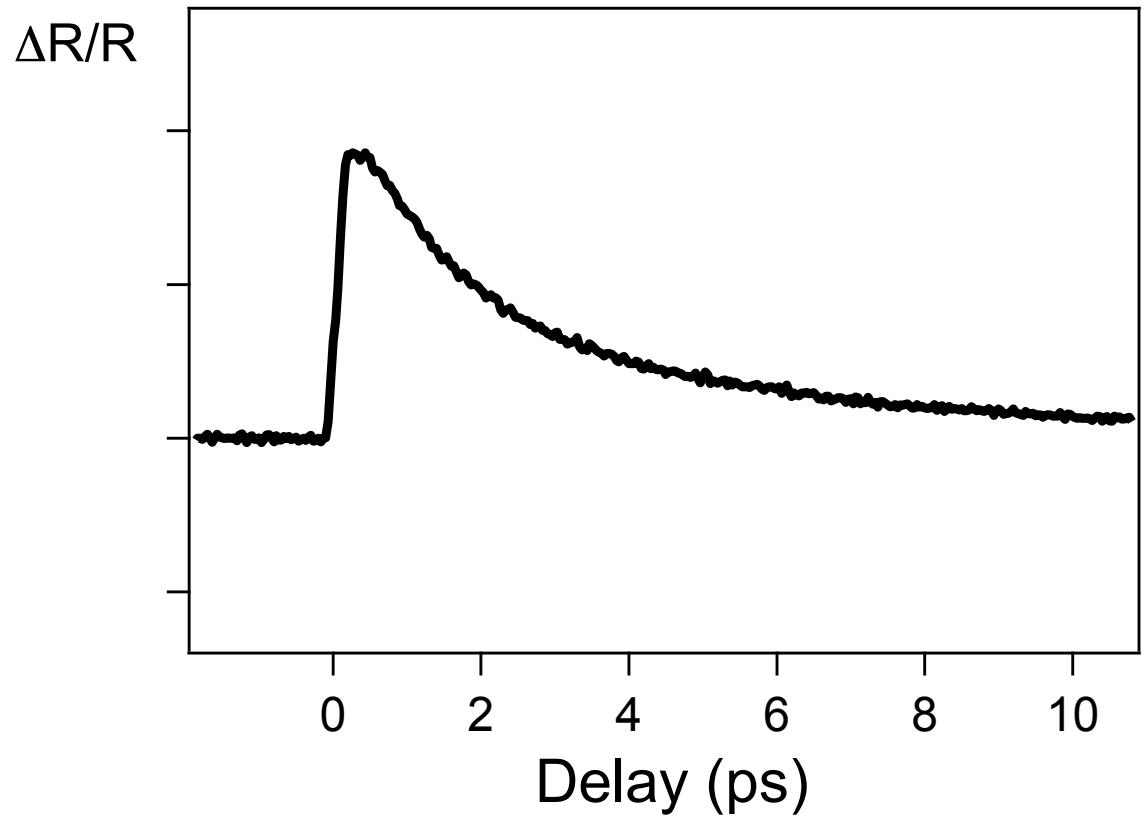
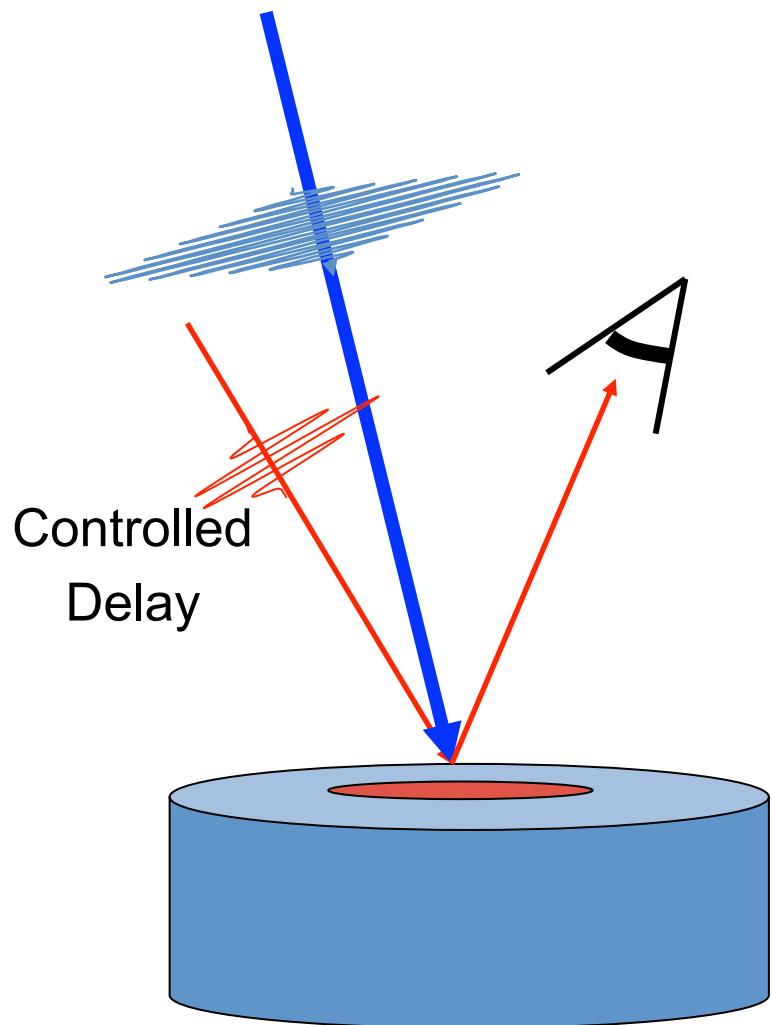
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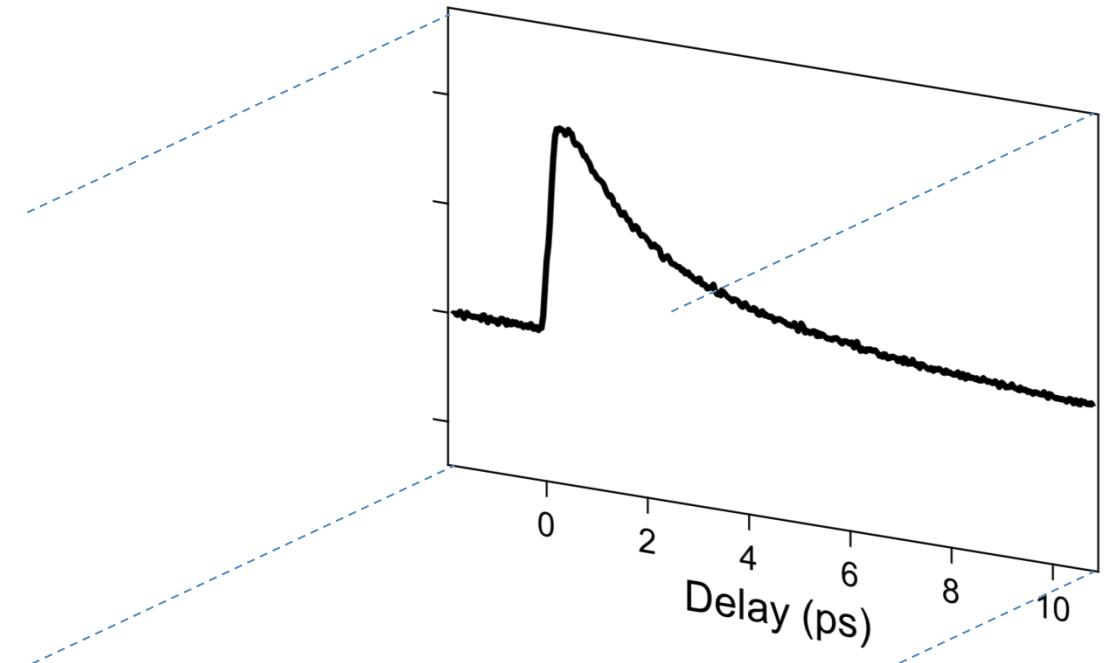
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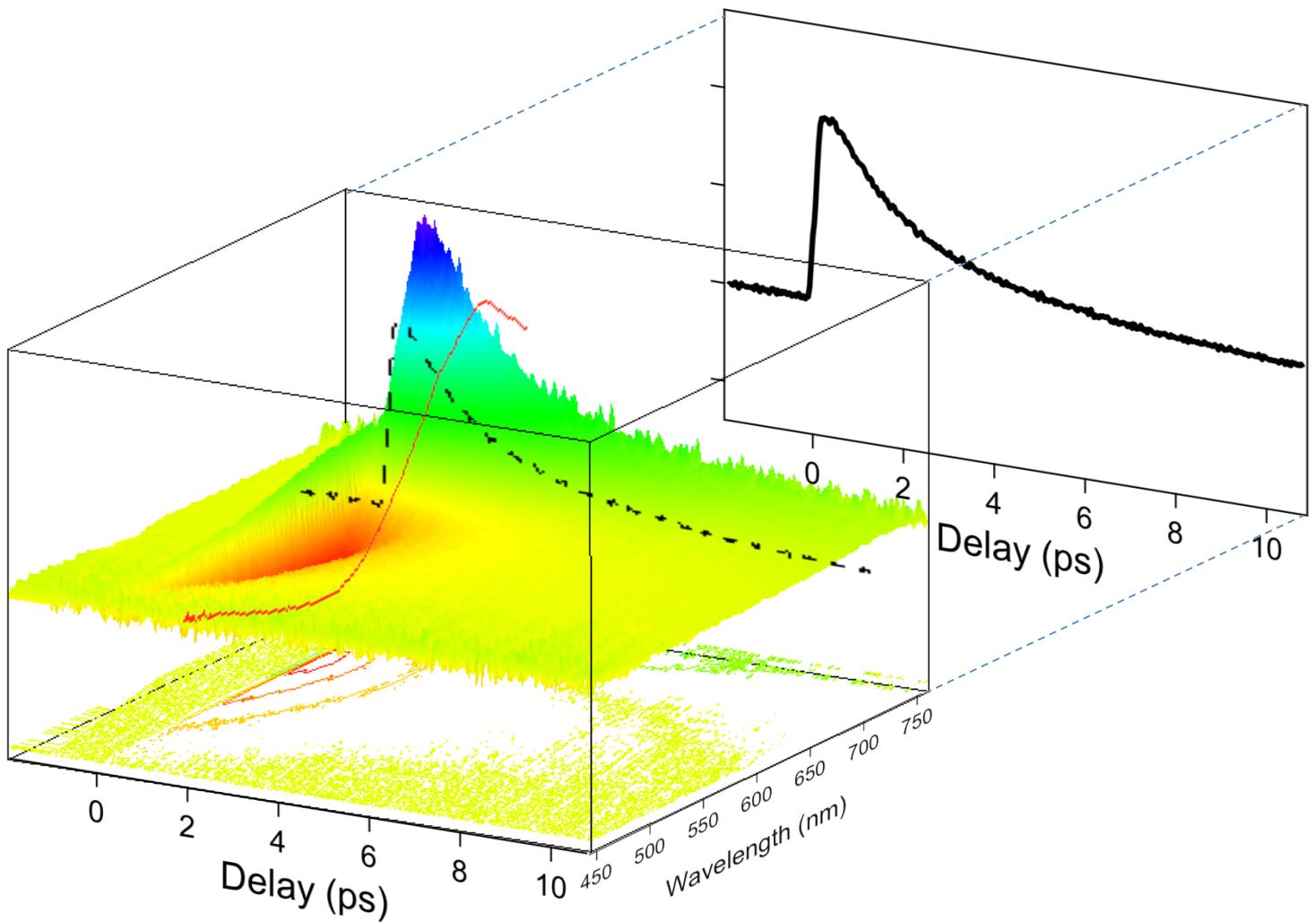
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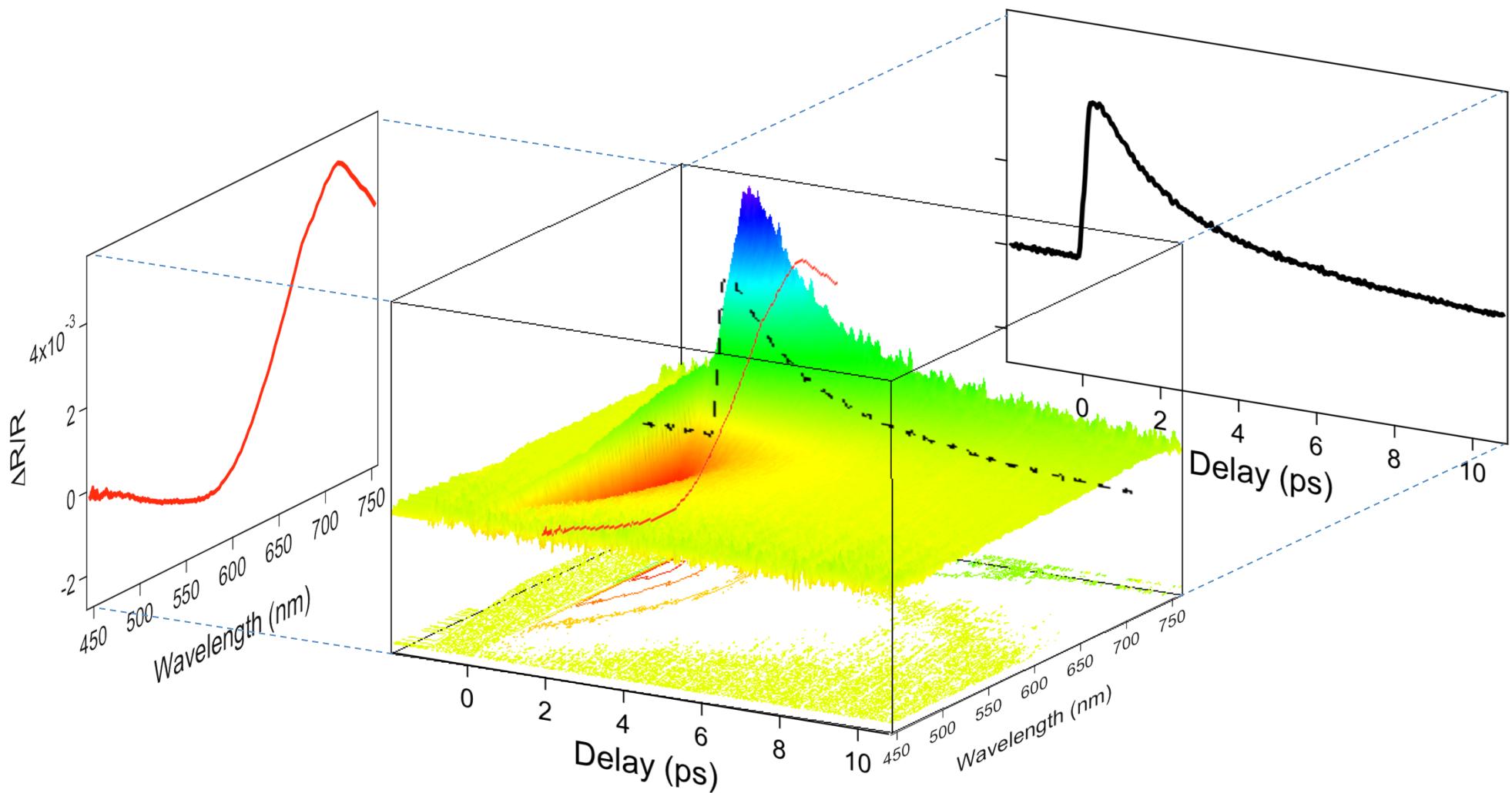
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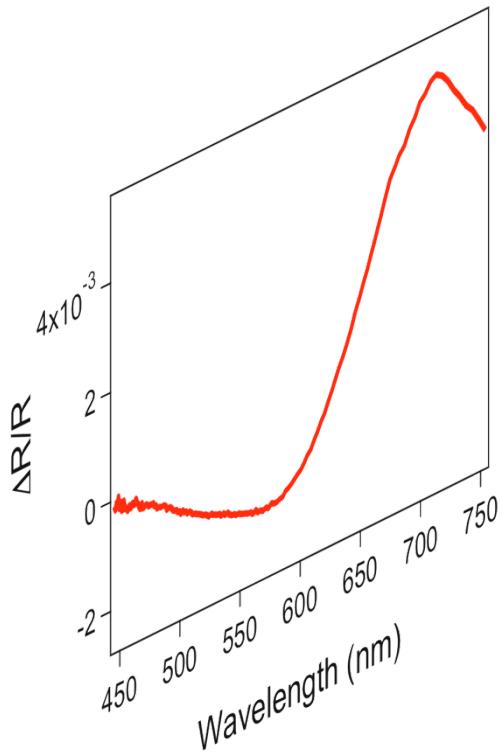
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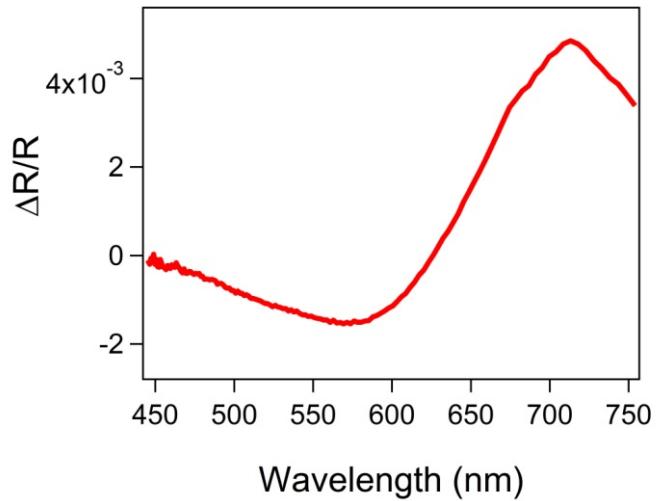
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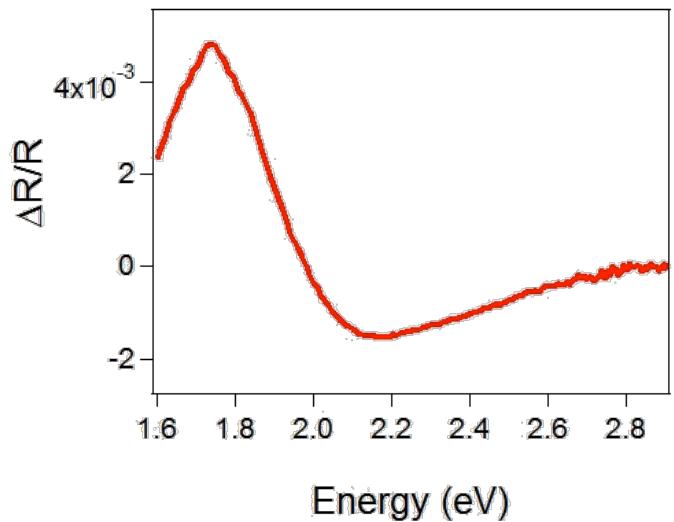
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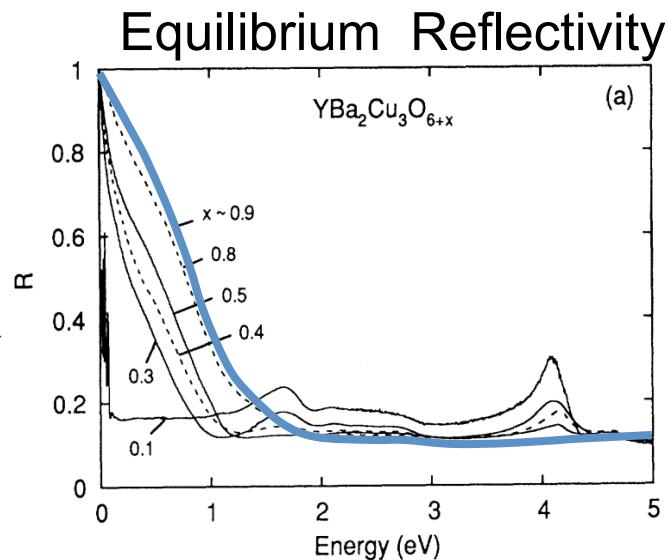
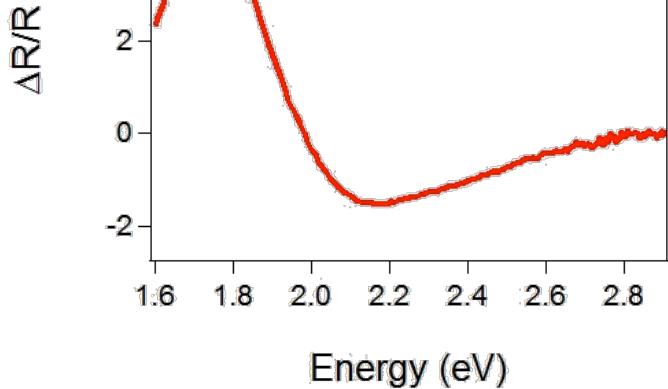


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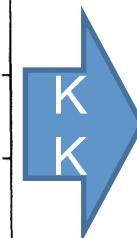
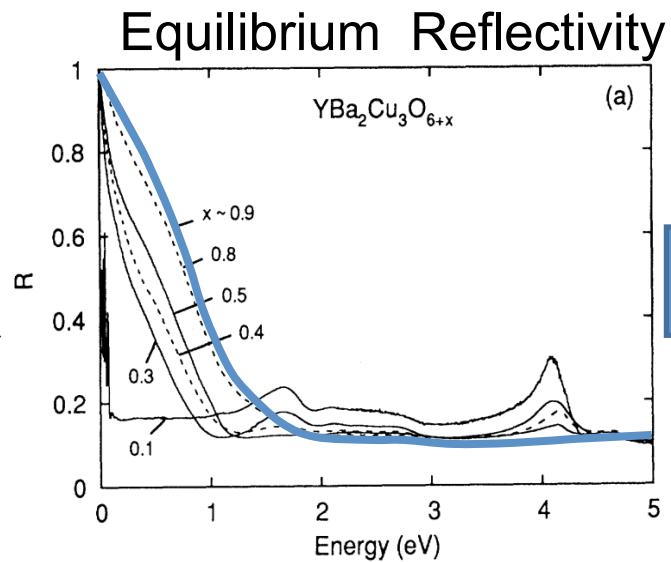
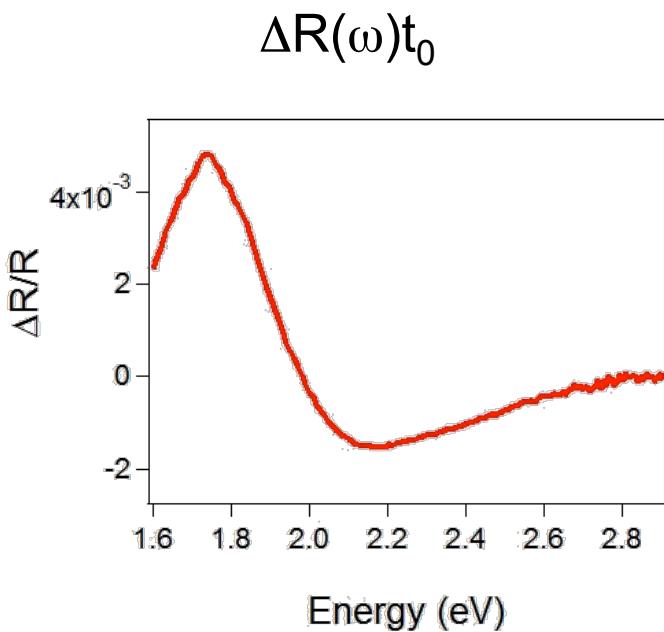


From P&P measurements to P&P spectroscopy

$\Delta R(\omega)t_0$

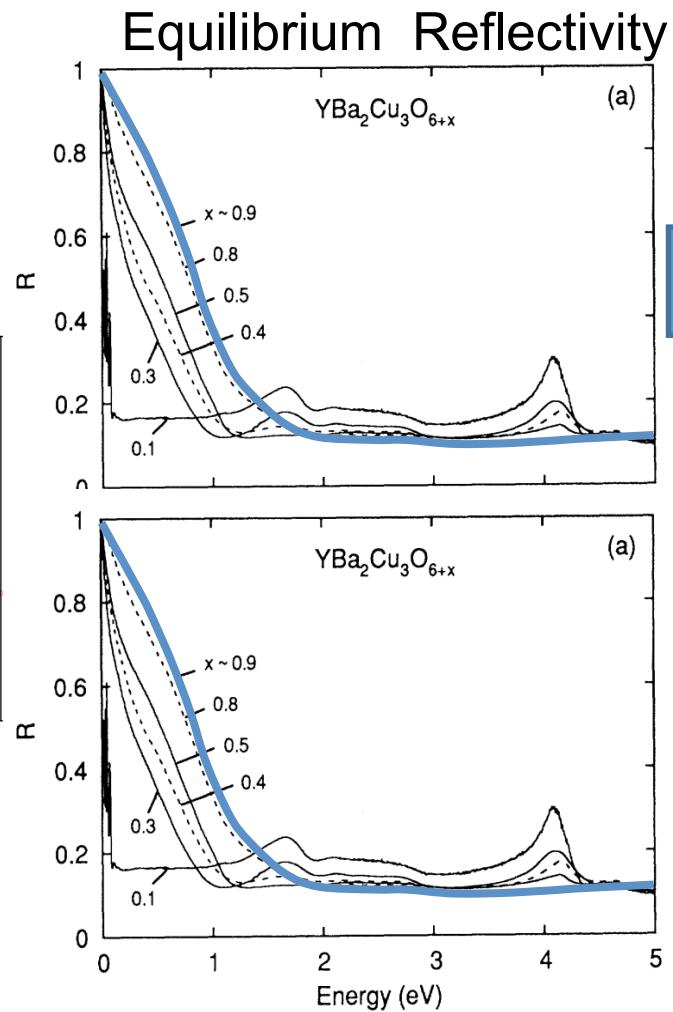
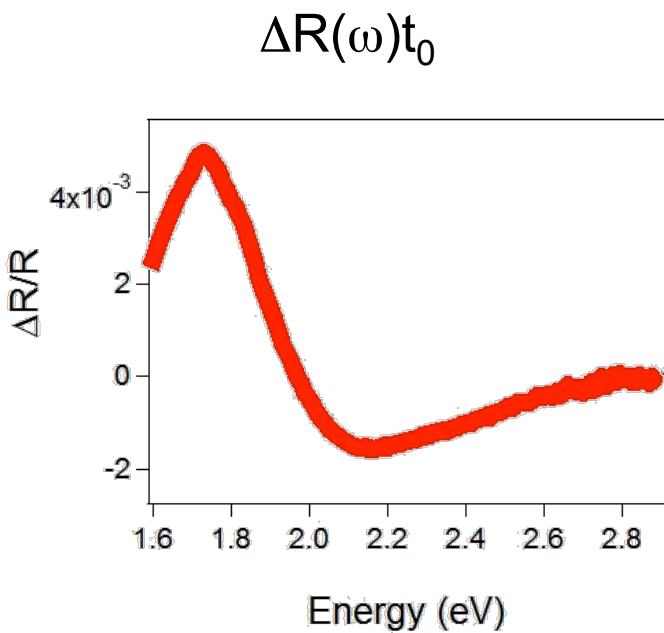


From P&P measurements to P&P spectroscopy



$\sigma_1(\omega)_{\text{eq}}, \sigma_2(\omega)_{\text{eq}}$

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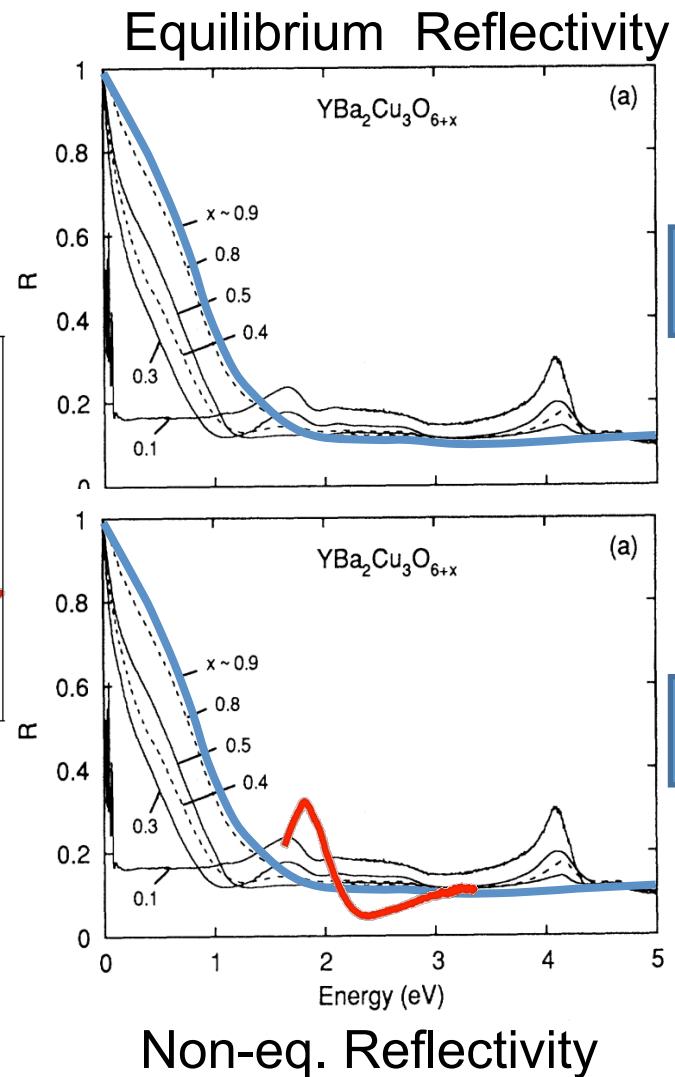
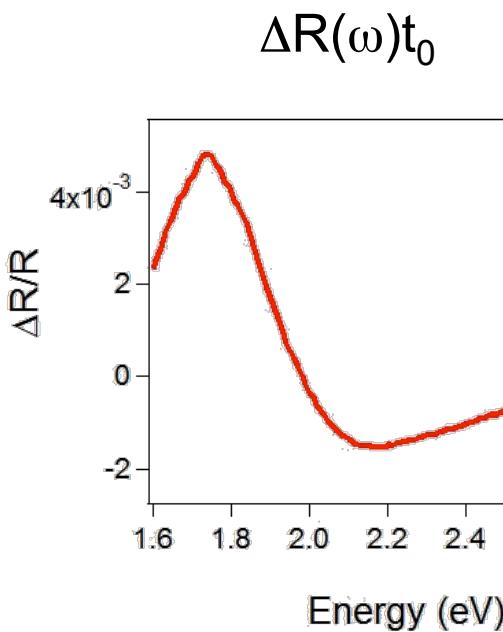


Non-eq. Reflectivity



$\sigma_1(\omega)_{\text{eq}}, \sigma_2(\omega)_{\text{eq}}$

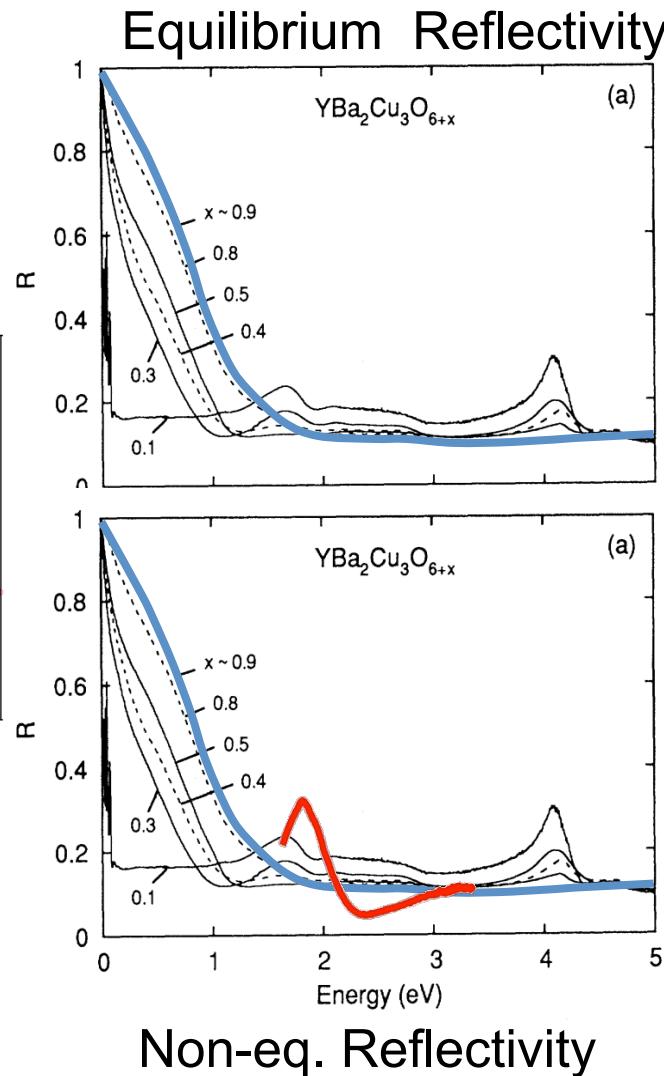
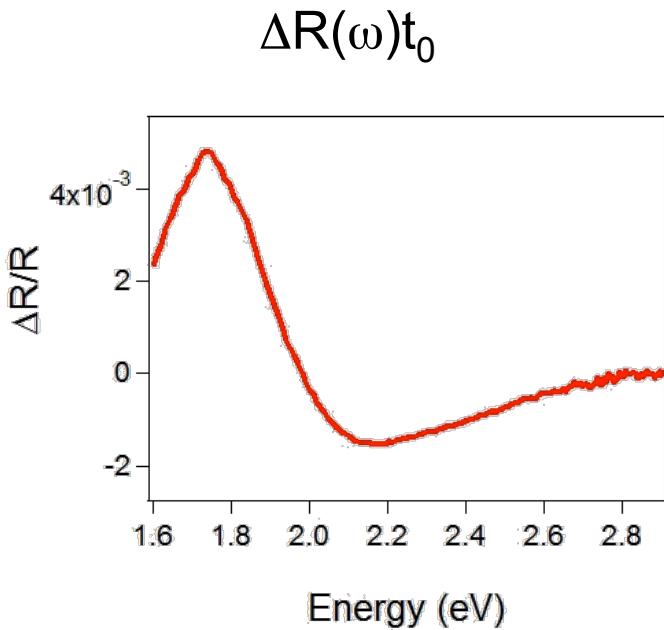
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$K \rightarrow \sigma_1(\omega)_{\text{eq}}, \sigma_2(\omega)_{\text{eq}}$

$K \rightarrow \sigma_1(\omega)_t, \sigma_2(\omega)_t$

From P&P measurements to P&P spectroscopy



$\rightarrow K$

$\sigma_1(\omega)_{\text{eq}}, \sigma_2(\omega)_{\text{eq}}$

$\rightarrow K$

$\sigma_1(\omega)_t, \sigma_2(\omega)_t$

$\Delta\sigma_1(\omega)_t, \Delta\sigma_2(\omega)_t$

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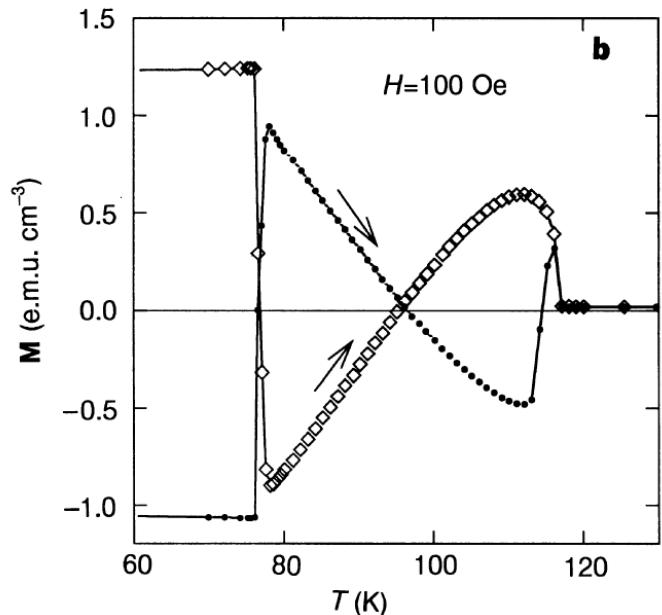
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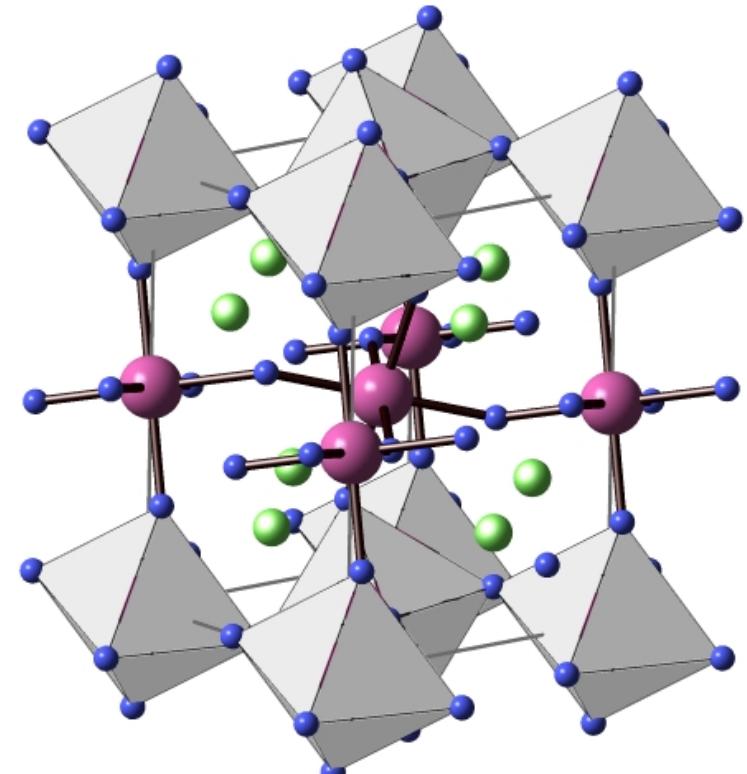
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YVO₃: Properties

- ✓ Layer compound of BaTiO₃-like distorted/tilted octahedra (Pbnm)
- ✓ V³⁺ → 3d₂
- ✓ Mott insulator
- ✓ Magnetization reversal
- ✓ Orbital/Spin orderings

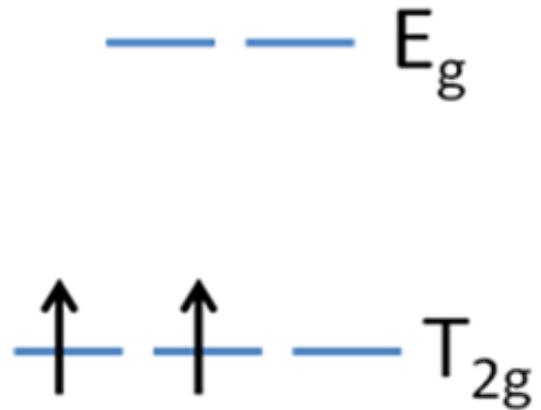
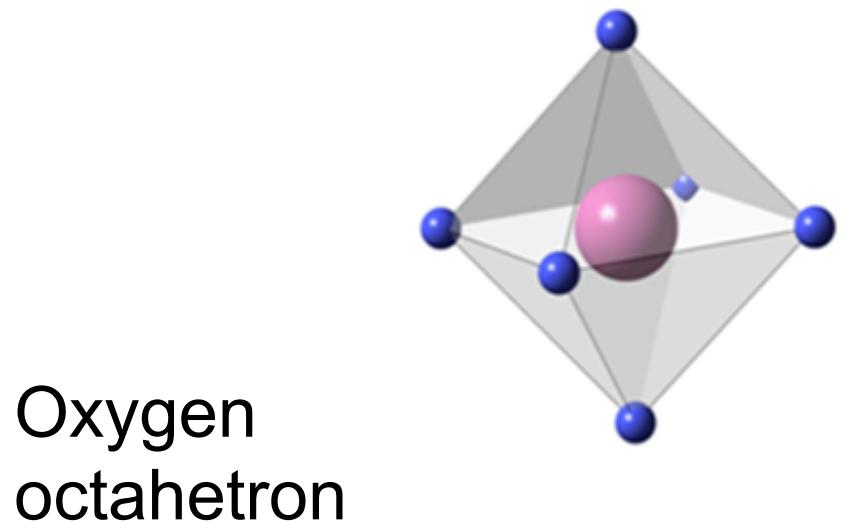


Y. Ren, Nature 396, 401, 1998

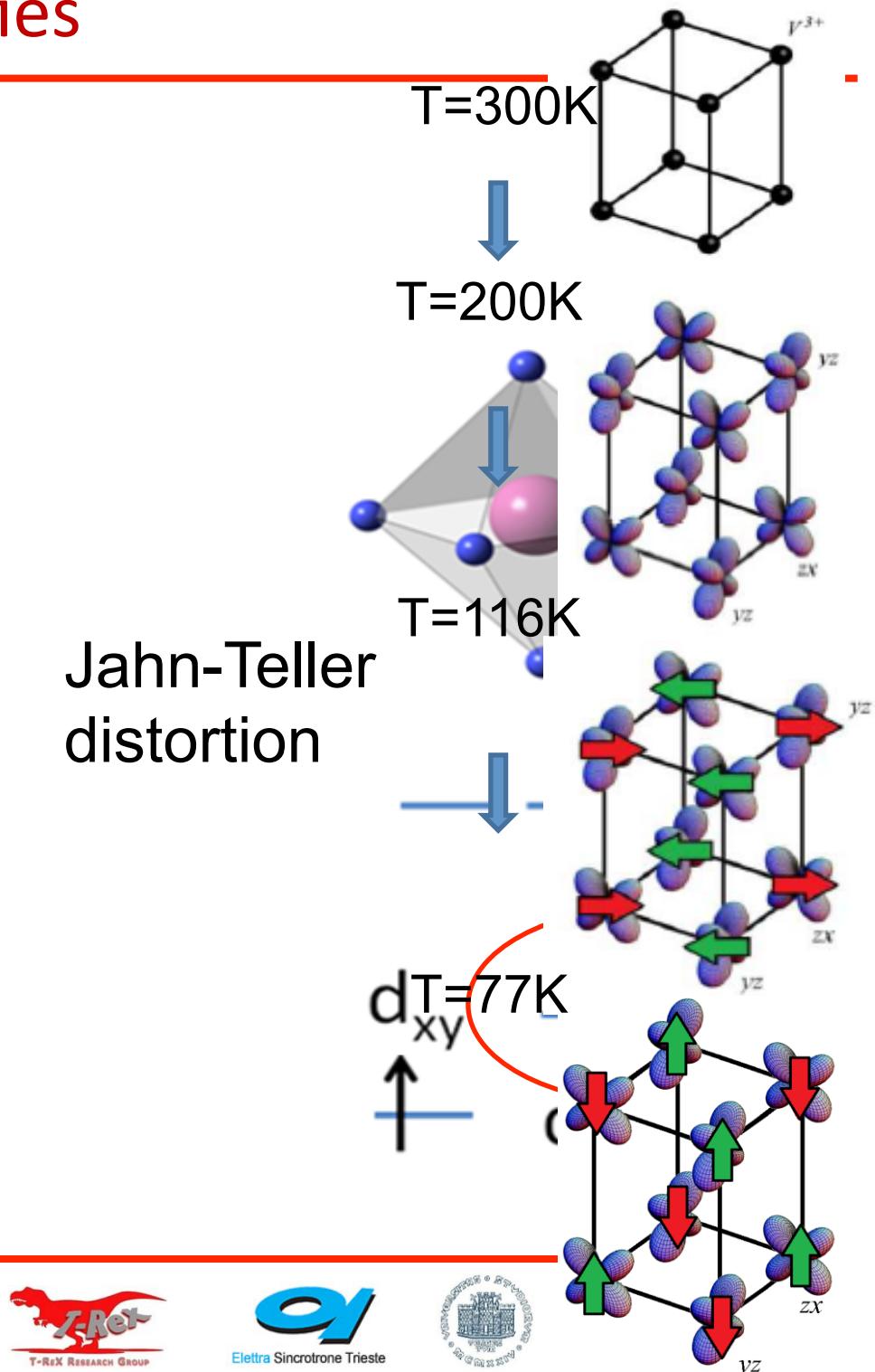


YVO_3 : Properties

Orbital Physics in V^{3+} (3d_2)

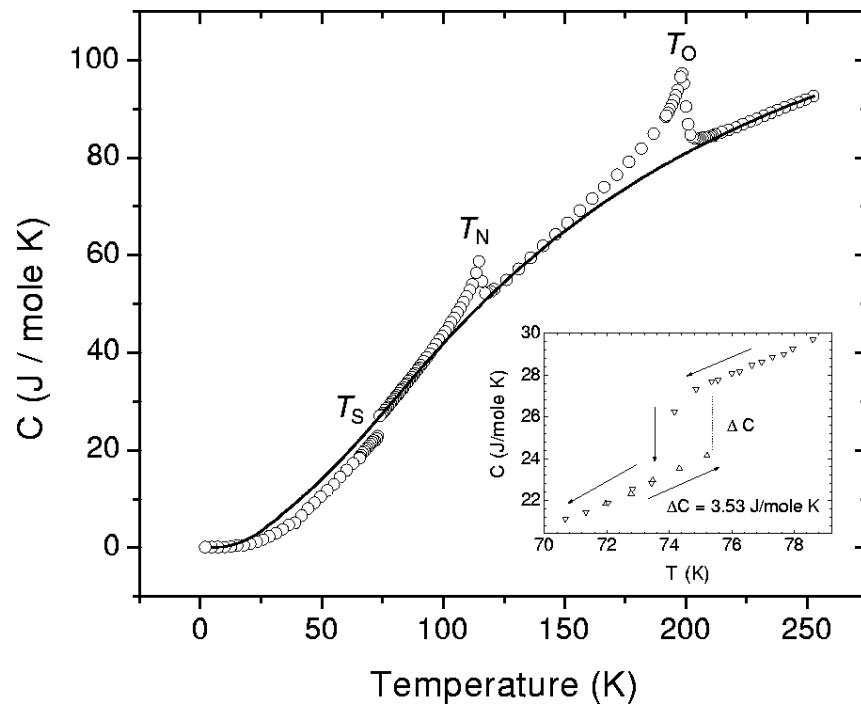


Jahn-Teller
distortion

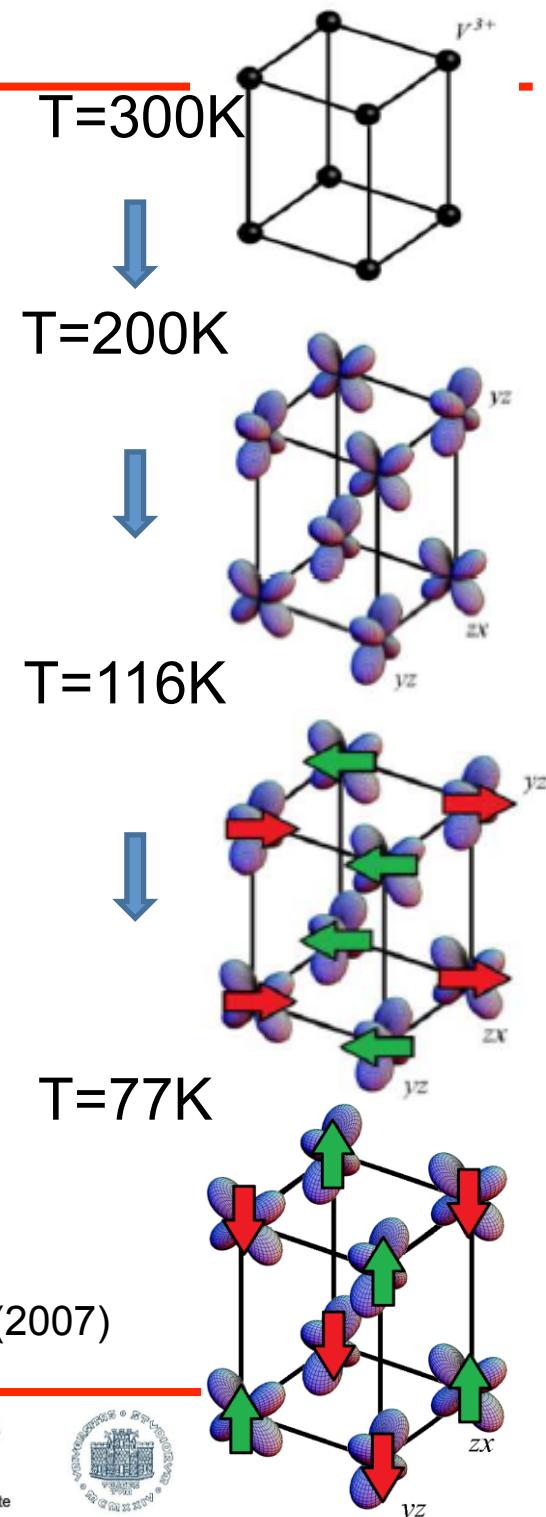


YVO_3 : Properties

- ✓ Mott insulator
Mott gap $\sim 1.2\text{eV}$
- ✓ Crystal field determined “mainly” by JT
- ✓ No Quantum fluctuation

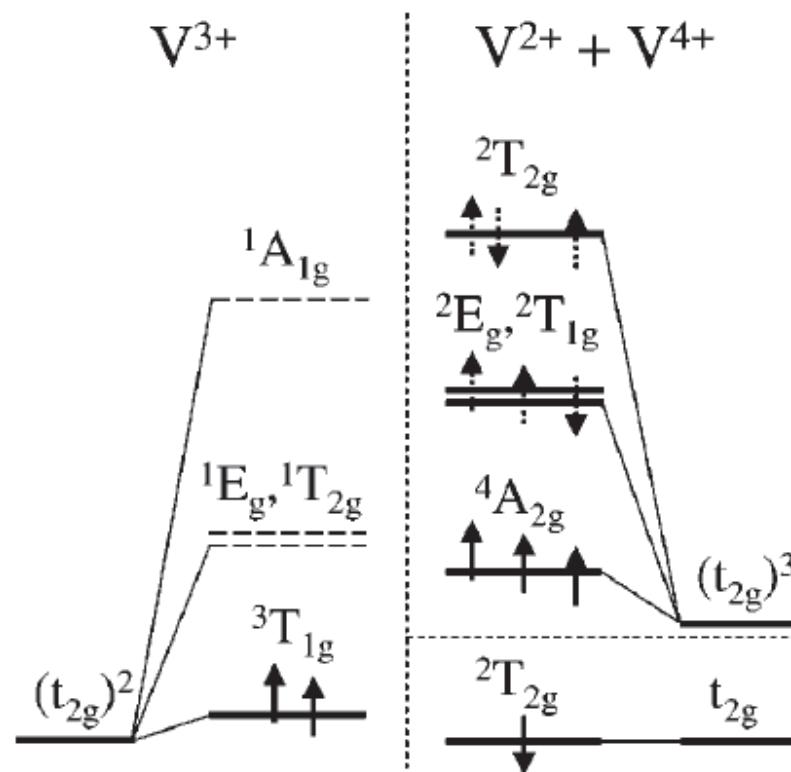


PRB, 65, 174112 (2002); PRL 91, 257202 (2003); PRL 99, 126402 (2007)

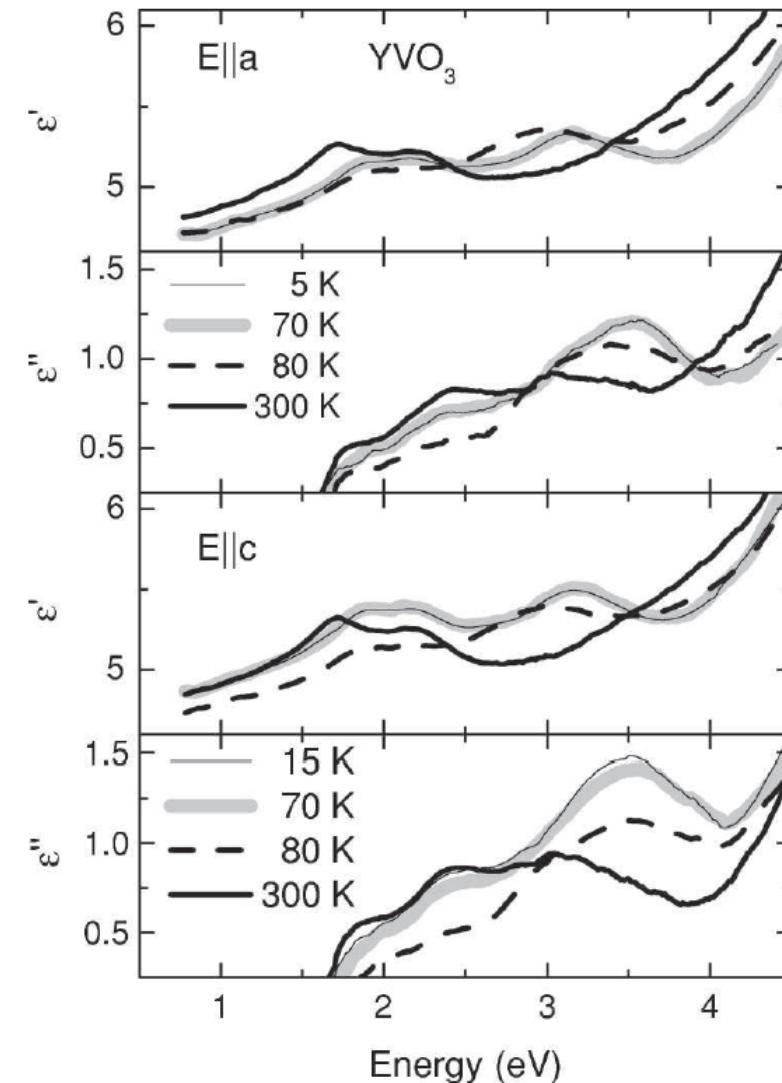


YVO_3 : Optical Properties

- ✓ $d_2d_2 \rightarrow d_1d_3$
 $\text{V}^{3+}\text{V}^{3+} \rightarrow \text{V}^{2+}\text{V}^{4+}$
- ✓ Multiplet Calculations

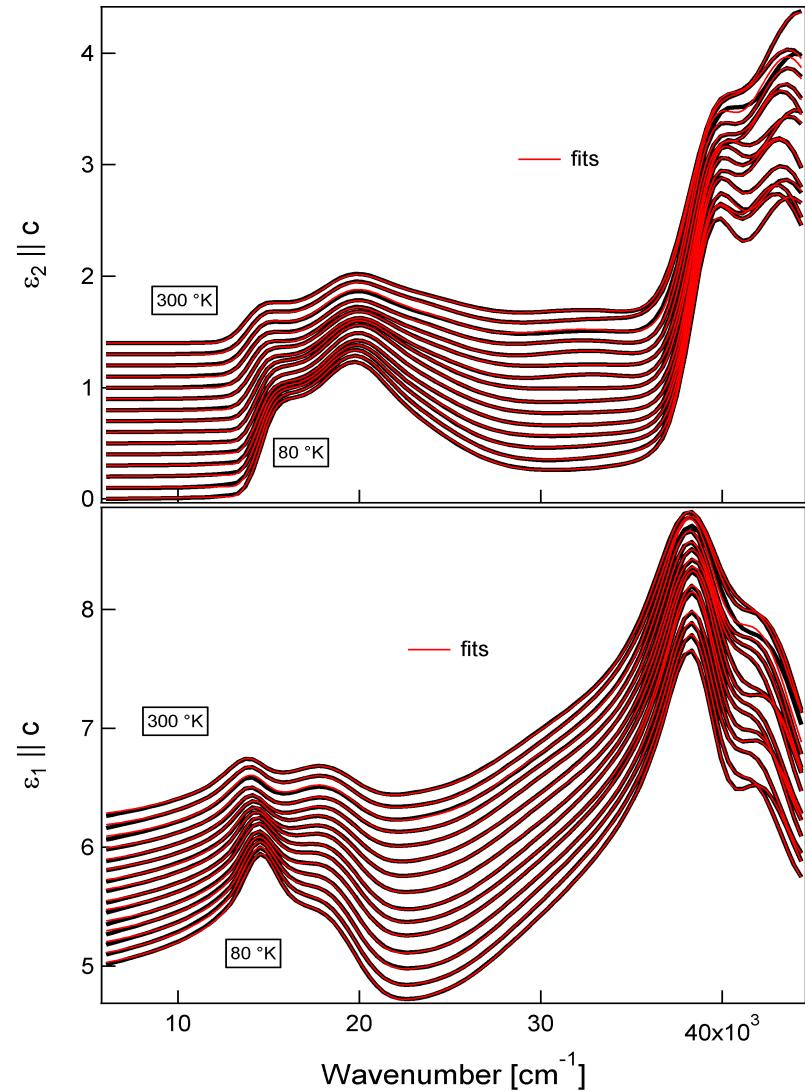


Tzvetkov A. & al. PRB, 69, 075110



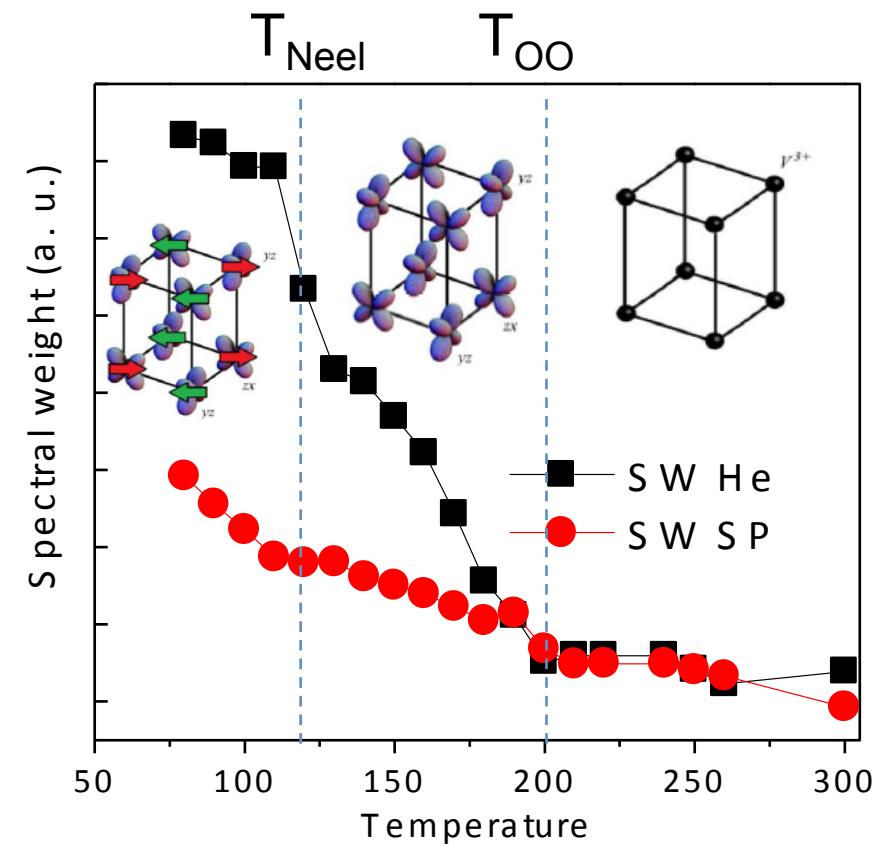
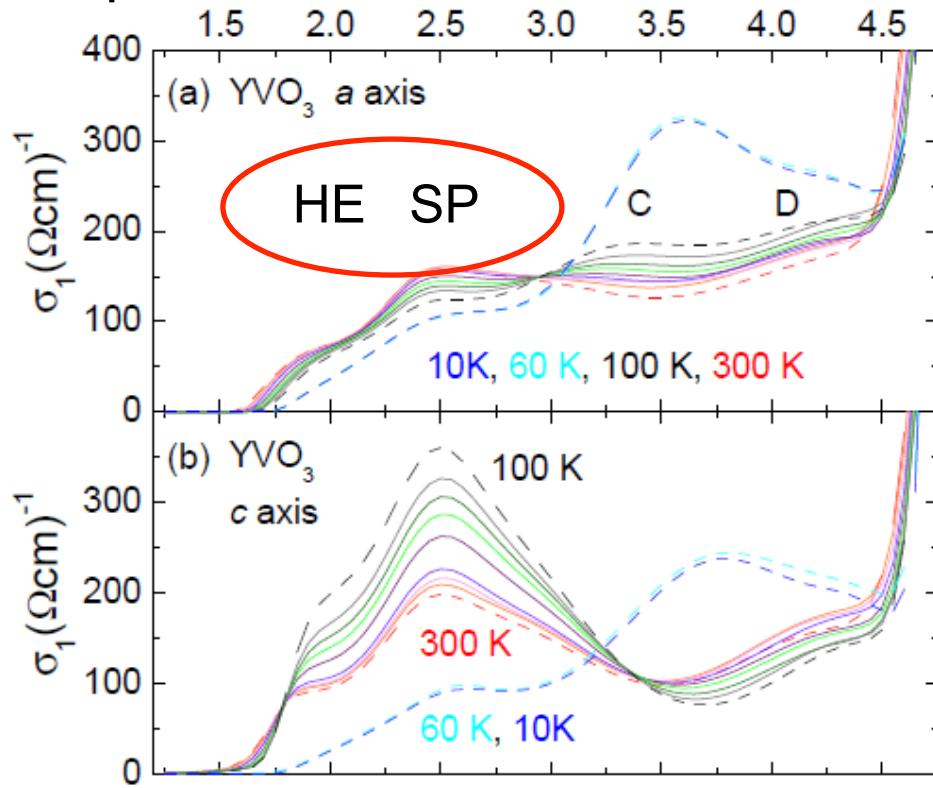
YVO_3 : Optical Properties

- ✓ 6 Gaussian and 1 Tauc-Lorentz oscillators



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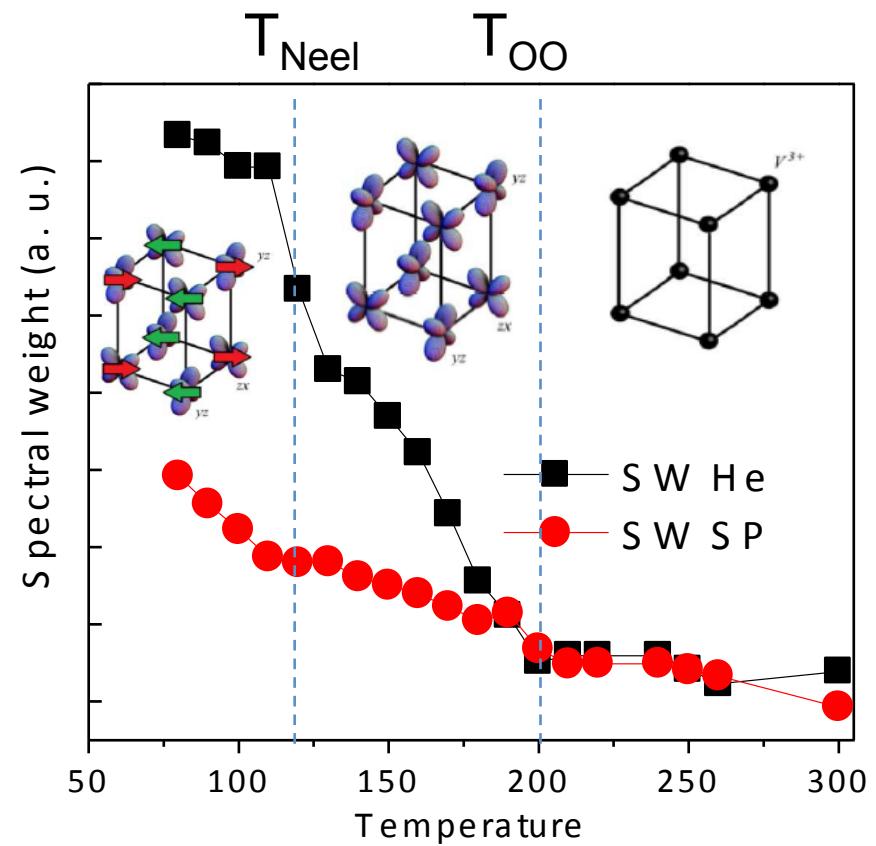
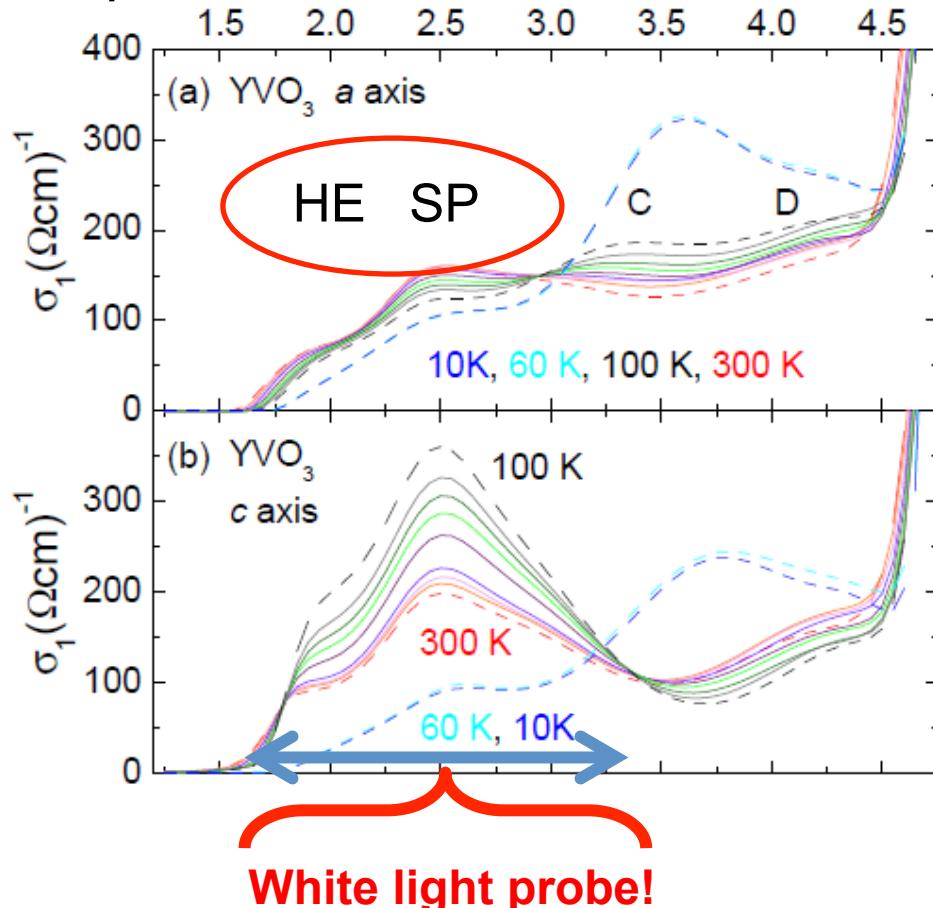
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Reul. J, Gruninger M. & PRB

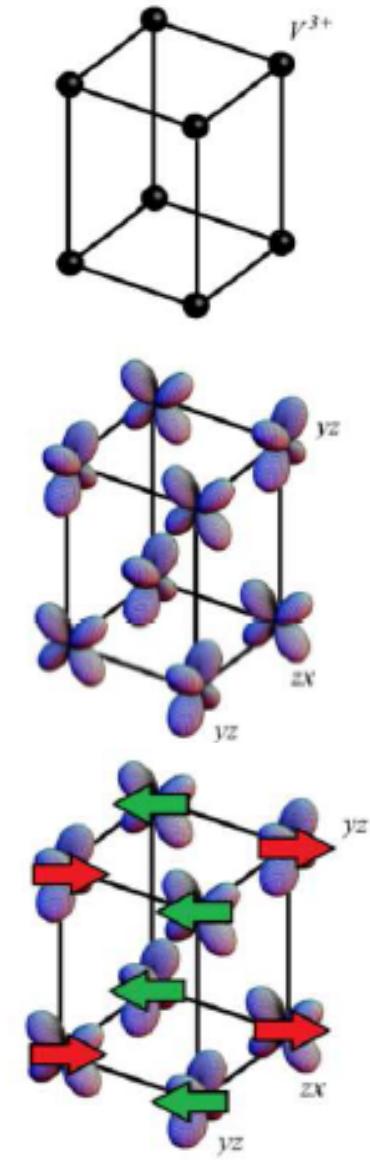
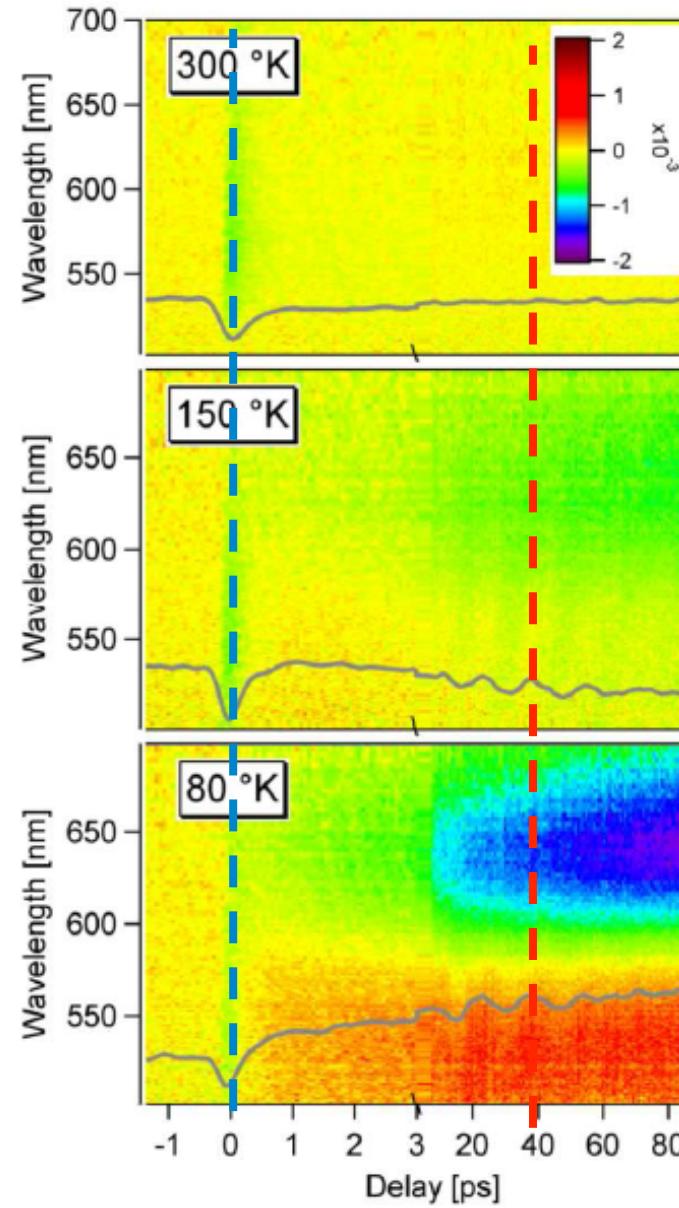
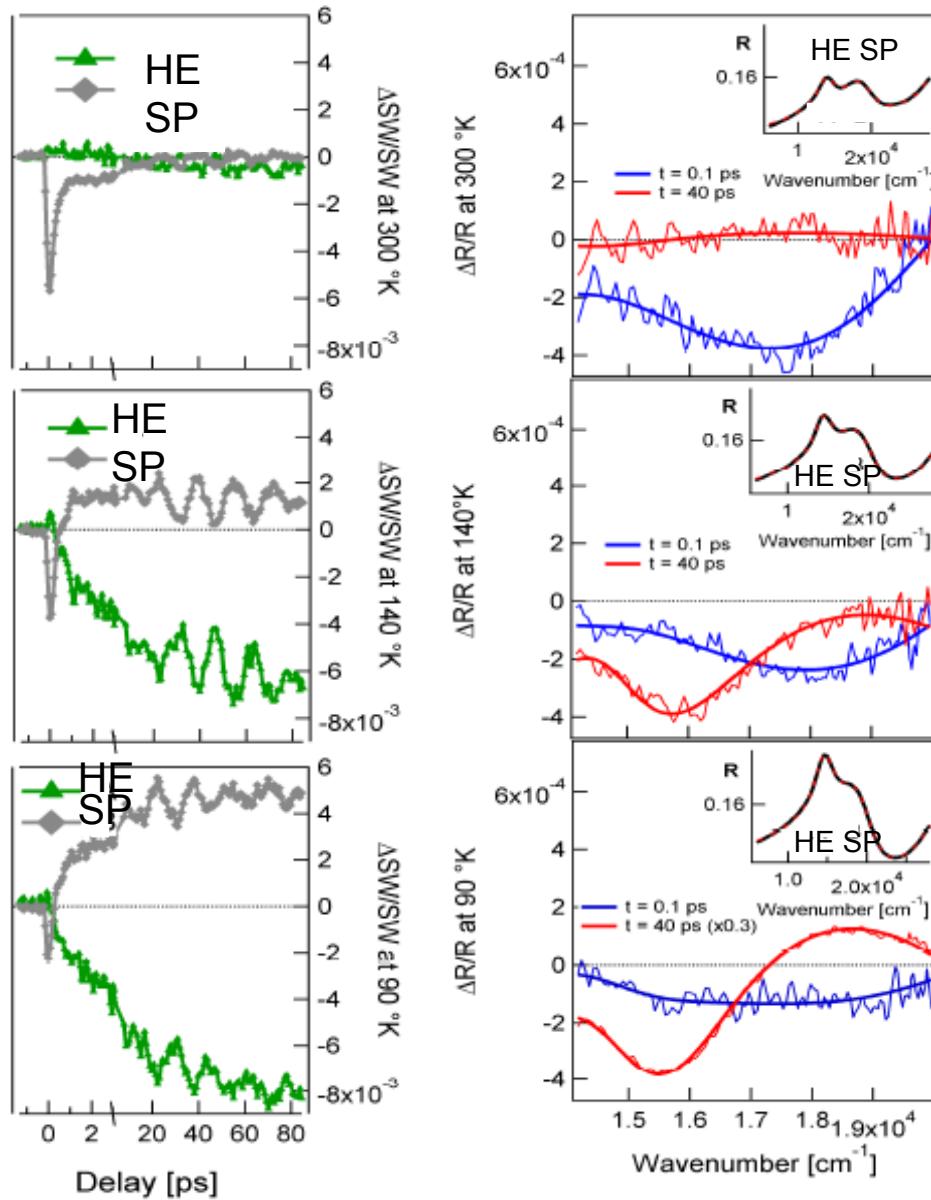
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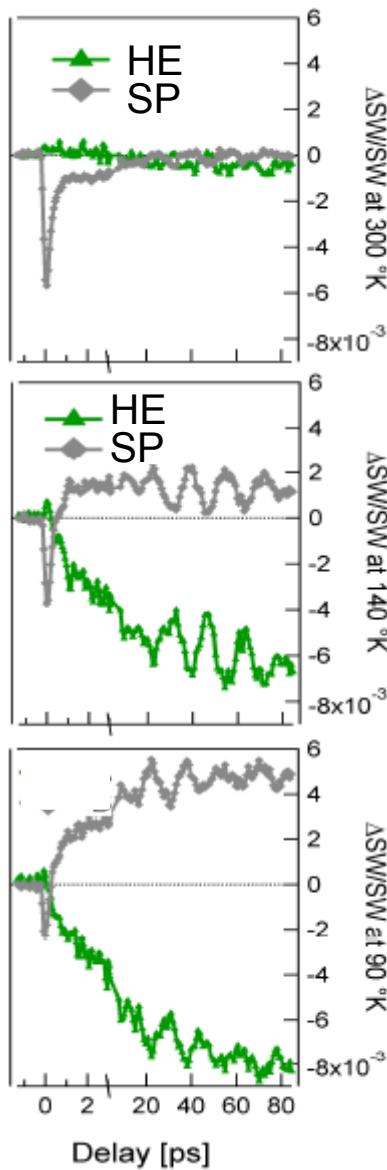


Reul. J, Gruninger M. & PRB

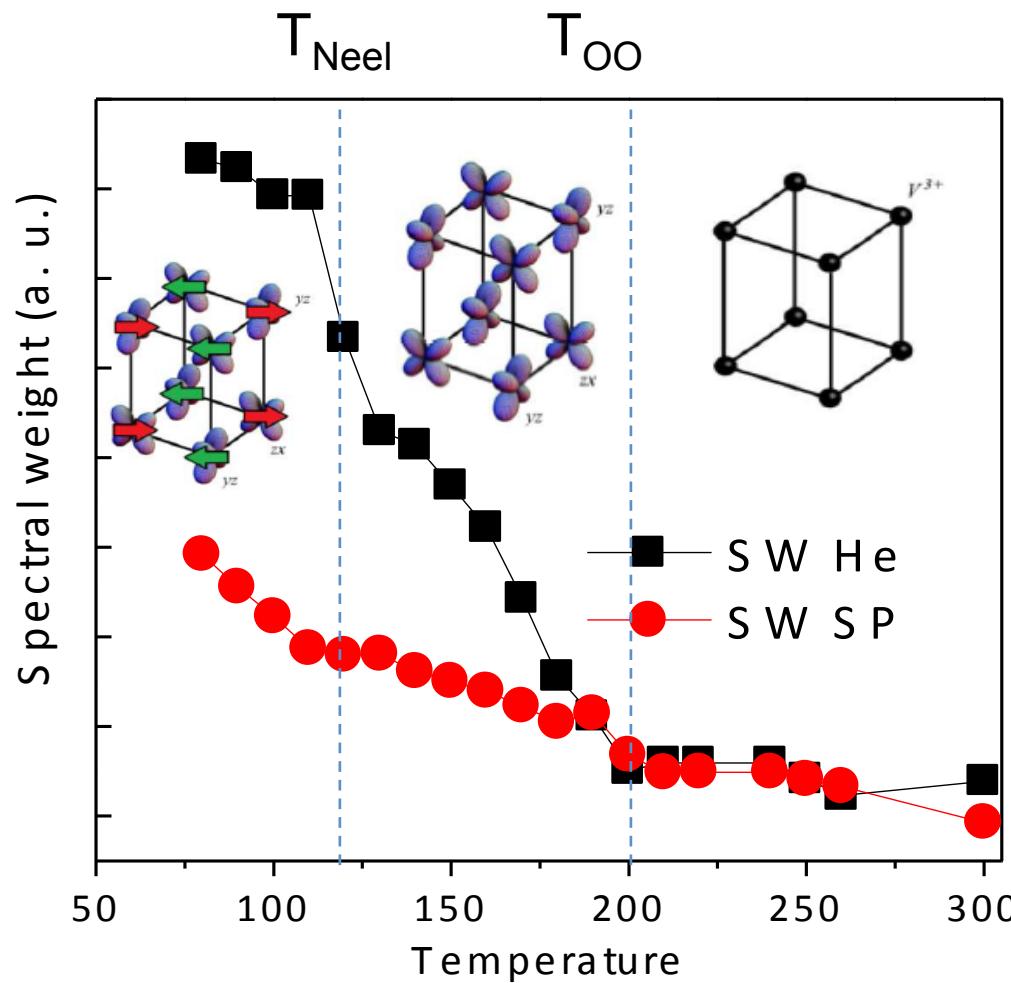
YVO_3 : Transient Optical Properties



Thermal Vs. Non-thermal SW



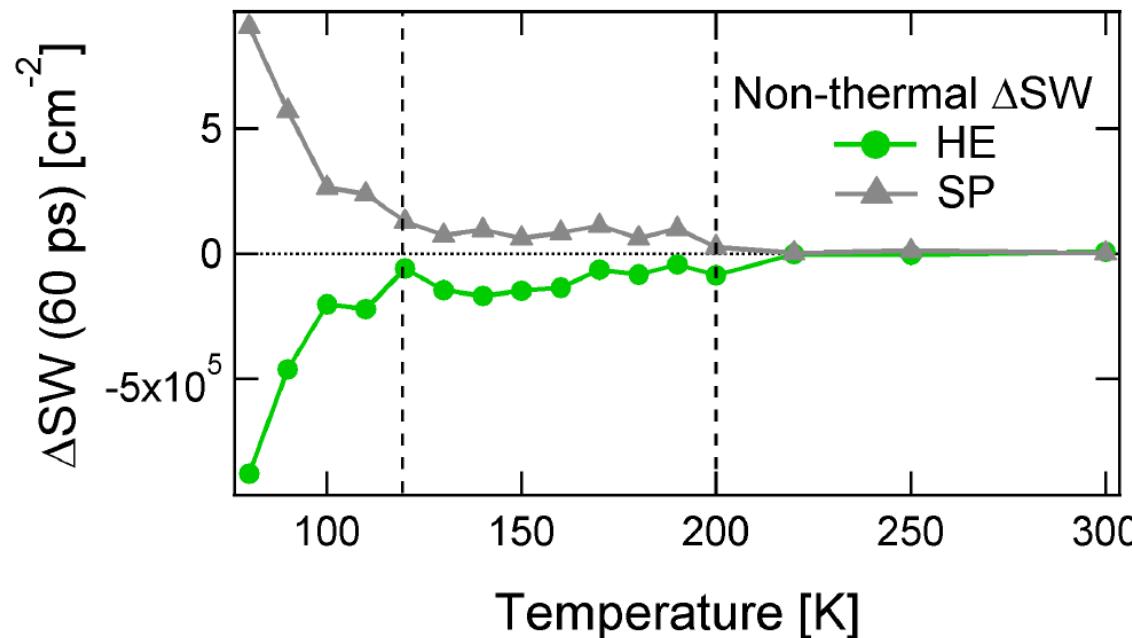
- ✓ Thermal benchmark
- ✓ Non-Thermal Thermal contribution



Thermal Vs. Non-thermal SW

- ✓ Thermodyn. estimate of the T variation in the photo-excited state(ΔT)
- ✓ Extrapolation of static optical properties ($T_x + \Delta T$)

$$\Delta T[K] = \frac{Q_{abs} \cdot N_A \cdot V}{S \cdot d \cdot u \cdot C_{mol}}$$



- ✓ Direct Exchange of SW between the two bands
i.e. **It is the same band!**

Hubbard Exciton?!

Phys. Rev. B 86, 165135 (2012)

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✓ Equilibrium Optical Spectroscopy

- The optical conductivity in the Visible and Near-IR
- What do we learn from optical conductivity?
- An example, metal insulator transition in complex oxides

✓ Non-equilibrium optical (visible near-IR) spectroscopy

- Pump&probe the main idea
- «Single color» Pump and probe
- Broadband P&P spectroscopy

✓ Self referential examples

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- The electron-phonon (EP) interaction in with strong electronic correlation

✓ Non-equilibrium Infrared spectroscopy

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- THz pump and optical probe spectroscopy
- Phonon pump optical probe spectroscopy (MidIR pulse generation)

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- Beyond classical spectroscopy
- Using the quantum state of light as a new spectroscopyc tool
- Table top Vs. FEL and sincrotrons

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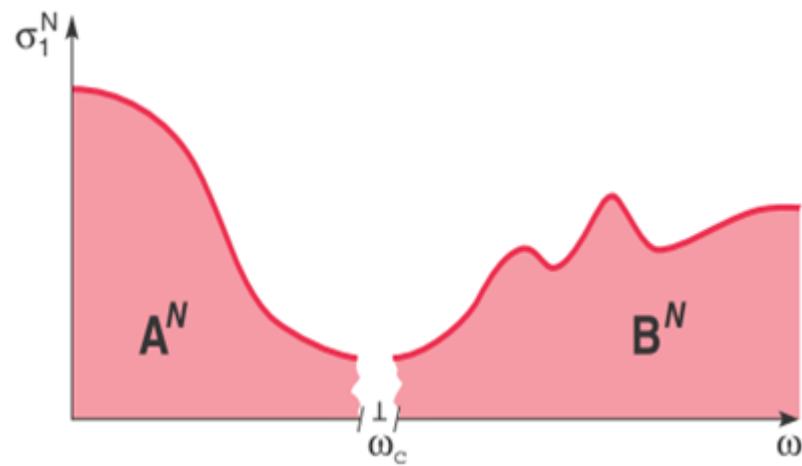
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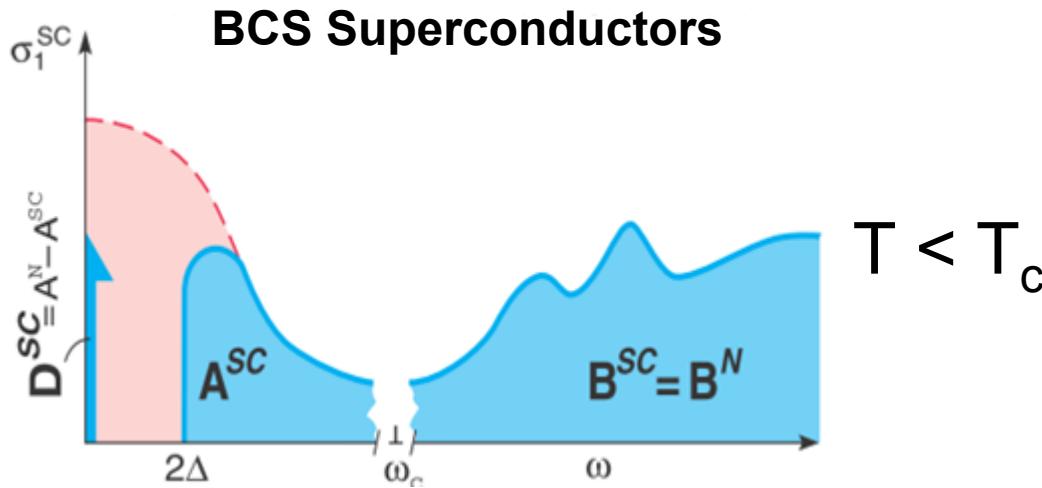
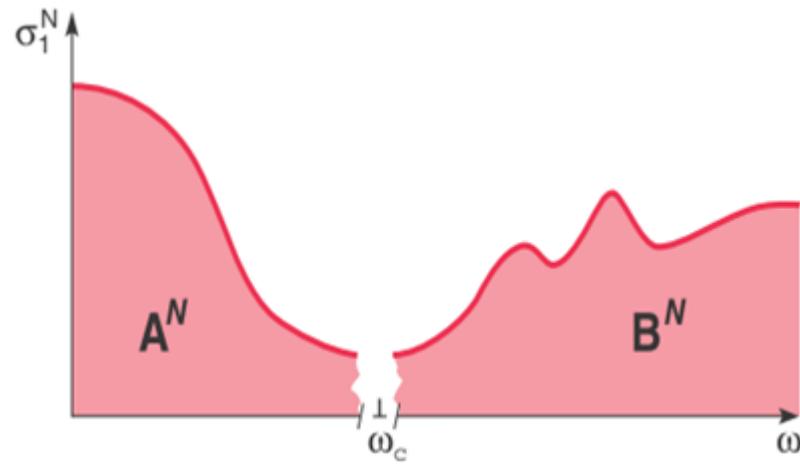
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Open problem in the cuprates



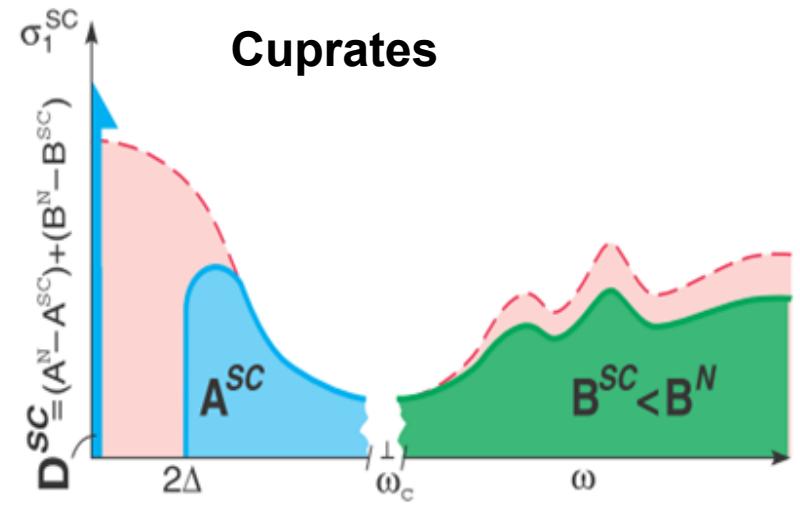
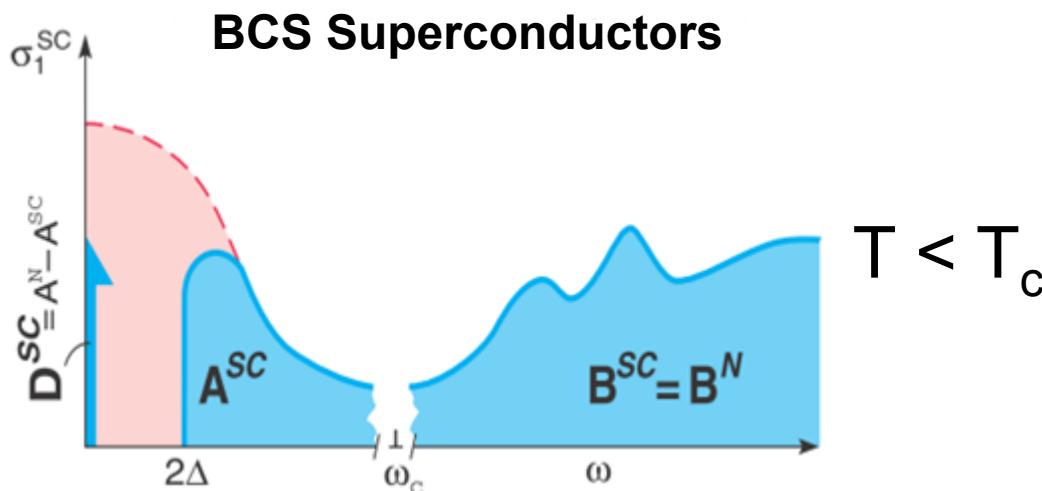
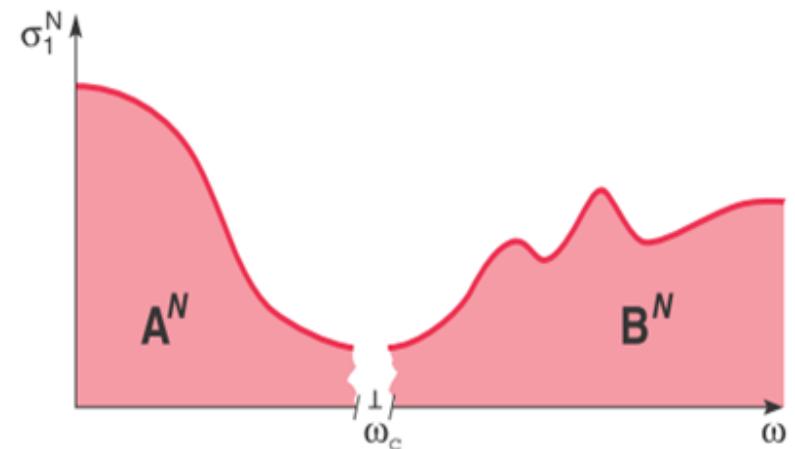
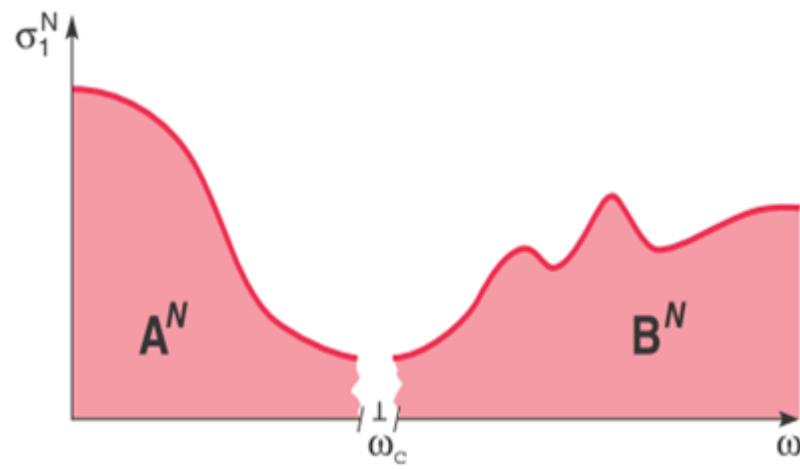
Rep.Prog.Phys. 66 1547 2003

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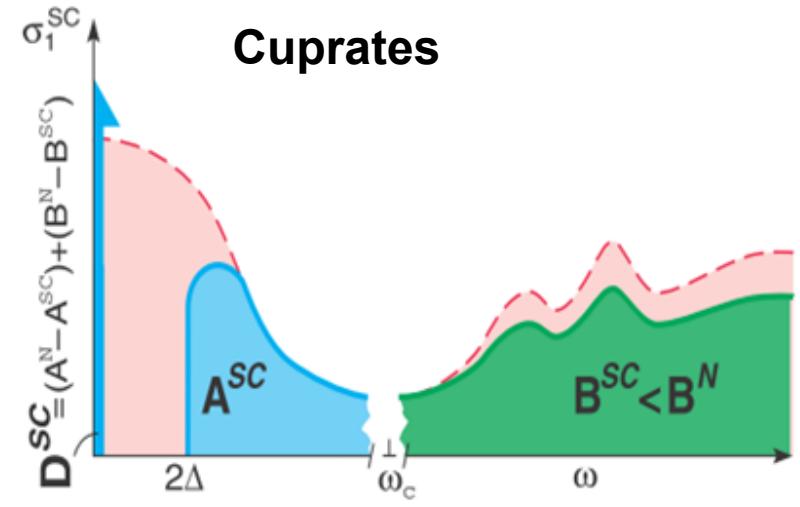
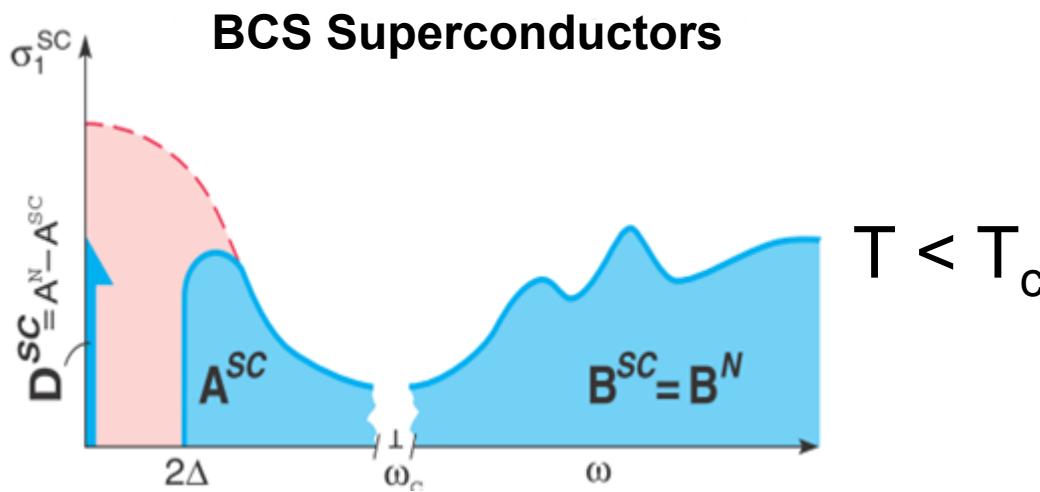
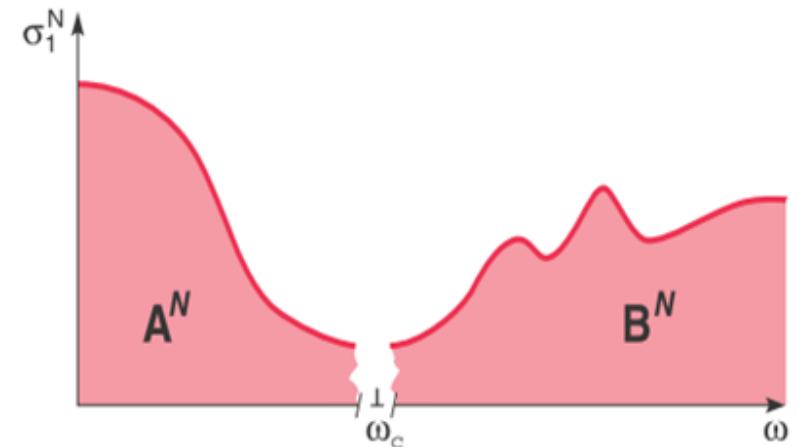
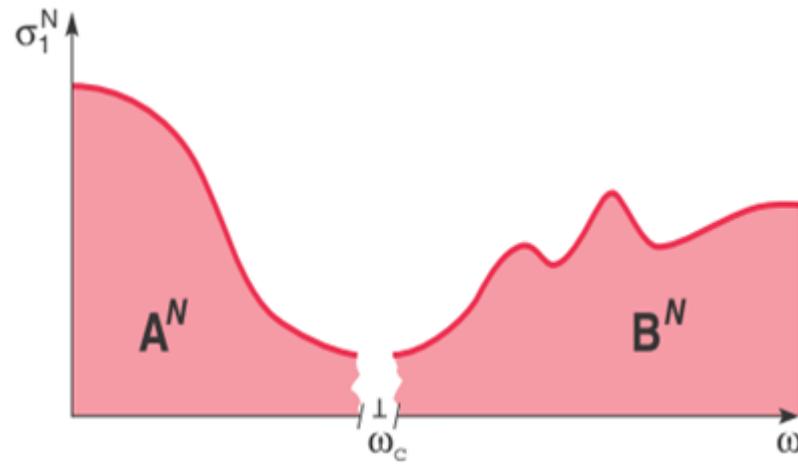
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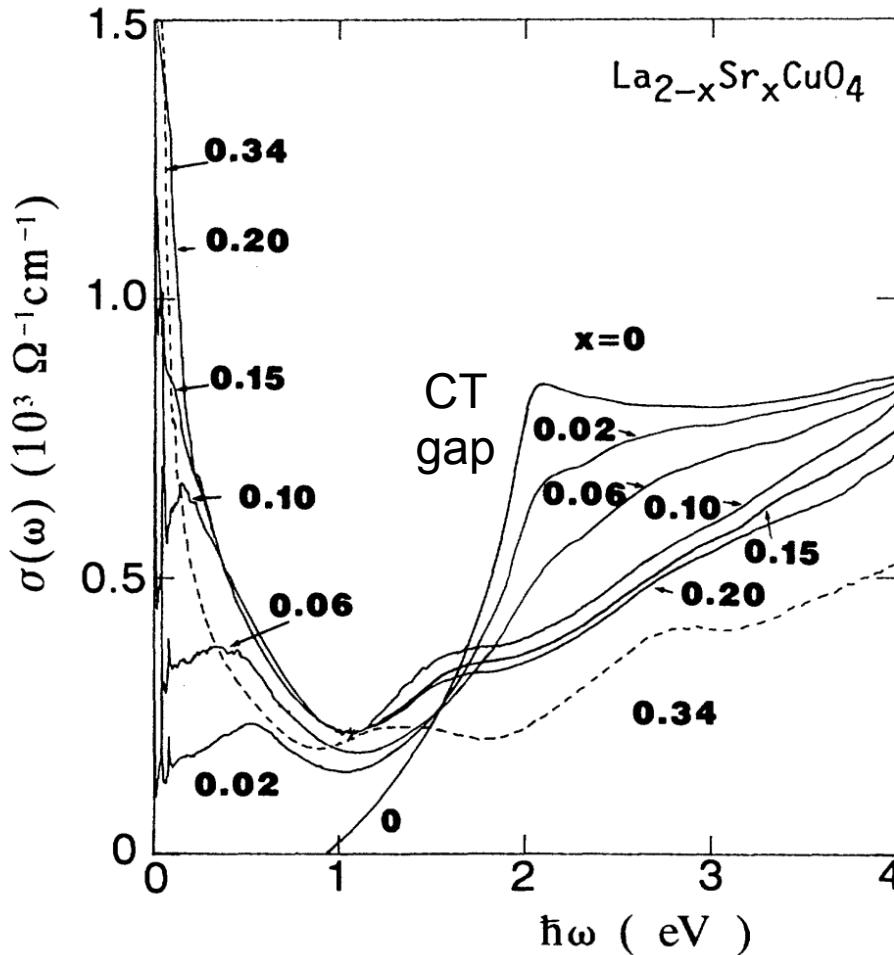
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What links the low energy physics of superconductivity to high energy?

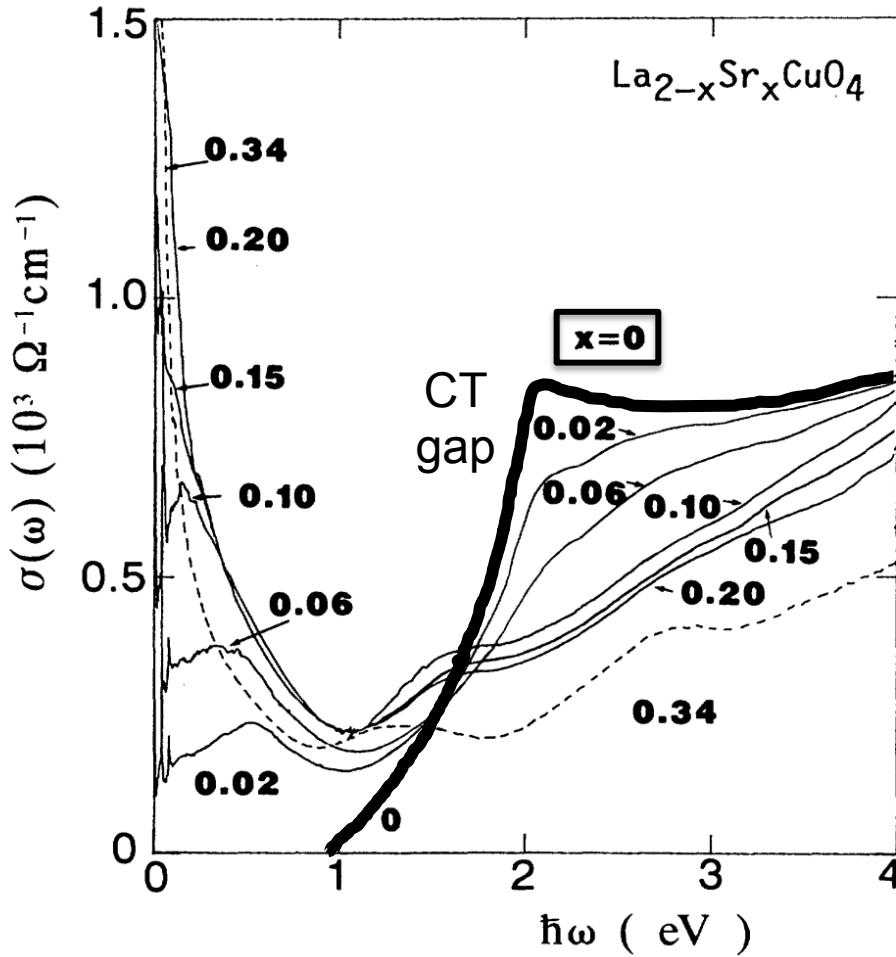
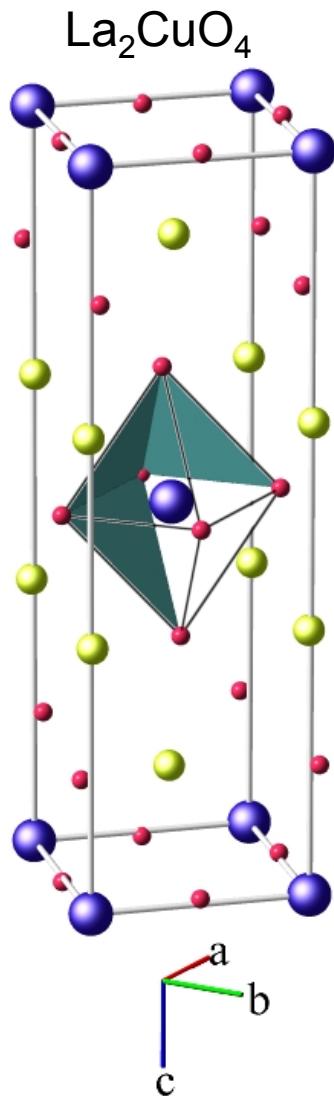
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Optical properties of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$



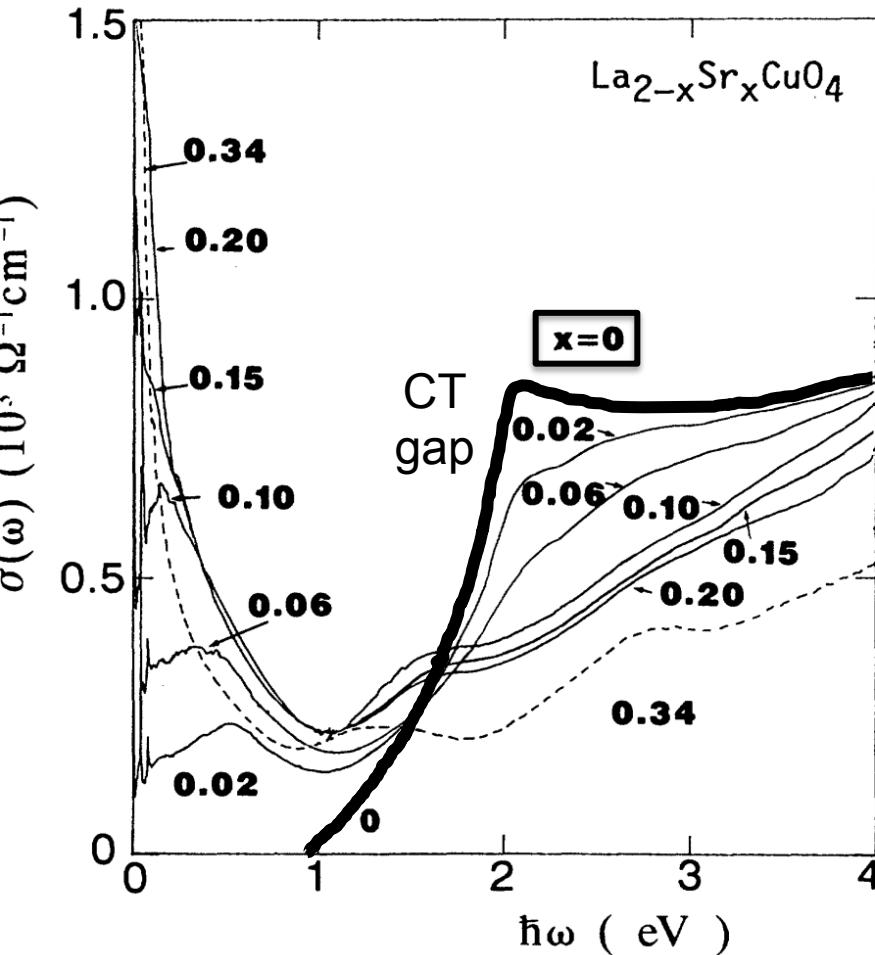
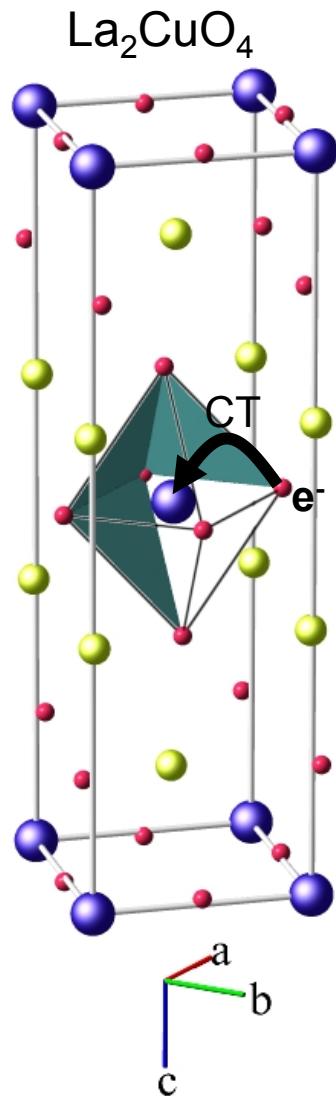
PRB 43 7942 1991

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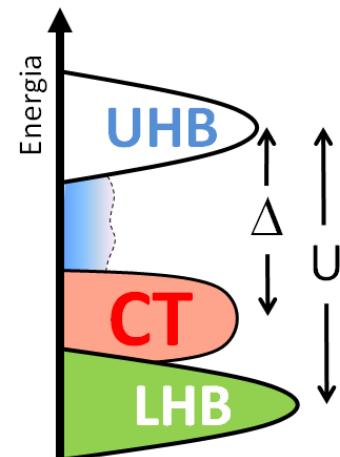


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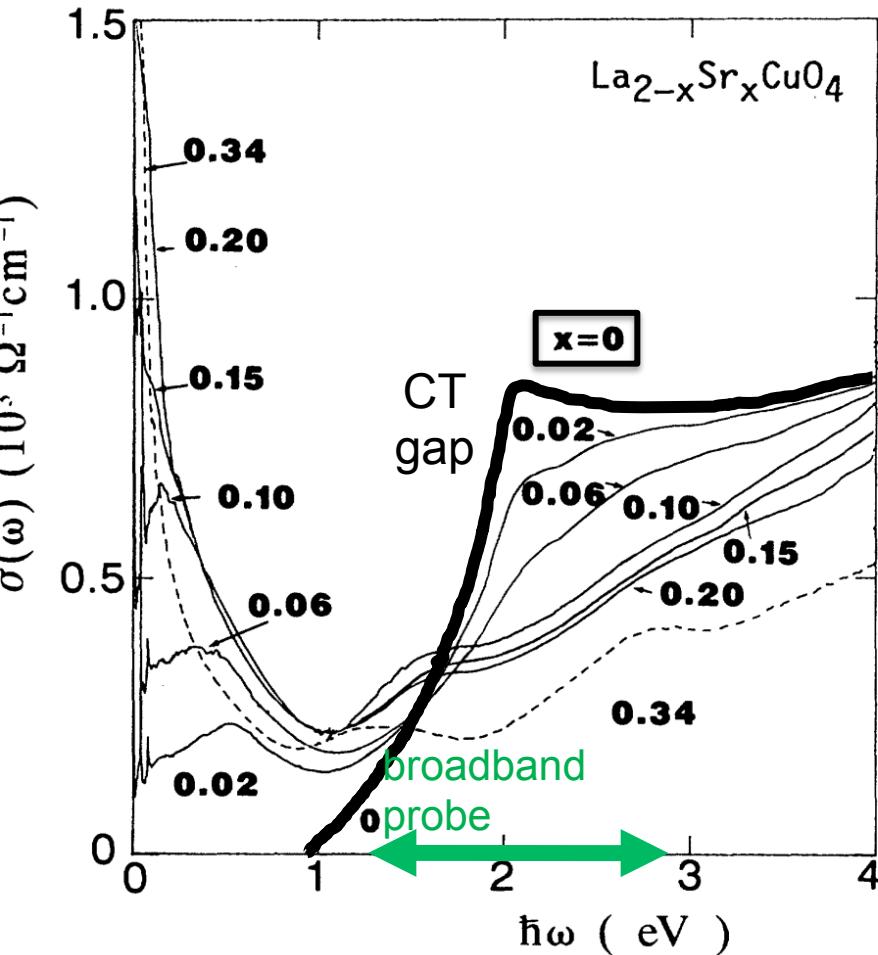
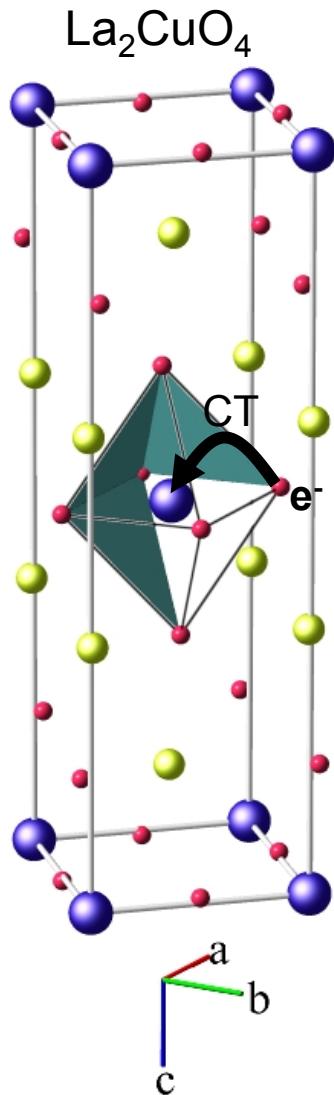
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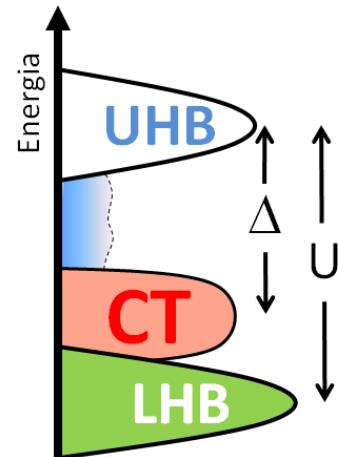
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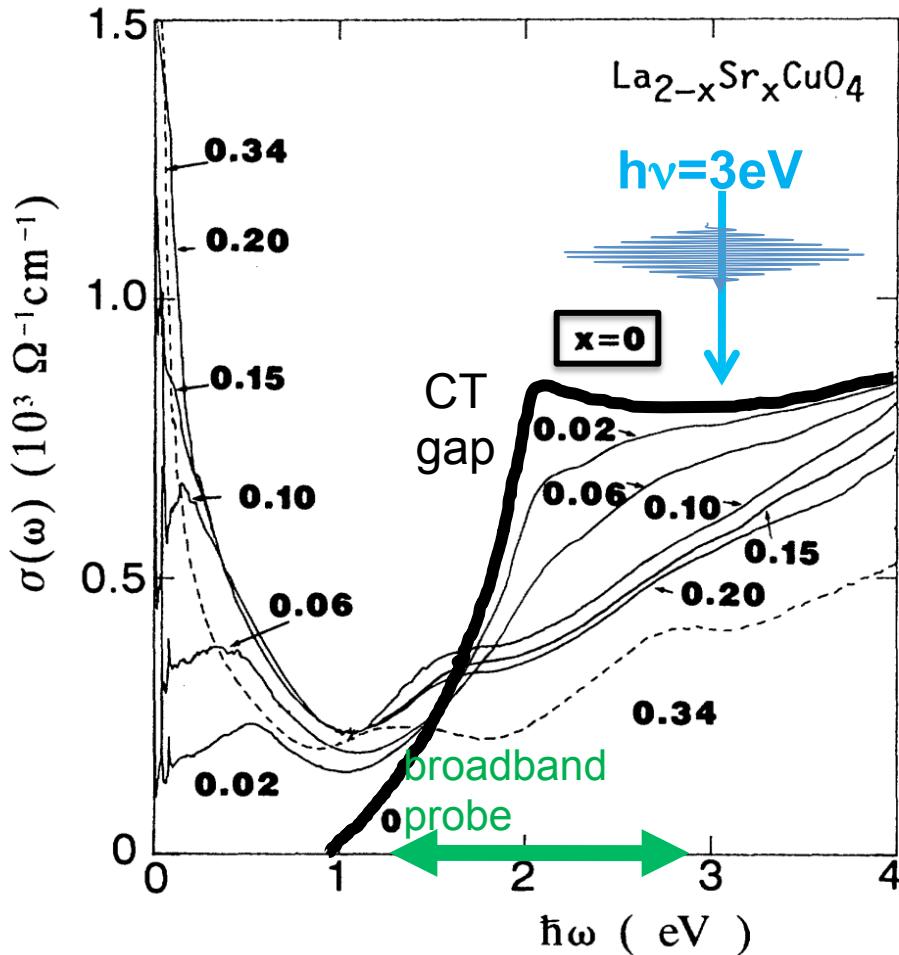
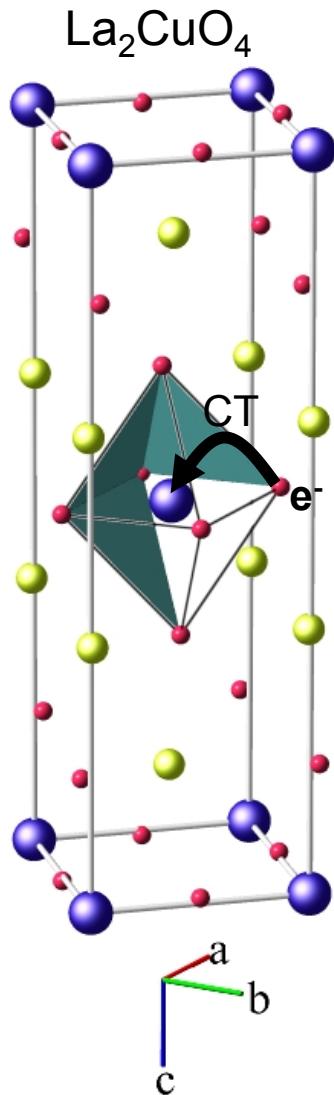
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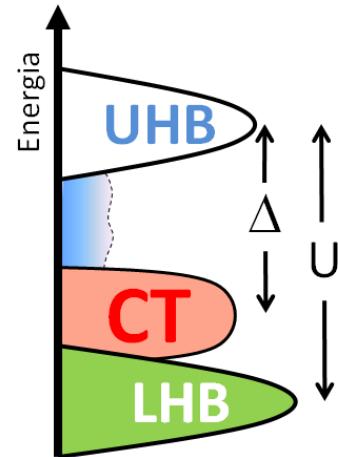
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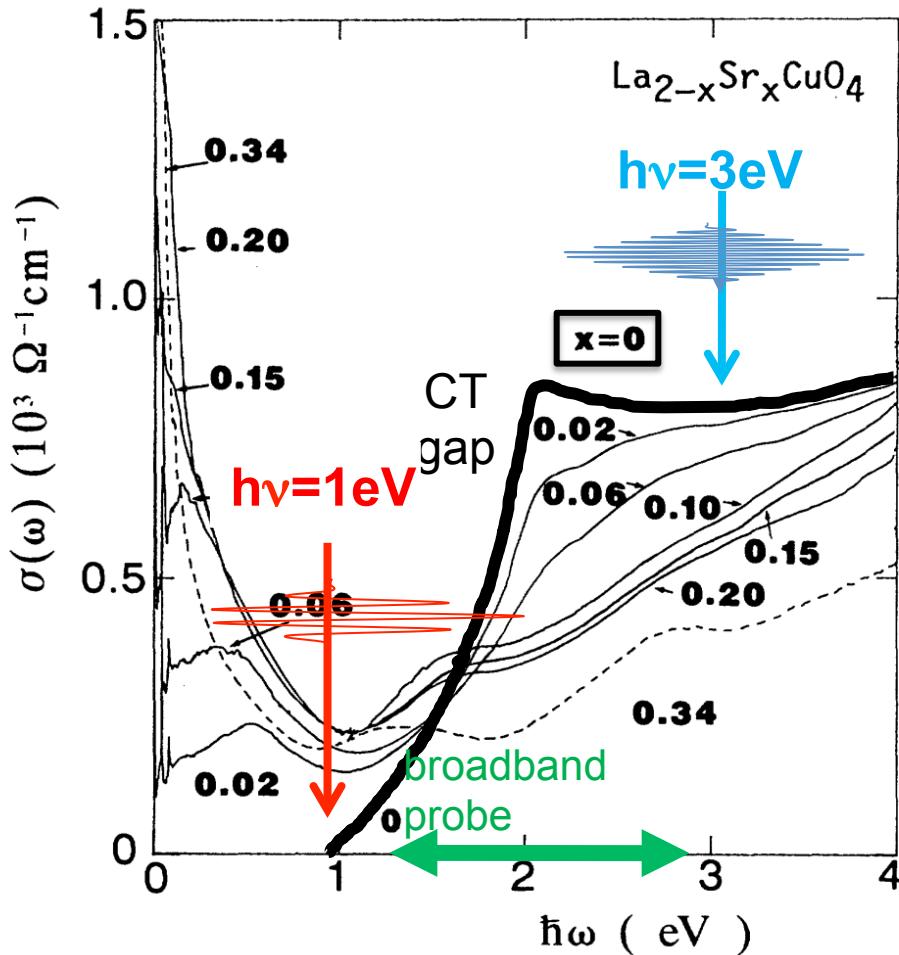
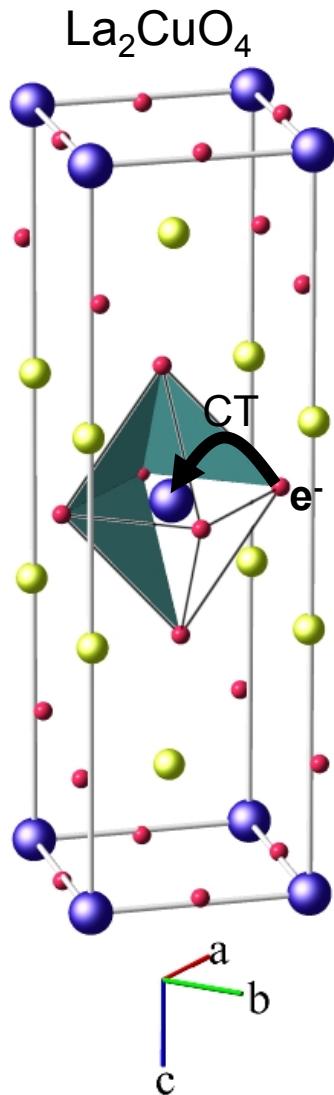
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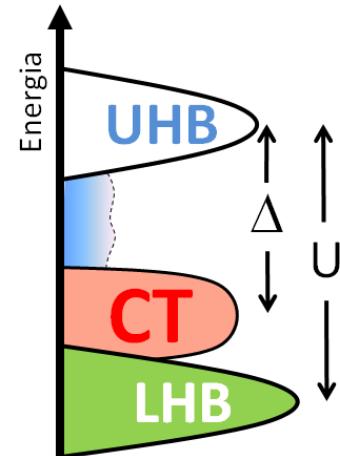
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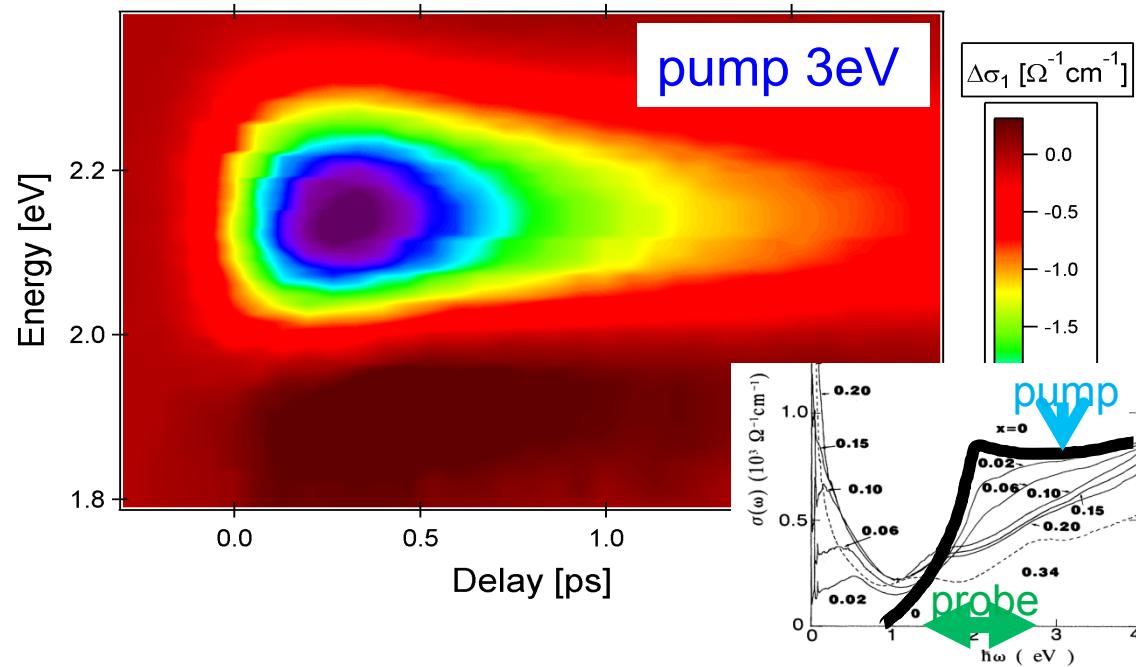
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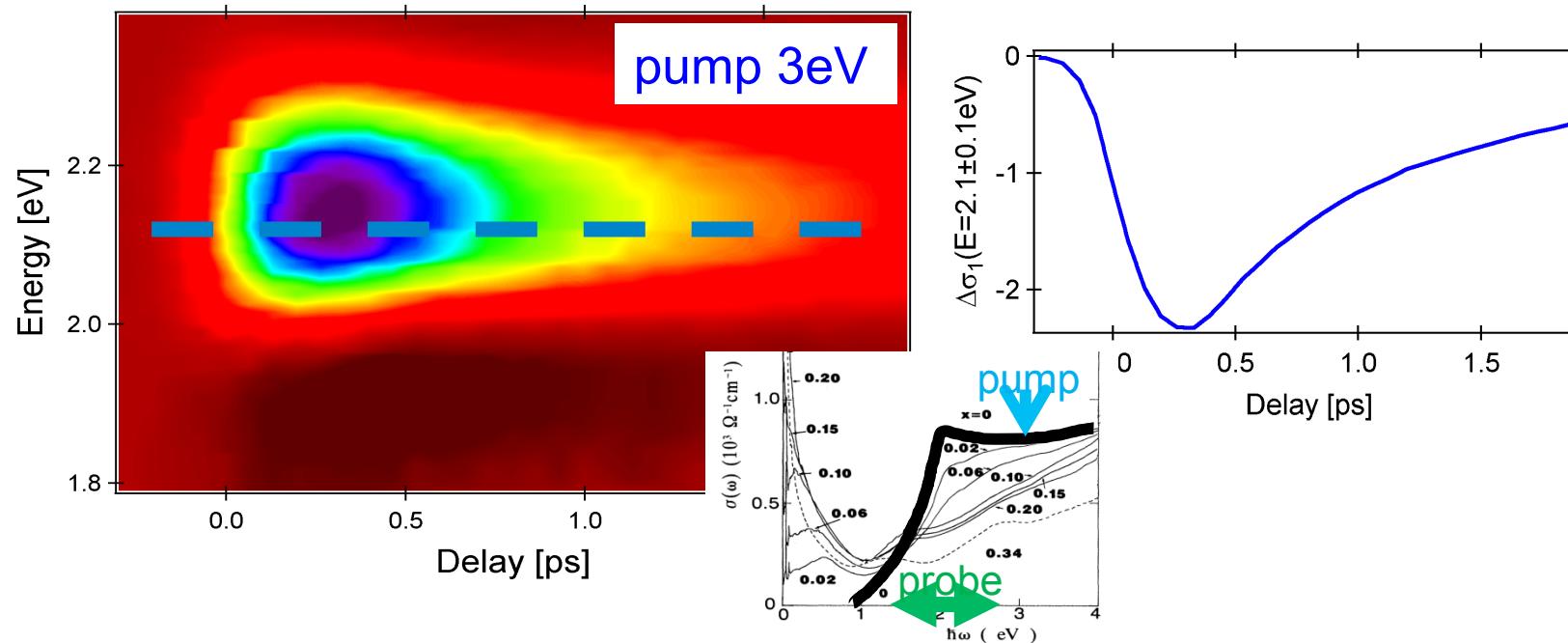
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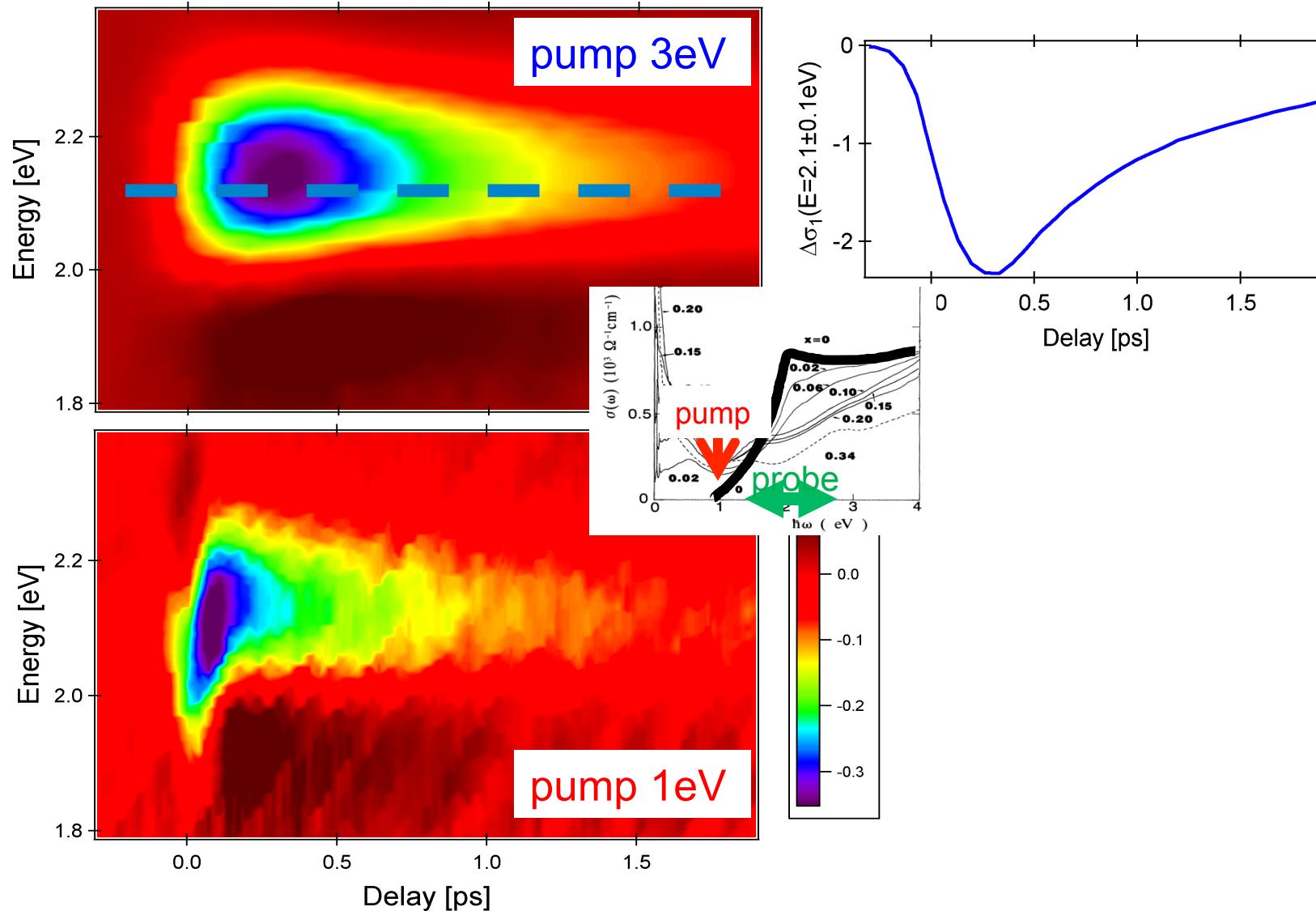
Selectivity of the excitation process



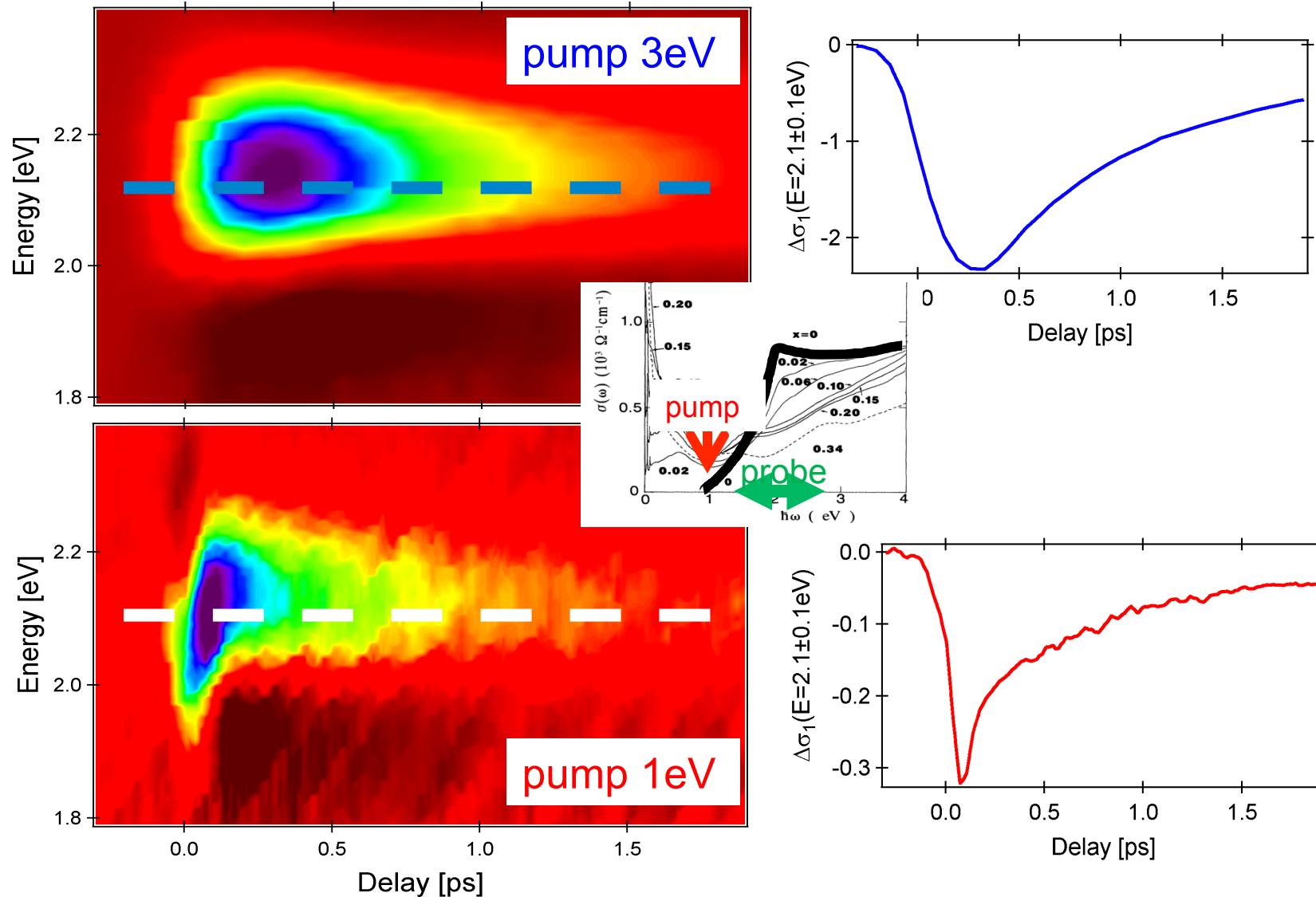
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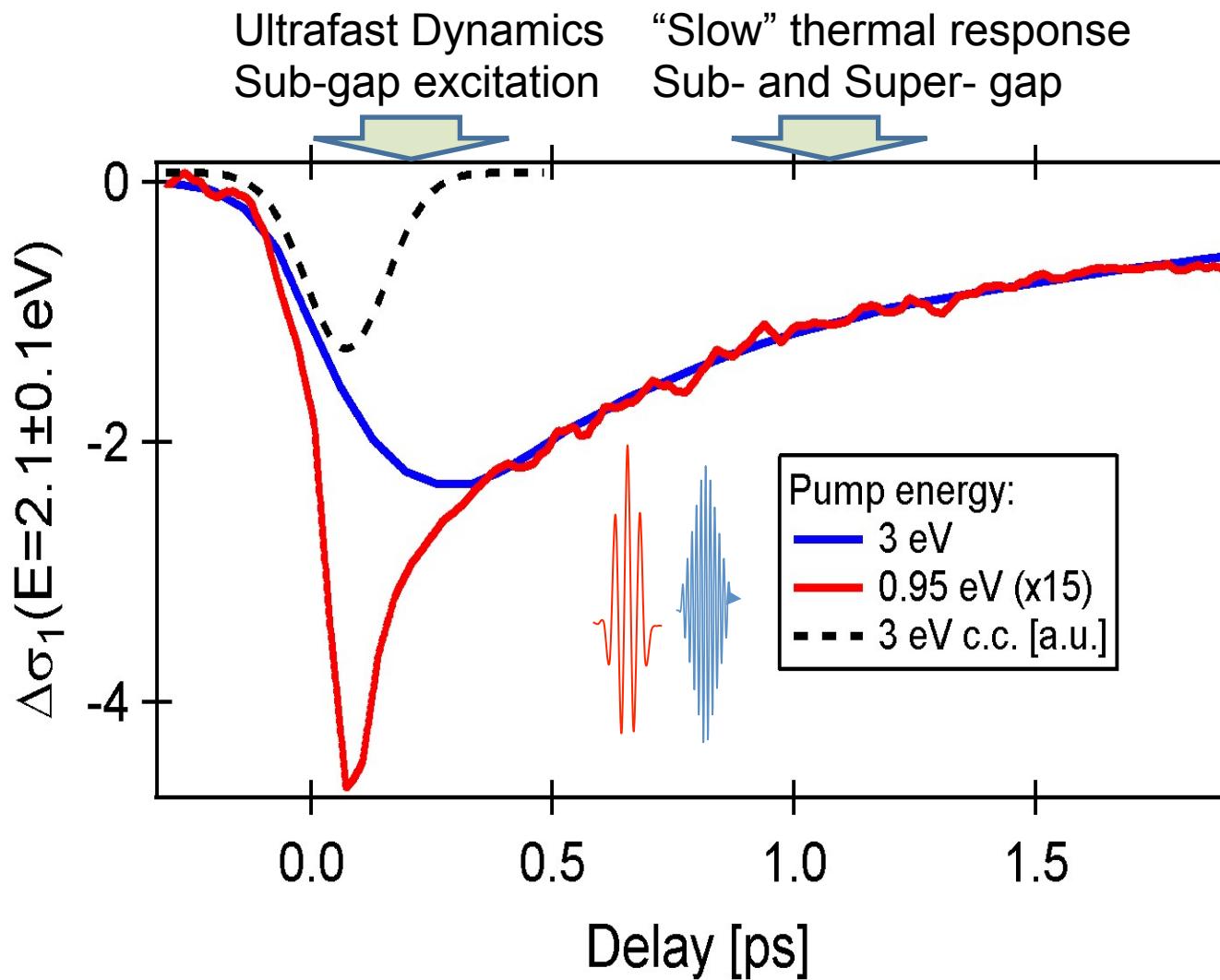
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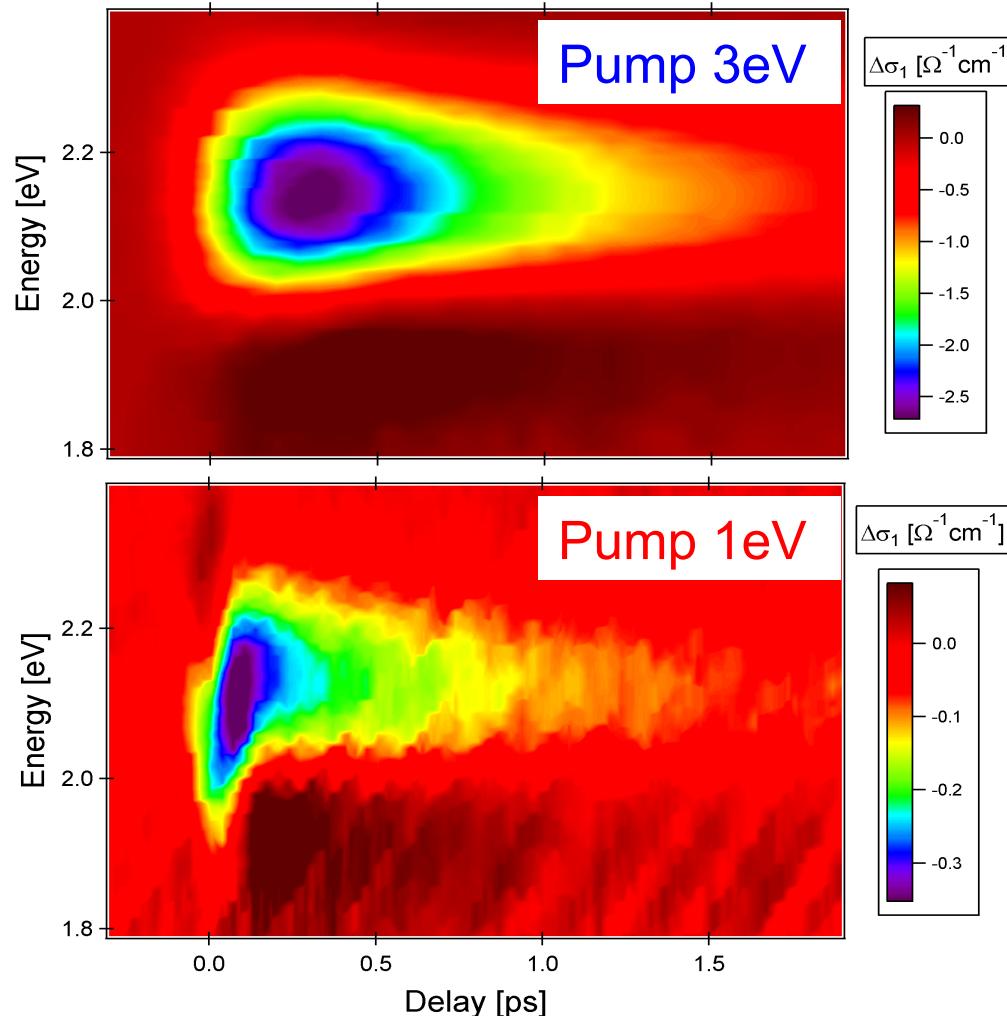
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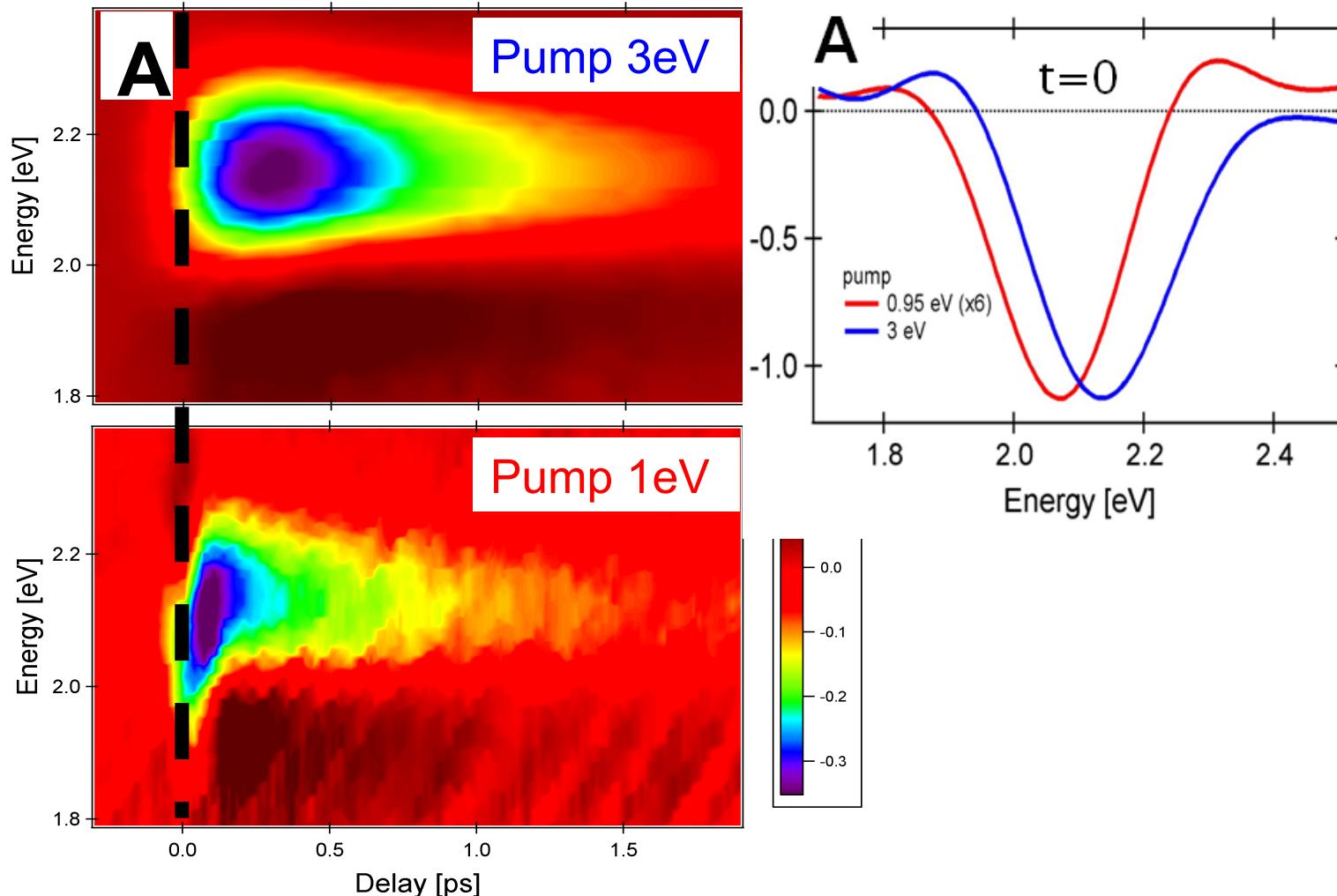
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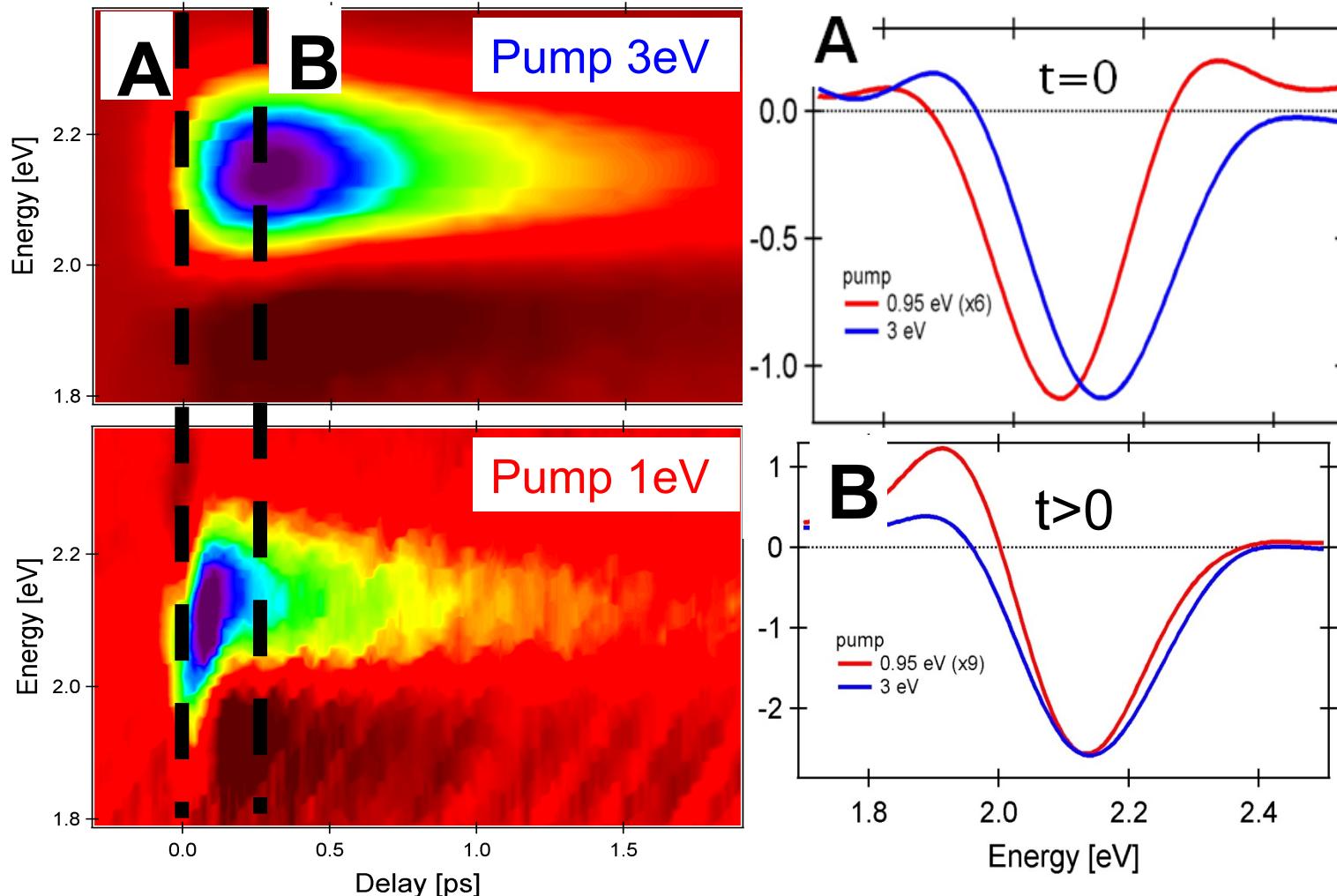
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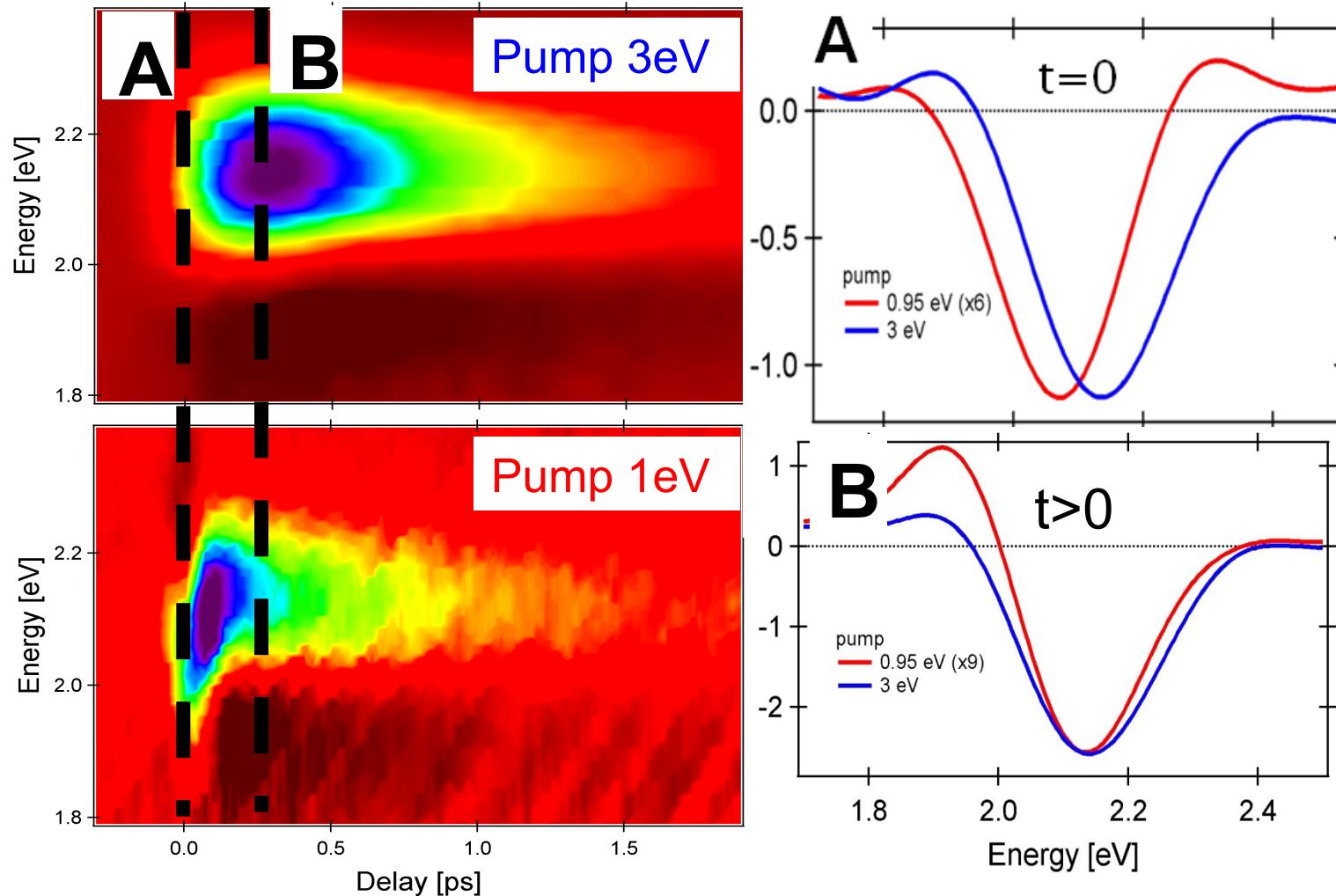
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Hubbard Holstein Hamiltonian

$$H = H_t + H_U + H_{EPI}$$

$$H_t = -t \sum_{i,\mu,\sigma} (c_{i+\mu,\sigma}^\dagger c_{i,\sigma} + H.c.),$$

Hopping

$$H_U = U \sum_i (n_{i,\uparrow} - \frac{1}{2})(n_{i,\downarrow} - \frac{1}{2}),$$

e-e repulsion

$$H_{EPI} = \omega_0 \sum_i a_i^\dagger a_i + g\omega_0 \sum_i (a_i^\dagger + a_i)(1 - n_i).$$

Boson coupling

collaboration with N. Nagaosa, A.
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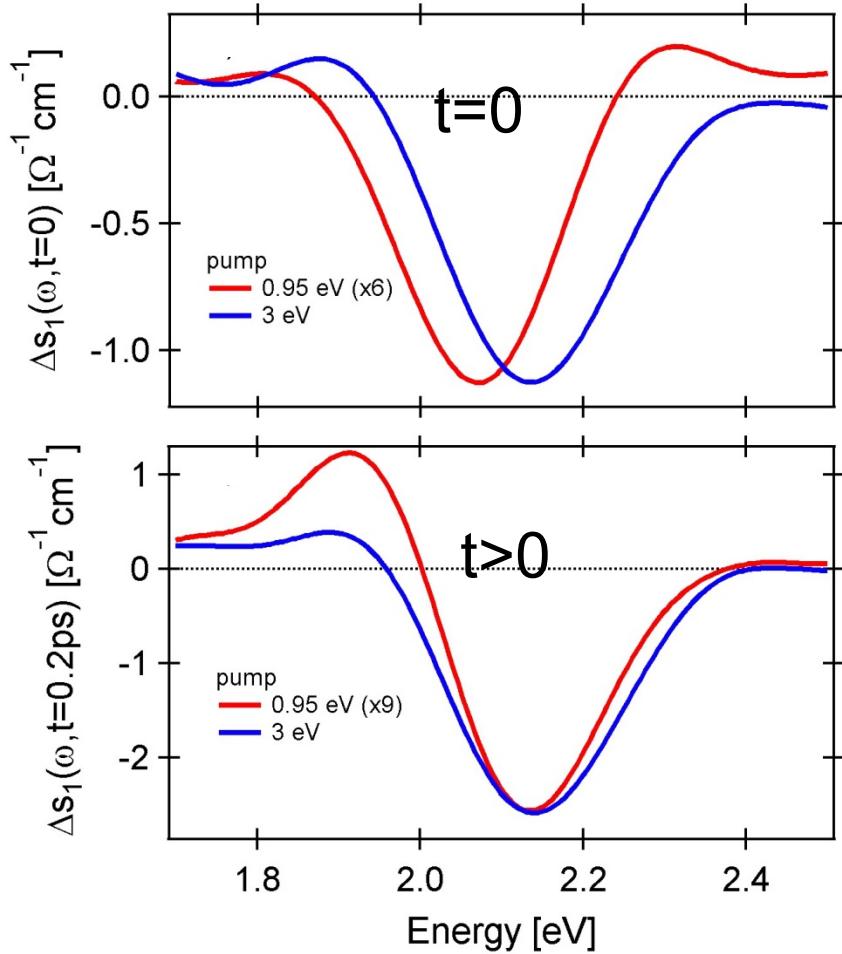
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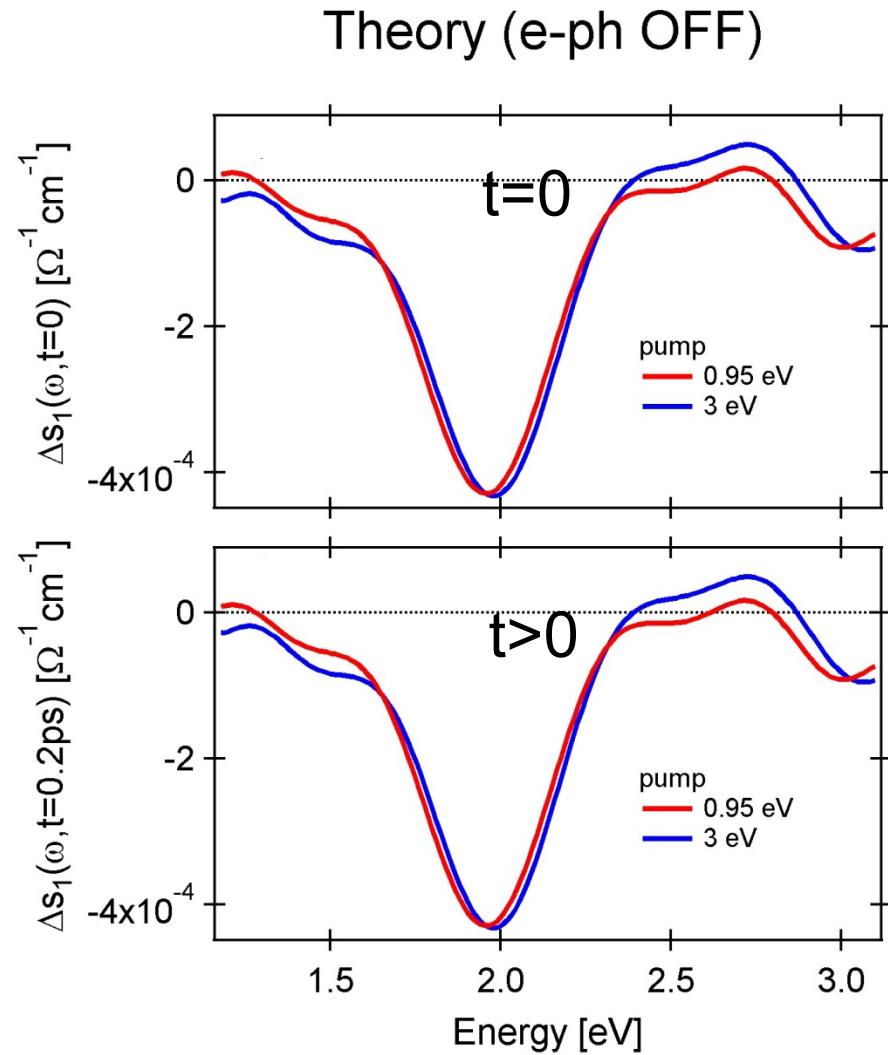
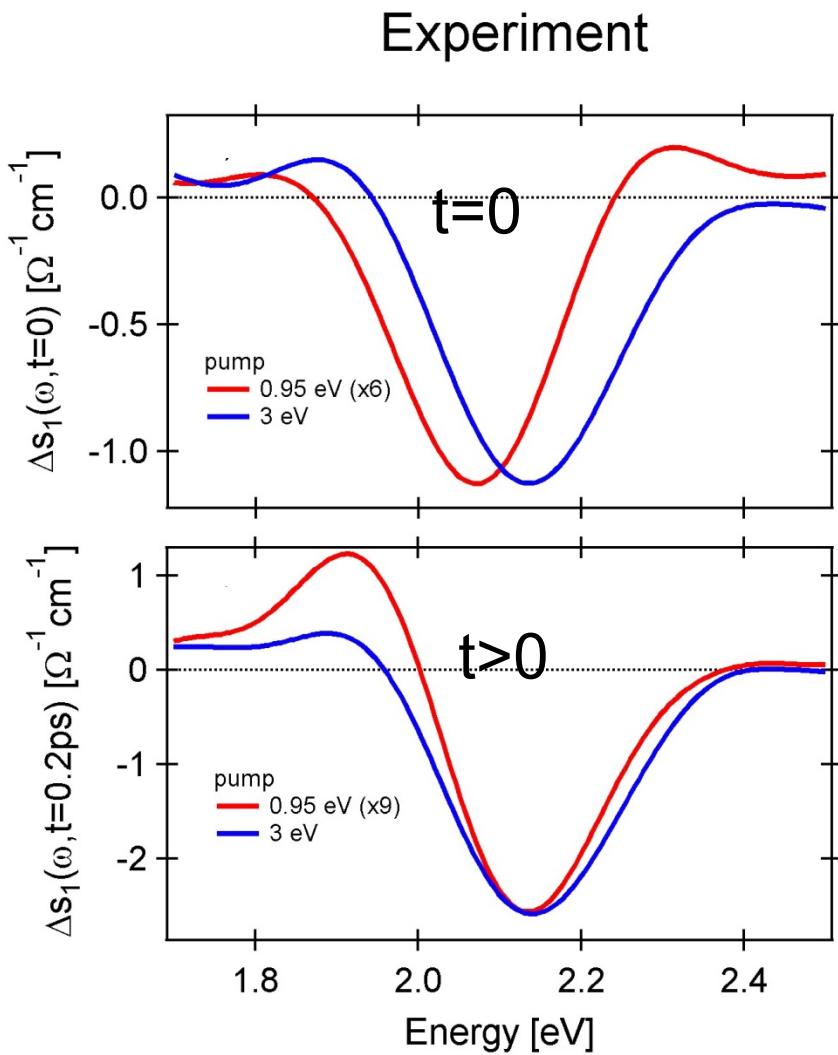
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Experiment Vs. Theory

Experiment

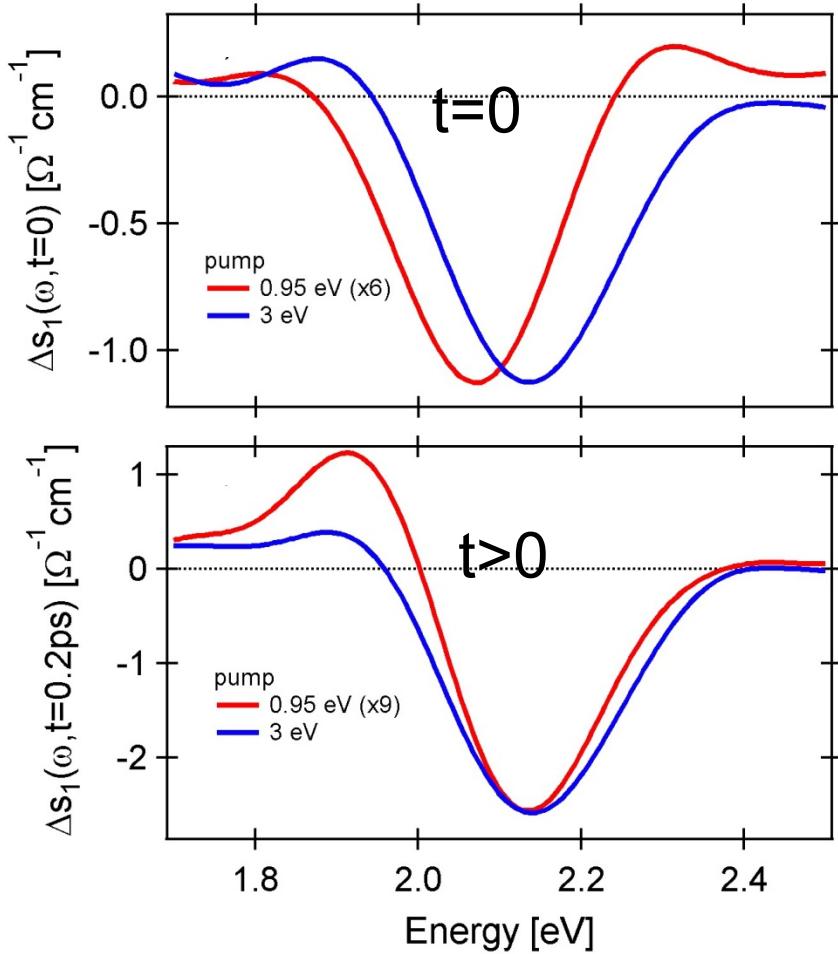


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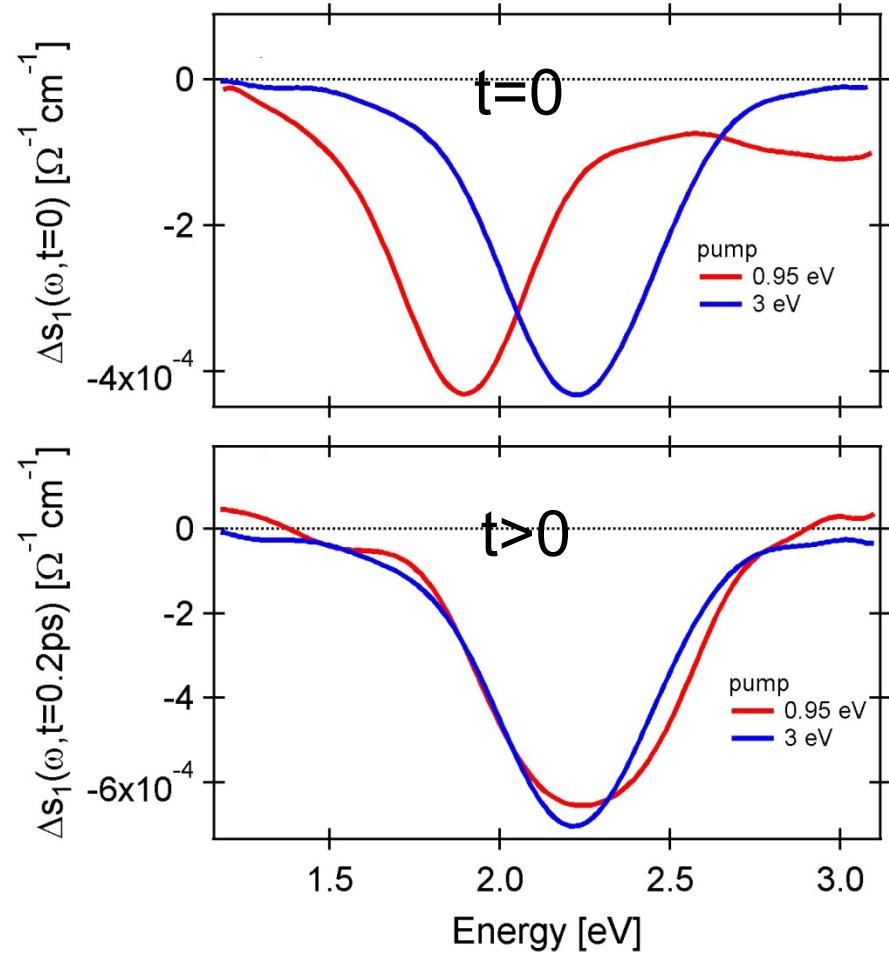


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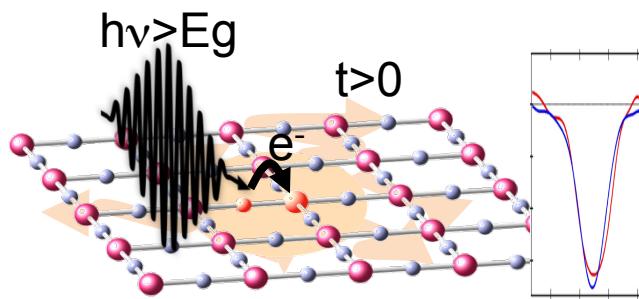
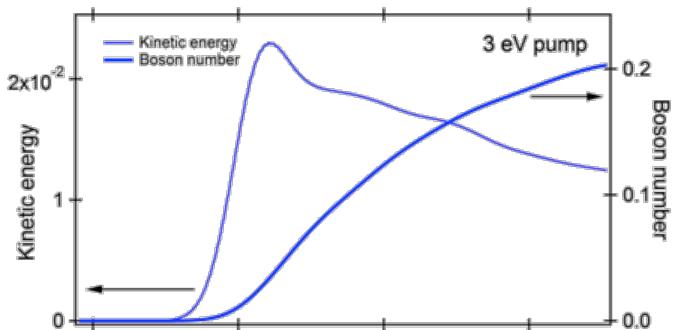
Experiment



Theory (e-ph ON)

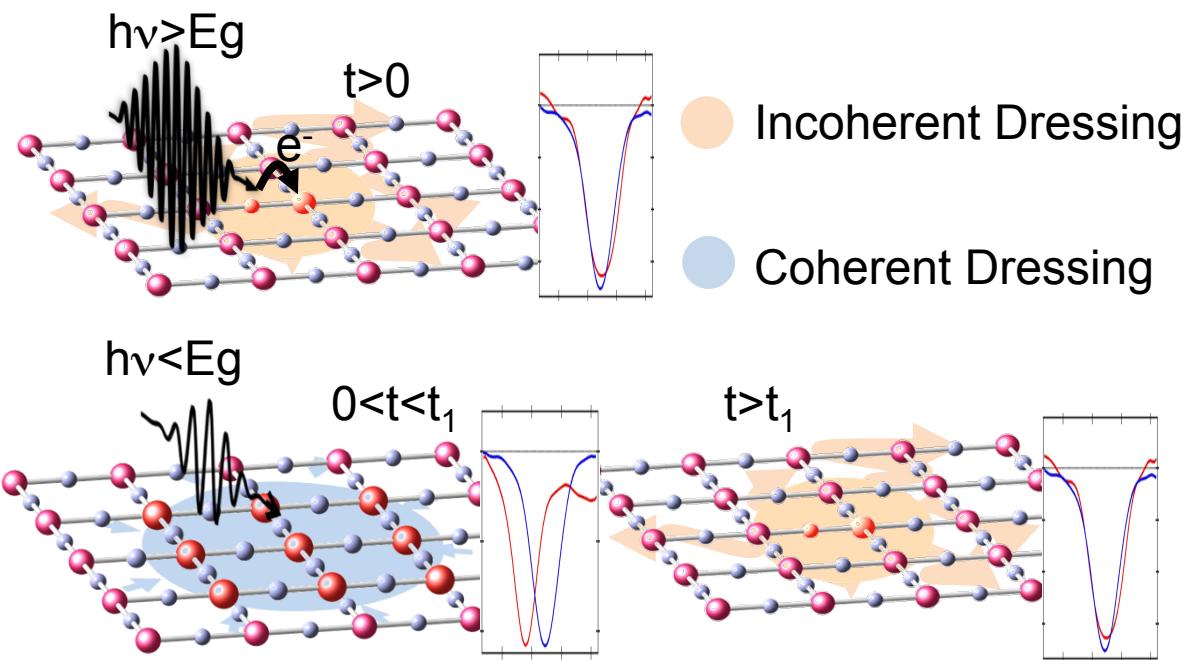
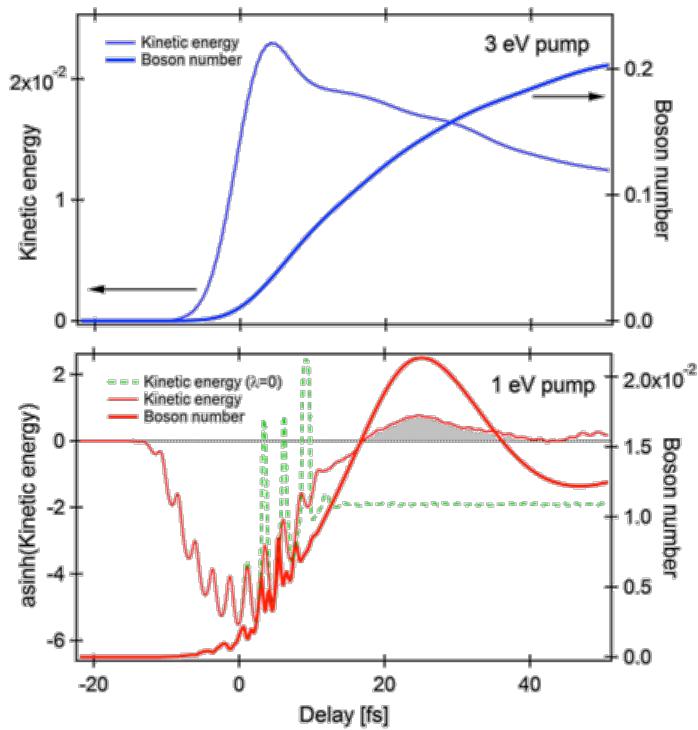


High Vs. Low-photon energy excitation

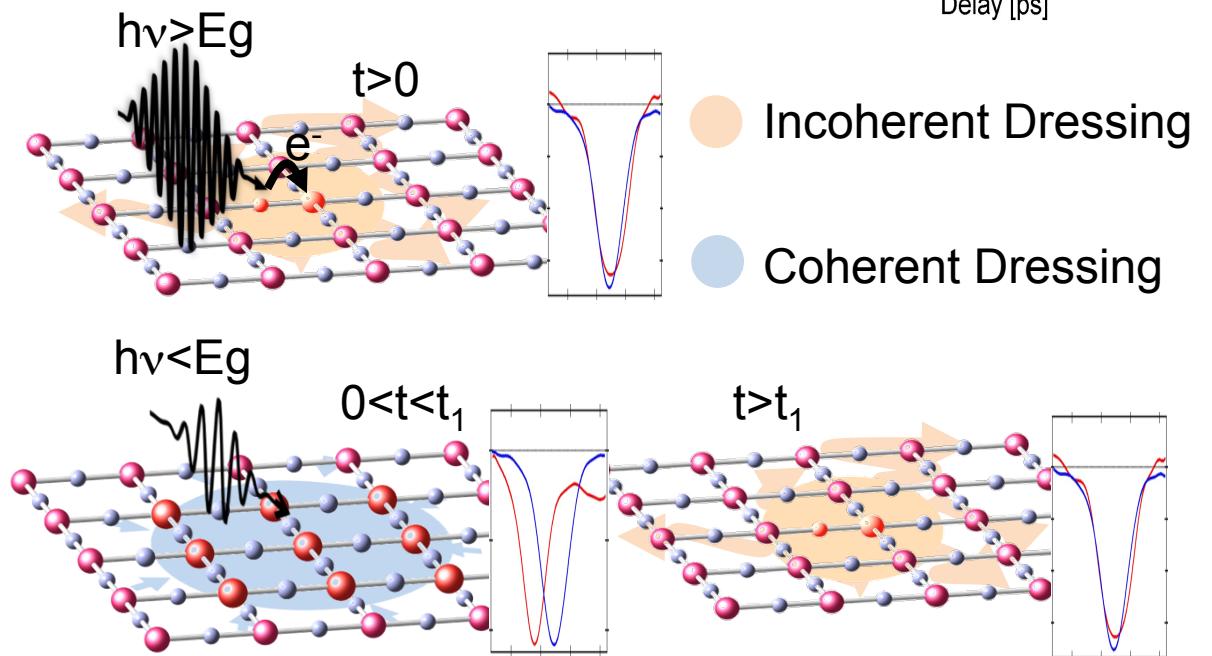
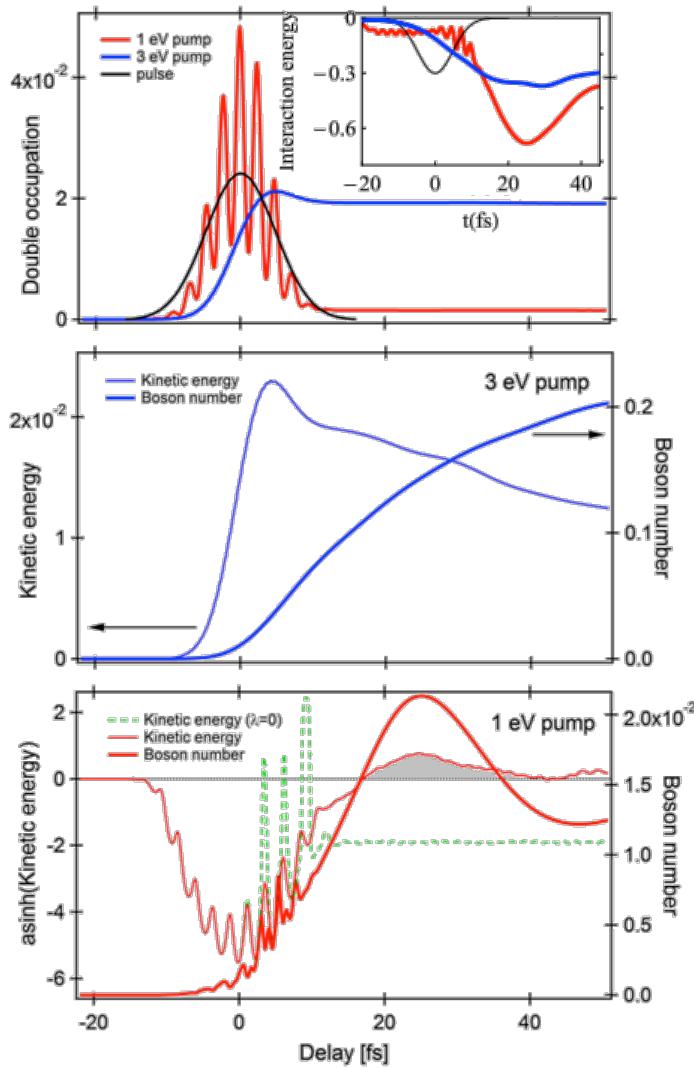


● Incoherent Dressing

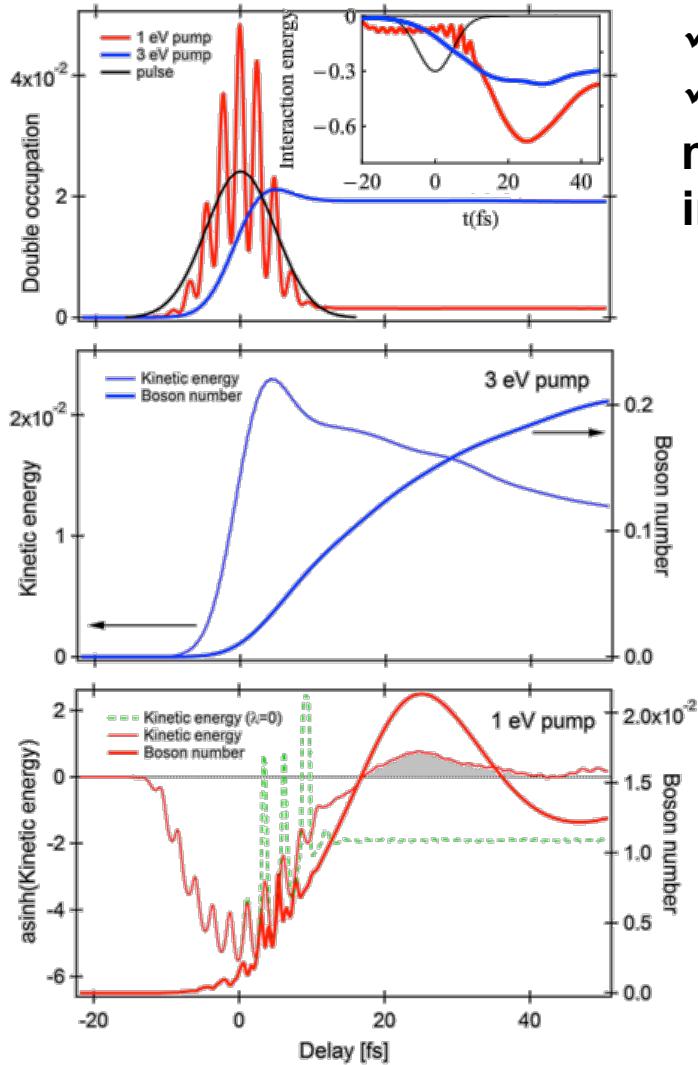
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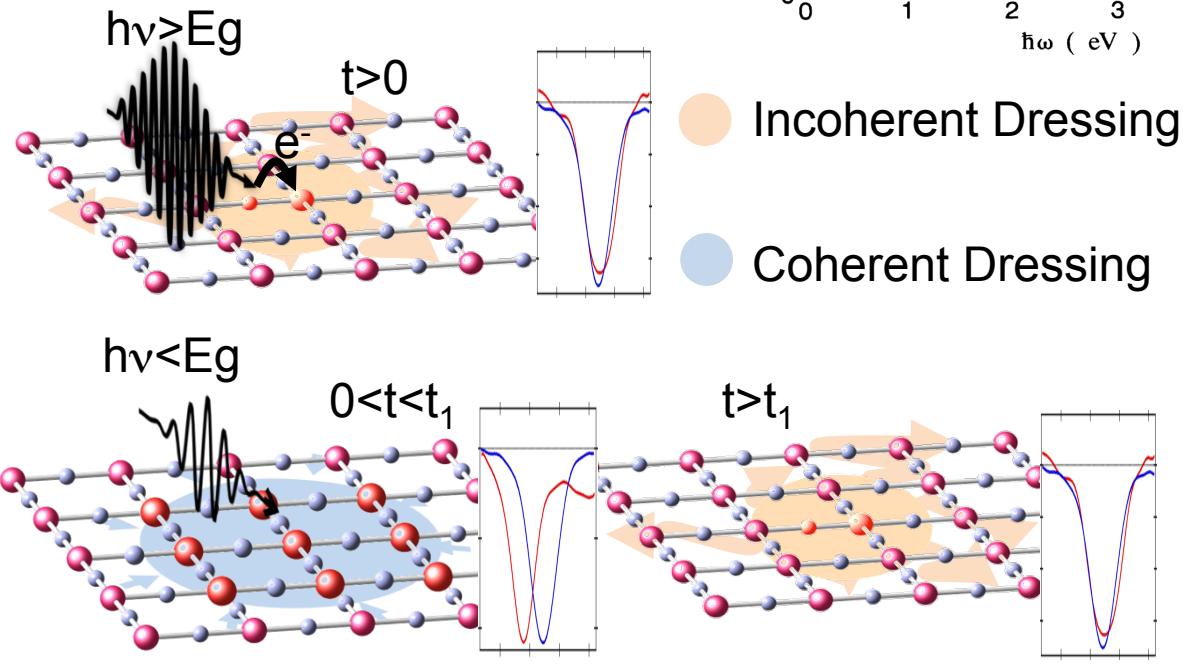
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High Vs. Low-photon energy excitation



- ✓ The boson is necessary
- ✓ Will this excitation mechanism influence response in conducting systems?



Nat. Comm. 5, 5112, 2014

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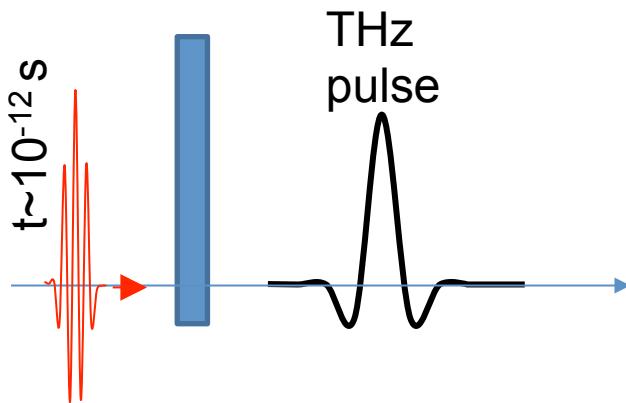
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Generation and measurements of ultrashort THz pulse

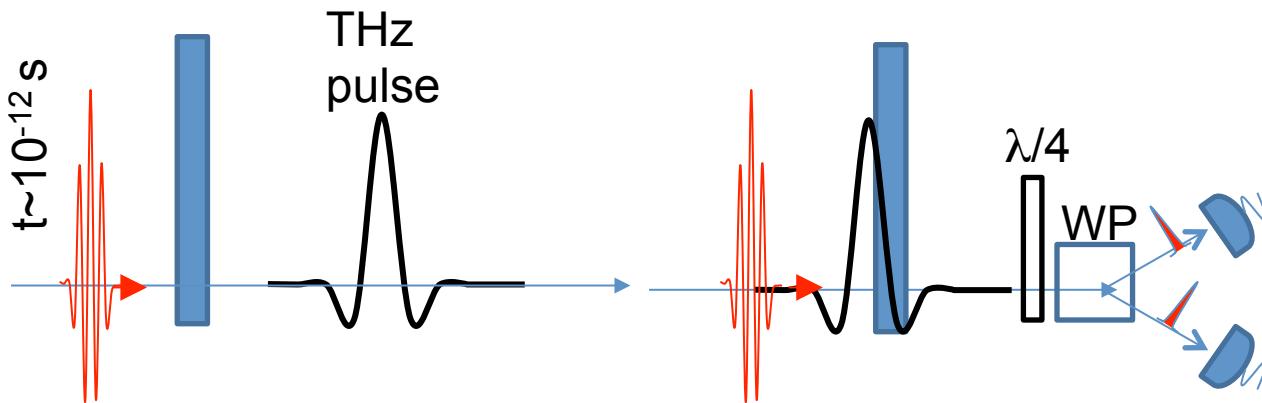
Generation:
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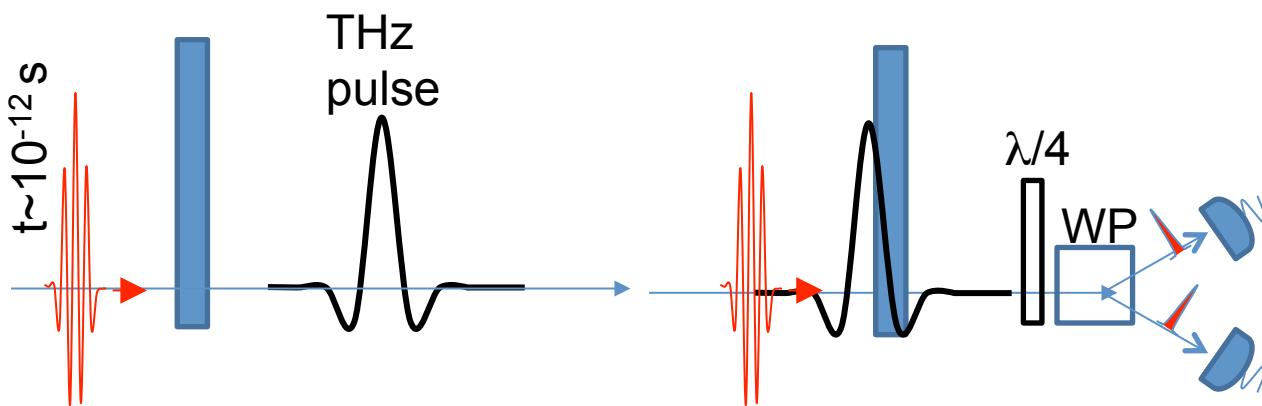
Generation:
Optical Rectification

Measurements:
Electro optic sampling

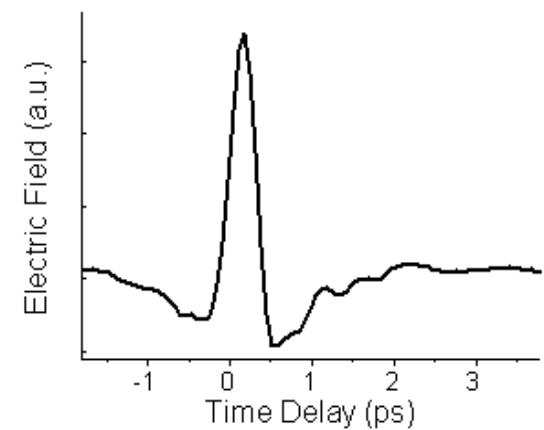


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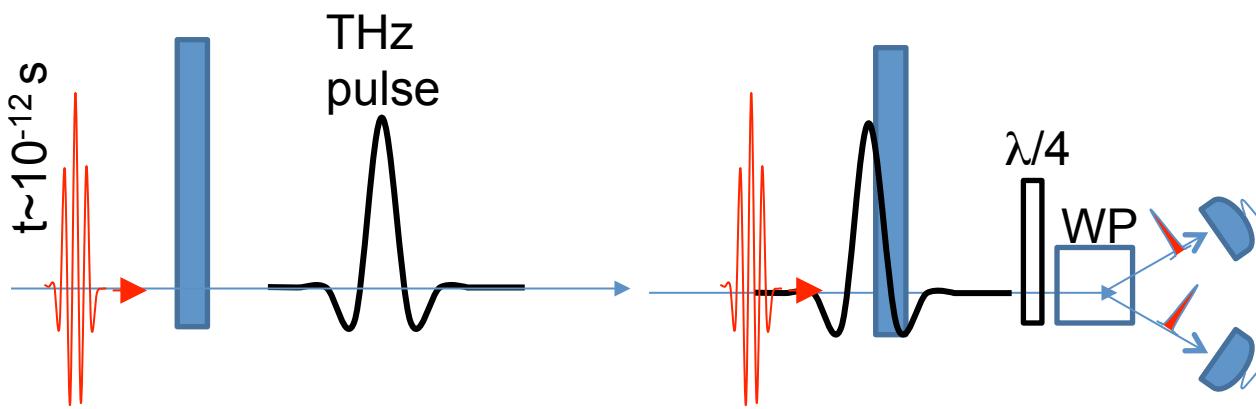


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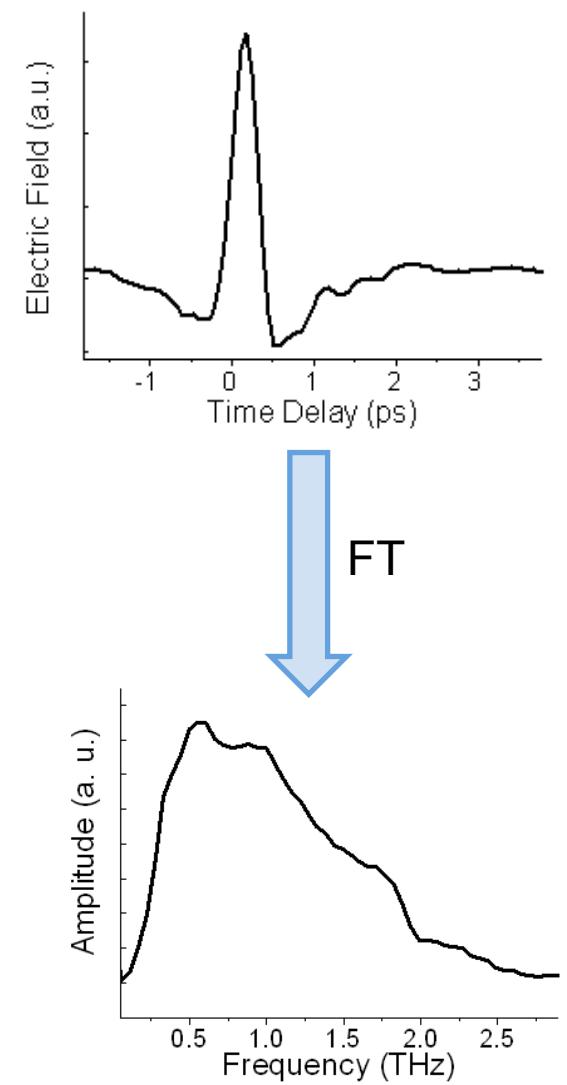


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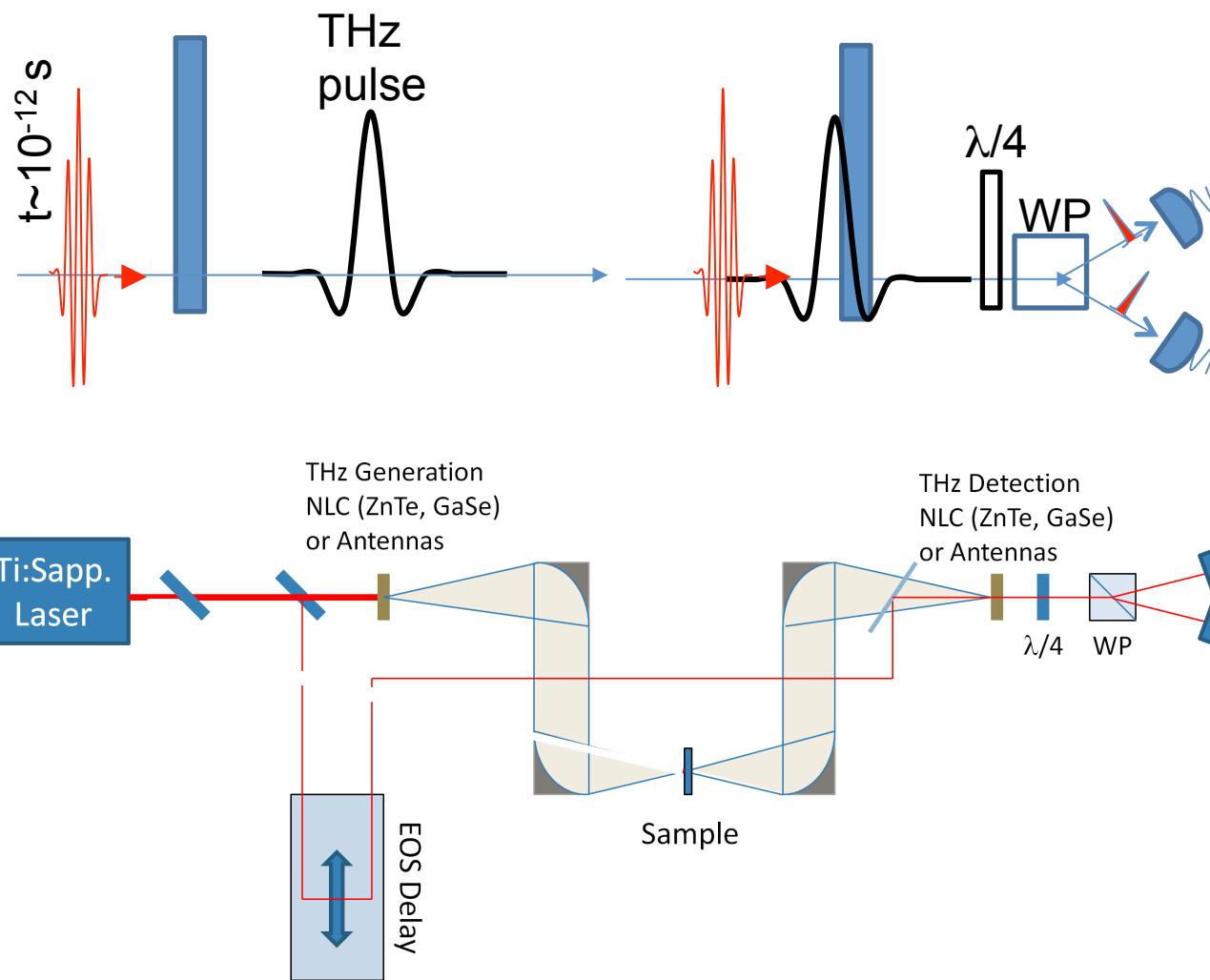


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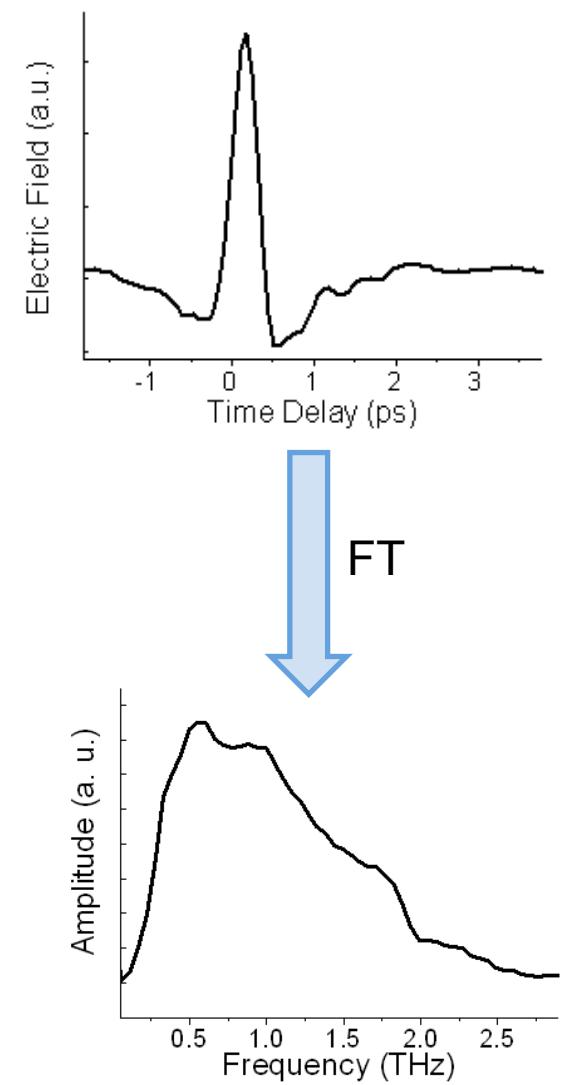


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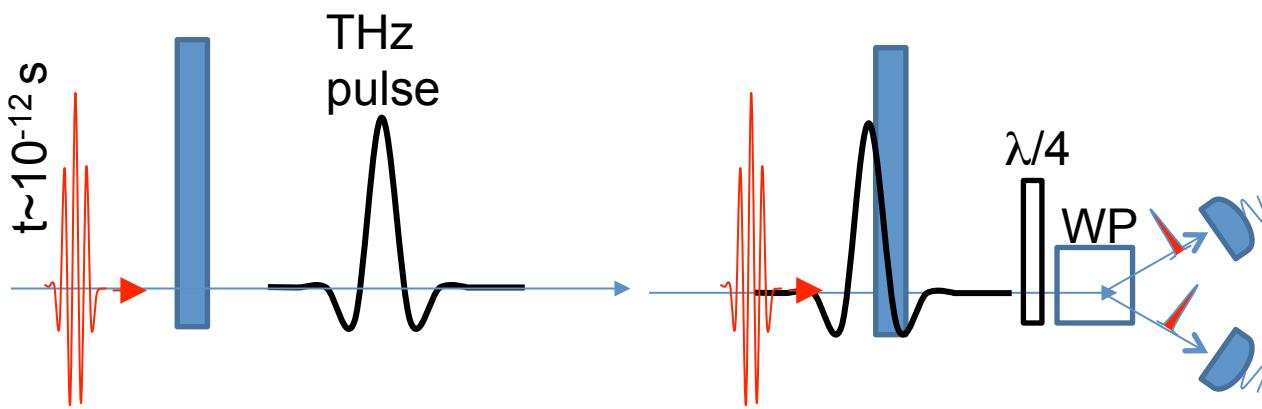


Measurements: Electro optic sampling

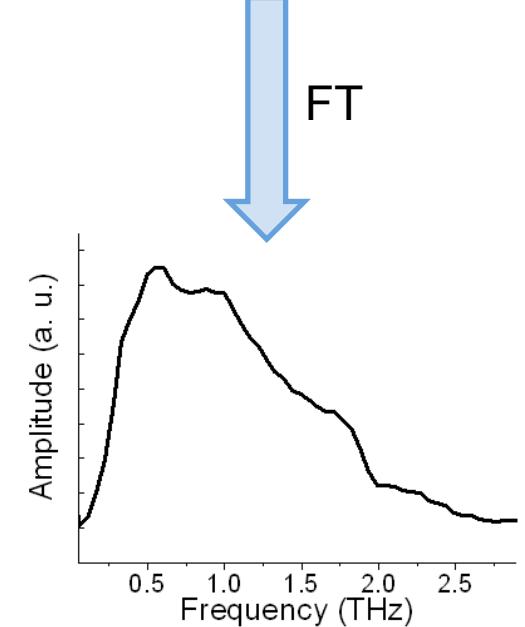
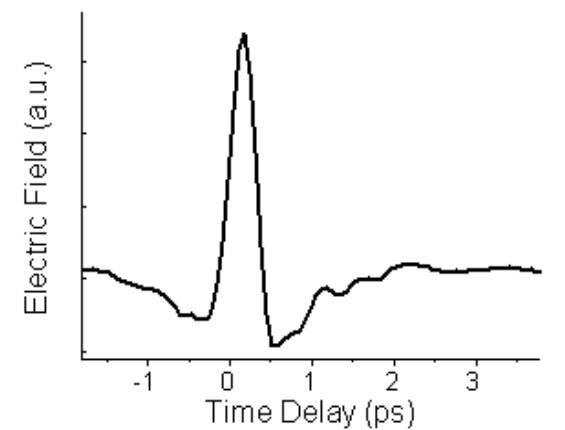


Generation and measurements of ultrashort THz pulse

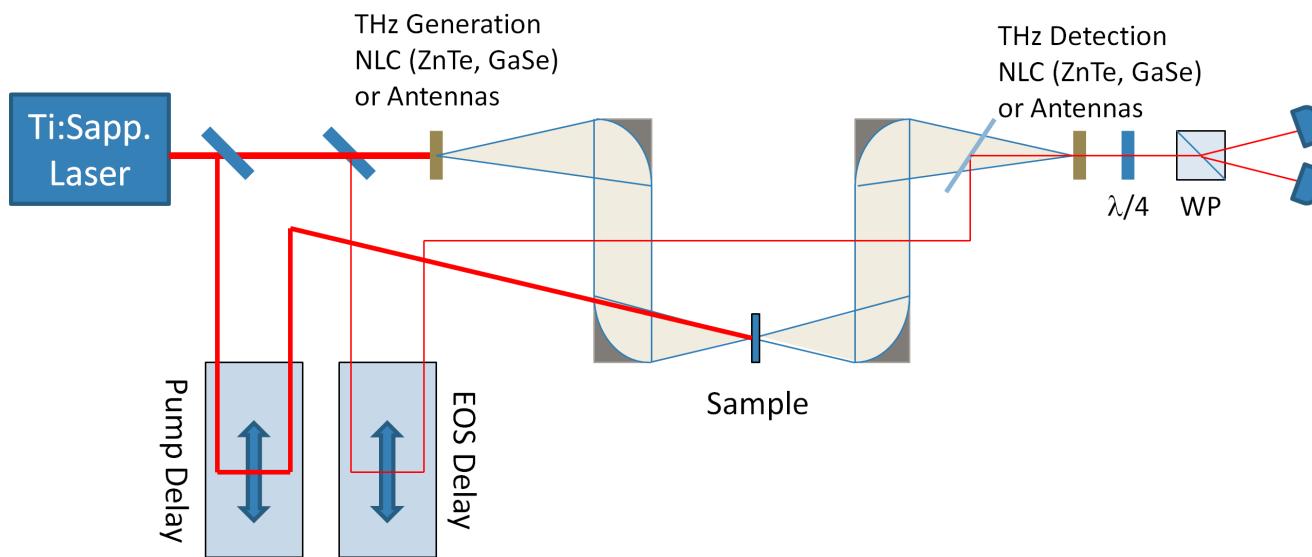
Generation: Optical Rectification



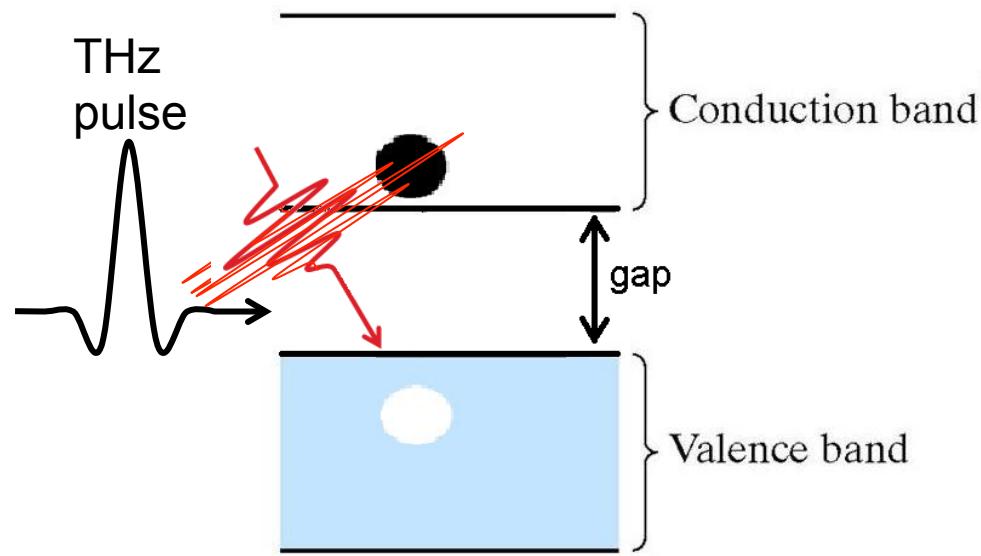
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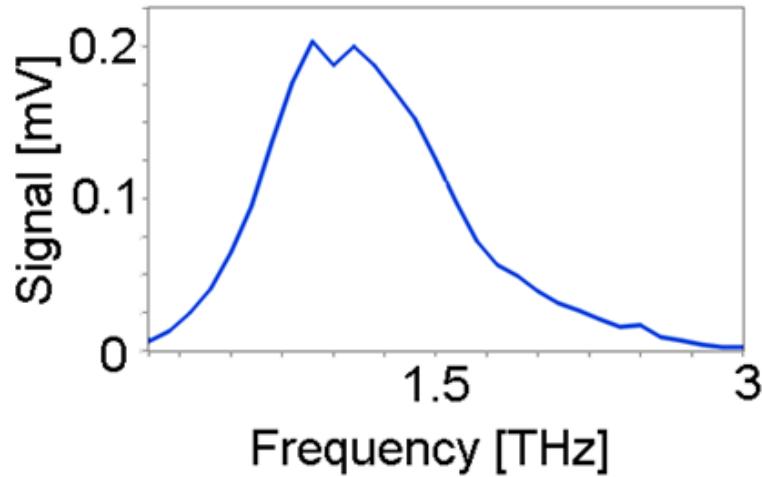
FT



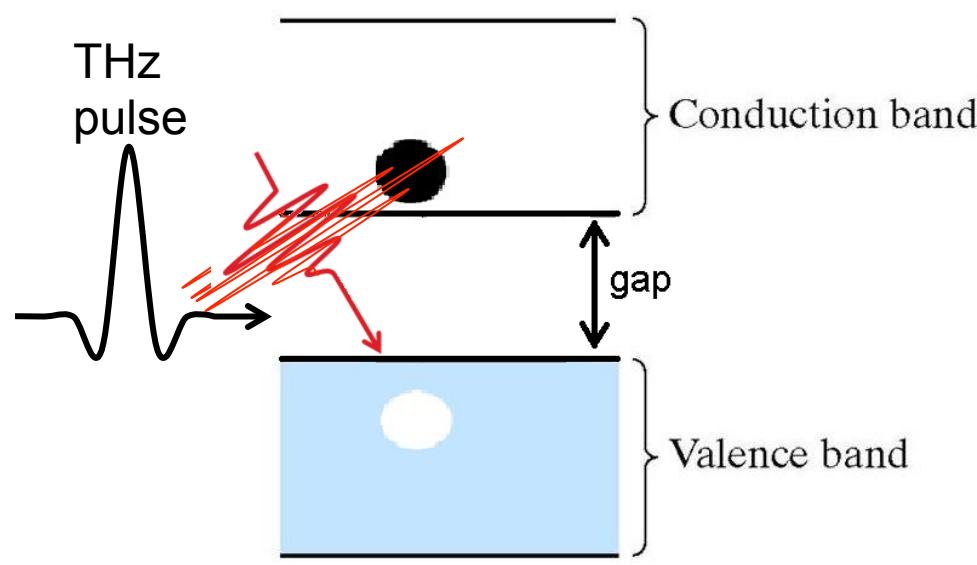
Test examples: Transmission on silicon



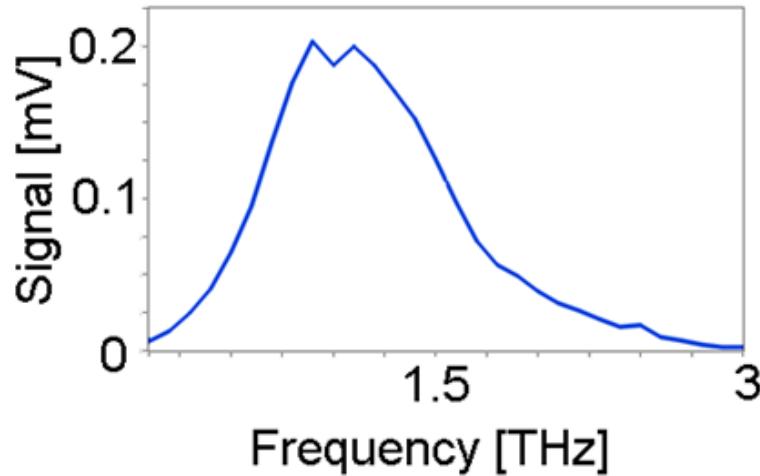
Equilibrium Transmission



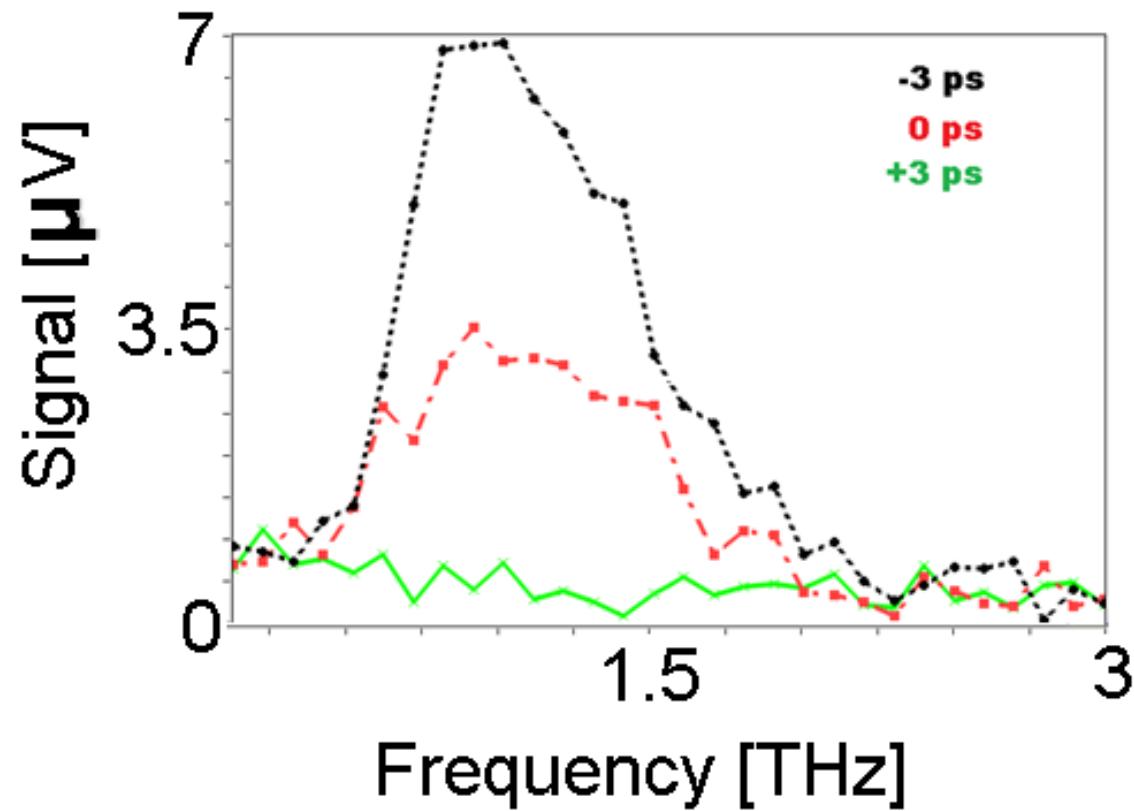
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Equilibrium Transmission



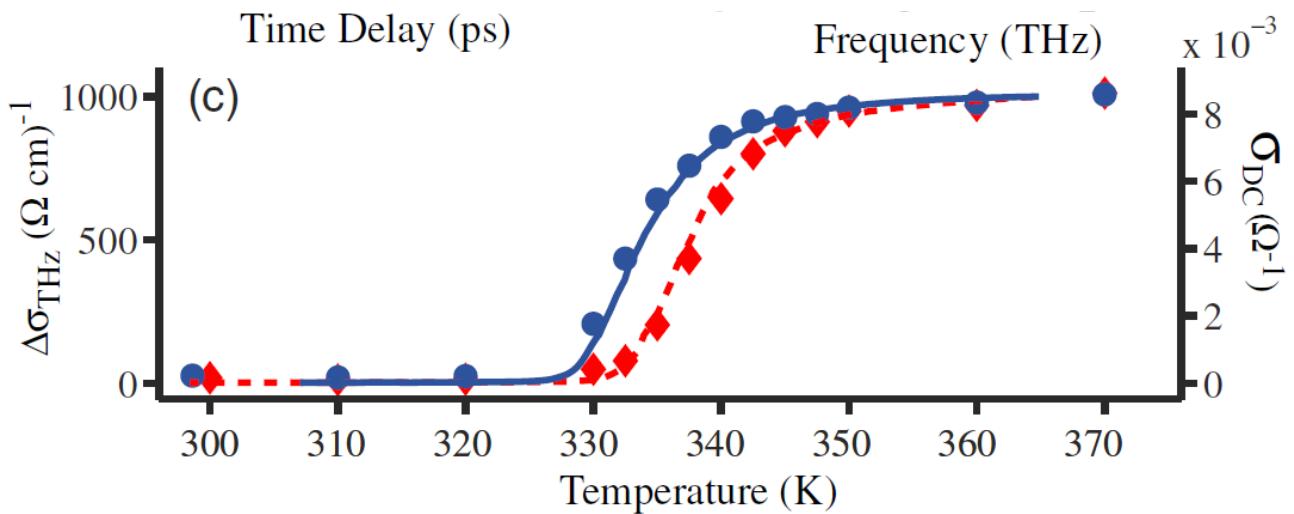
Time domain changes in Transmission



Fabio Novelli Phd Thesis

Metal-insulator transition in VO₂ single crystal

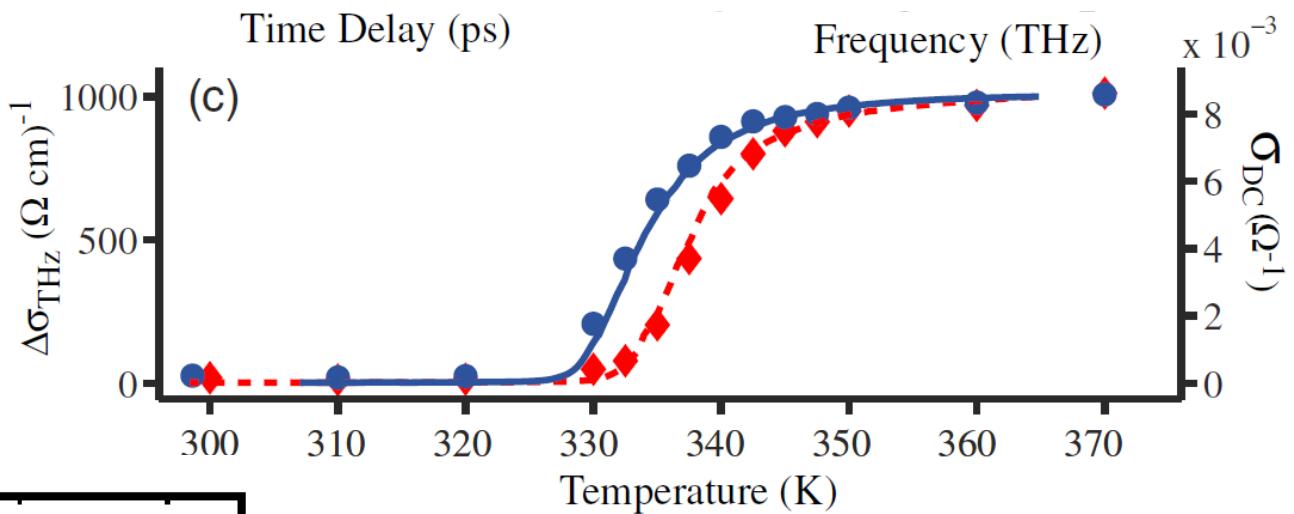
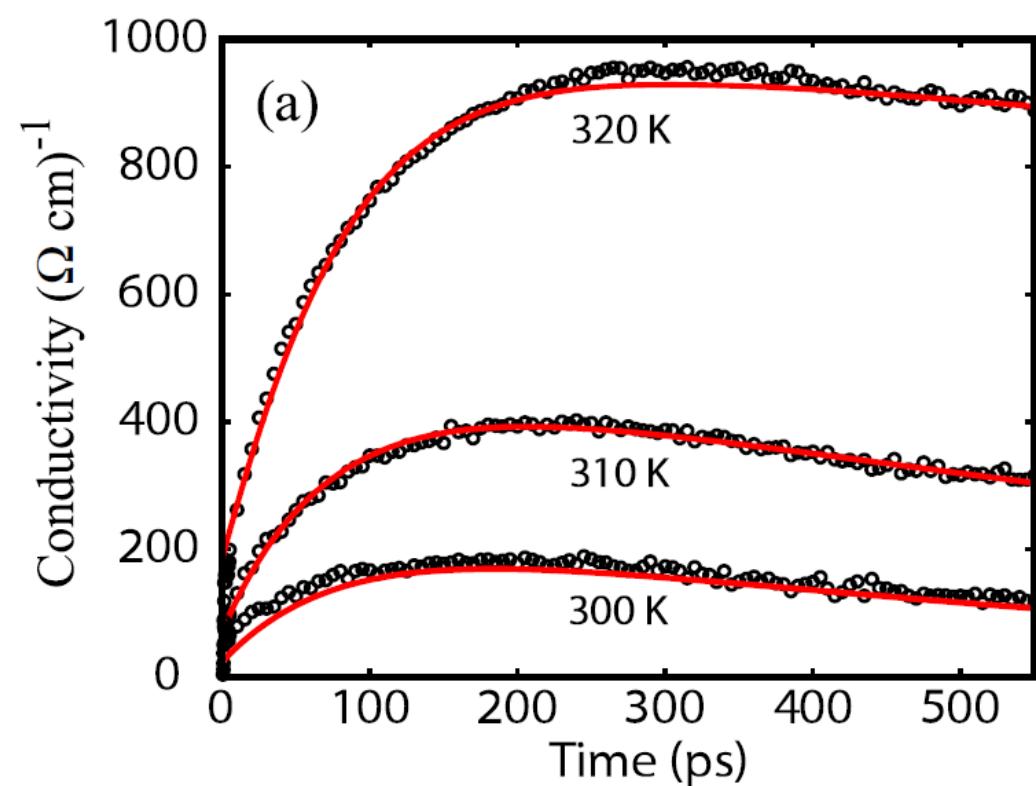
- ✓ Static changes in THz transmittivity
- ✓ Temperature dependence of DC conductivity



PRL, 226401 (2007)

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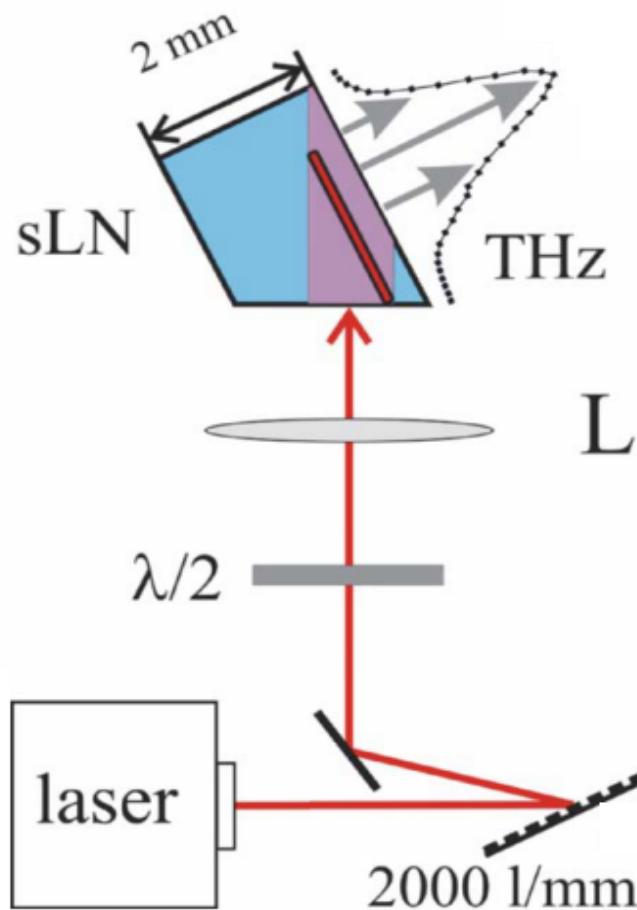
- ✓ Light driven formation of a metallic phase
- ✓ Detail studies of the nucleation processes

PRL, 226401 (2007)

Can we use THz pulses as pump?

Using THz as a pump

The table-top intense THz Source



Zs.Bor and B.Racz, *Opt.Comm.* **54**, 165(1985)
J.Hebling et al., *J.Opt.Soc.Am.B* **25**,B6(2008)

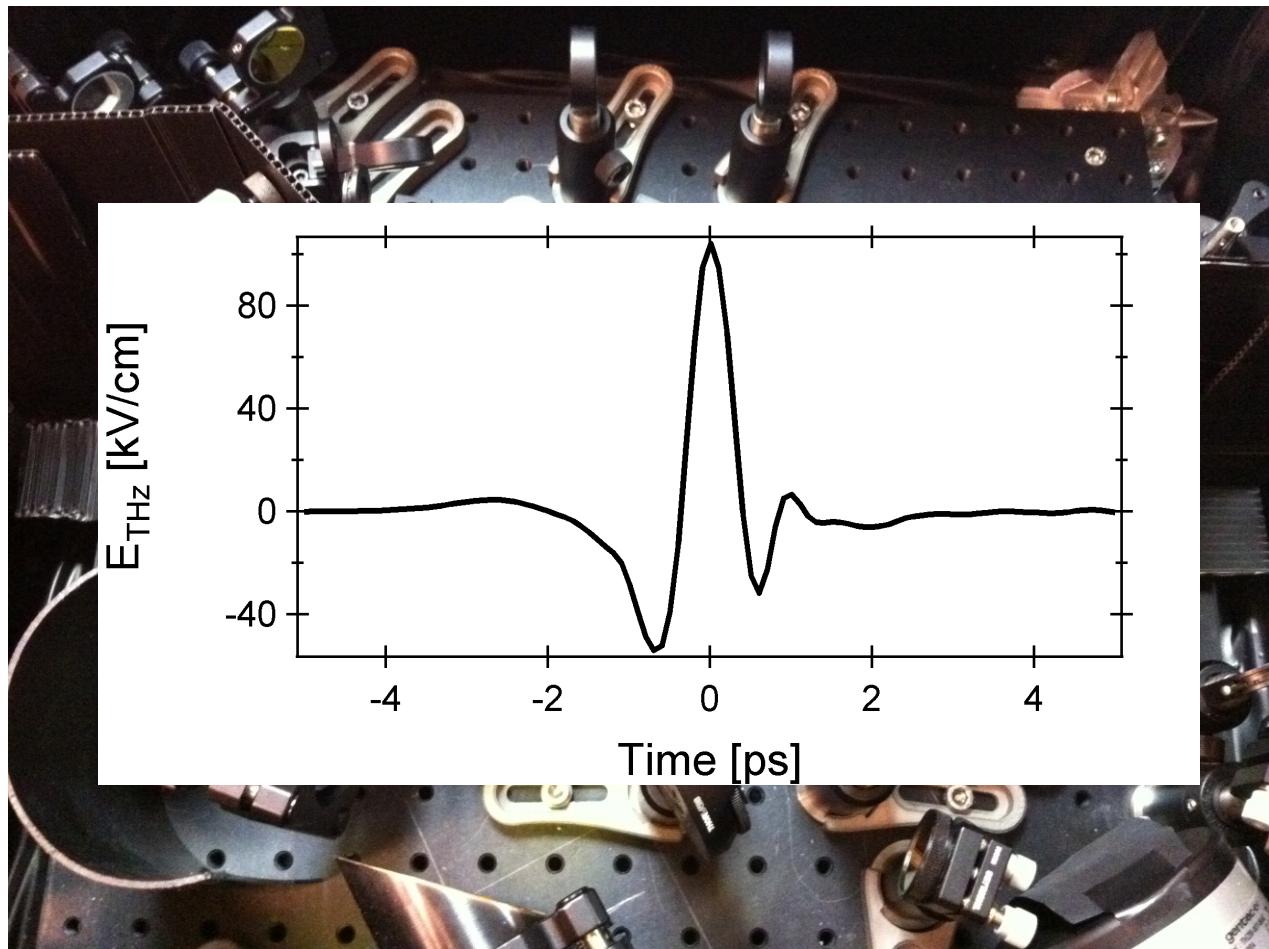
The table-top THz Source

$\eta \geq 10^{-4}$

$\geq 100 \text{ nJ/pulse}$

$\geq 10 \mu\text{J/cm}^2$ fluence

$E_{\text{THz}} \approx 100 \text{ kV/cm}$



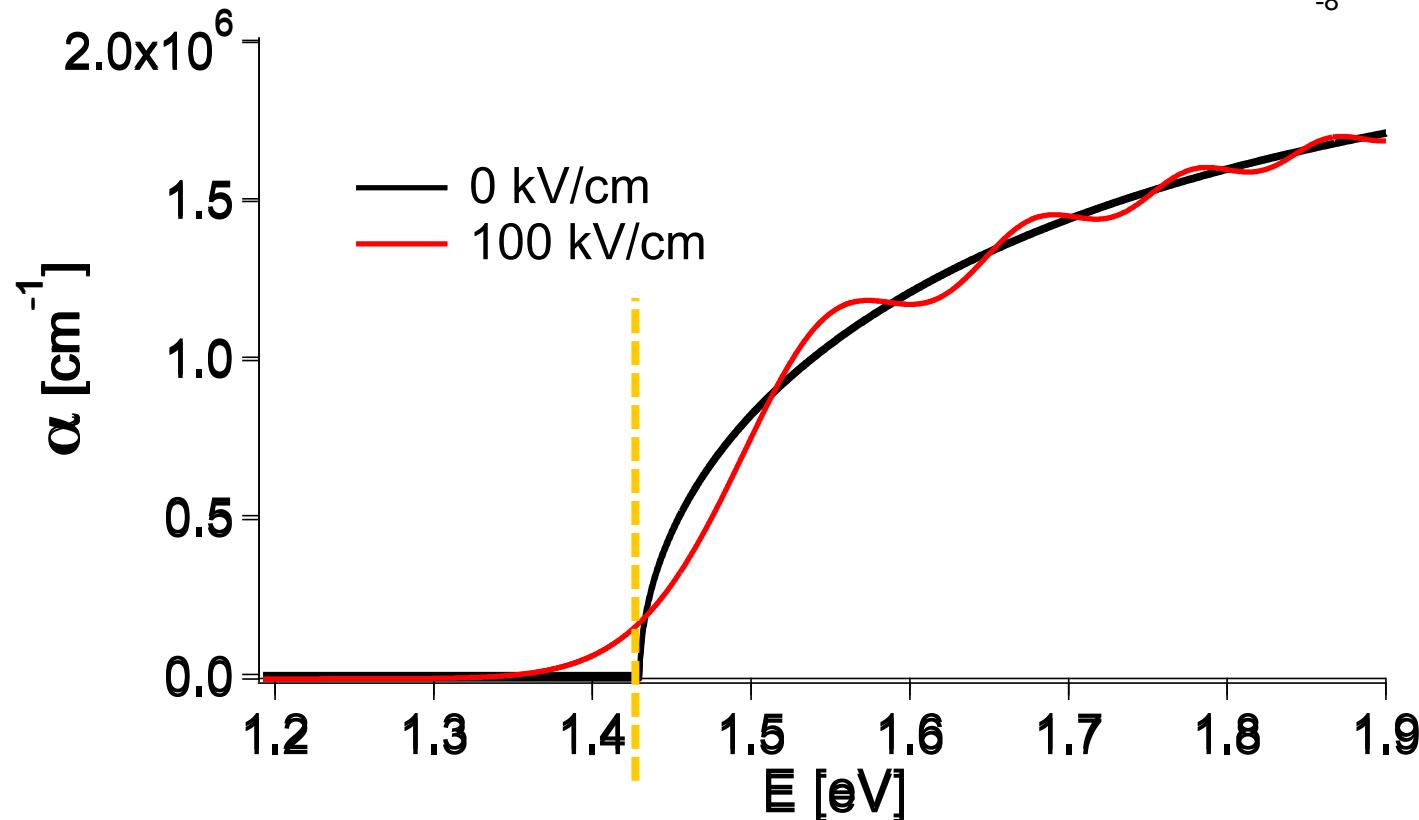
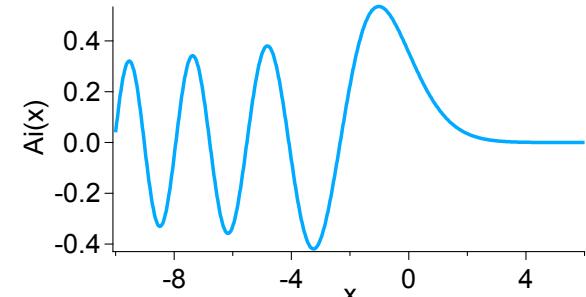
The Franz-Keldysh effect

✓ Renormalization of the semiconductor gap with the application of a static field.

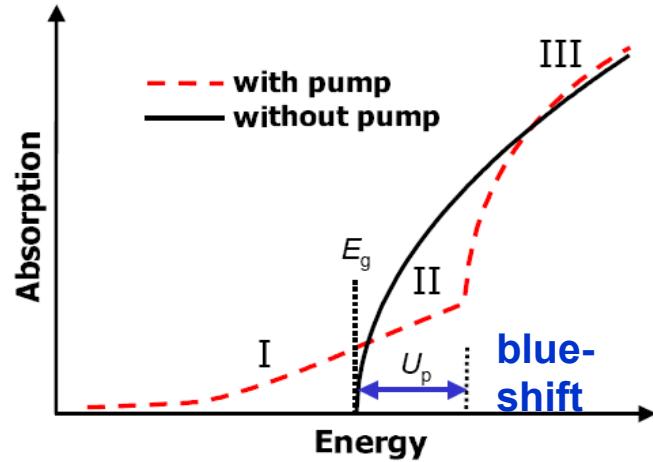
$$\left(-\frac{e^2}{2\mu} \nabla_r^2 - eEz \right) \varphi(r) = E_r \varphi(r)$$

$$\varphi_z(\xi) \propto Ai(\xi)$$

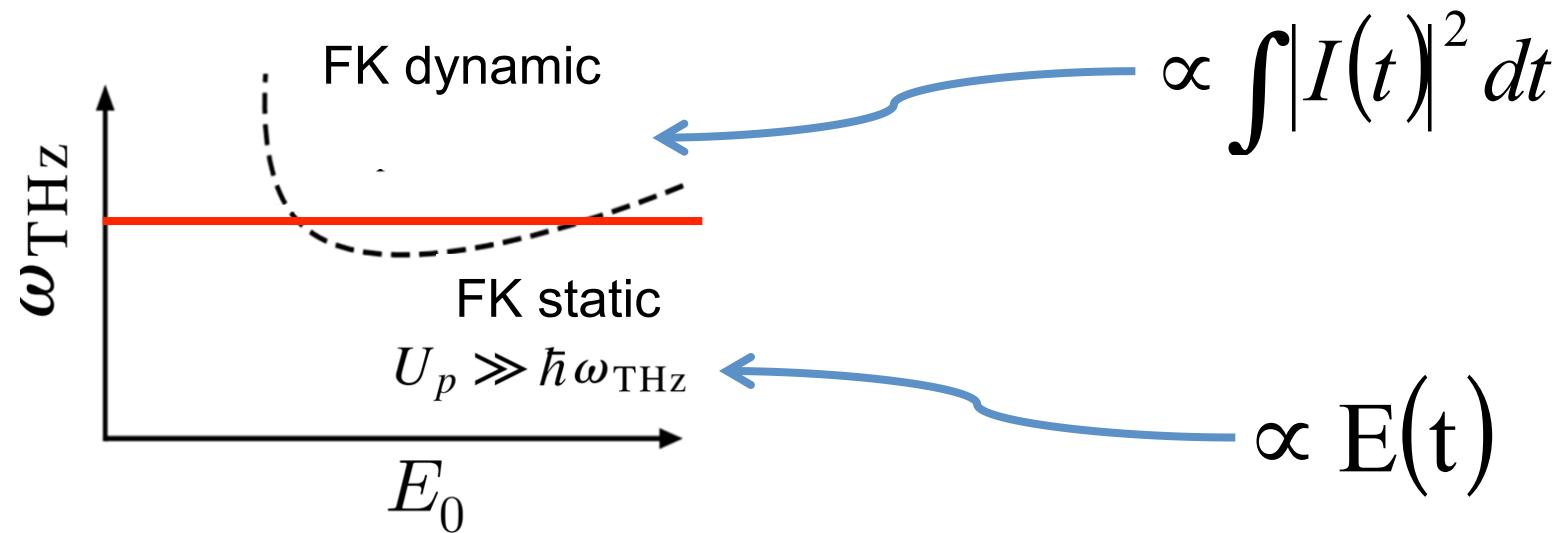
$$\xi = \left(\frac{2\mu}{e} |E| \right)^{1/3} \left(z + \frac{E_z}{eE} \right)$$



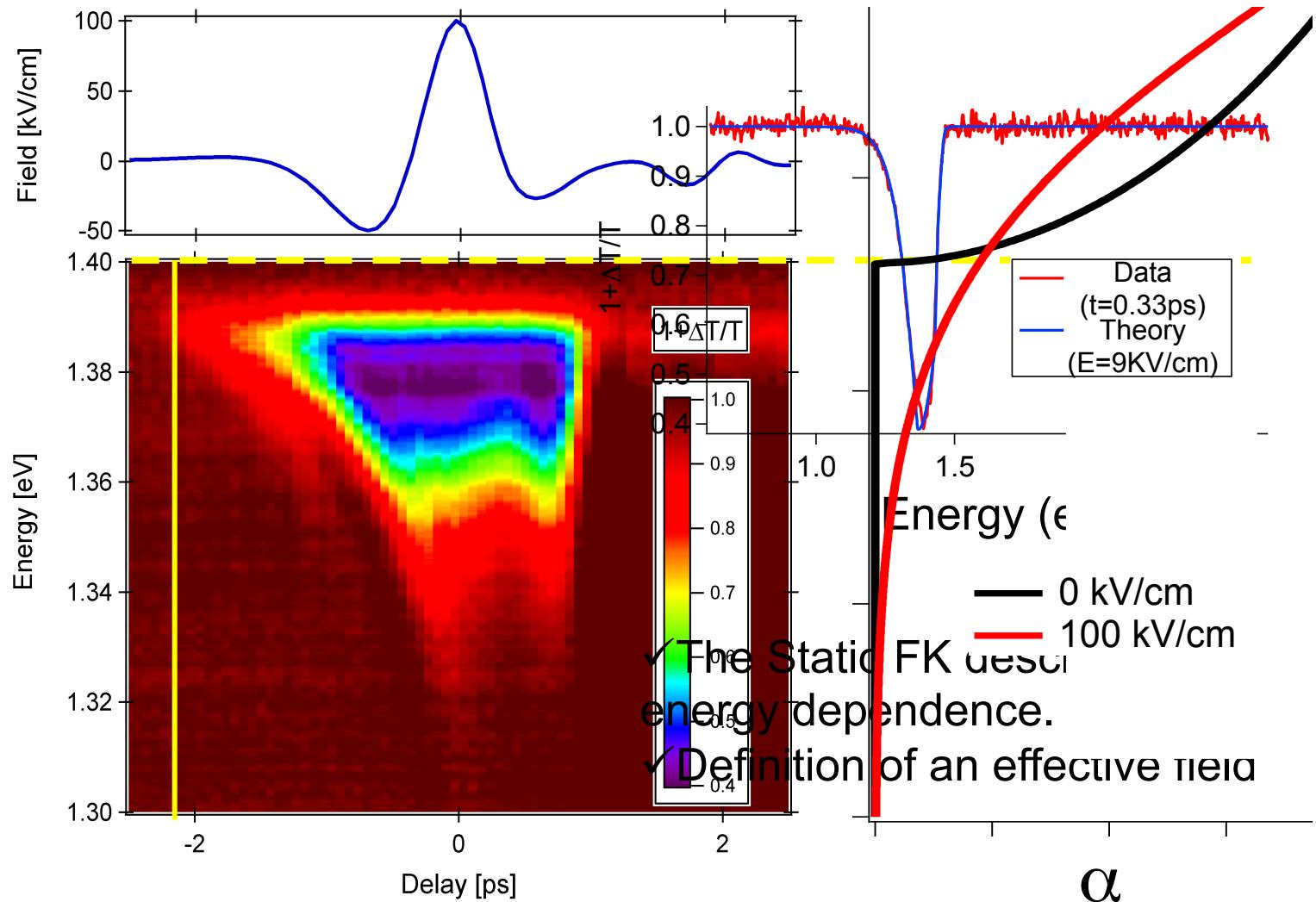
Static Vs. Dynamic Franz-Keldysh effect



$$U_p = \frac{e^2 E_0^2}{4m \omega_{THz}^2} = \omega_{THz} \gamma$$



The Franz-Keldysh effect



Outline

✓ Equilibrium Optical Spectroscopy

- The optical conductivity in the Visible and Near-IR
- What do we learn from optical conductivity?
- An example, metal insulator transition in complex oxides

✓ Non-equilibrium optical (visible near-IR) spectroscopy

- Pump&probe the main idea
- «Single color» Pump and probe
- Broadband P&P spectroscopy

✓ Self referential examples

- Revealing the excitonic nature of excitation (Hubbard Exciton)
- The electron-phonon (EP) interaction in with strong electronic correlation

✓ Non-equilibrium Infrared spectroscopy

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✓ Perspectives

- Beyond classical spectroscopy
- Using the quantum state of light as a new spectroscopyc tool
- Table top Vs. FEL and sincrotrons

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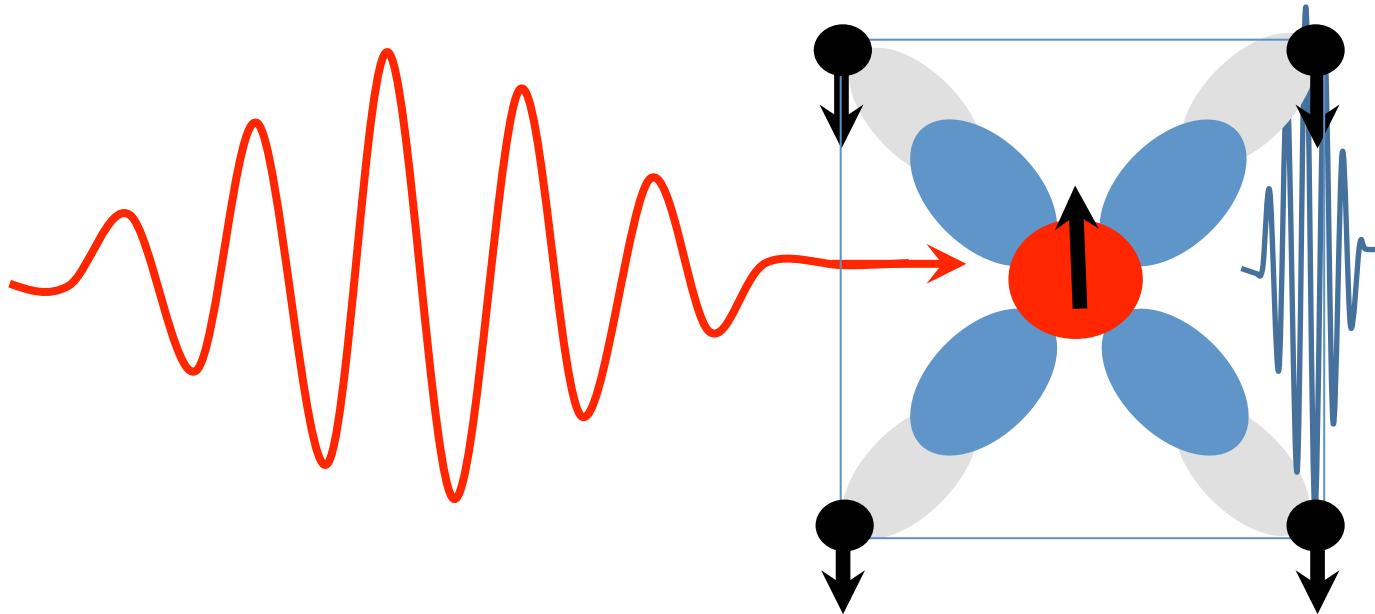
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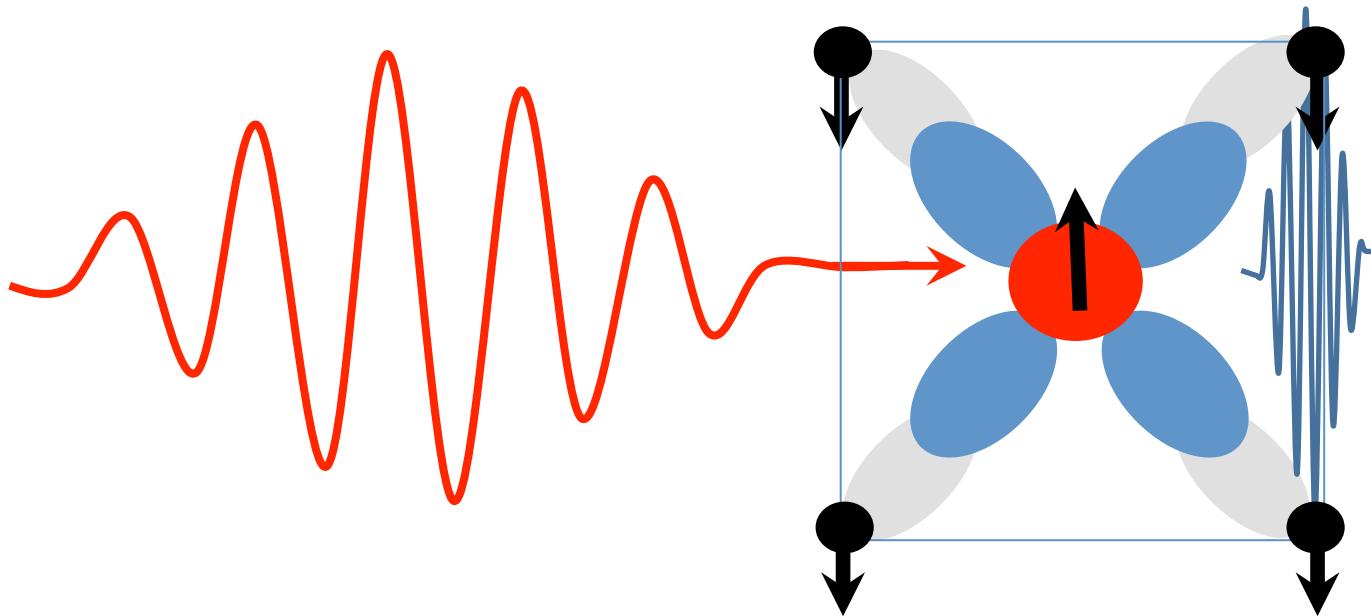
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Can we resonantly excite phonon modes?



Can we resonantly excite phonon modes?

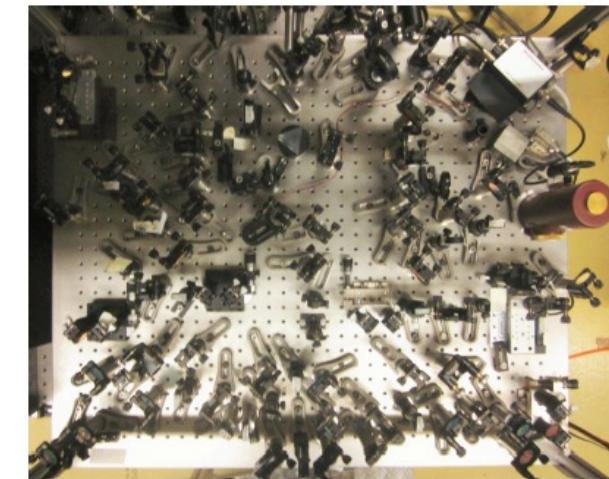
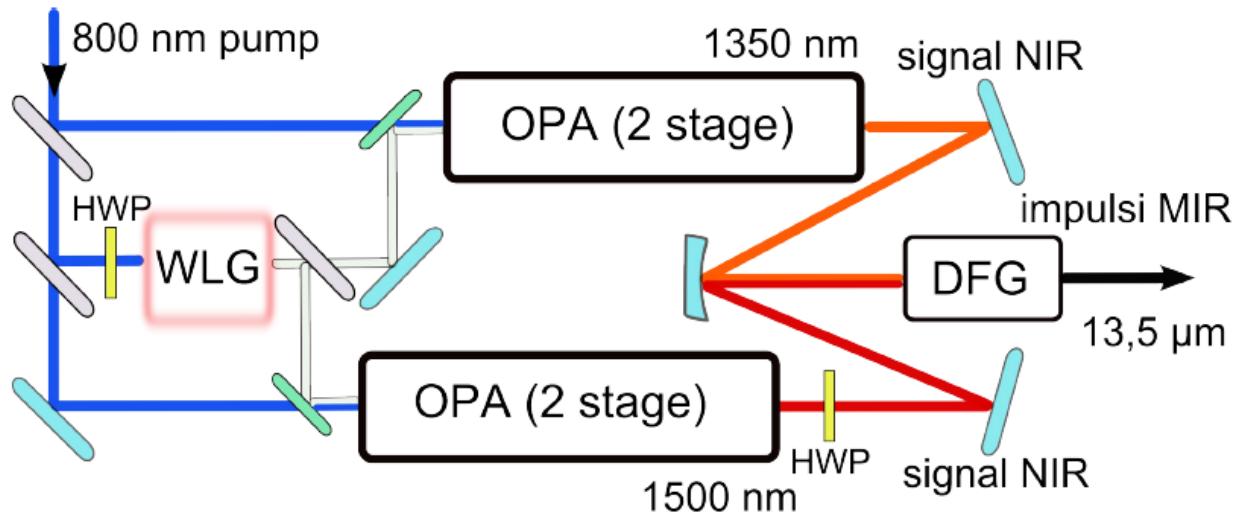


- ✓ Revealing the non-adiabatic response of electron
- ✓ Amplitude and phase resolution
- ✓ **Optical control of material properties**

Science 331, 189 (2011)*; Nature Material 12, 882 (2013)
Nat. Photonics 5,485 (2011), Nat. Materials 12, 535 (2013)

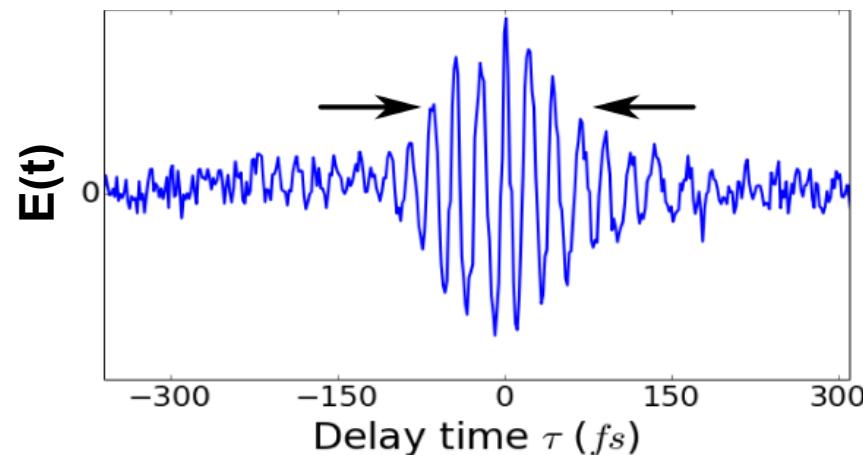
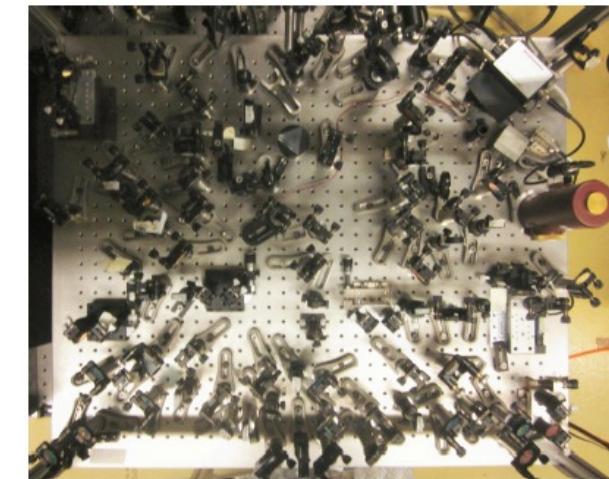
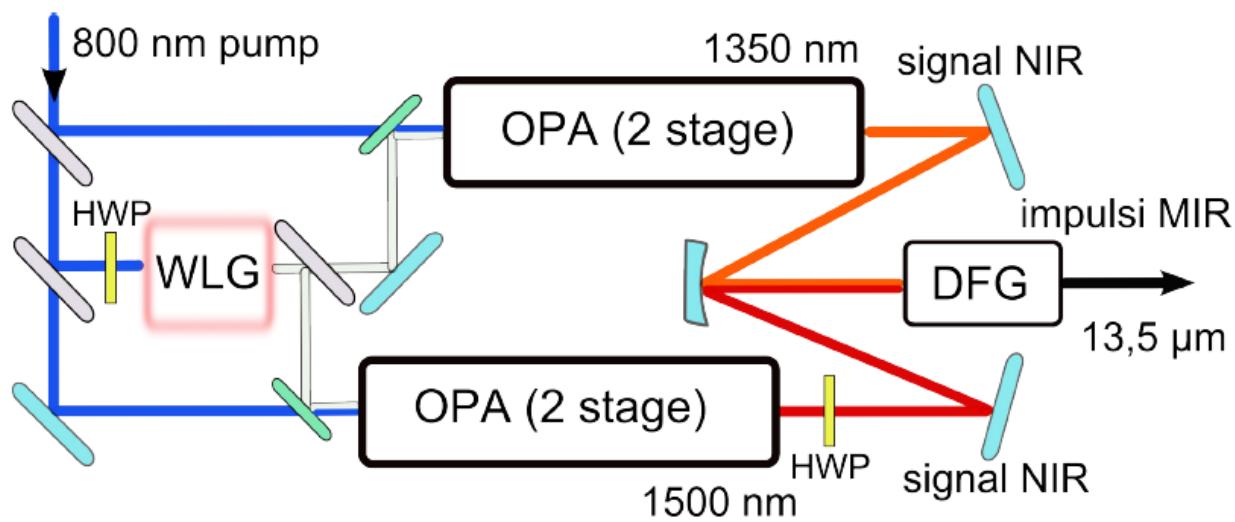
Can we resonantly excite phonon modes?

A cascade of non-linear processes to generate carrier envelope phase stable mid-IR pulses?



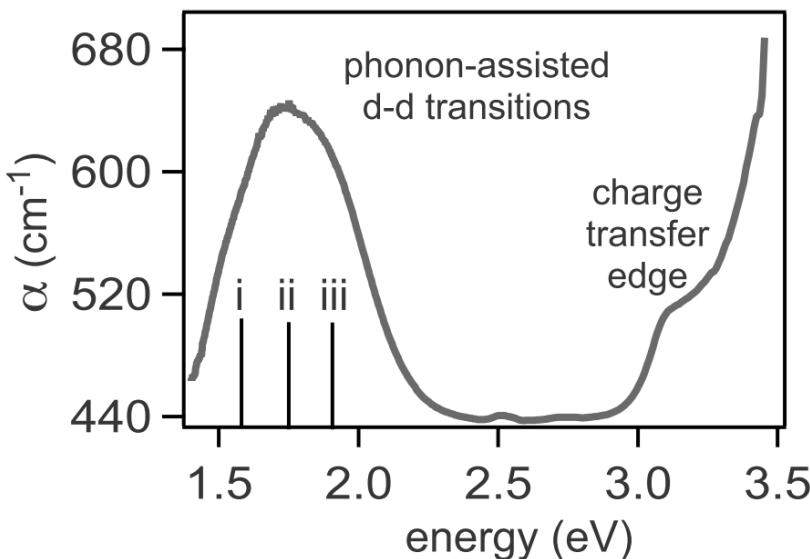
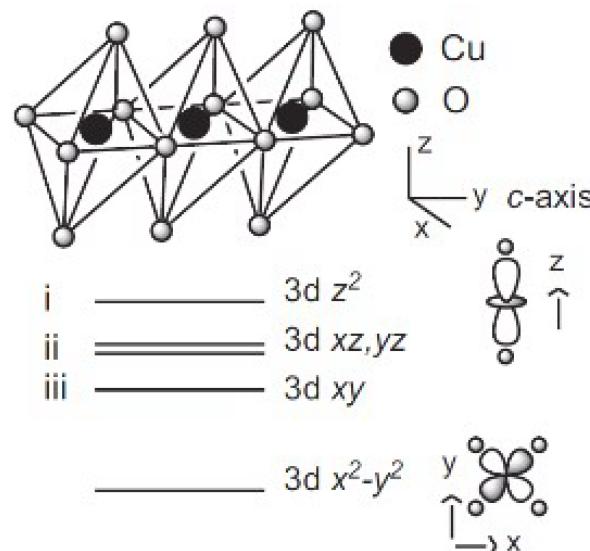
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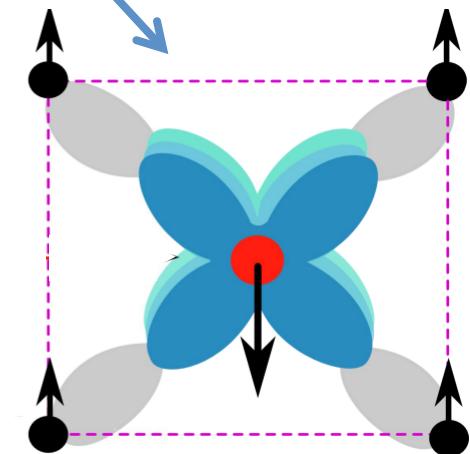
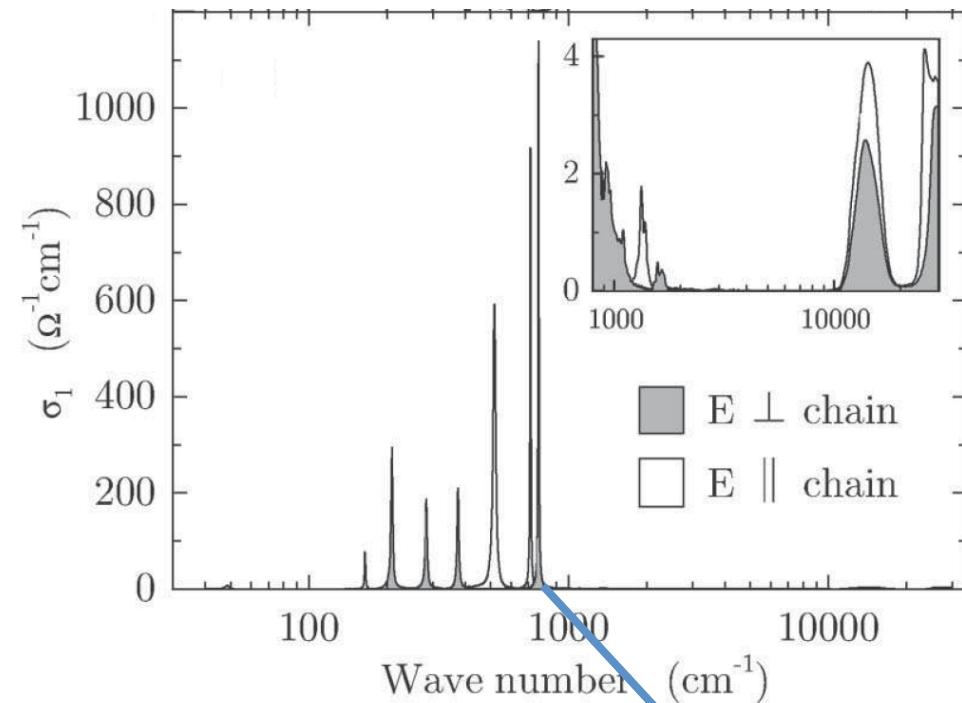
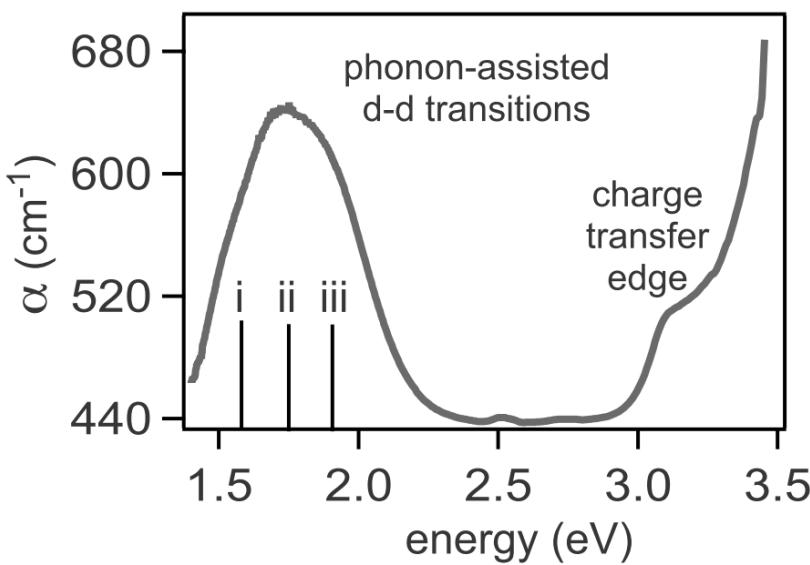
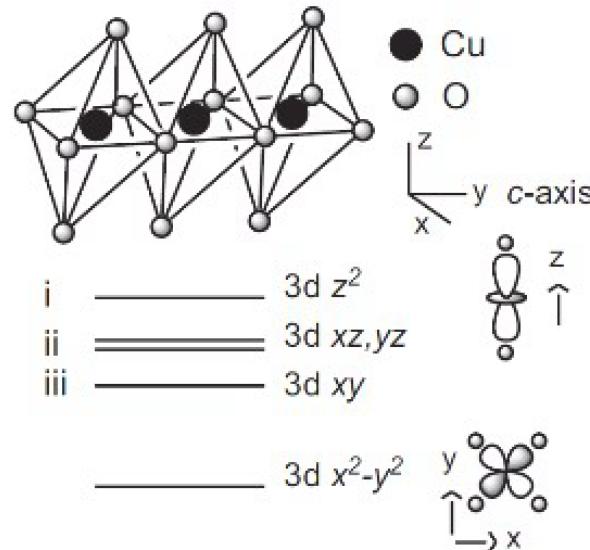
- ✓ Ultrashort pulses in the mid-IR
- ✓ $5 < \lambda < 16 \mu\text{m}$
- ✓ $E > 1 \mu\text{J}/\text{pp}$

Phonon Pump in CuGeO₃



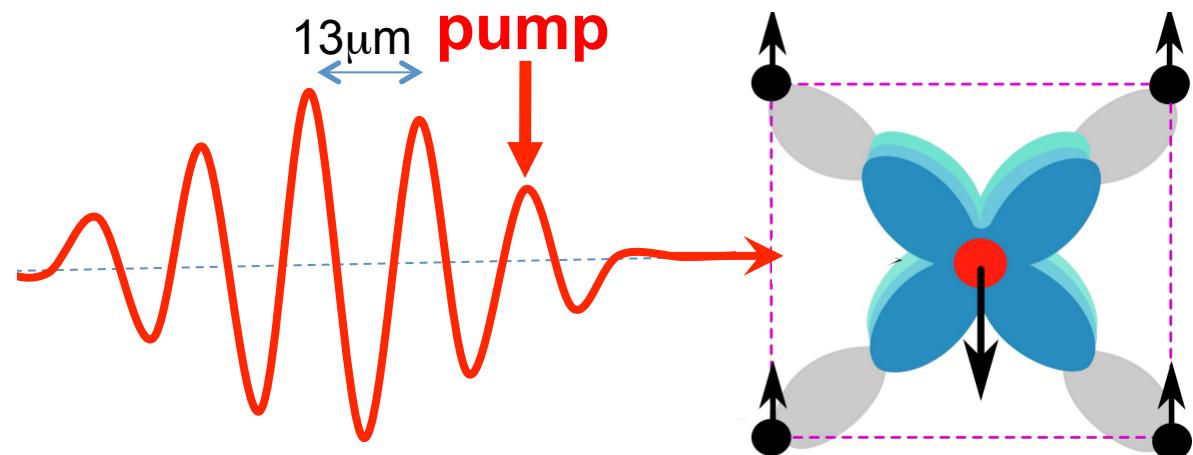
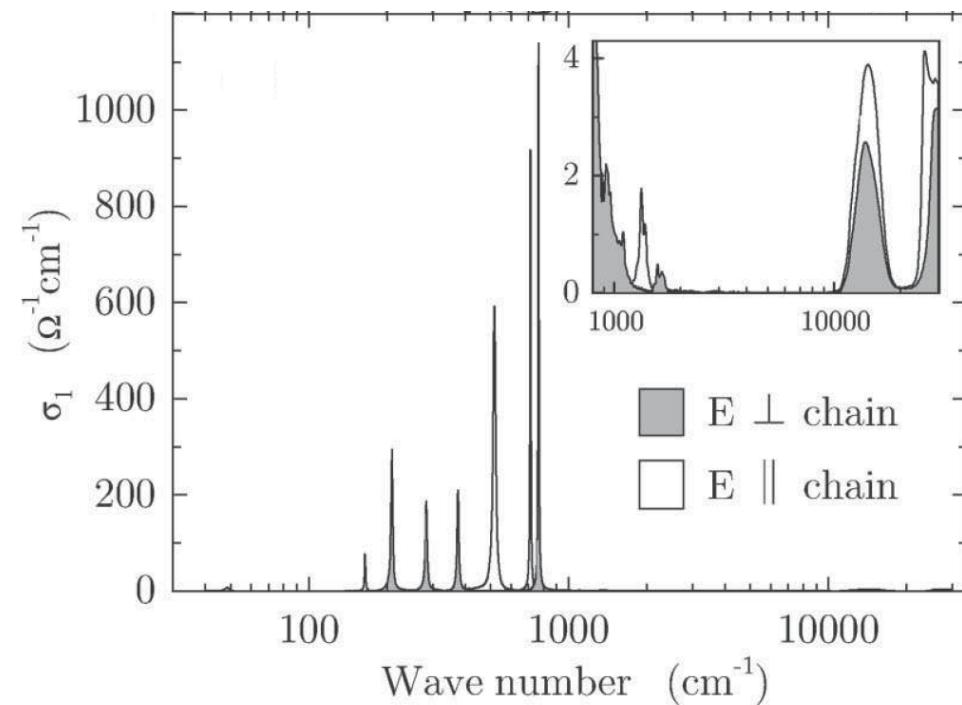
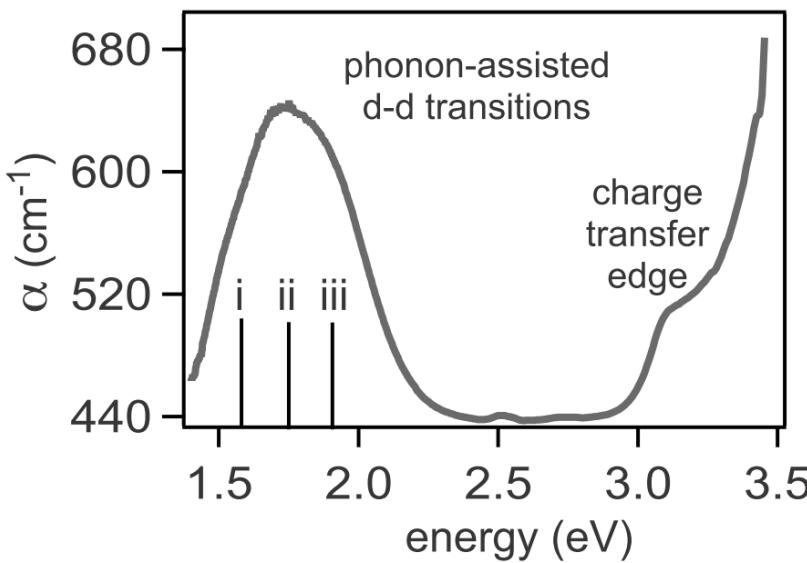
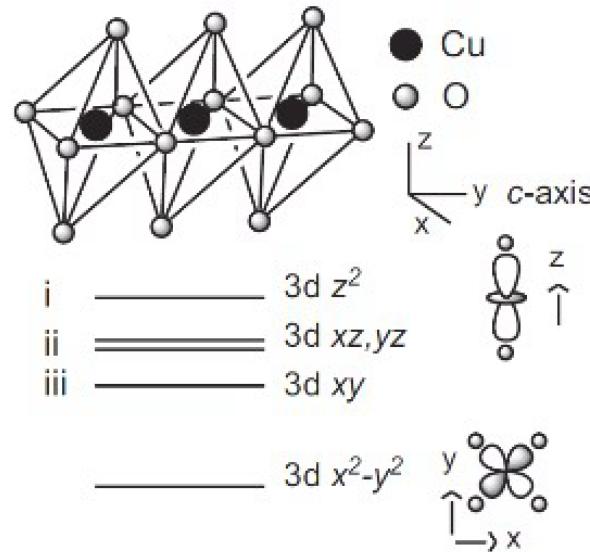
Prb, 80, 235139

Phonon Pump in CuGeO₃



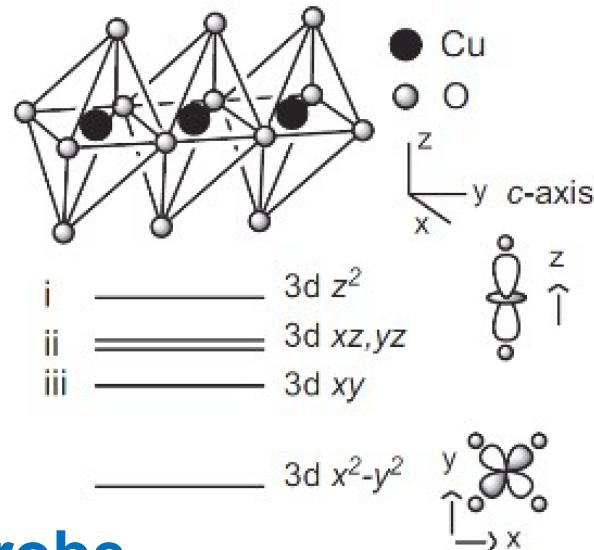
Prb, 80, 235139; Prb, 61, 12063

Phonon Pump in CuGeO₃

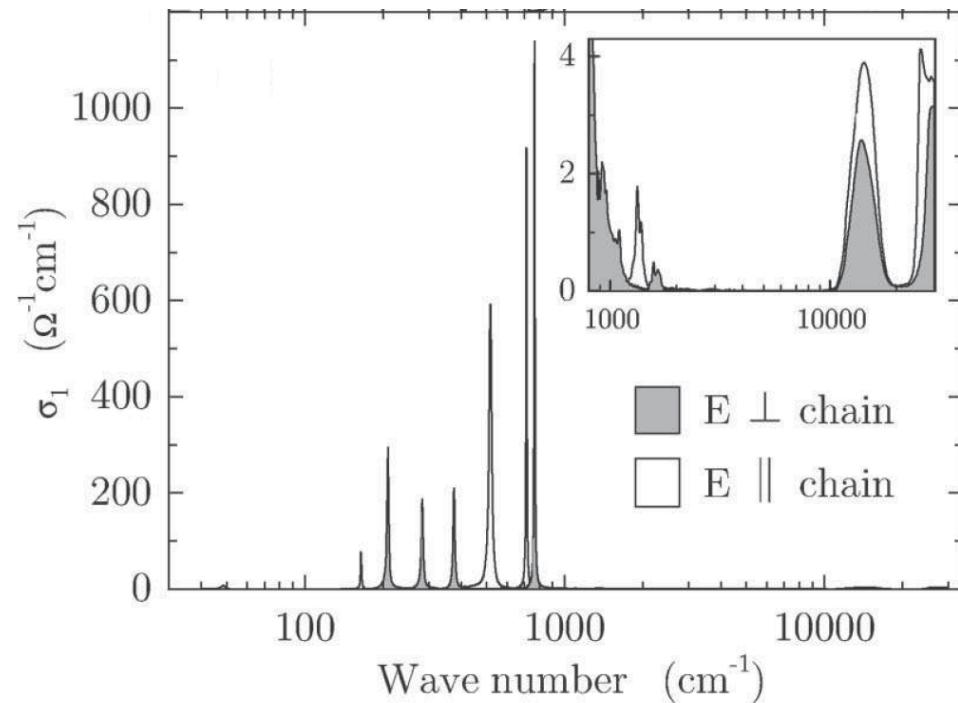
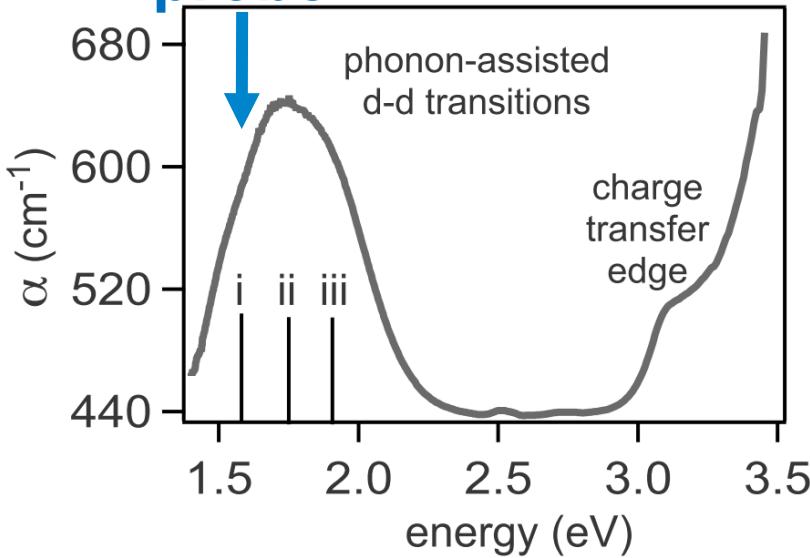


Prb, 80, 235139; Prb, 61, 12063

Phonon Pump in CuGeO₃

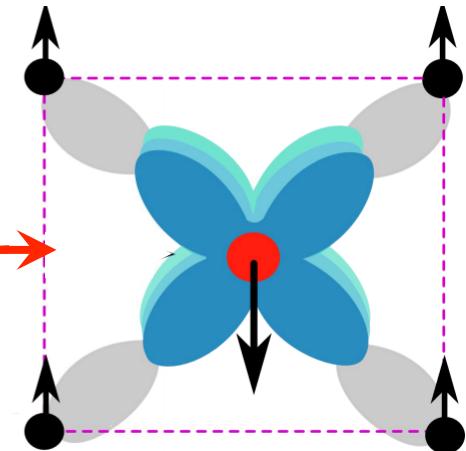


probe



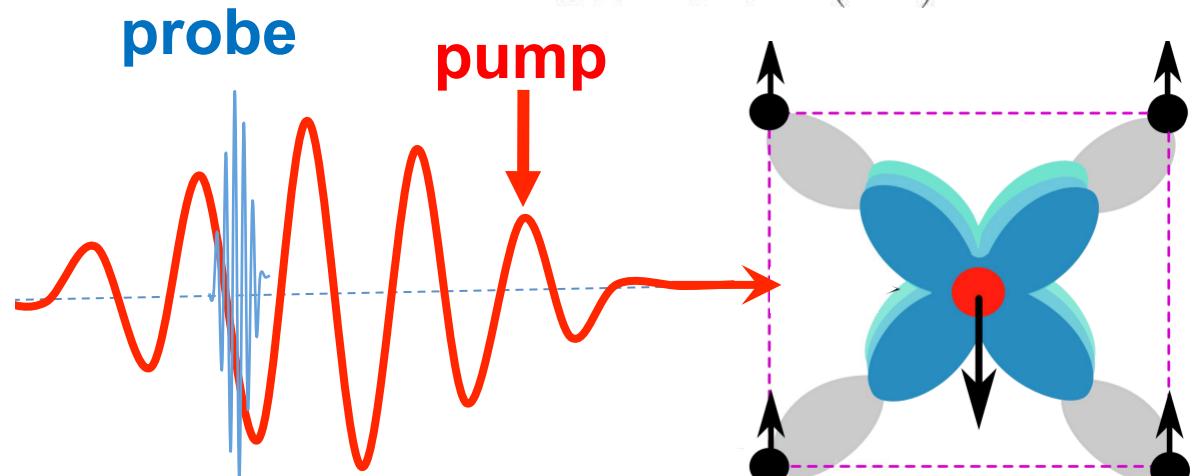
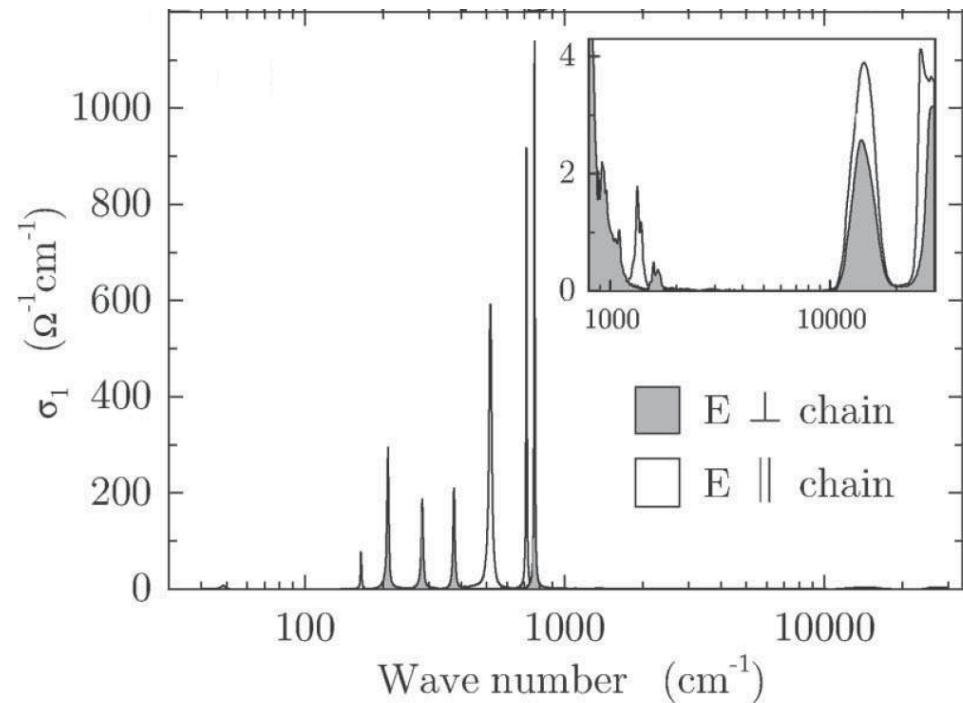
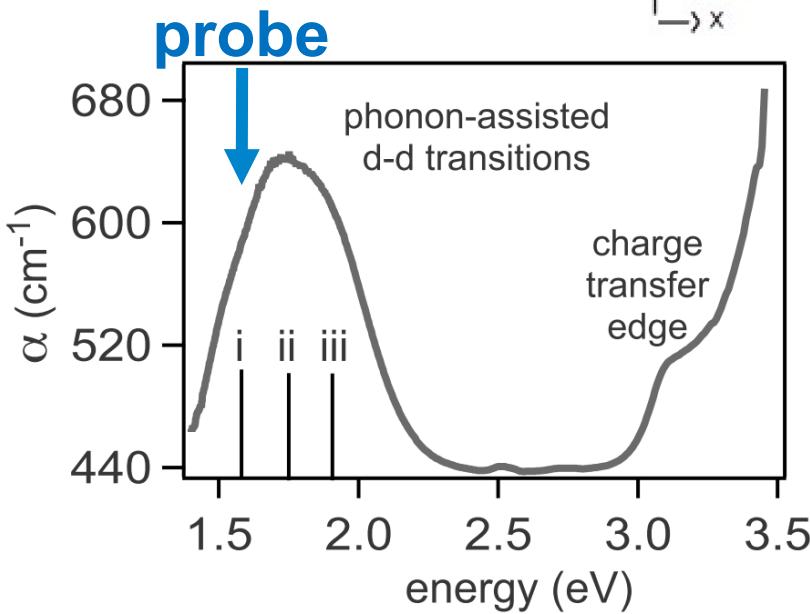
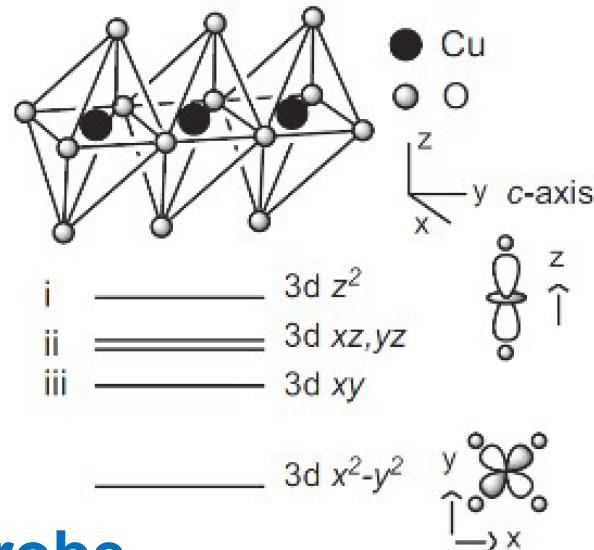
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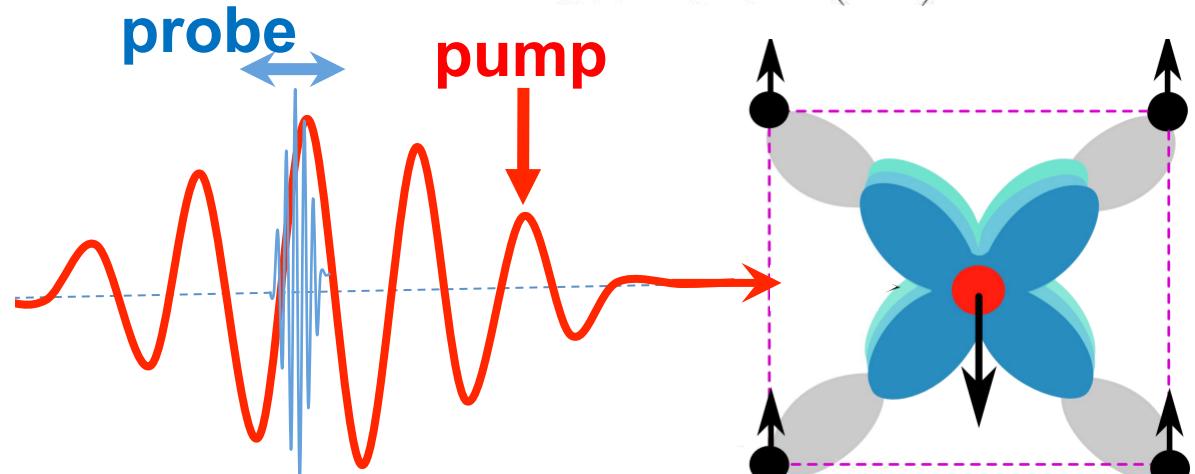
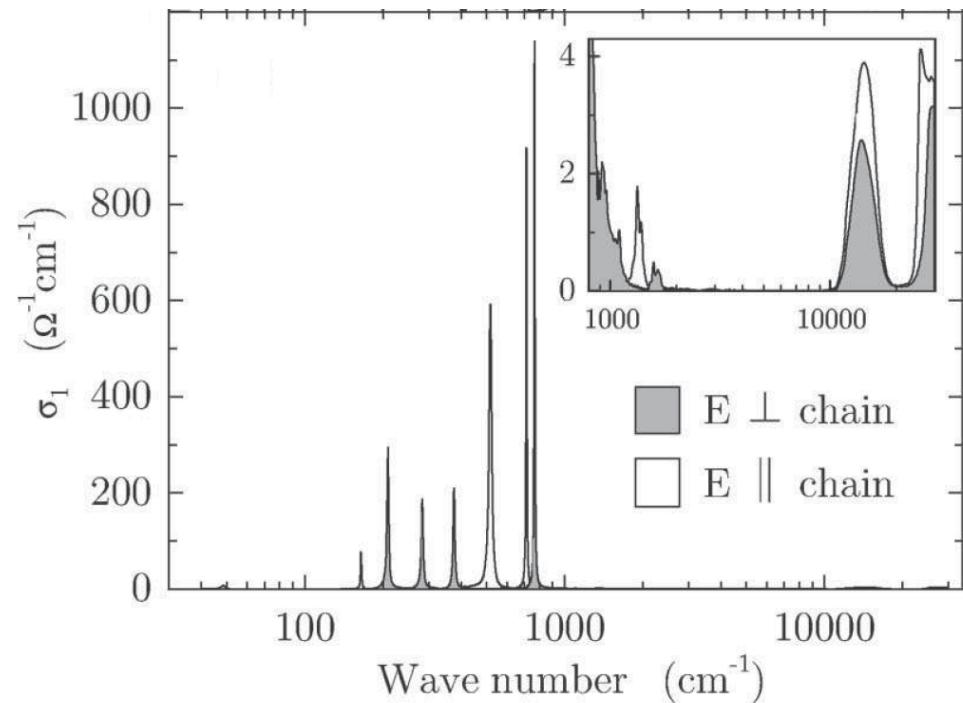
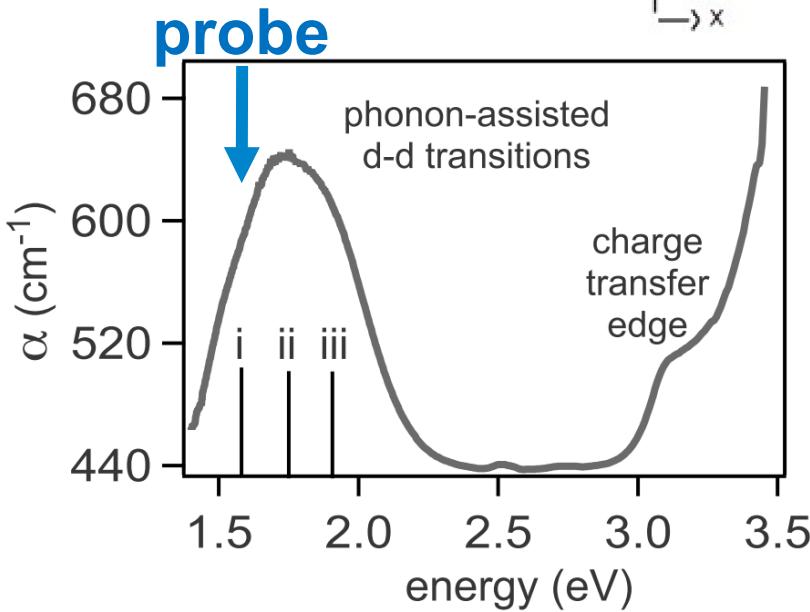
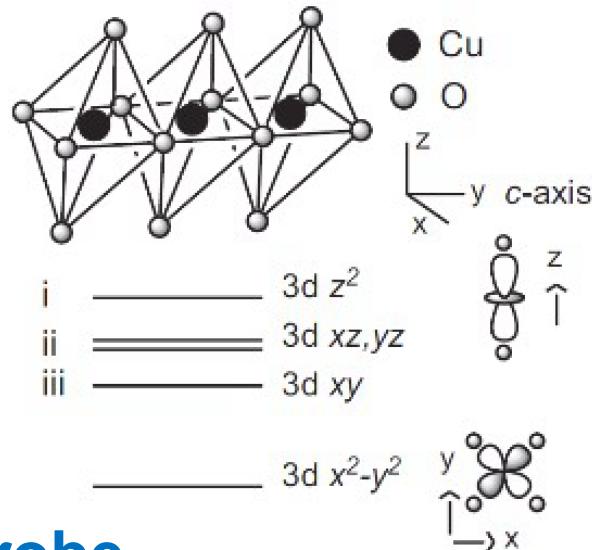
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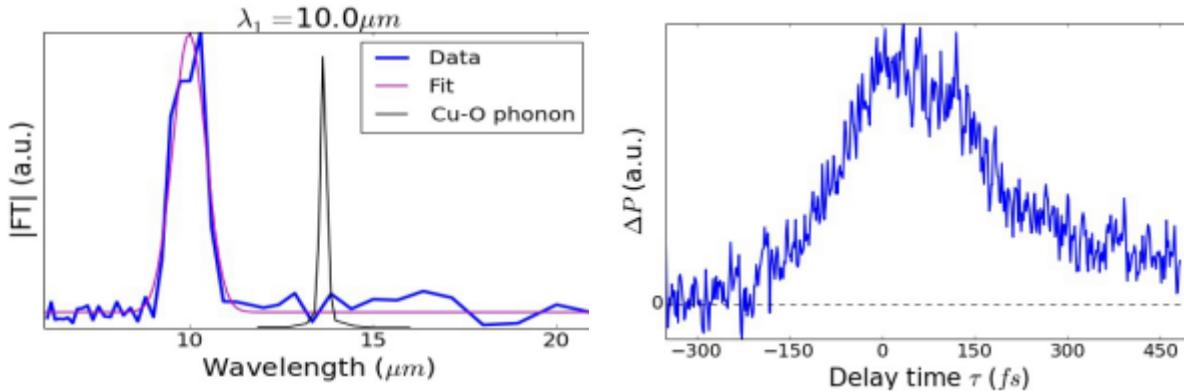
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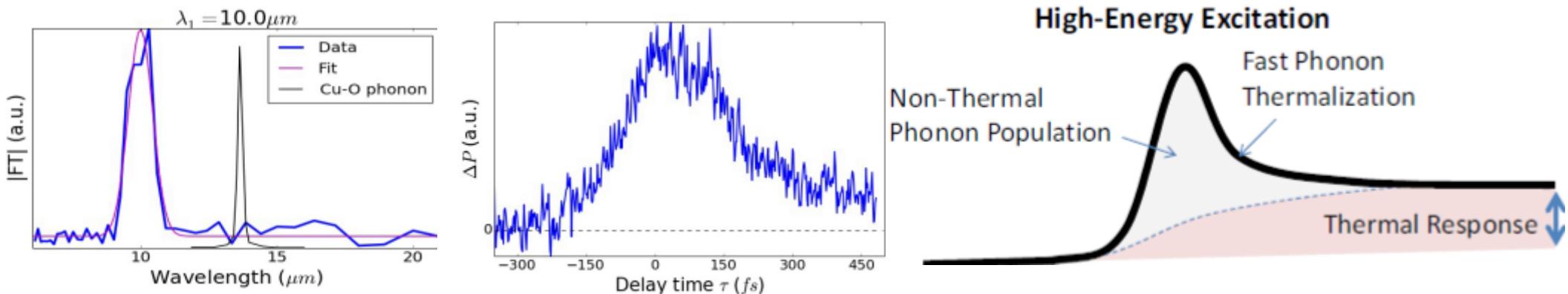


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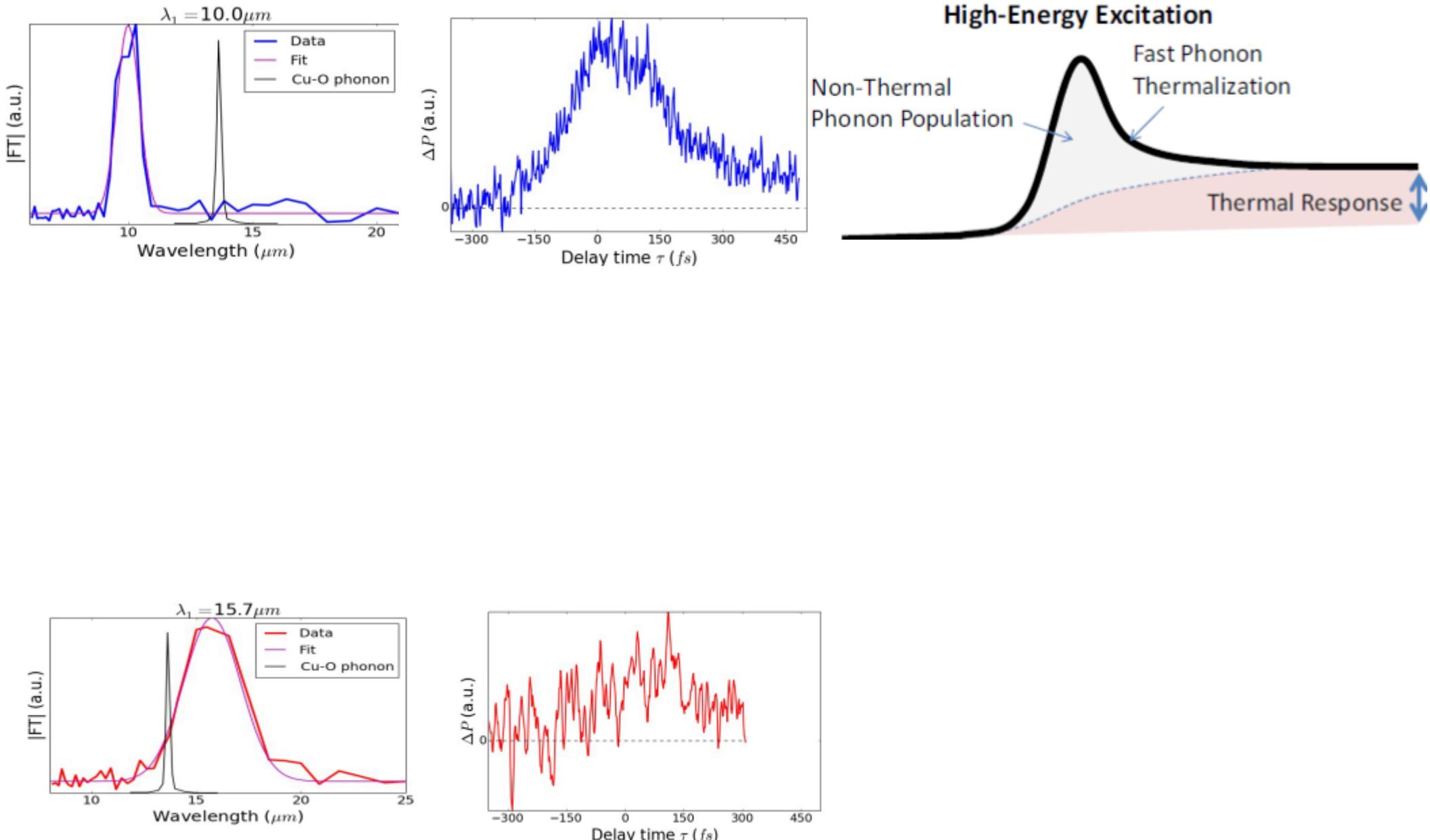
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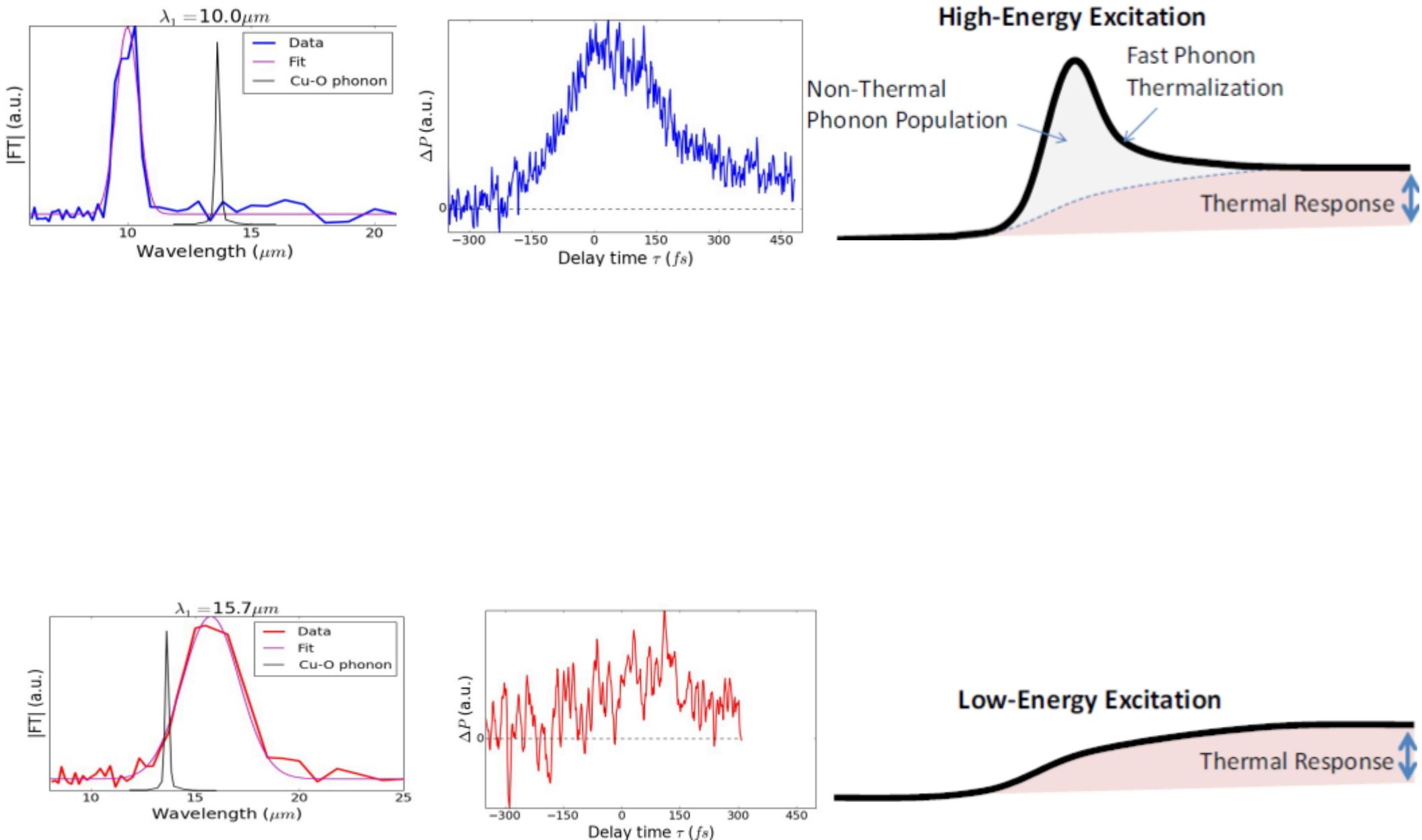
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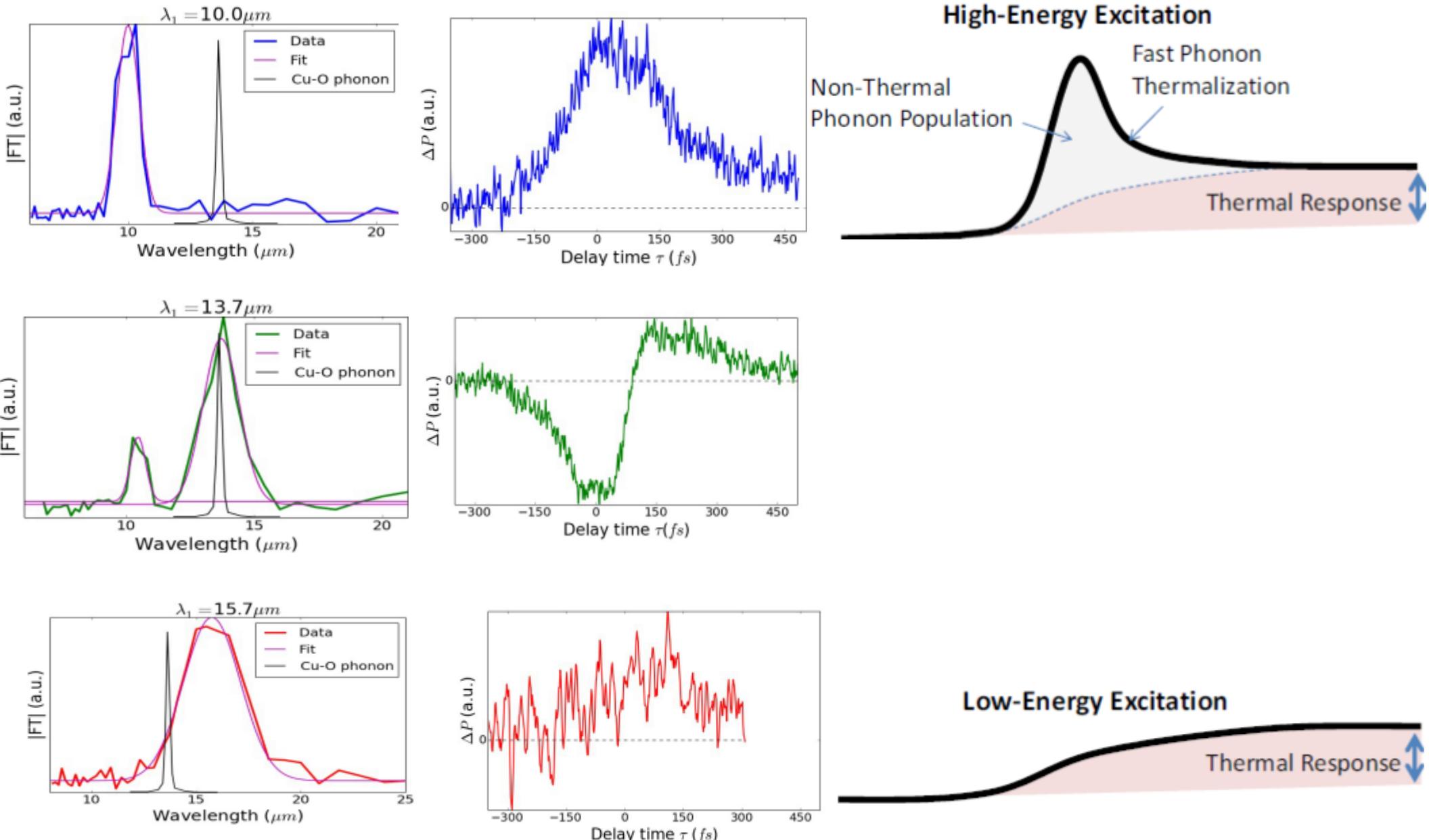
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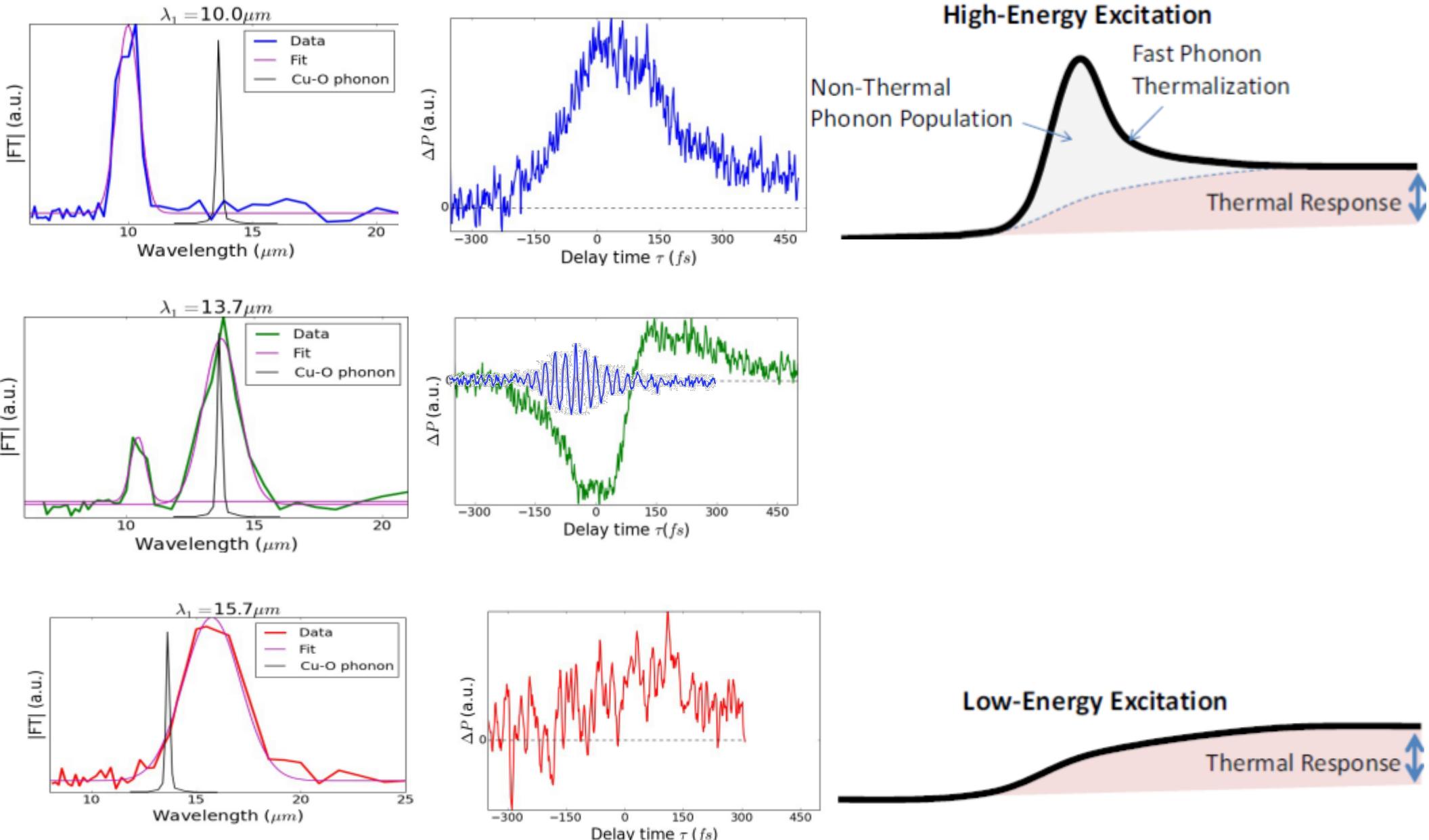
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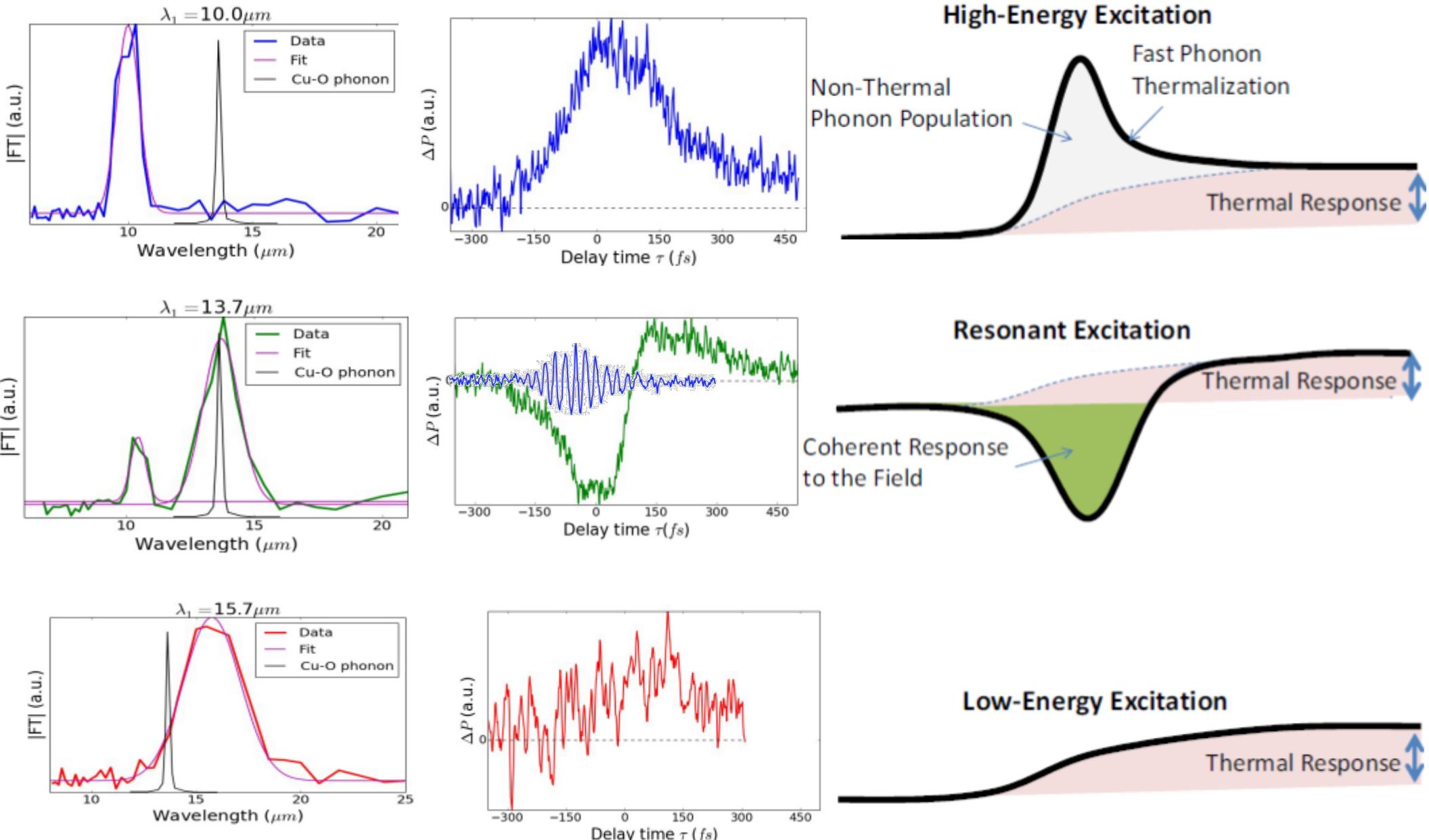
Phonon Pump in CuGeO₃



Phonon Pump in CuGeO₃



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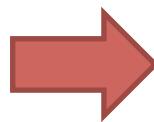
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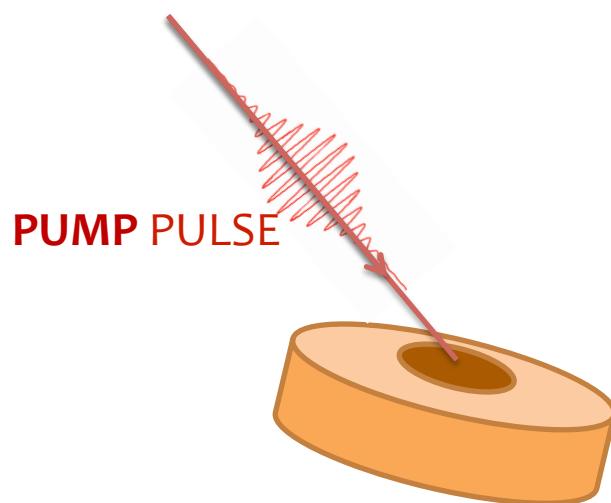
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Basic idea

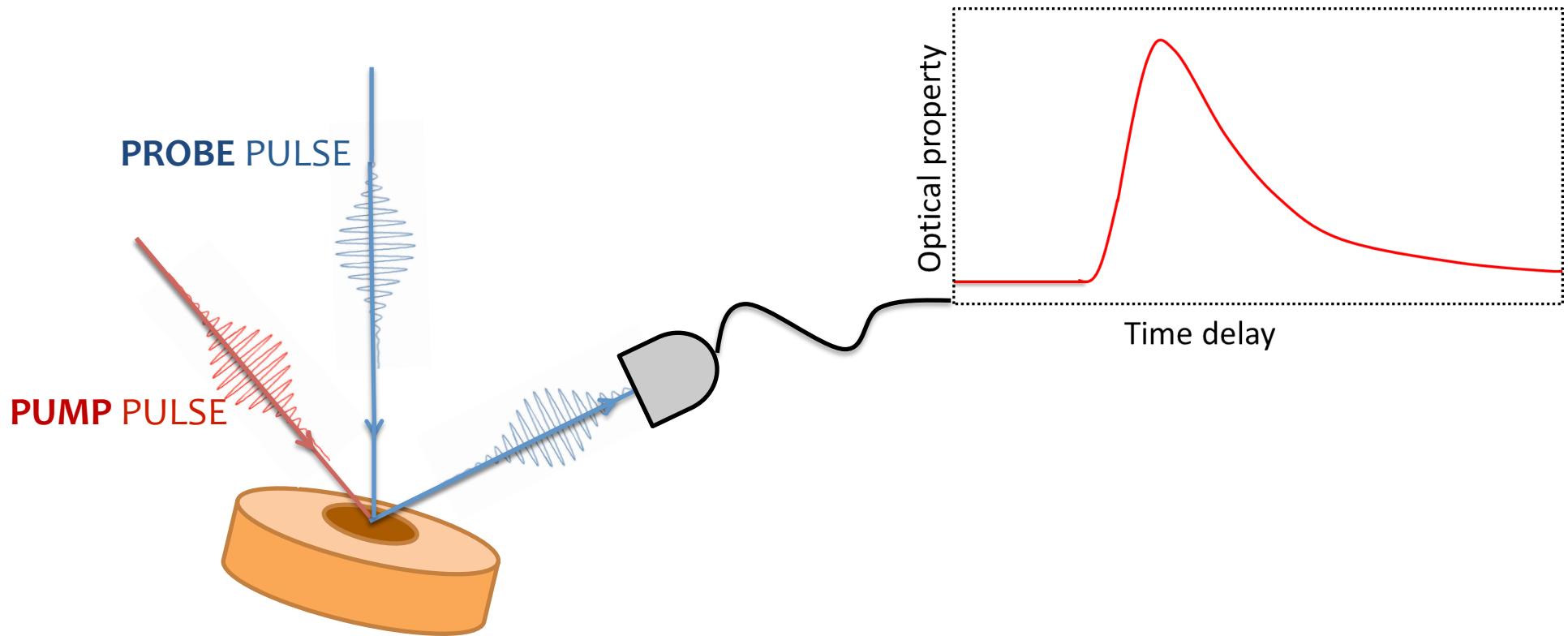
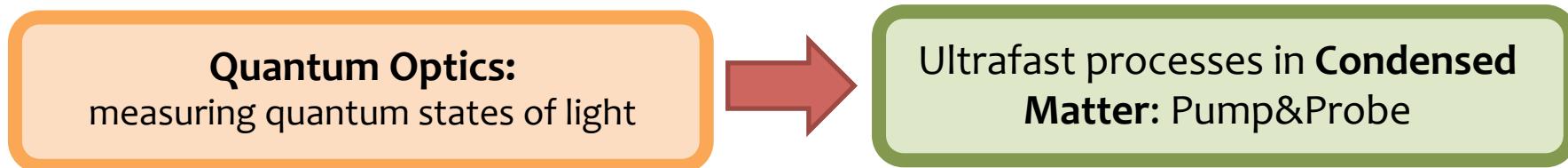
Quantum Optics:
measuring quantum states of light



Ultrafast processes in **Condensed Matter**: Pump&Probe



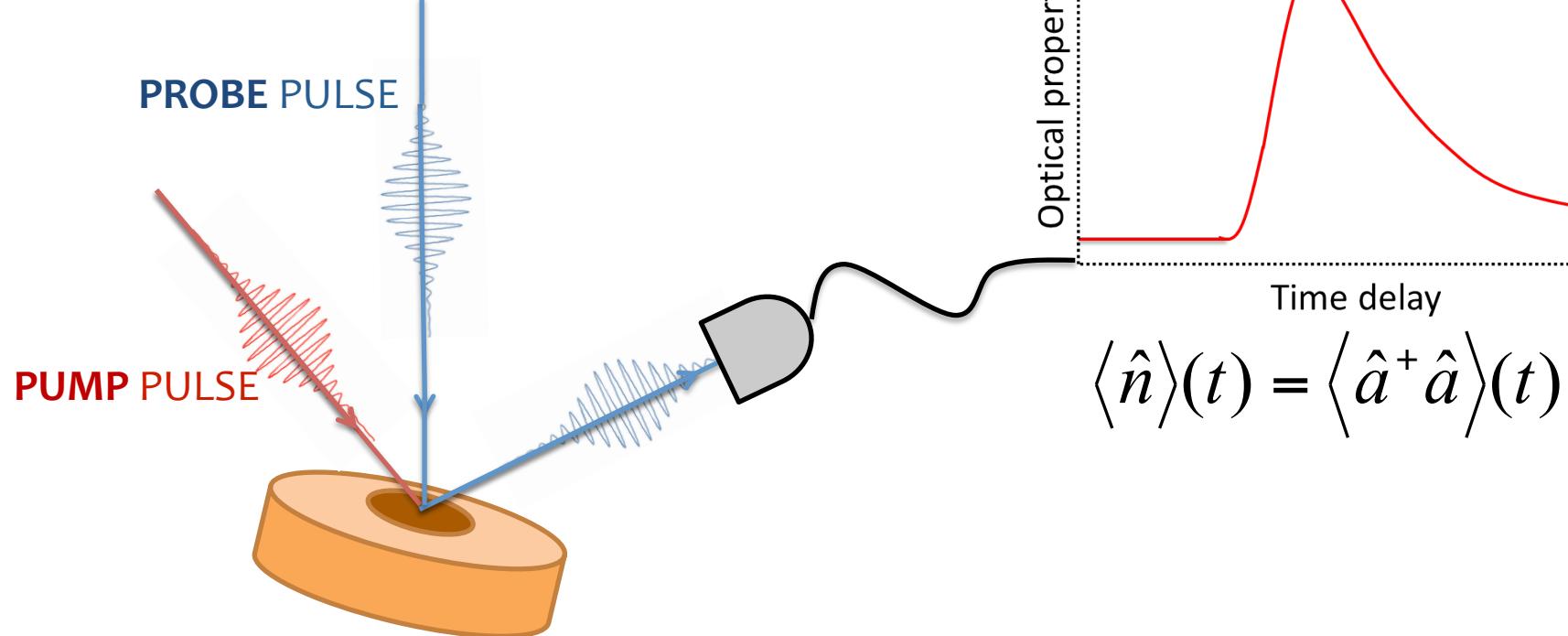
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Basic idea

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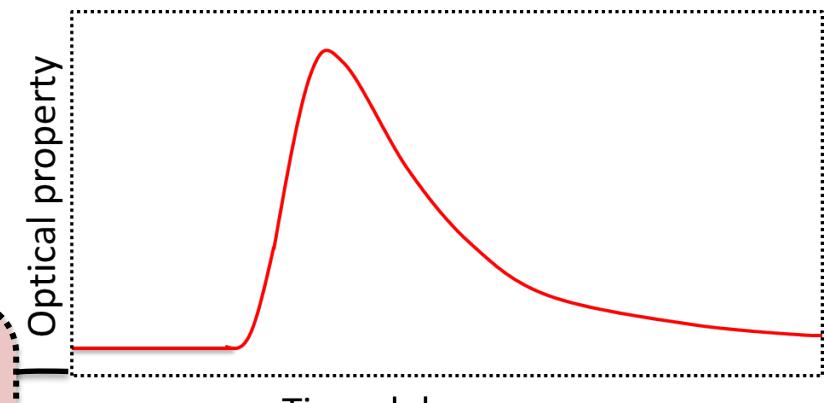
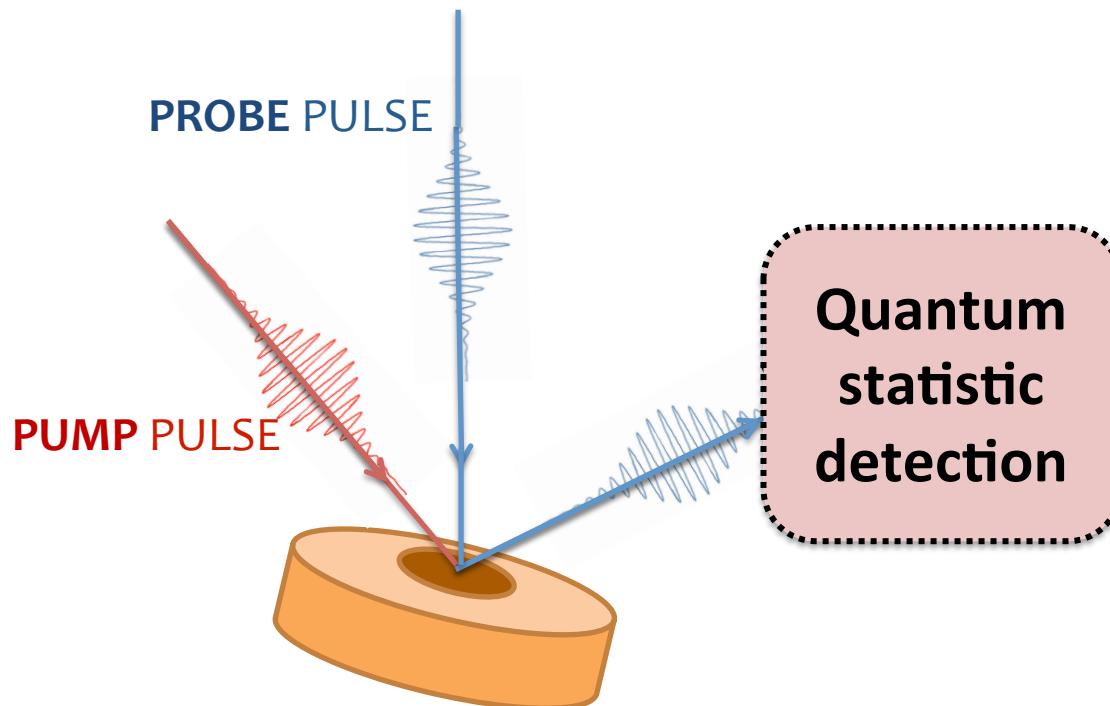
Ultrafast processes in Condensed
Matter: Pump&Probe



Basic idea

Quantum Optics:
measuring quantum states of light

Ultrafast processes in Condensed Matter: Pump&Probe



$$\langle \hat{n} \rangle(t) = \langle \hat{a}^\dagger \hat{a} \rangle(t)$$

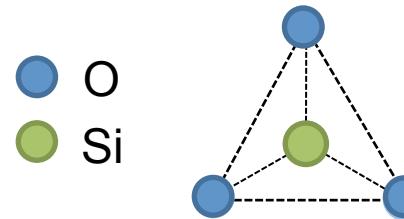
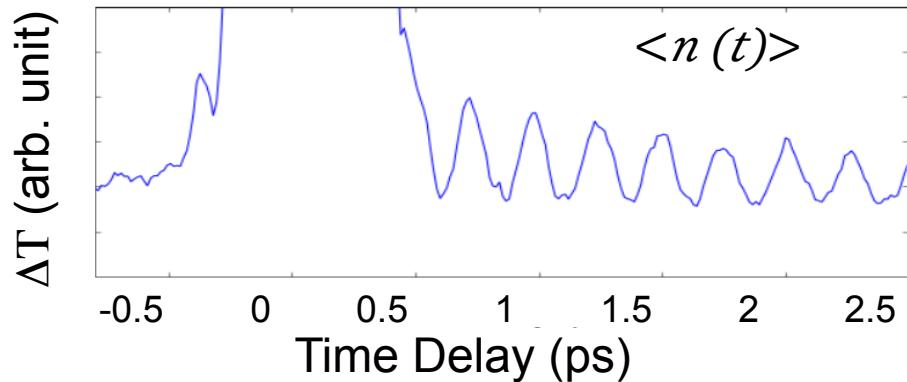
$$\langle \hat{n}^2 \rangle(t)$$

⋮

$$\langle \hat{n}^k \rangle(t)$$

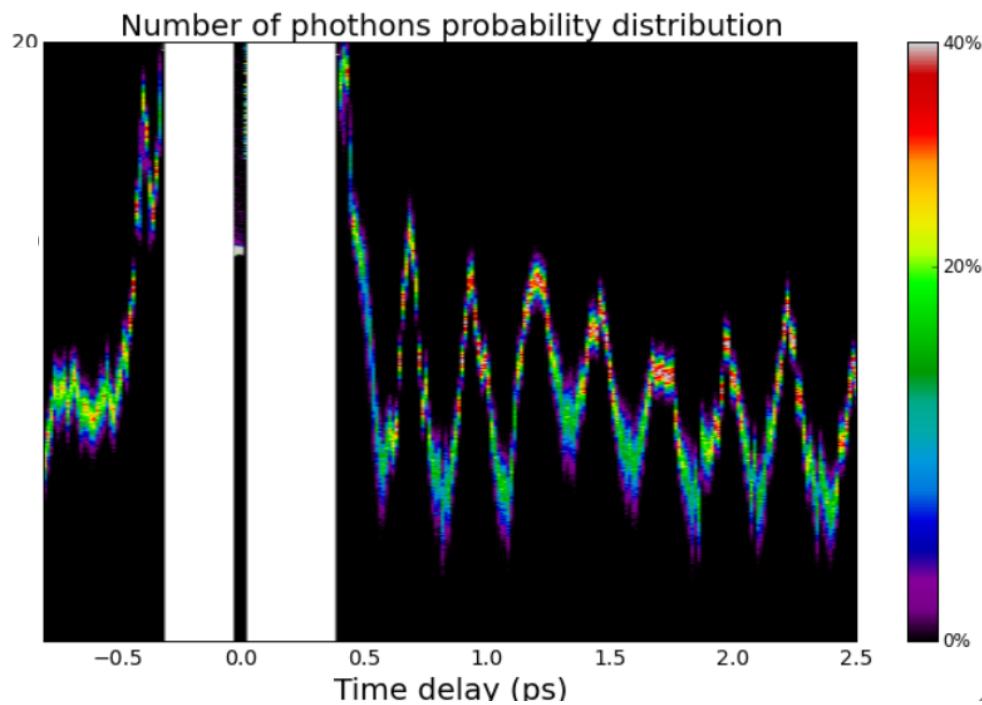
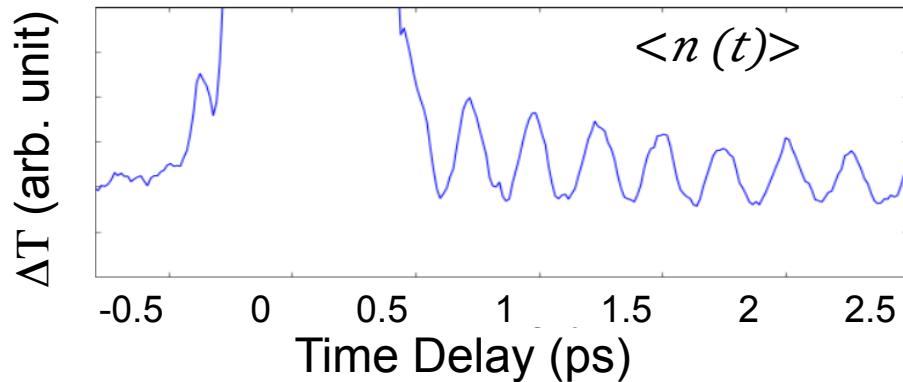
Nature Comm. 6, 10249 (Dec. 2015)

Higher order: Pump and Probe α -quartz



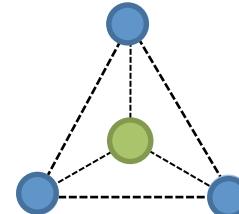
Coherent Motion

Higher order: Pump and Probe α -quartz

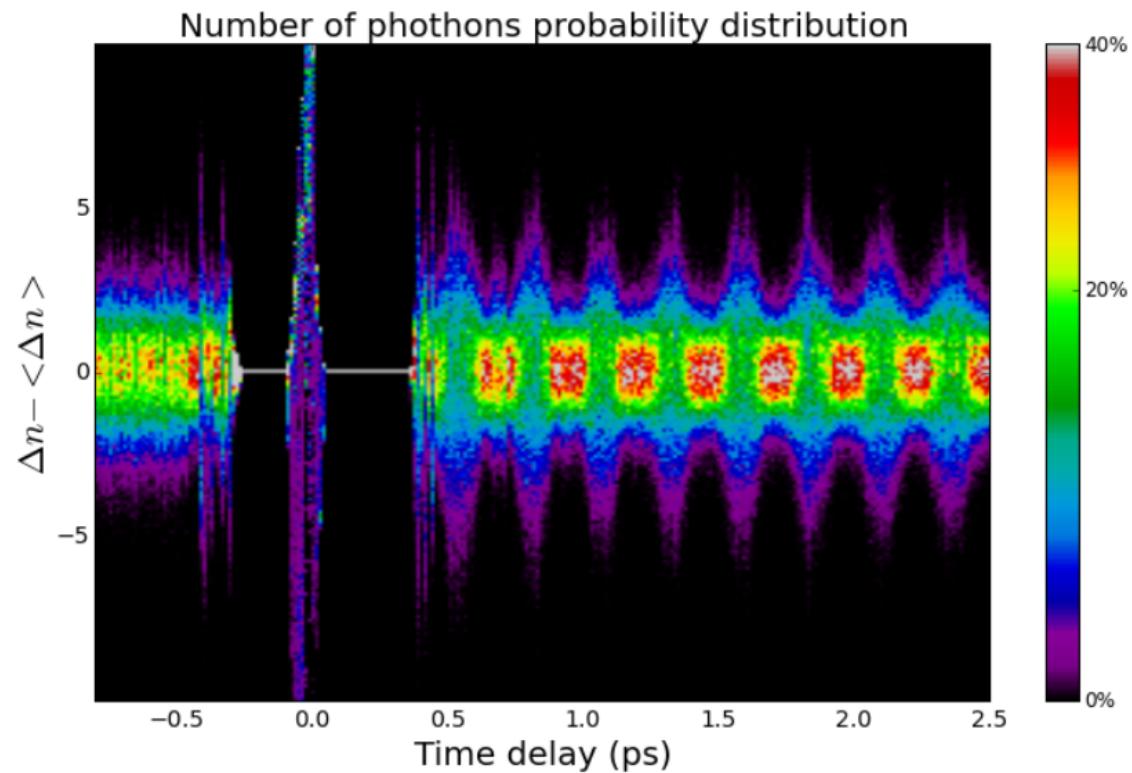
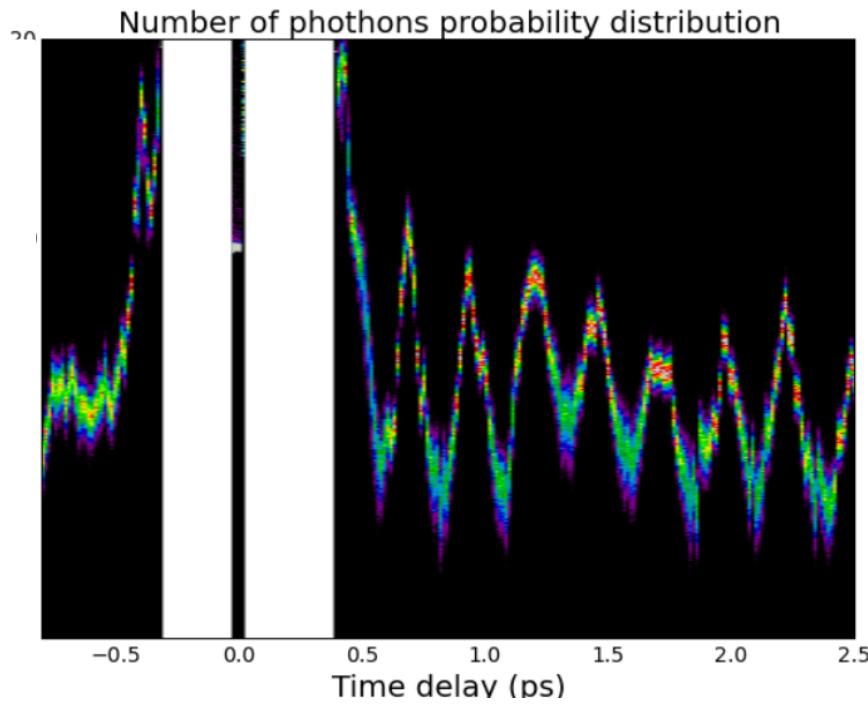
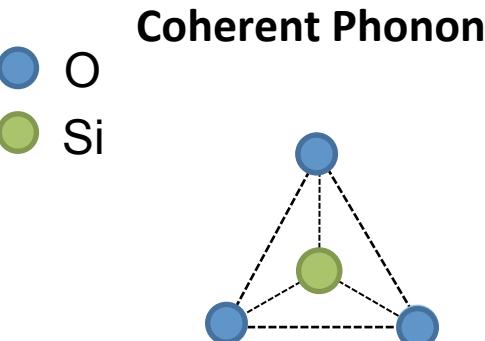
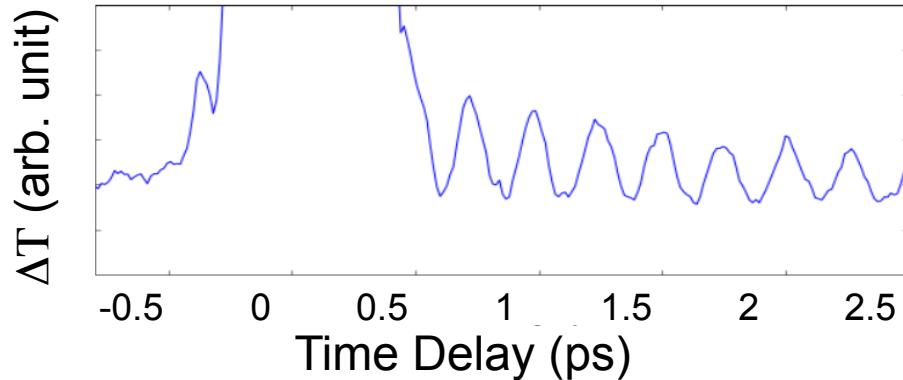


Coherent Phonon

● O
● Si

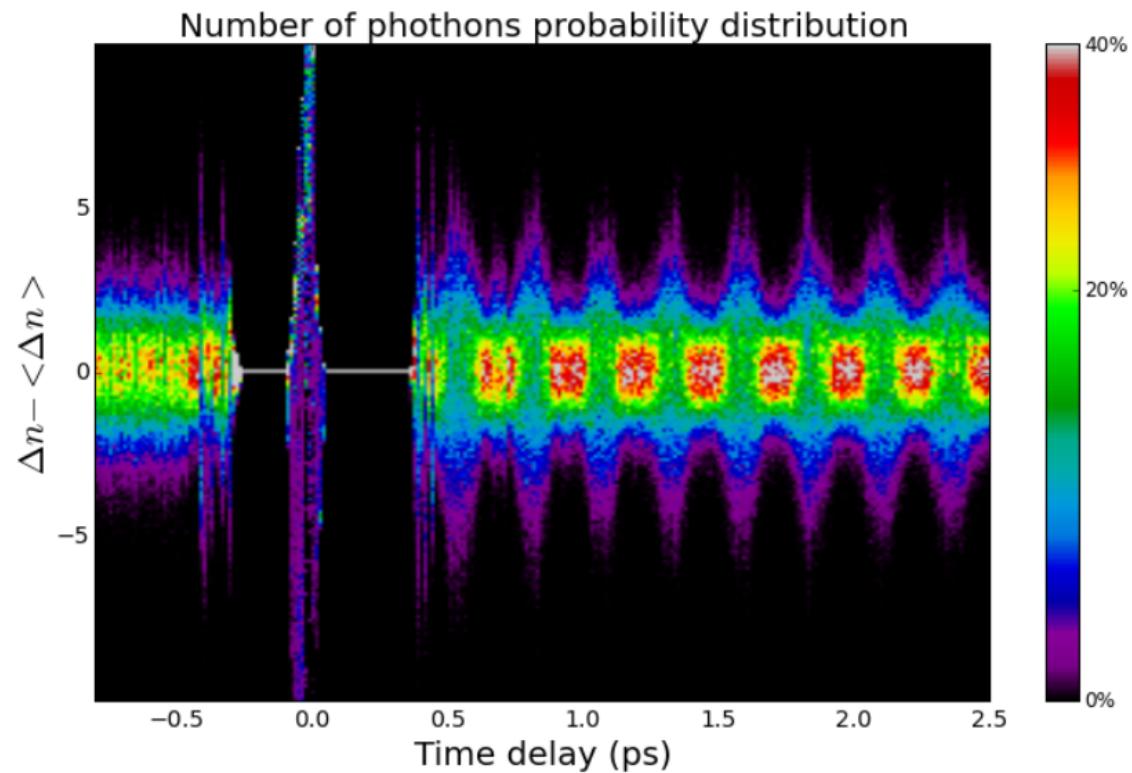
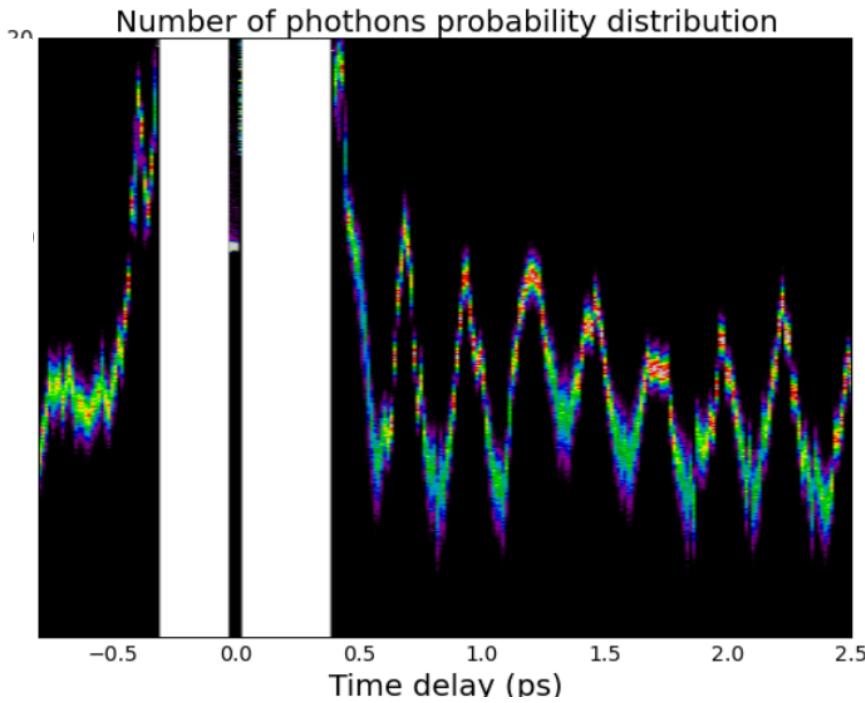
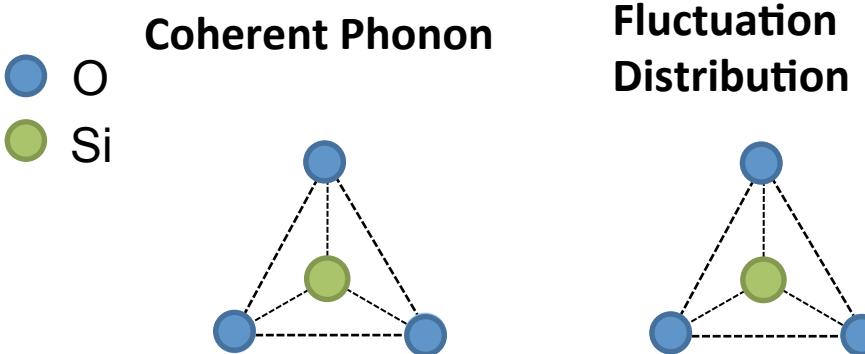
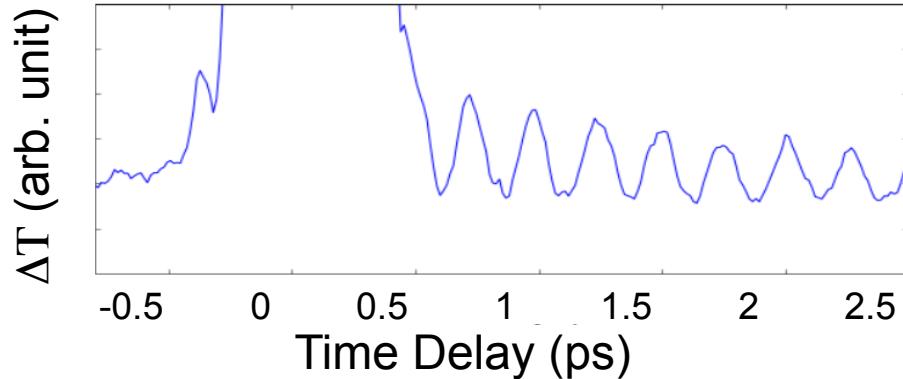


Higher order: Pump and Probe α -quartz



Nature Comm. 6, 10249 (Dec. 2015)

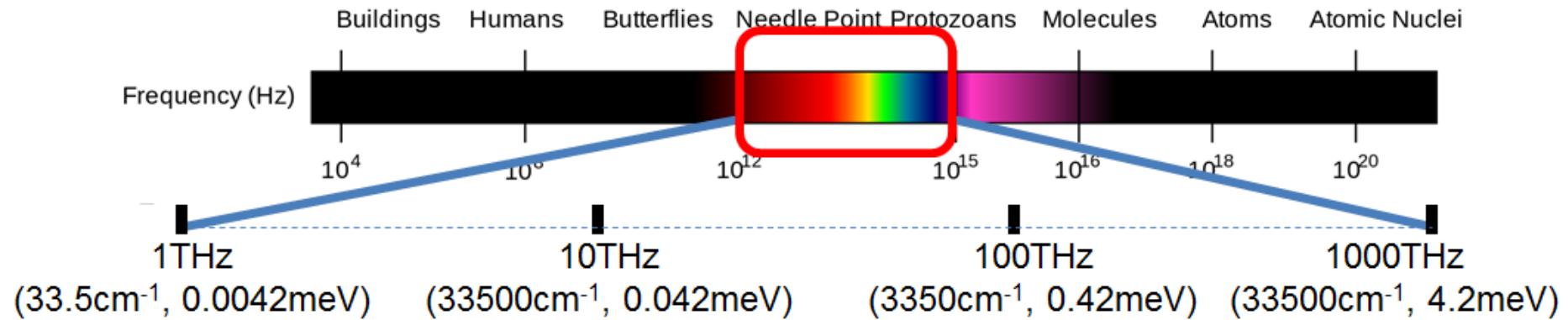
Higher order: Pump and Probe α -quartz



Nature Comm. 6, 10249 (Dec. 2015)

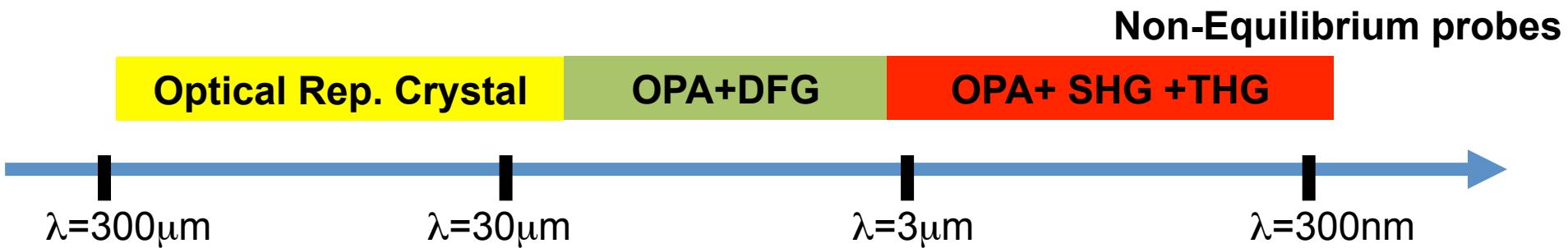
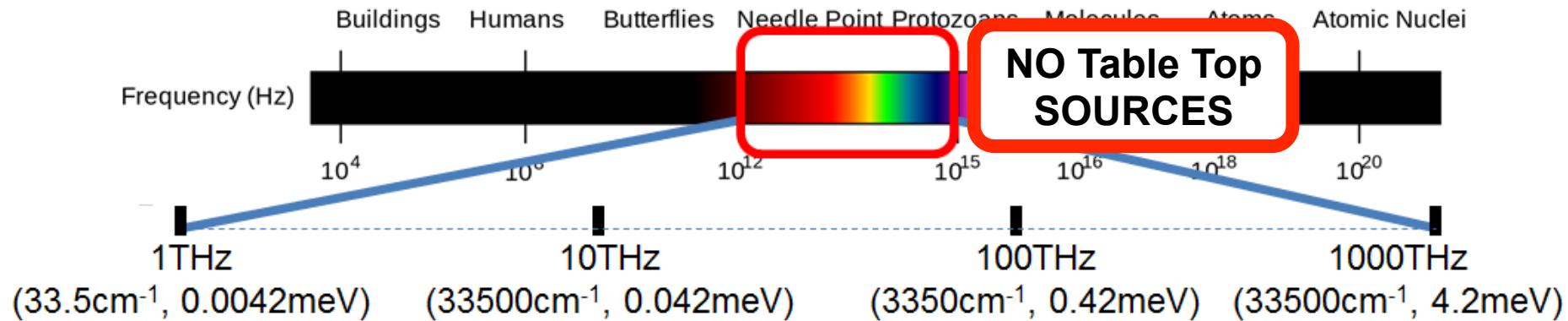
Technology overview

Table top techniques are a nice complement to synchrotron and FEL facilities



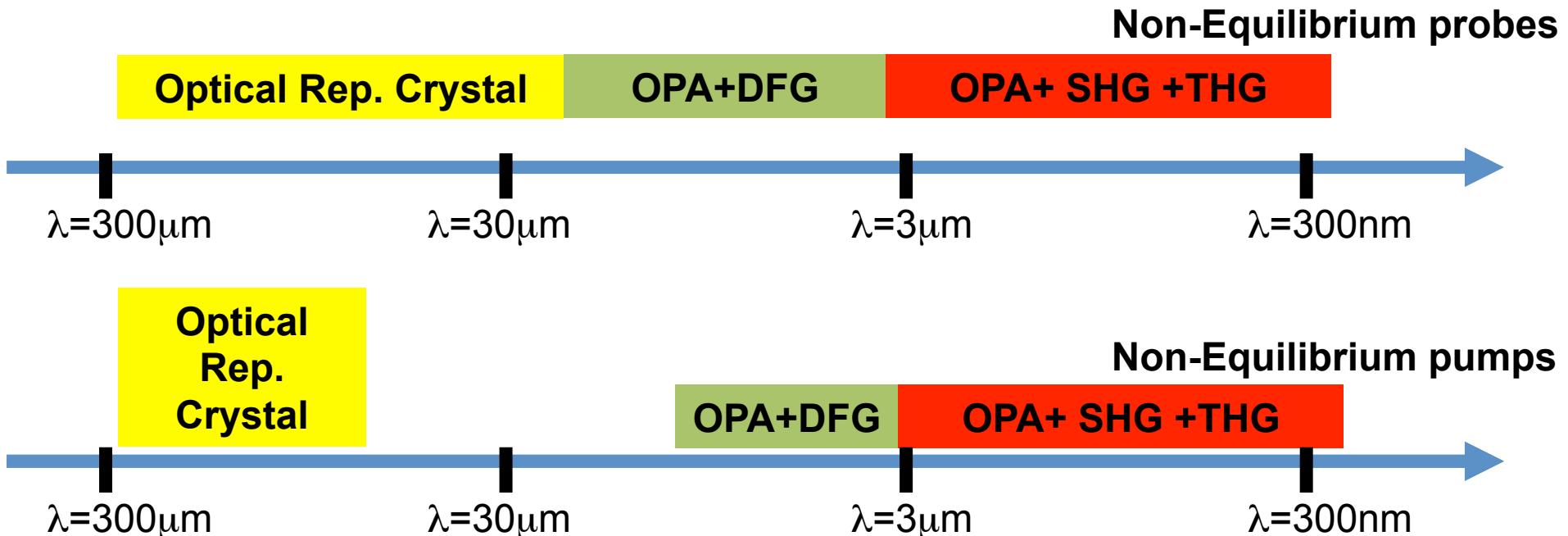
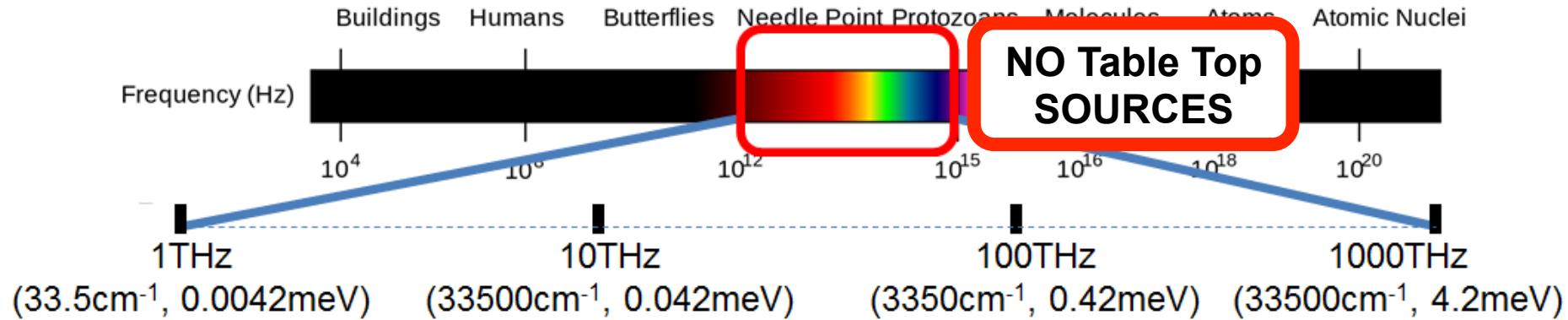
Technology overview

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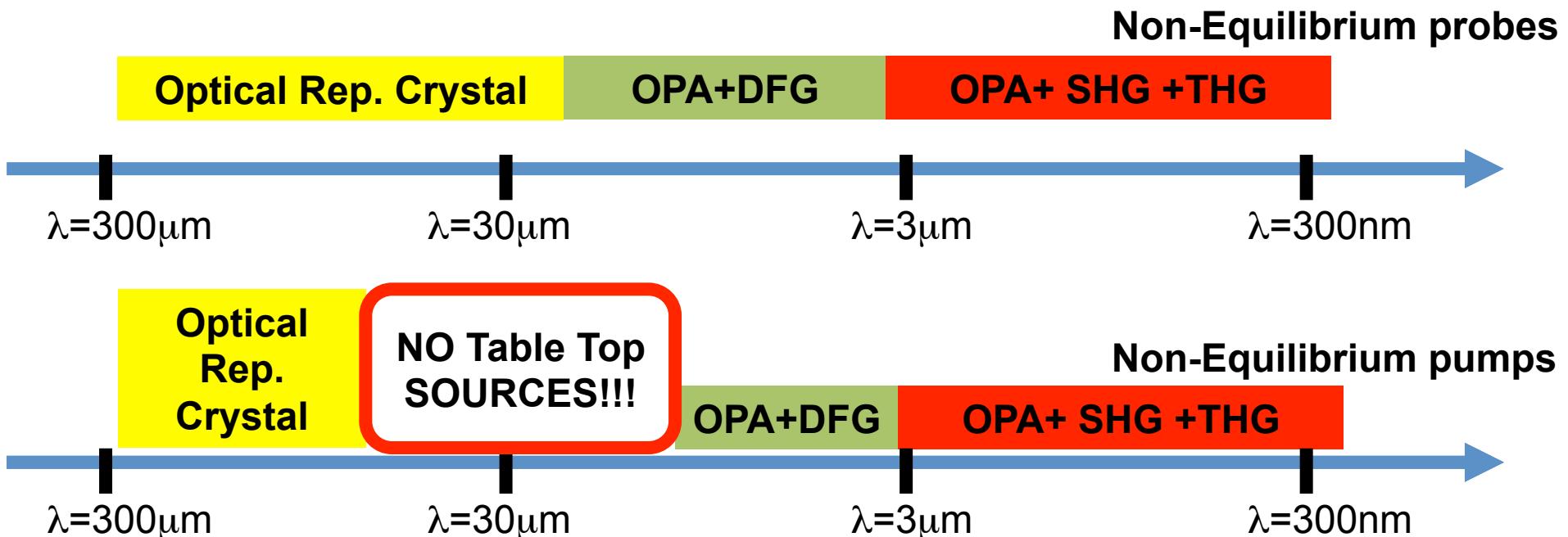
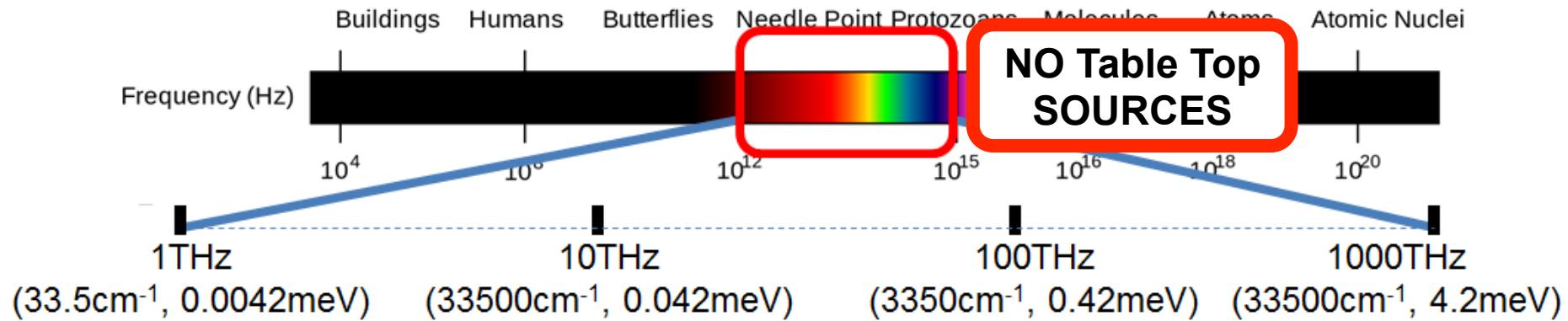
Technology overview

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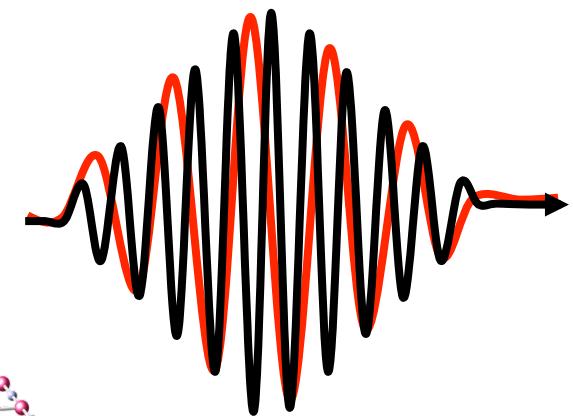
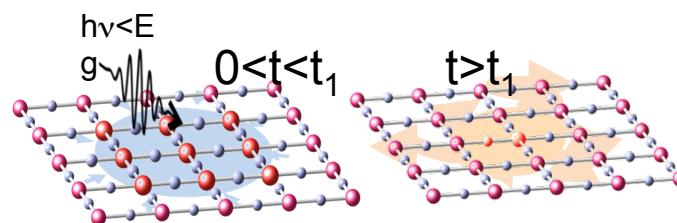
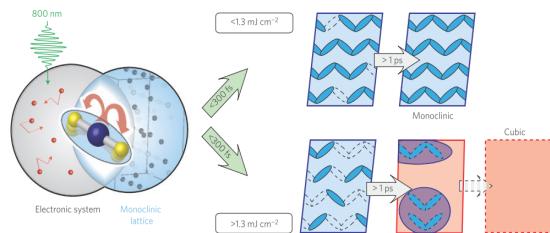


Conclusions

✓ The light pulses do not “just” inject energy into the system

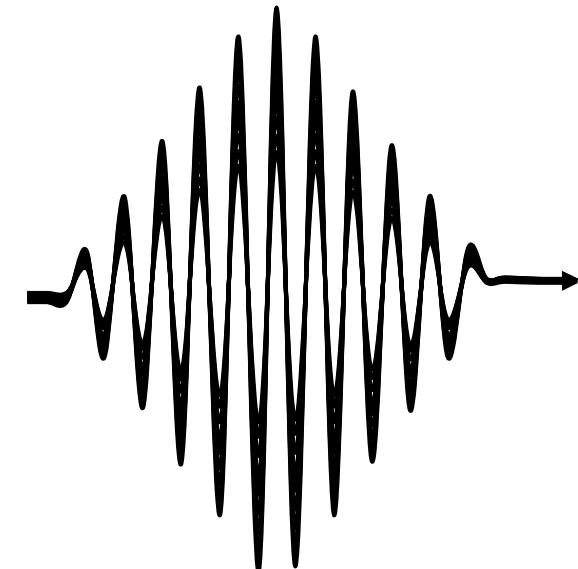
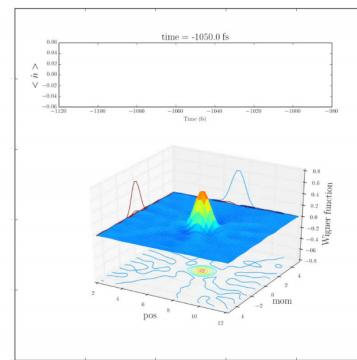
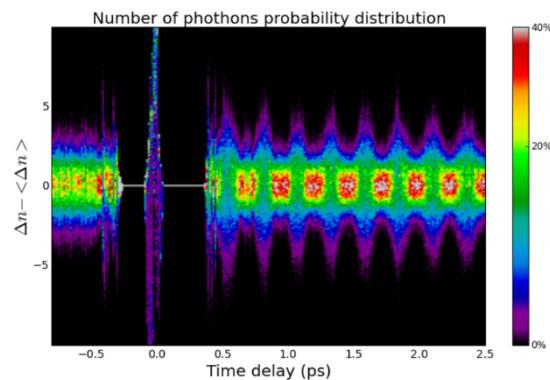
✓ Classical Parameters of the light pulses:

- Spectrally selective excitation to understand equilibrium
- Selective excitation to control functionalities



✓ Use the **Quantum State** of light pulses as a spectroscopic tool

- Photon number fluctuation (Coh. Vs. Squeezed states)
- Quantum state reconstruction



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Tom Fennell (PSI, Villigen)
Franziska Hammerath (IFW, Dresden)
Oleg Janson (TU Wien)
Mathieu Le Tacon (MPIKF, Stuttgart)
Matteo Mitrano (University of Illinois UC)
Marco Moretti Sala (ESRF, Grenoble)
Yusuke Nomura (Ecole Polytechnique, Palaiseau)
Gerald Knizia (Penn State University)
Suchitra Sebastian (Cambridge University)
Lev Vidmar (Penn State University)
Simon Wall (ICFO, Barcelona)
Cedric Weber (King's College, London)

DATES

June 15: Abstract submission
July 1: Acceptance notification
July 15: Early bird registration
September 1: Final registration

ORGANIZERS

Daniele Fausti (Elettra, Trieste)
Adriano Amaricci (SISSA, Trieste)
Michael Sentef (MPSD, Hamburg)
Edwin Kermarrec (LPS, Orsay)

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