Quantum Condensed Matter Dynamics

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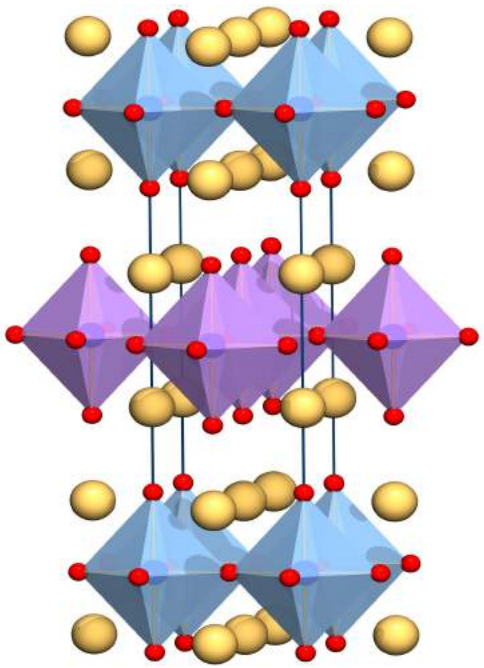
Lecture 1: Techniques

Quantum Condensed Matter

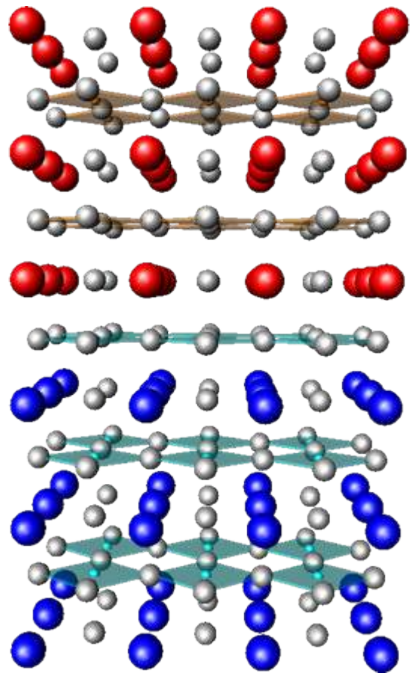
Quantum materials are a group of solids in which we cannot, even qualitatively, describe collective behavior using classical physics

Complex Oxides



$La_{(2-x)}Sr_xCuO_4$

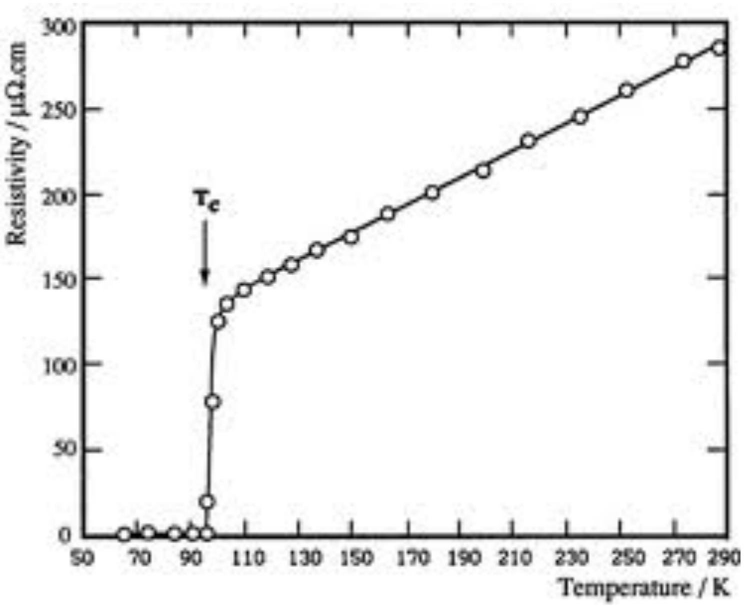
Artificial Heterostructures



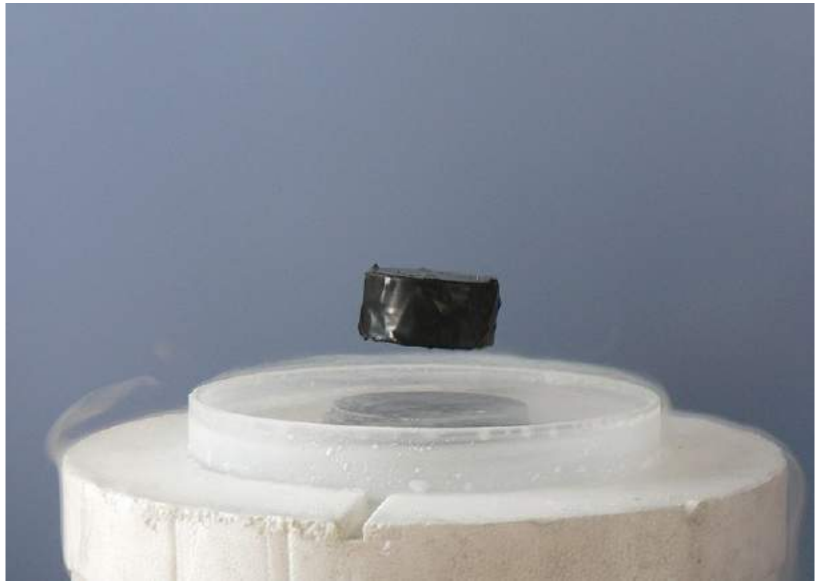
$LaAlO_3 / SrTiO_3$

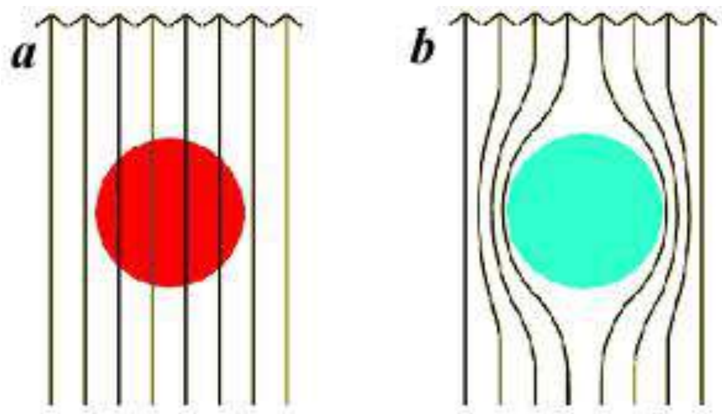
Superconductivity

Zero DC resistance



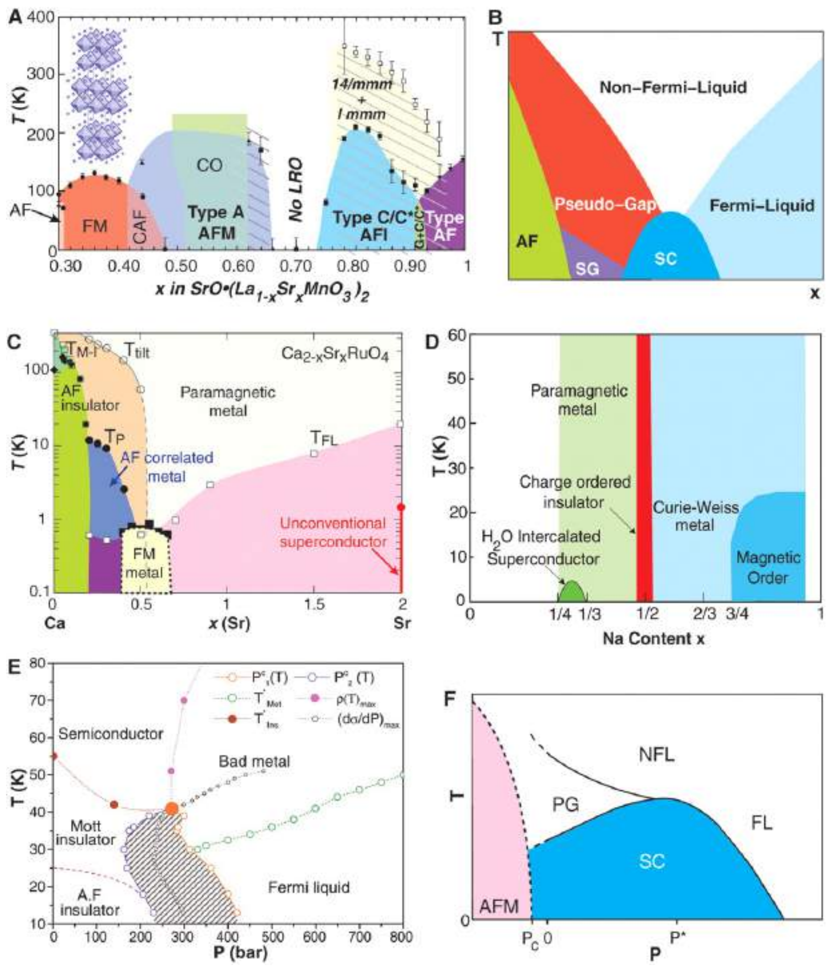
Meissner effect





Quantum Materials do "Big Things"

Highly nonlinear materials: they often possess a variety of competing phases with unconventional properties



Metal-insulator transitions

Colossal magnetoresistance

High-temperature superconductivity

E. Dagotto, Science 309, 257 (2005)

Quantum Materials are “Complex”

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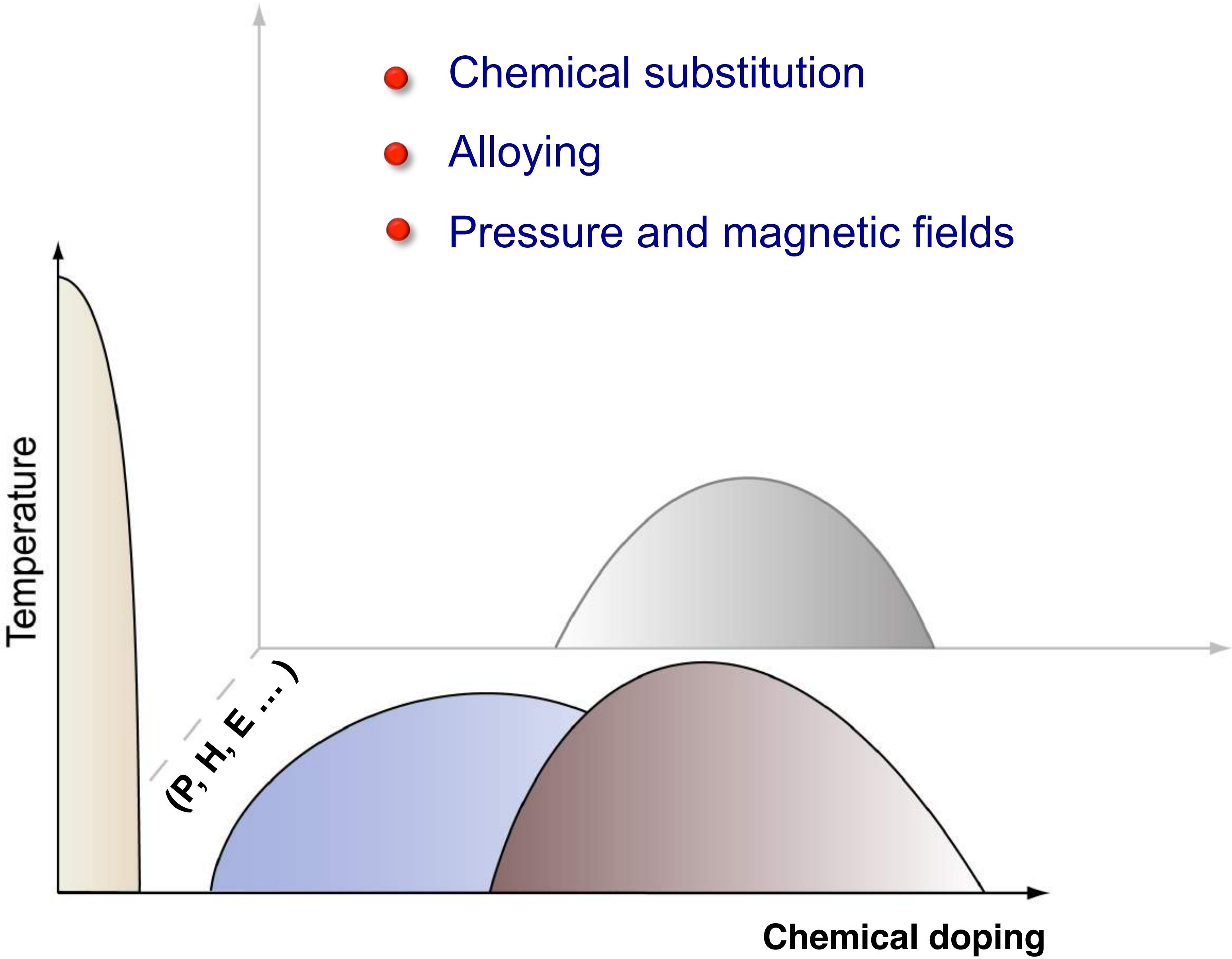
Strong correlations produce collective giant responses to small external perturbations.

Such responses are often functionally relevant.

One important goal is to CONTROL complex materials, induce these phenomena at higher temperatures or amplify their responses

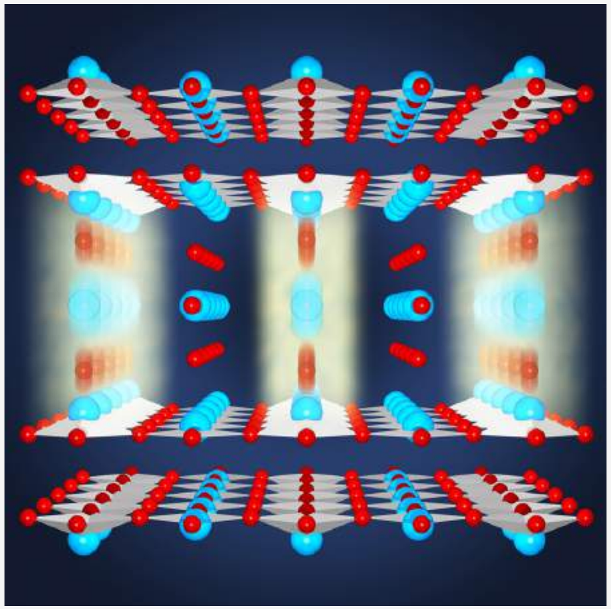
Control and Optimization: Established Routes

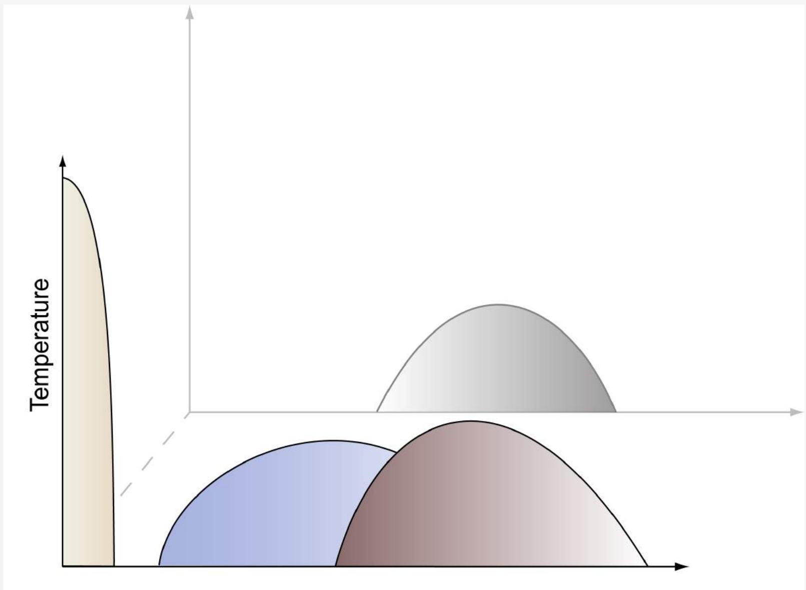
- Chemical substitution
- Alloying
- Pressure and magnetic fields



New physics away from equilibrium

Dynamical modulation can create effective Hamiltonians with new stable states





Driven systems: new energy landscapes

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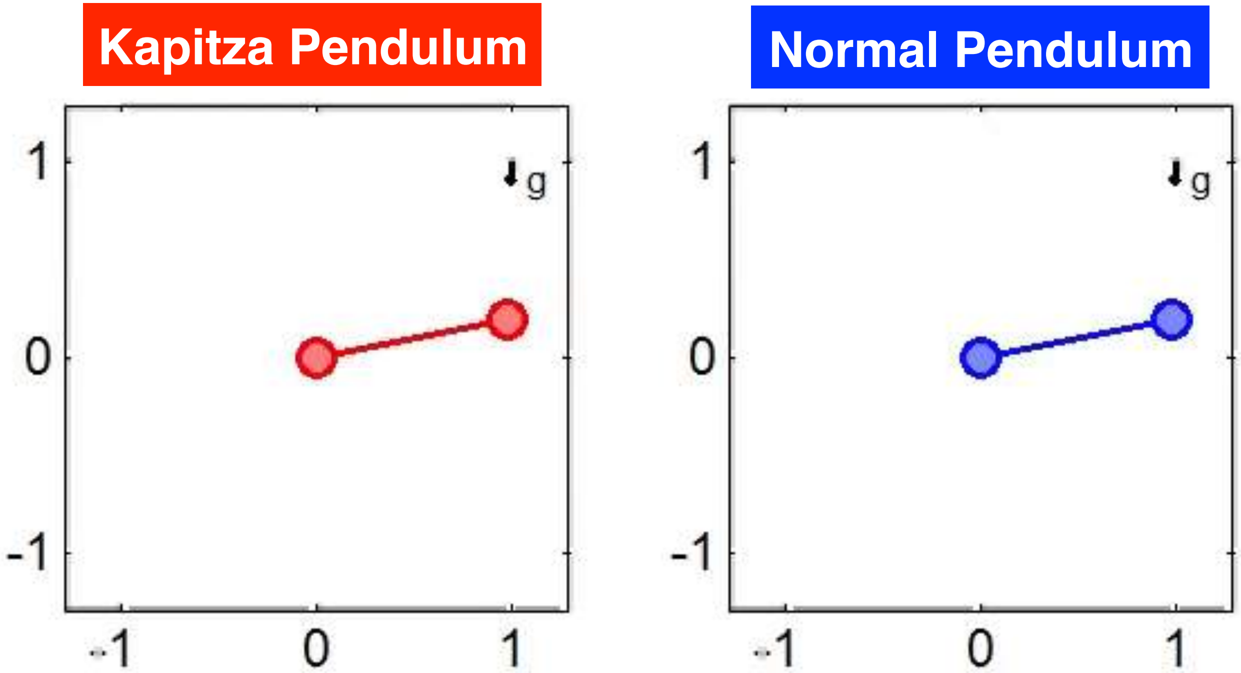
Take a pendulum and vibrate its pivot point:

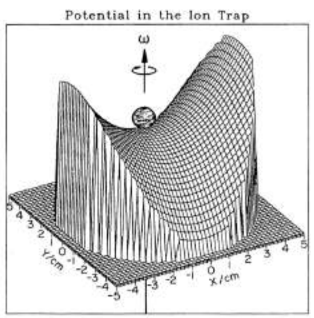


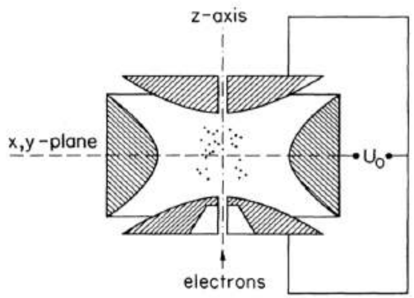
P.L. Kapitza, "Dynamic stability of a pendulum with an oscillating point of suspension,"
Zh. Eksp. Teor. Fiz. 21, 588 (1951)

L.D. Landau and E.M. Lifschitz *Mechanics* (Pergamon, Oxford 1976)

Driven systems: new energy landscapes

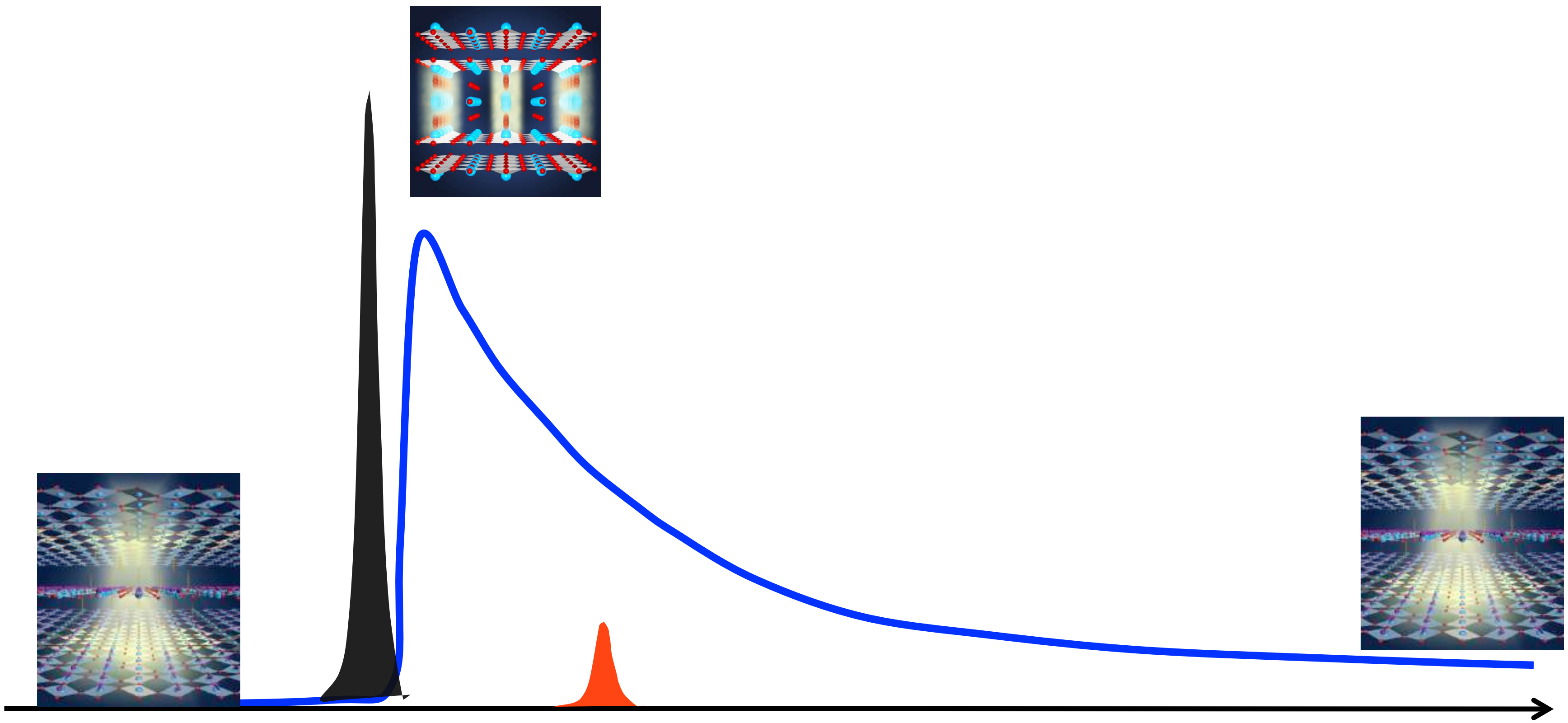
Take a saddle and spin it:





W. Paul, Nobel Lecture in *Rev. Mod. Phys. 62, 531 (1990).*

The Pump Probe Technique



The Pump

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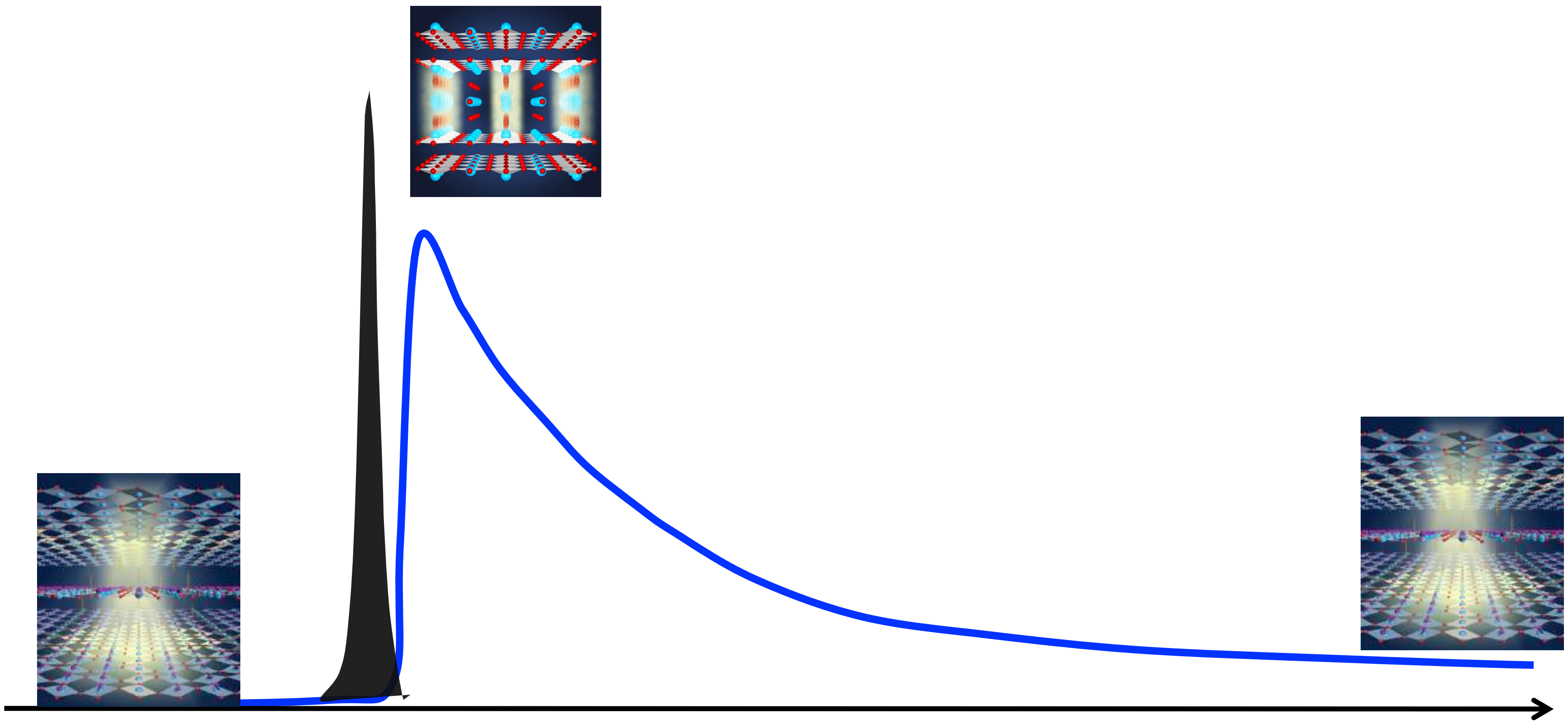
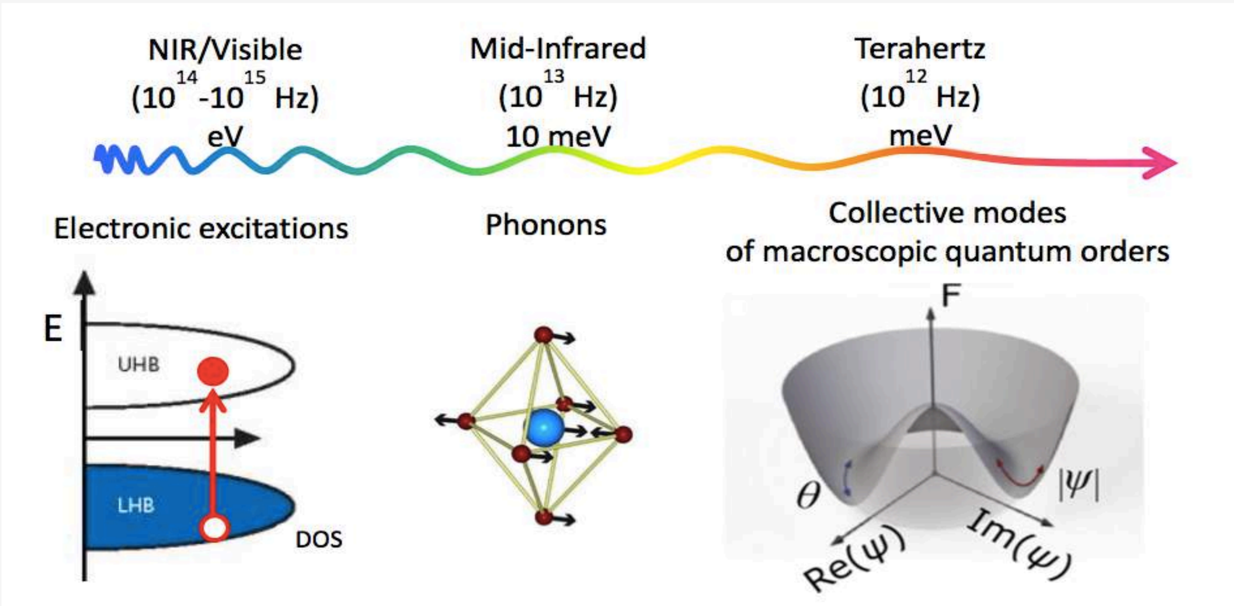


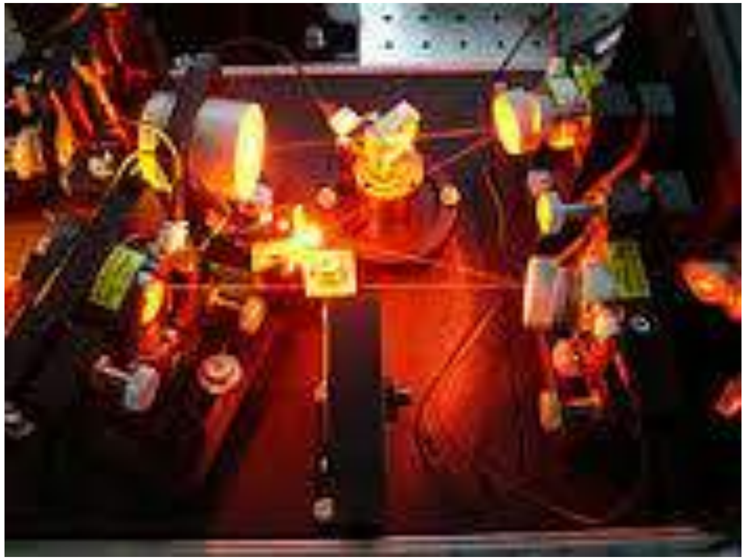
Excitation of solids: frequency scales

Driving frequency

We want to excite collective modes of quantum materials beyond their linear response



The early days: the Dye Laser



Weak

Unstable

Temperamental

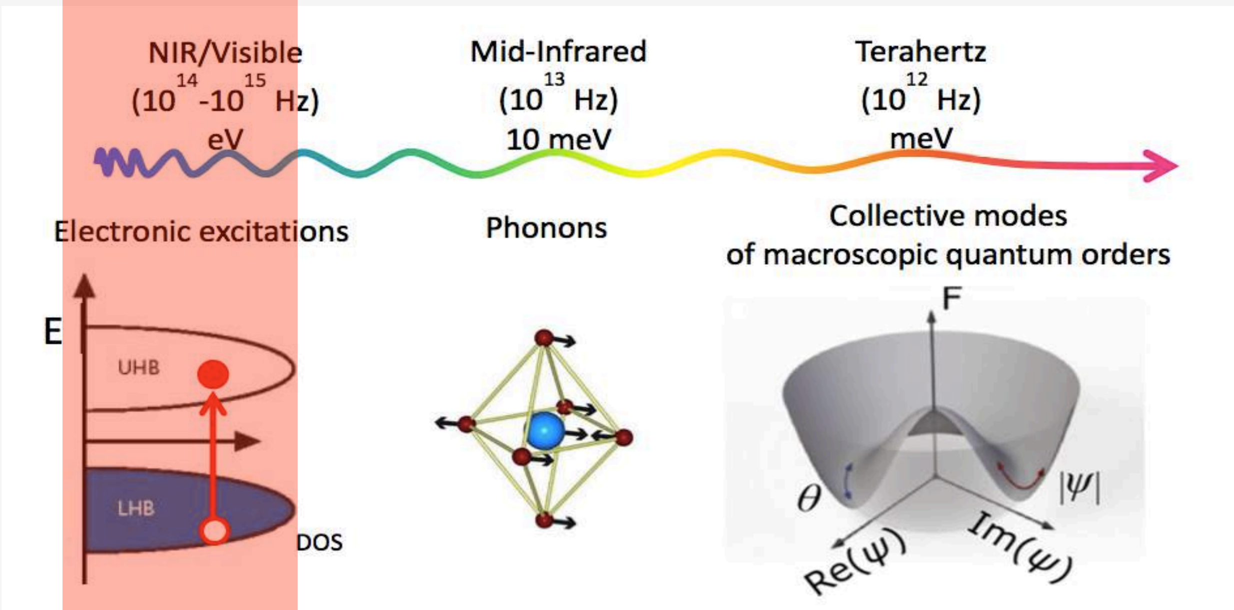
2.1 eV photon energy

………

The early days

Driving frequency

We want to excite collective modes of quantum materials beyond their linear response



Early days: optical melting of semiconductors

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VOLUME 51, NUMBER 10 PHYSICAL REVIEW LETTERS 5 SEPTEMBER 1983

Femtosecond-Time-Resolved Surface Structural Dynamics of Optically Excited Silicon

C. V. Shank, R. Yen, and C. Hirlimann
Bell Telephone Laboratories, Holmdel, New Jersey 07733

(Received 9 May 1983)

The dynamics of the structural changes that take place on a silicon surface following excitation with an intense optical pulse are observed with 90-fs time resolution. The threefold rotational symmetry of the silicon ⟨111⟩ surface becomes rotationally isotropic within a picosecond after excitation consistent with a transition from the crystalline to the liquid molten state.

PACS numbers: 68.20.+t, 64.70.Dv

VOLUME 50, NUMBER 6 PHYSICAL REVIEW LETTERS 7 FEBRUARY 1983

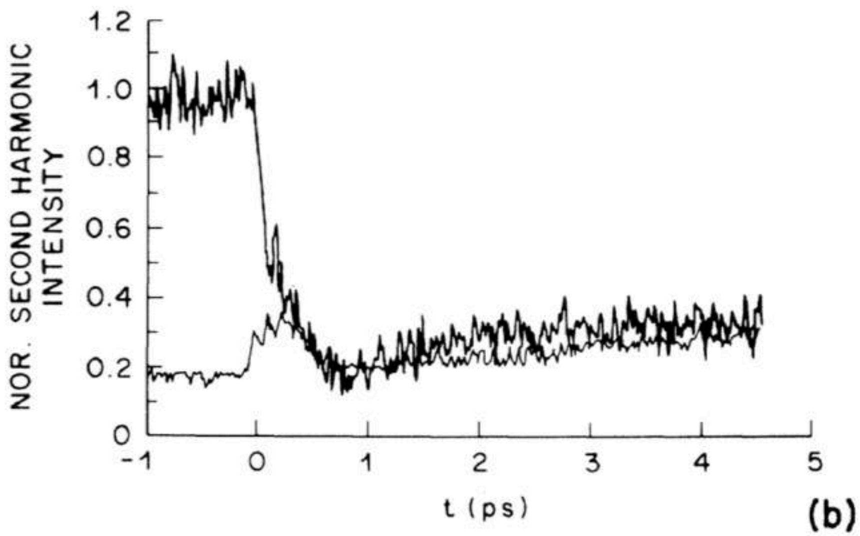
Time-Resolved Reflectivity Measurements of Femtosecond-Optical-Pulse-Induced Phase Transitions in Silicon

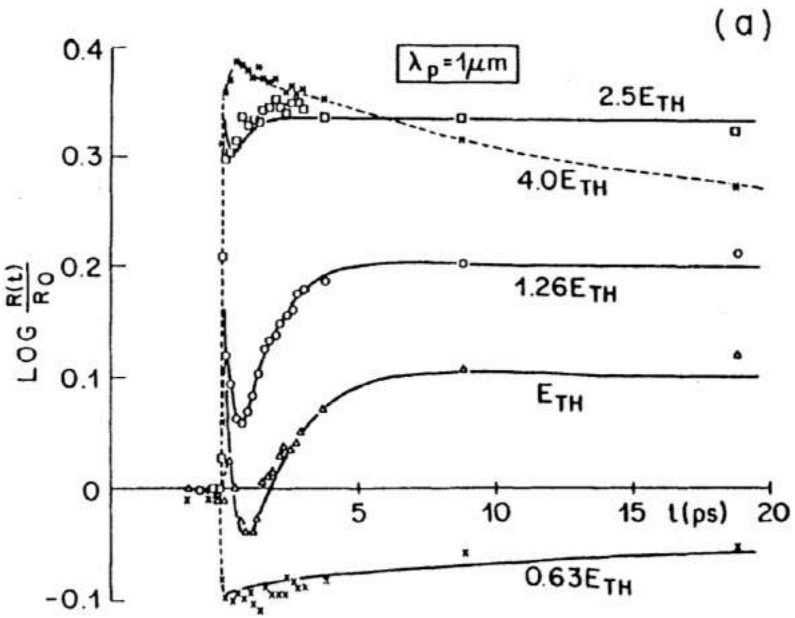
C. V. Shank, R. Yen, and C. Hirlimann
Bell Telephone Laboratories, Holmdel, New Jersey 07733

(Received 29 November 1982)

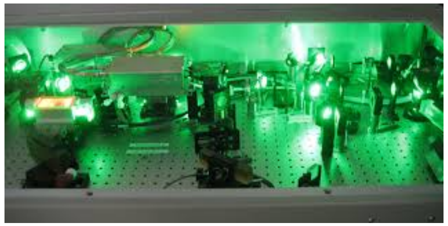
The reflectivity of silicon has been measured following excitation with intense 90-fsec optical pulses. These measurements for the first time clearly resolve in time the process of energy transfer to the crystal lattice and the dynamics of the phase transition to the melted state.

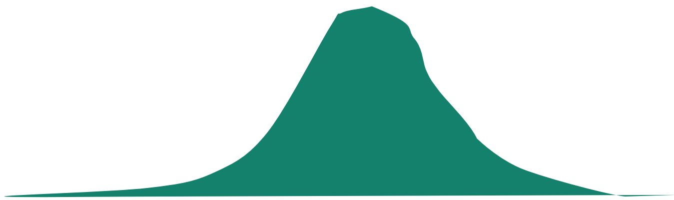
PACS numbers: 78.20.Dj, 64.70.Dv, 81.40.Tv

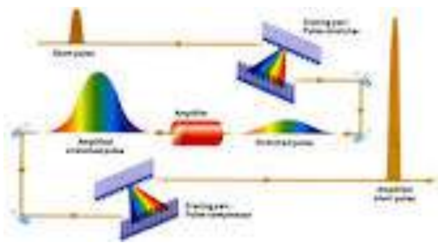




Chirped Pulse Amplified Ti:Sa Laser





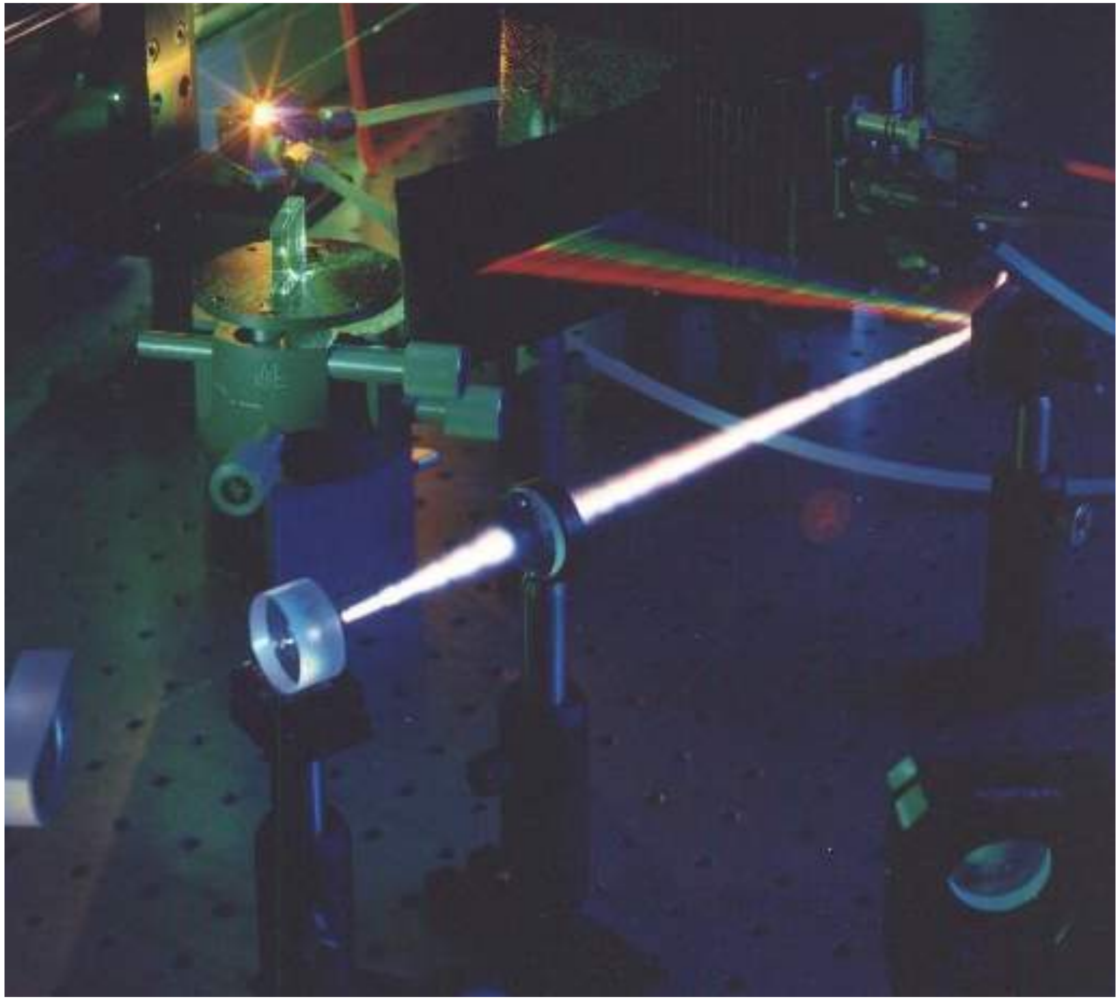


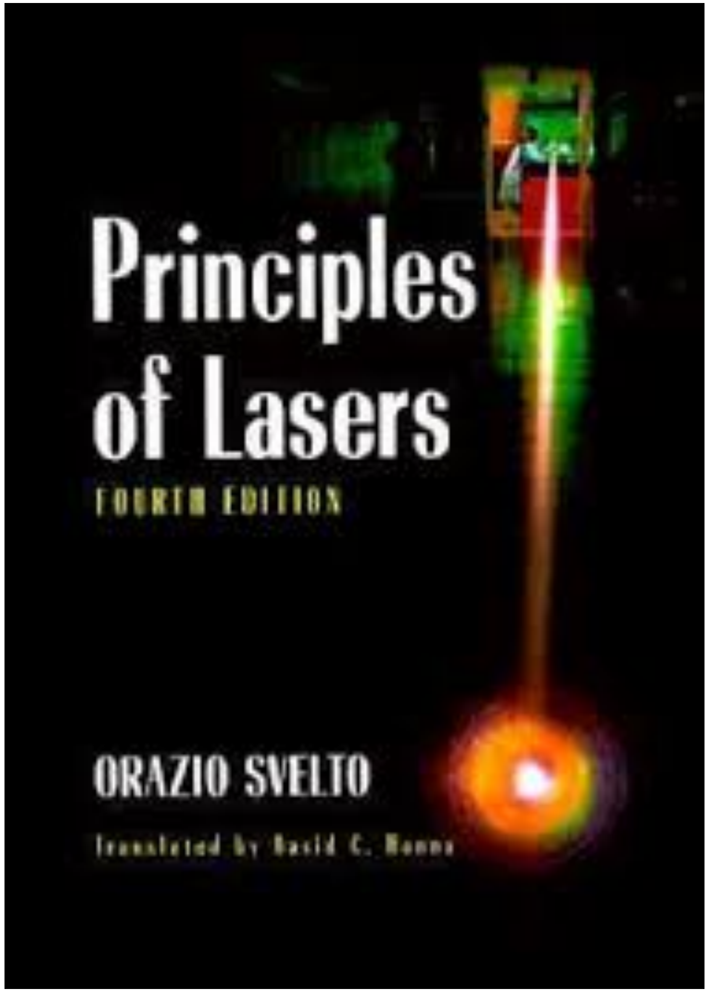
800 nm

1.5 eV

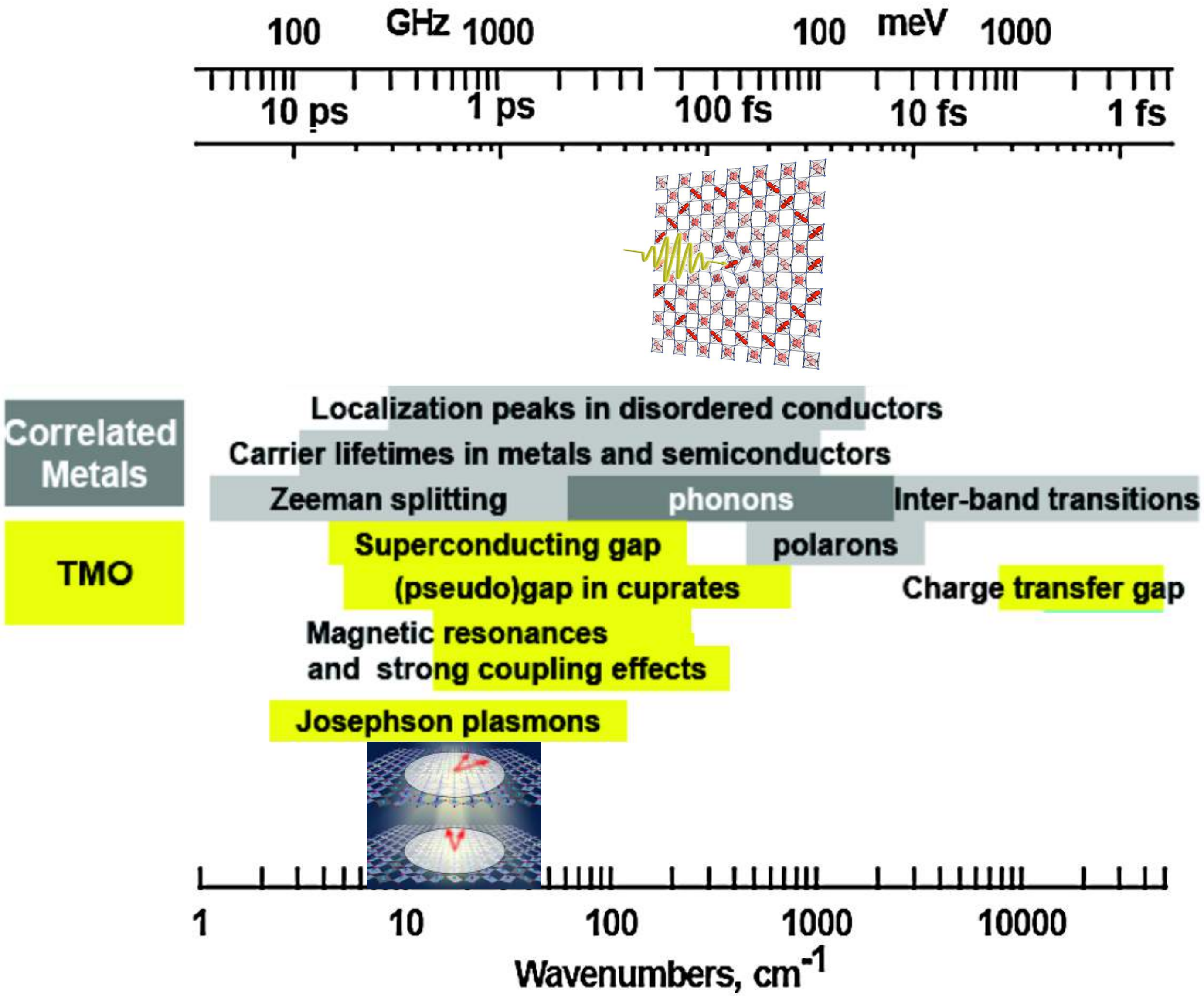
Strickland and Mourou *Opt. Comm.* 56, 219 (1985)

Intense and Stale lasers for frequency conversion





Examples given in lectures 2,3,4



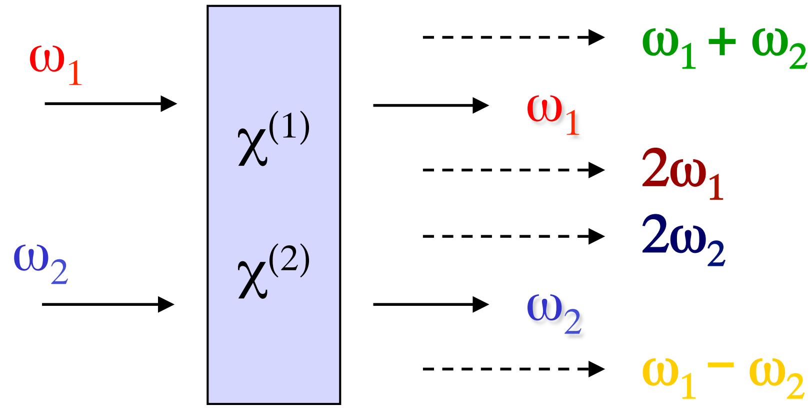
D. N. Basov *et al.*, Rev. Mod. Phys. 83,471 (2011)

Second order Nonlinear Optics

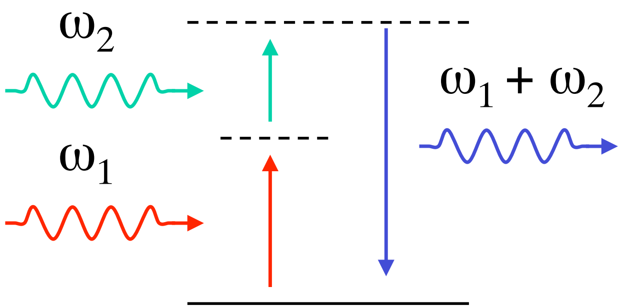
High-intensity fields

$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 + \dots$$

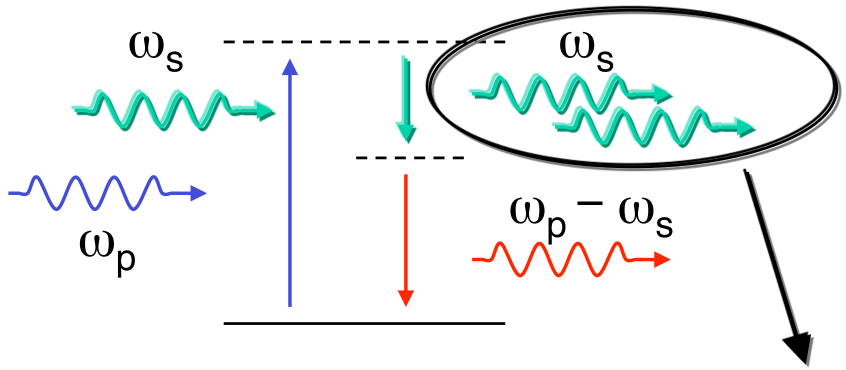
Weak fields



SFG - Sum Frequency Generation

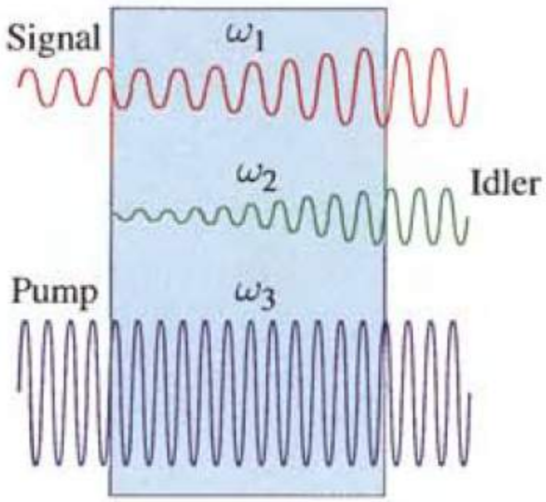


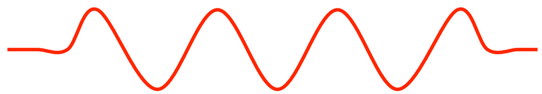
DFG - Difference Frequency Generation



OPA - Optical Parametric Amplification

Optical Parametric Amplification





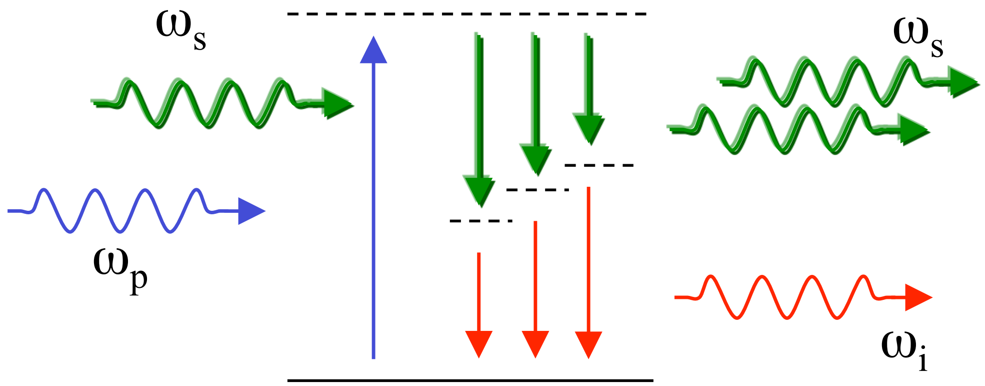
High-intensity, high-frequency *"pump"* beam

amplifies lower intensity, lower frequency *"signal"* beam

and generates third *"idler"* beam

Optical Parametric Amplification: can be tuned

mpsd



- Second order nonlinear processes require:
  - ✓ Energy conservation $\hbar\omega_s + \hbar\omega_i = \hbar\omega_p$
  - ✓ Momentum conservation $\hbar\vec{k}_s + \hbar\vec{k}_i = \hbar\vec{k}_p$

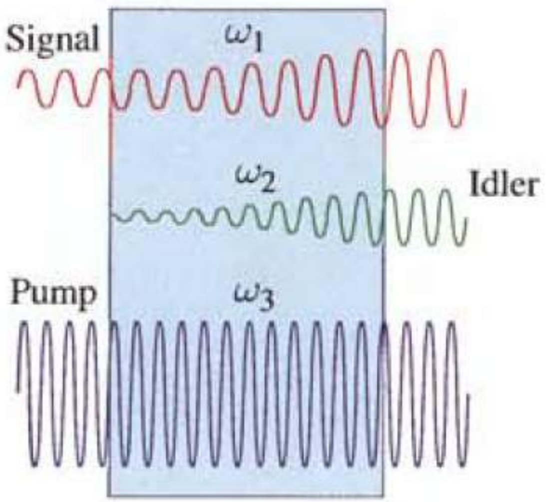
- Phase-mismatch: $\Delta\vec{k} = \vec{k}_p - \vec{k}_s - \vec{k}_i$

  - ✓ When detuning the signal frequency by $\Delta\omega$

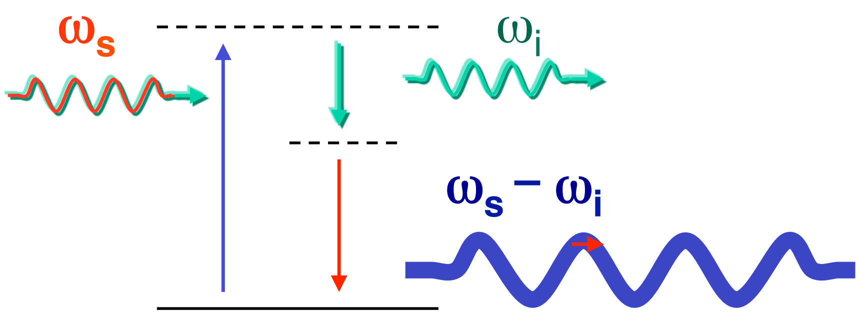
$$\Delta k = -\frac{\partial k_s}{\partial\omega}\Delta\omega + \frac{\partial k_i}{\partial\omega}\Delta\omega = \left(\frac{1}{v_{gs}} - \frac{1}{v_{gi}}\right)\Delta\omega$$

OPA + DFG

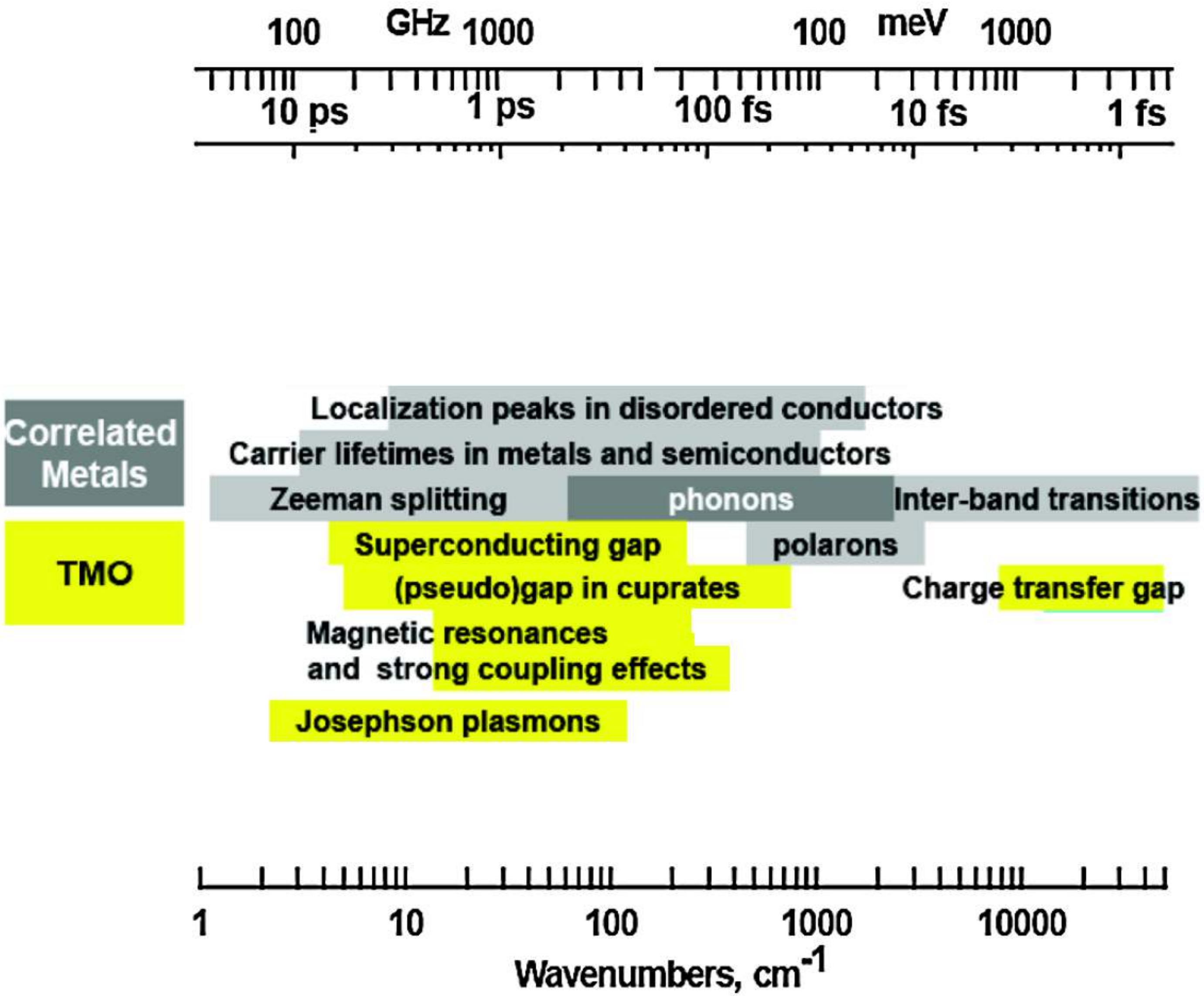
OPA – Optical Paramtriic Amplificationion



DFG - Difference Frequency Generation

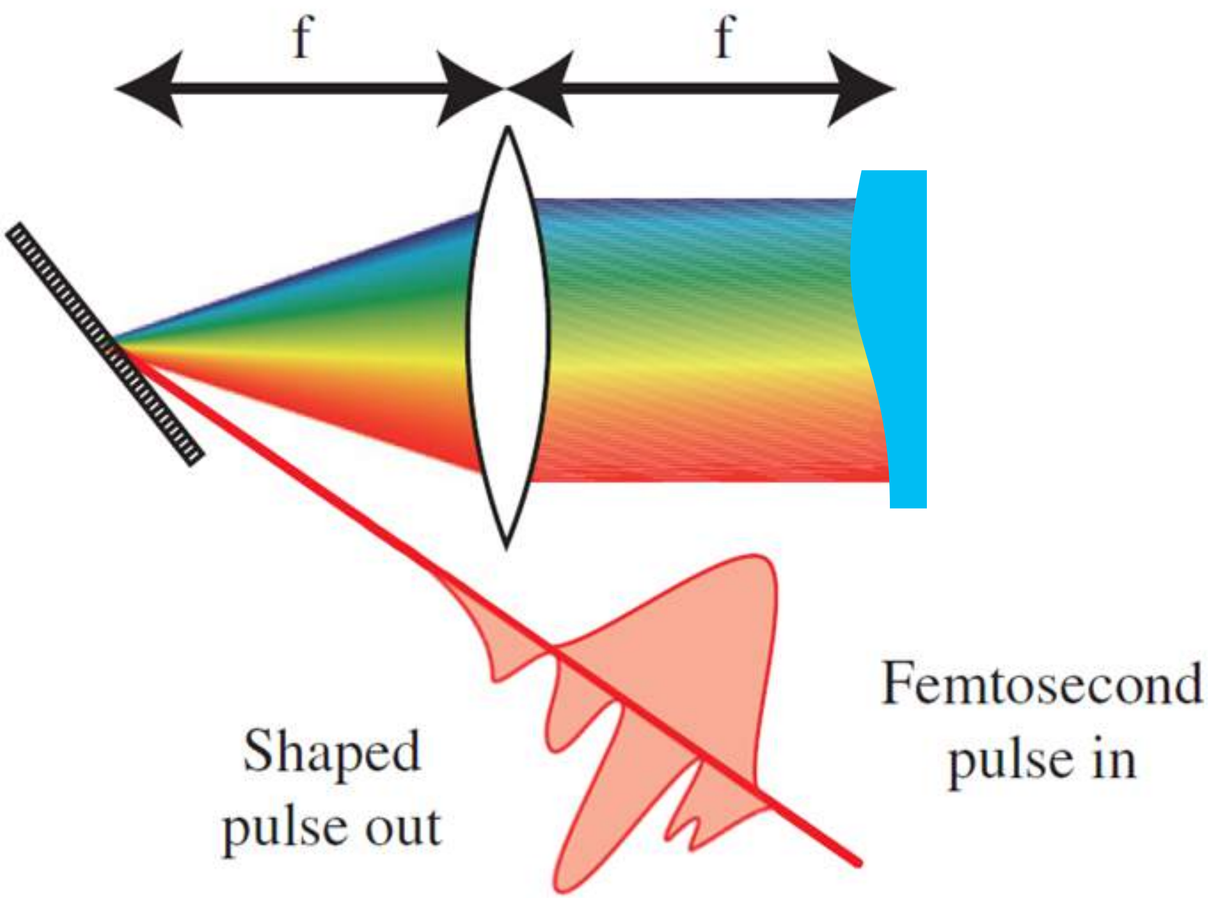


OPA + DFG cover the full spectrum



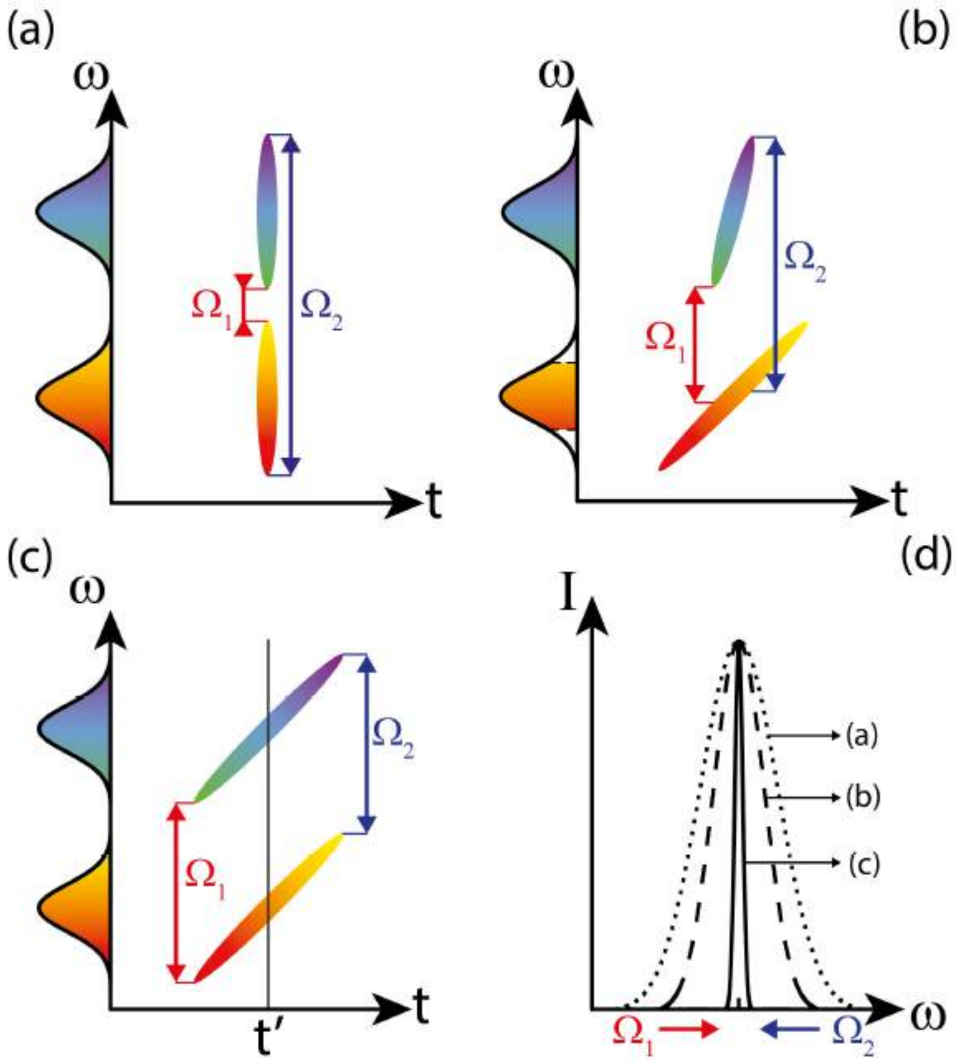
D. N. Basov *et al.*, Rev. Mod. Phys. 83,471 (2011)

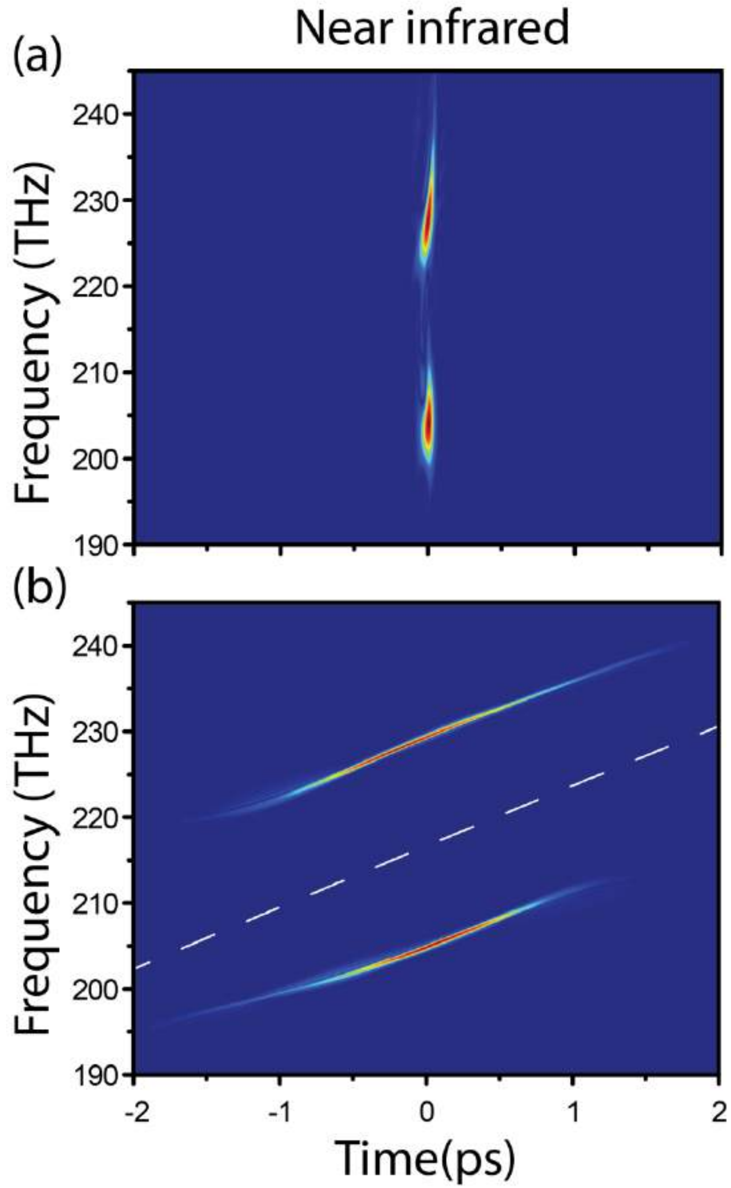
Pulses can be manipulated and sculpted



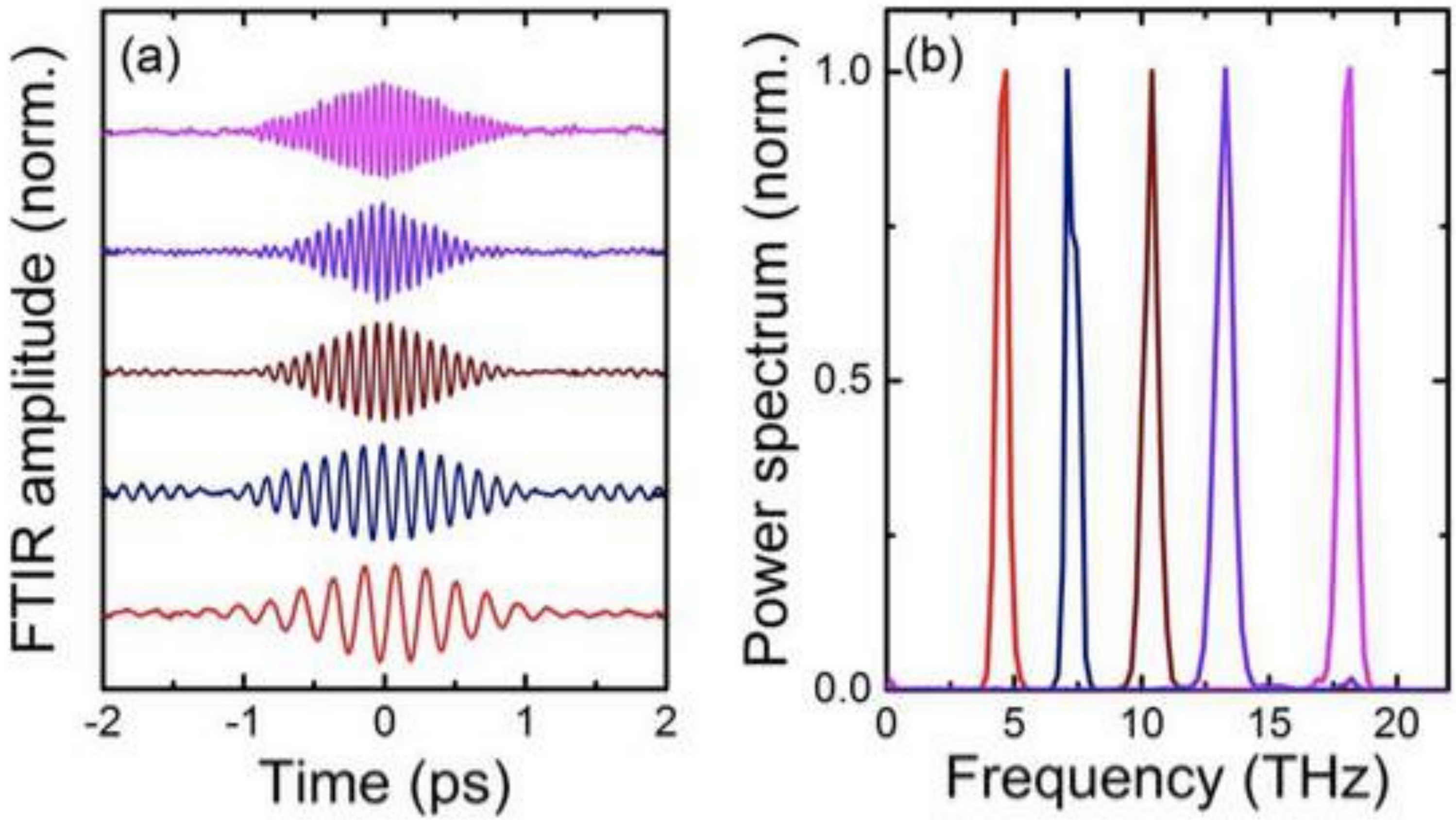
A. Cartella et et al. *Opt. Lett.* 39, 1485 (2014)

Can be made narrowband



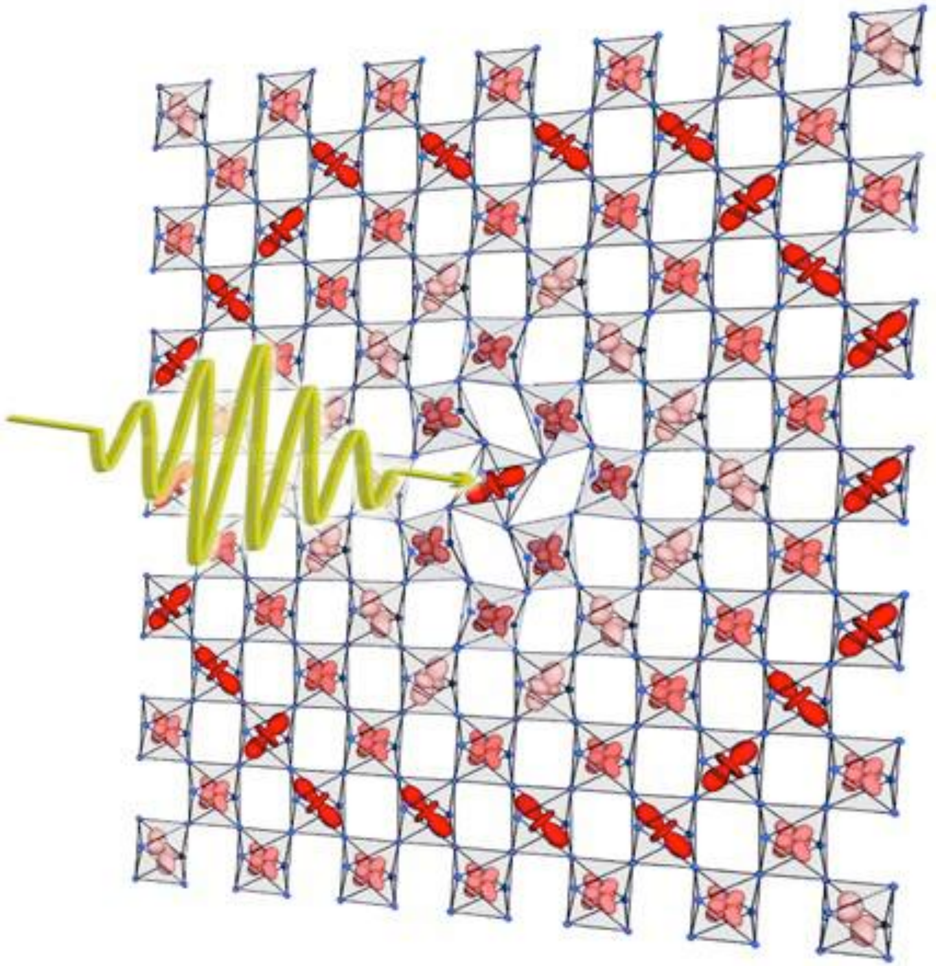


Can be made narrowband and tuned

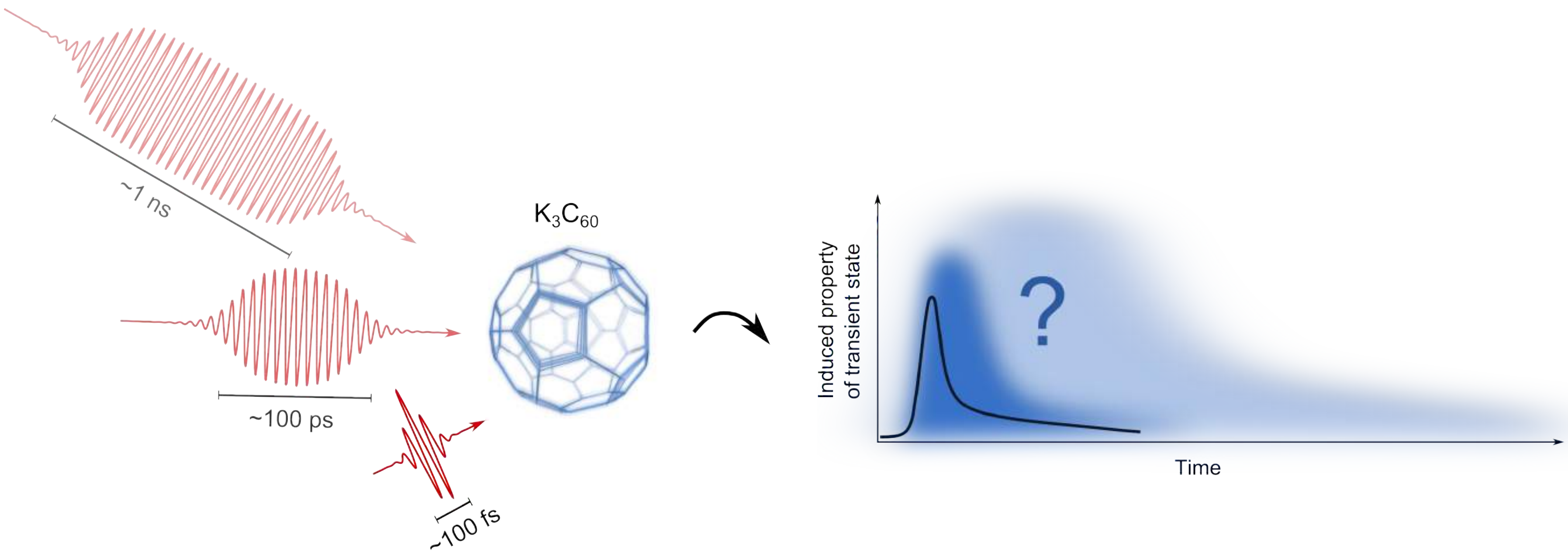


B. Liu et al. *Opt. Lett.* 41, 129 (2017)

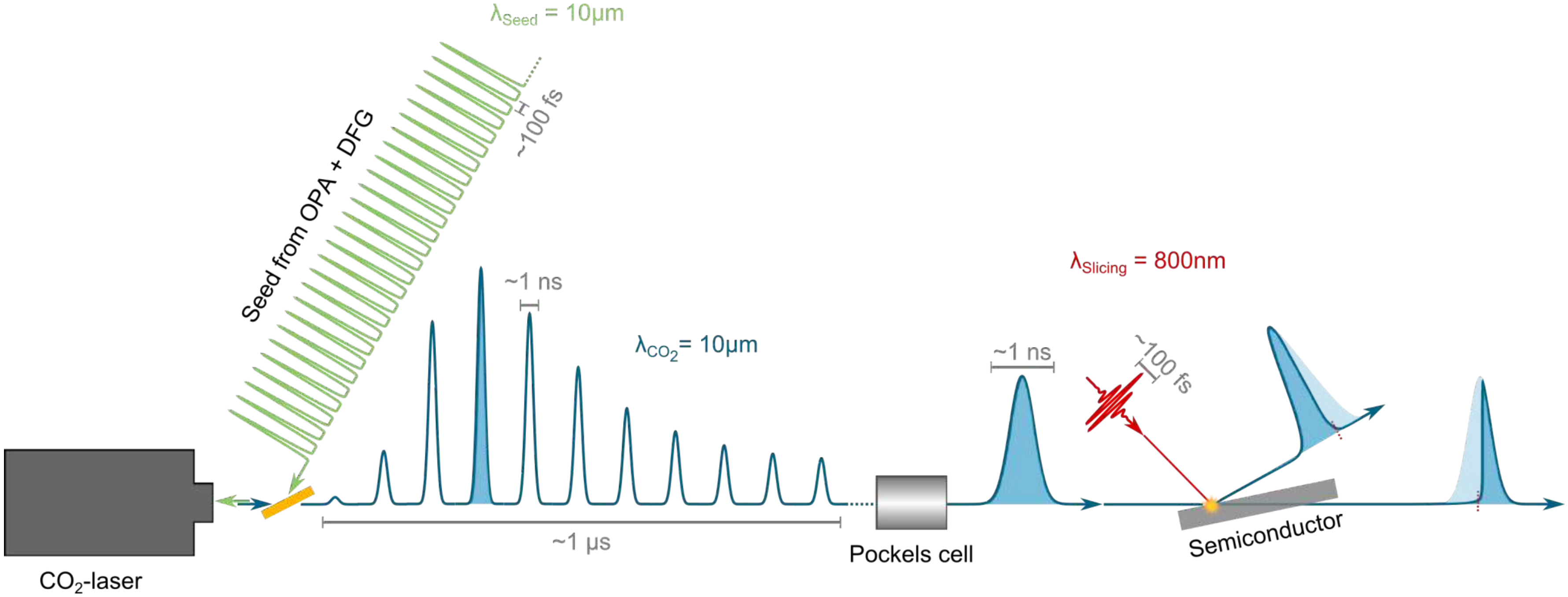
Manipulate low frequency excitations



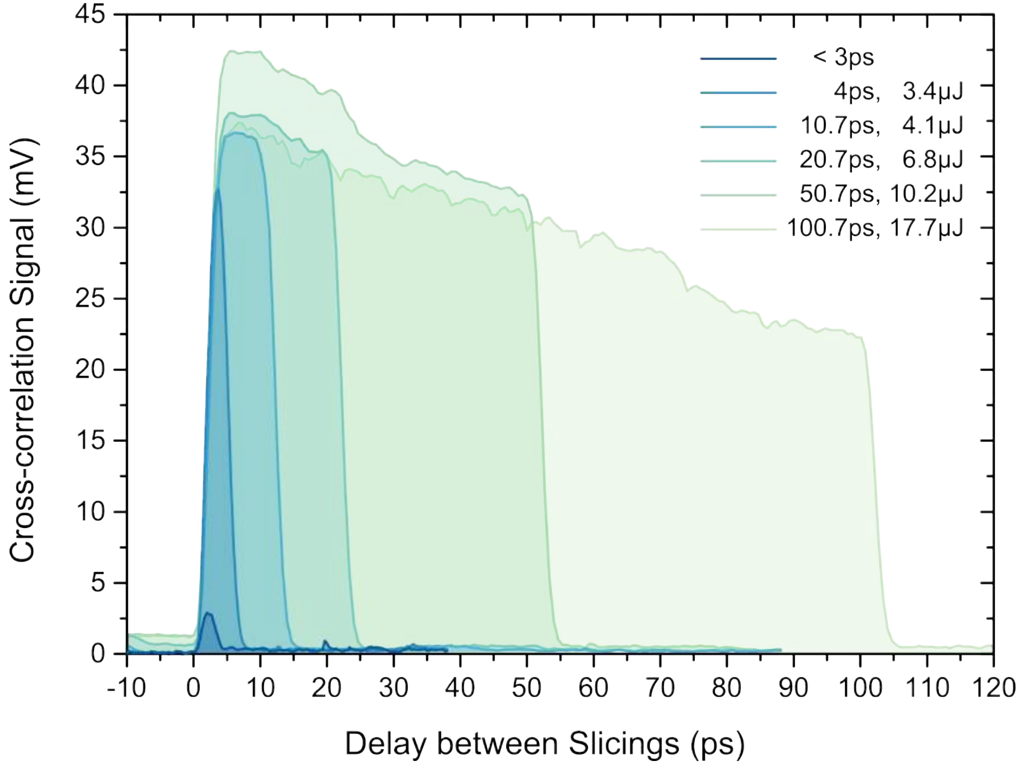
Low frequency excitations driven continuously

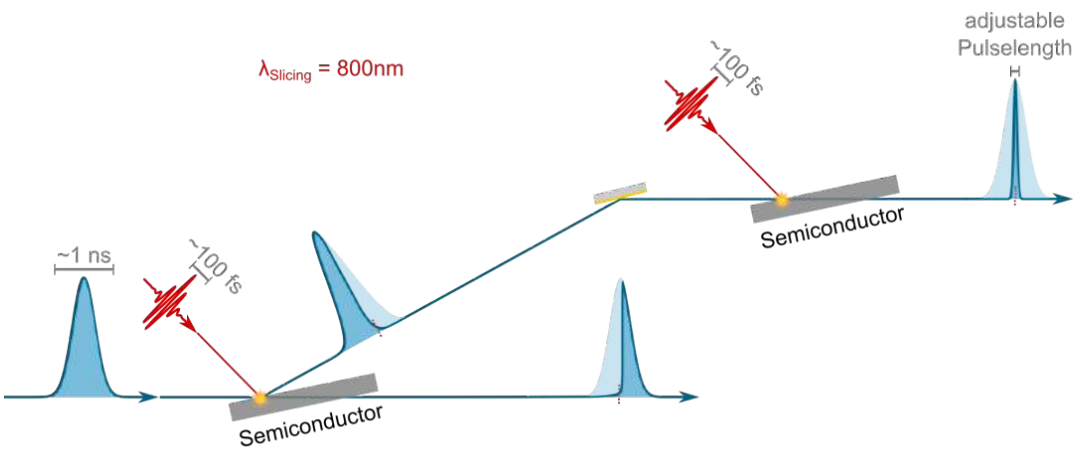


Hybrid techniques: seeded gas lasers

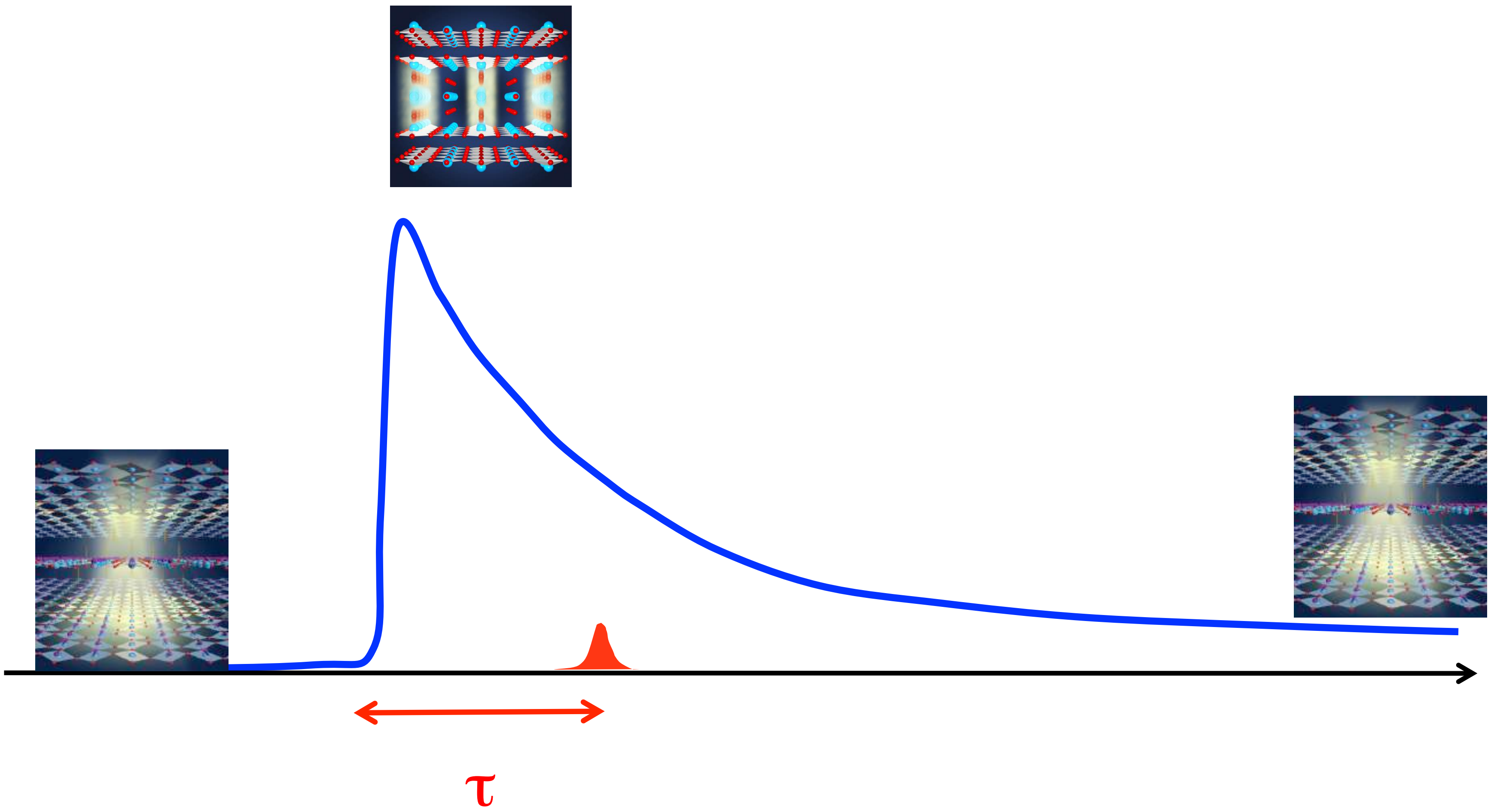


Hybrid techniques: seeded gas lasers + slicing

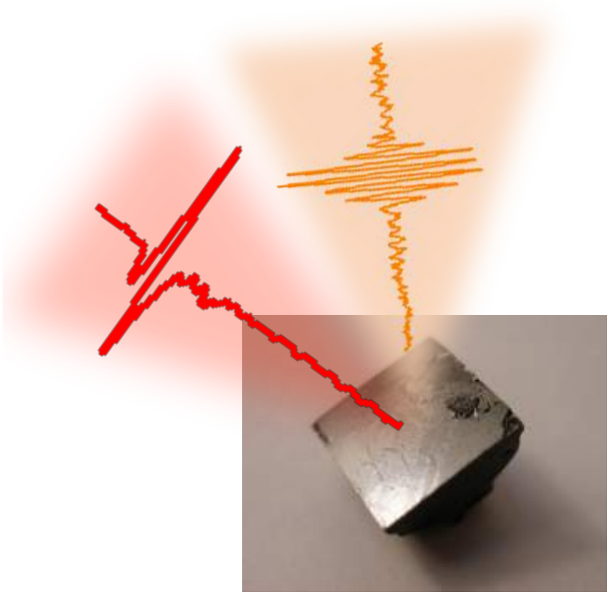


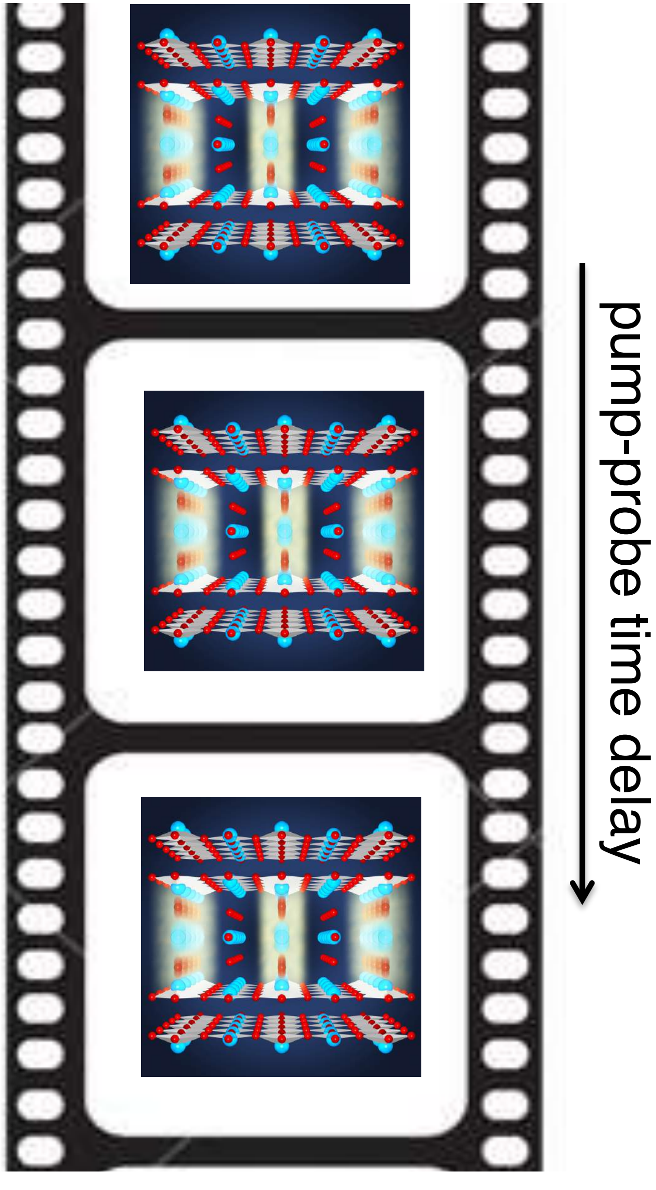


The probe



Taking snapshots of a non-equilibrium dynamics





At equilibrium: people can probe ….

1) Optical Spectrum with infrared optics

2) Spectral function with photo-emission

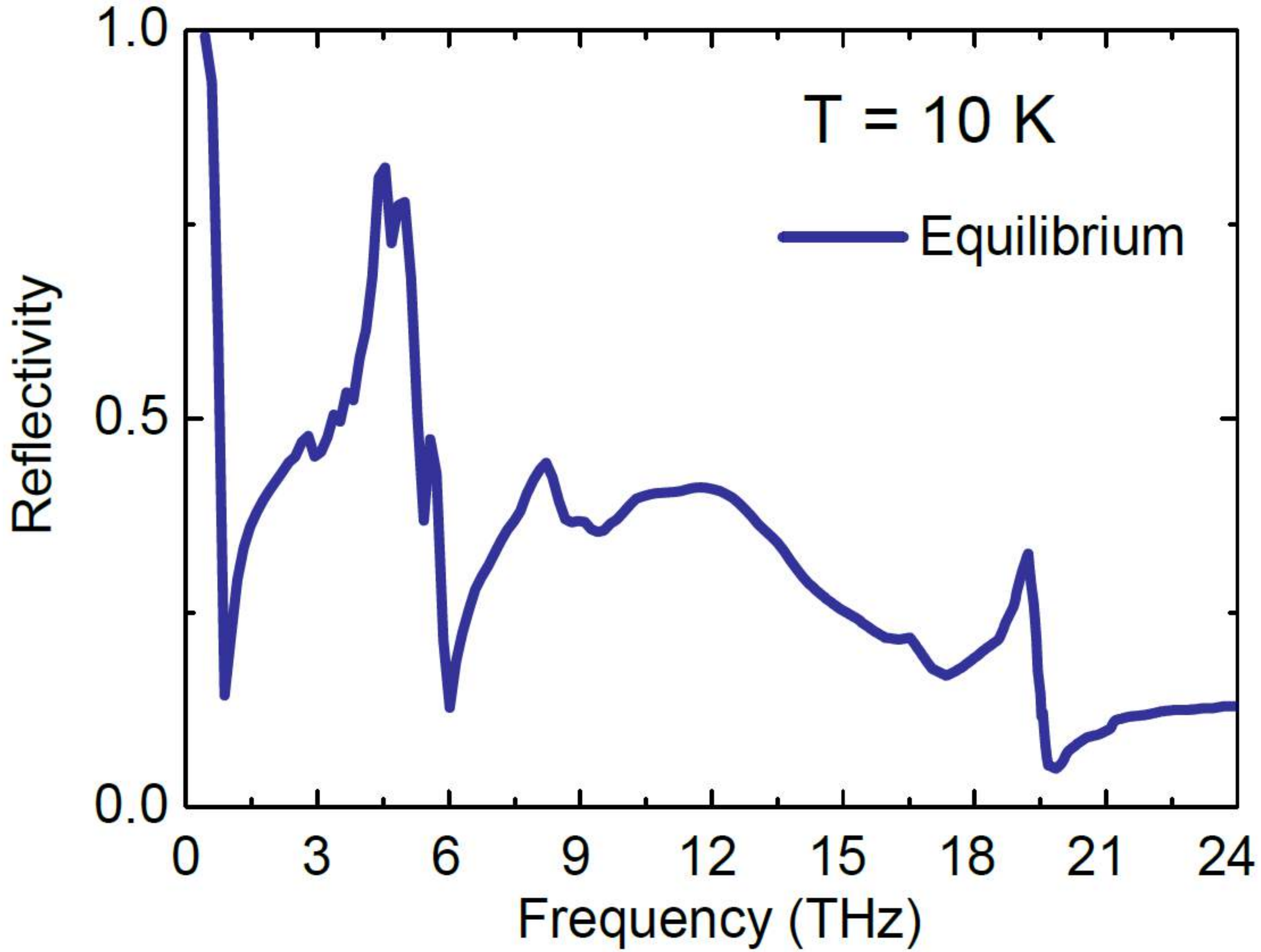
3) Density of states with Scanning Tunnellng Microscopy

4) Atomic structures with hard x-rays

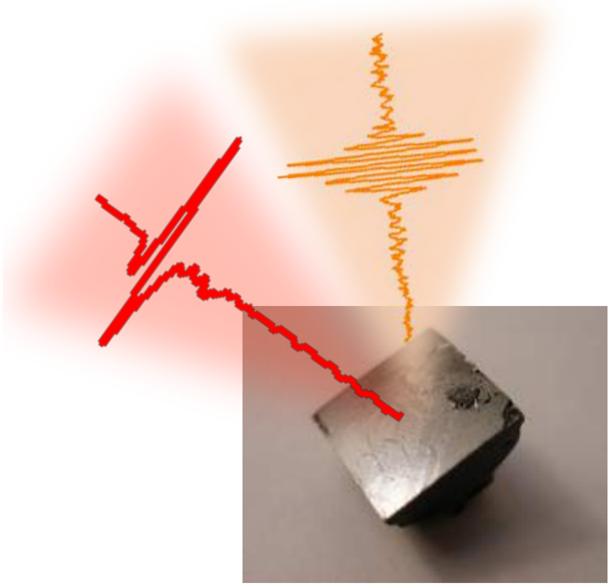
5) Magnetic order with Neutron or Resonant x-ray scattering

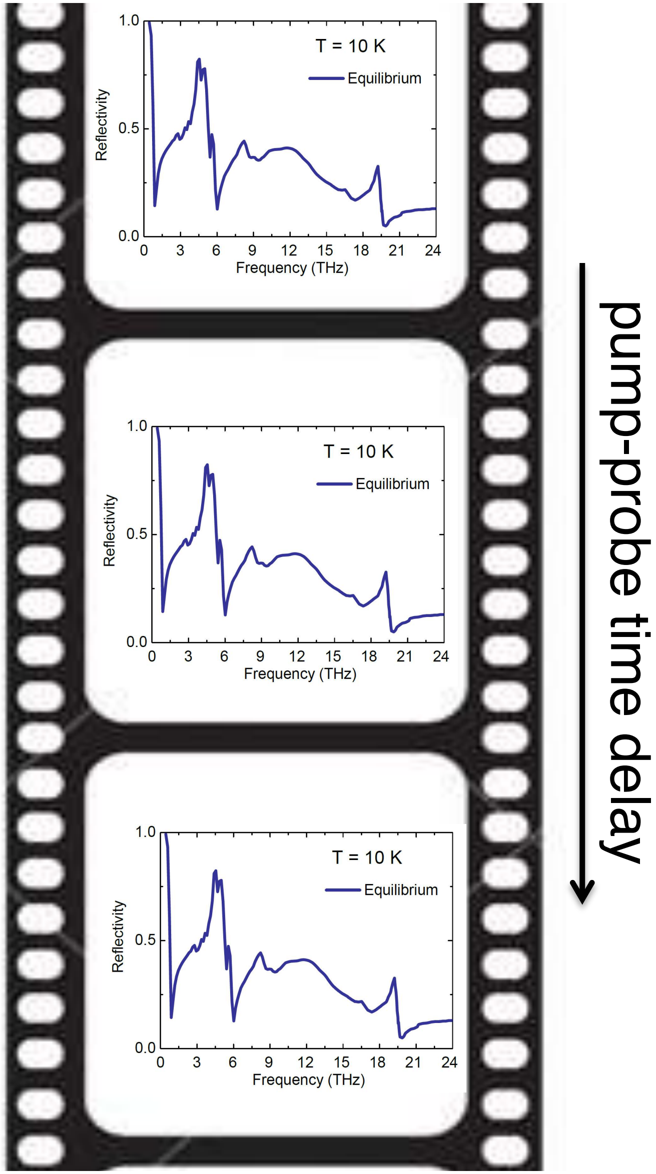
…………….

The optical spectrum of a superconductor



Broadband optics

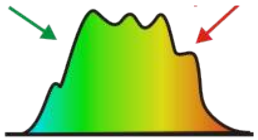


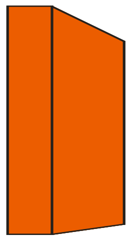


Generating THz Probe Pulses

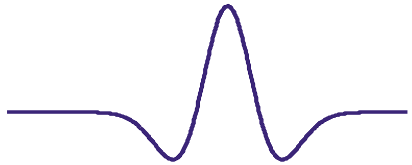
femtosecond laser pulse

nonlinear medium ($\chi^{(2)}$)



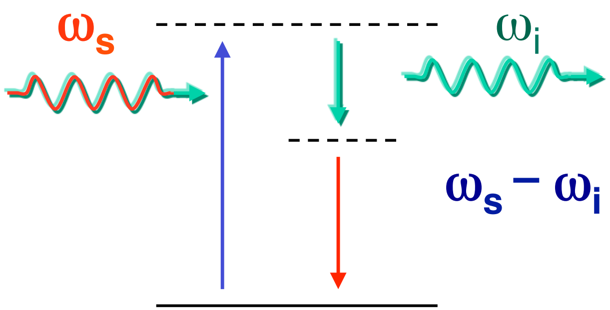


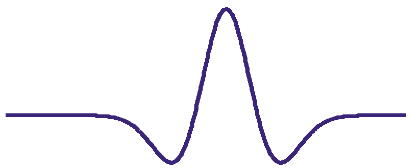
single cycle THz pulse



Intra-pulse Difference Frequency Radiation

Optical Rectification

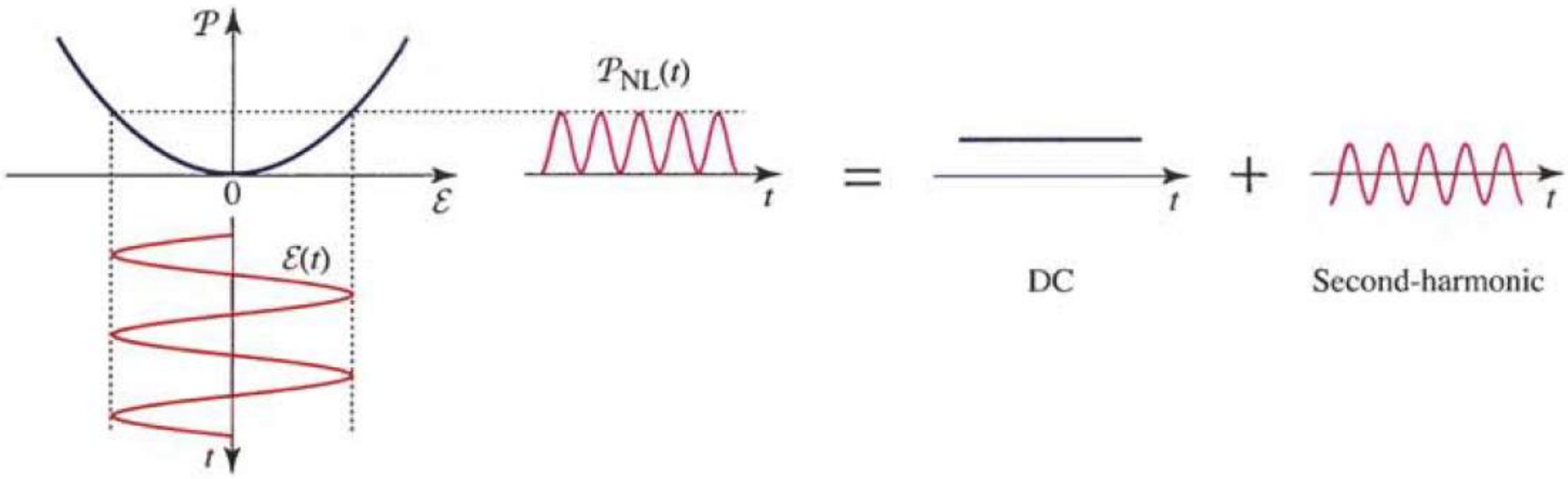




Optical Rectification

$$E(t) = E_0 \cos(\omega t)$$

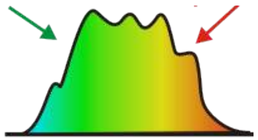
$$\begin{aligned} P_{NL}^{(2)}(t) &= \varepsilon_0 \chi^{(2)} E_0^2 \cos^2(\omega t) \\ &= \tfrac{1}{2} \varepsilon_0 \chi^{(2)} E_0^2 \left[1 + \cos(2\omega t)\right] \\ &= P_{NL}^{(2)}(0) + P_{NL}^{(2)}(2\omega) \cos(2\omega t) \end{aligned}$$

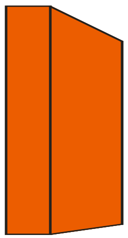


Generating THz Probe Pulses

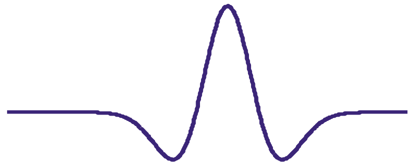
femtosecond
laser pulse

nonlinear
medium ($\chi^{(2)}$)



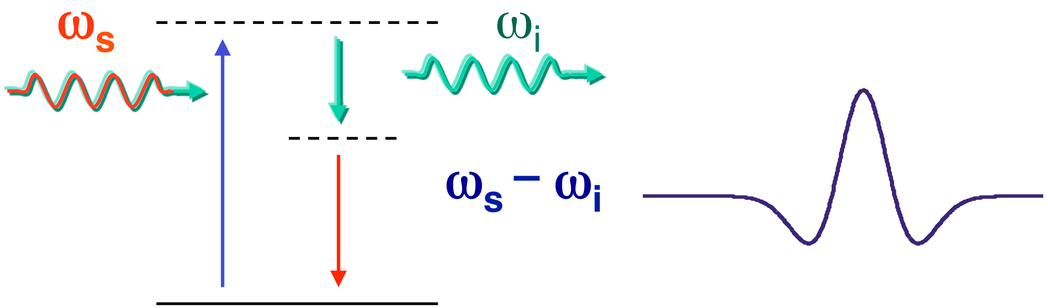


single cycle THz pulse

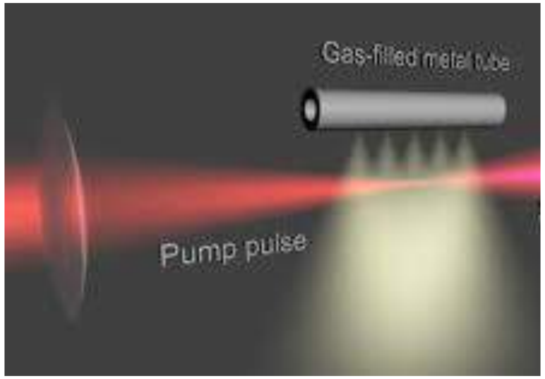


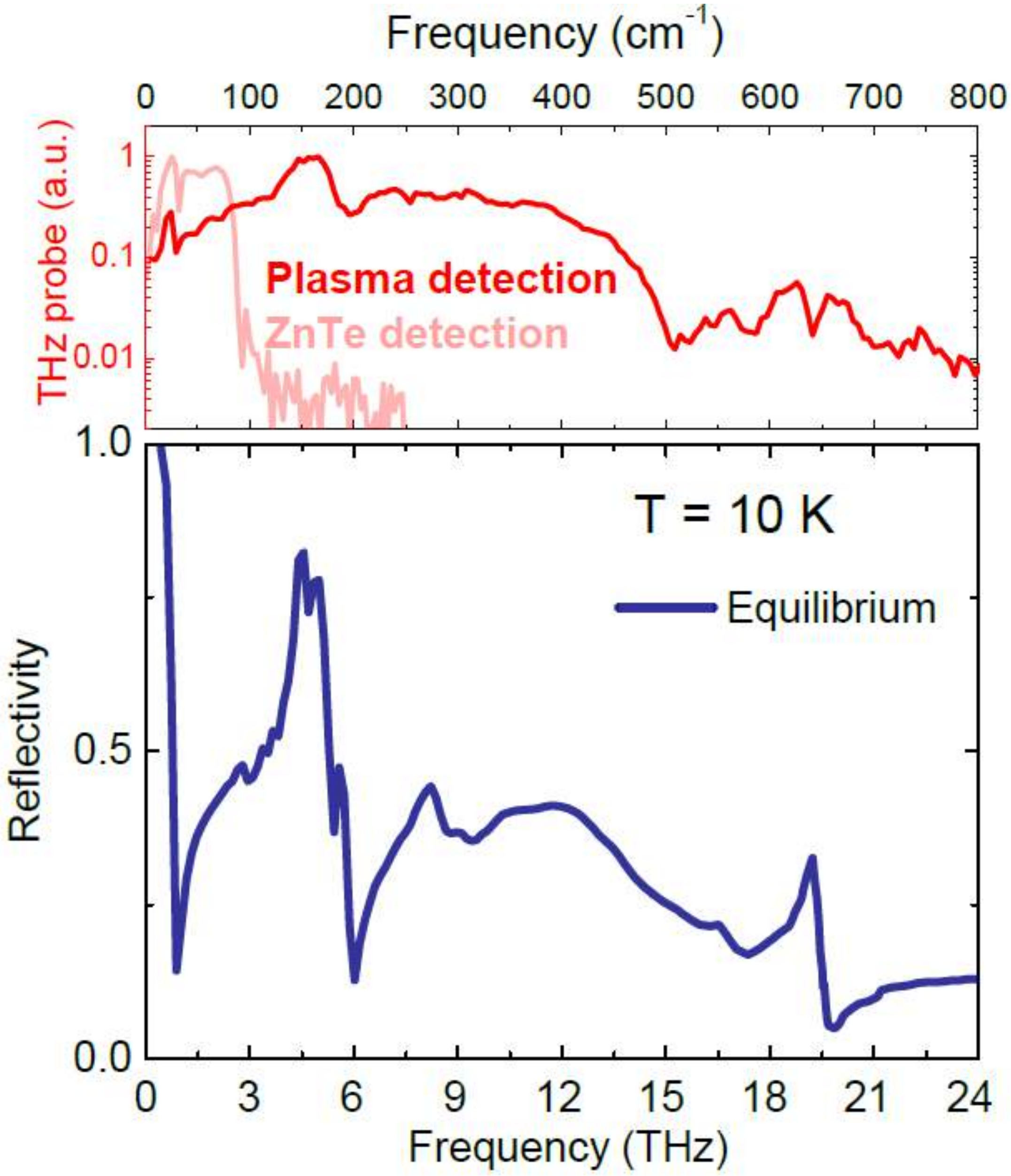
Intra-pulse Difference Frequency Radiation

Optical Rectification

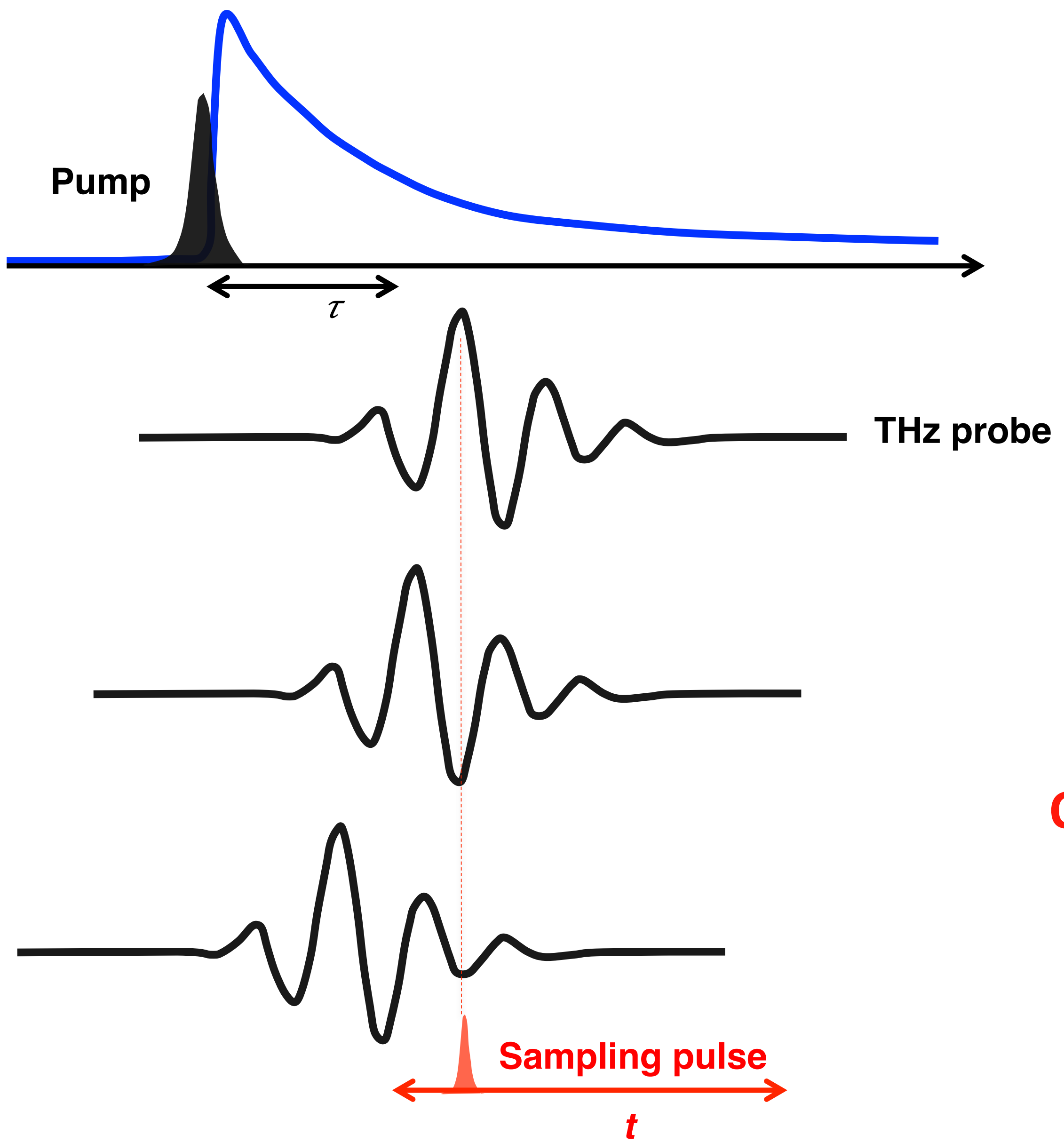


Narrowband vs Boradband THz





Measuring spectra as a function of time

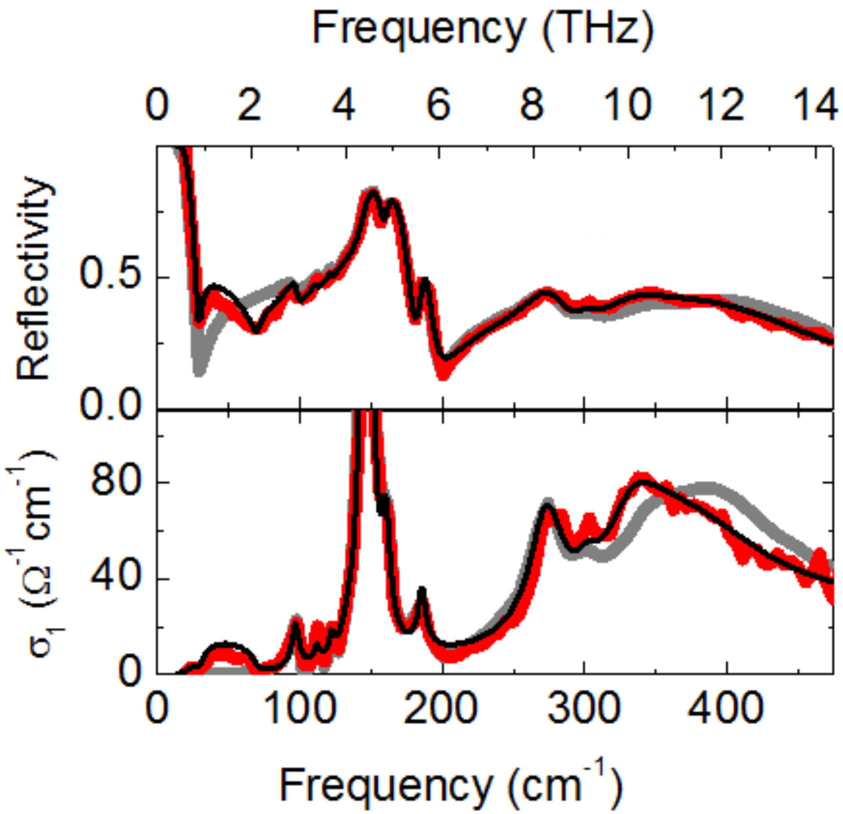


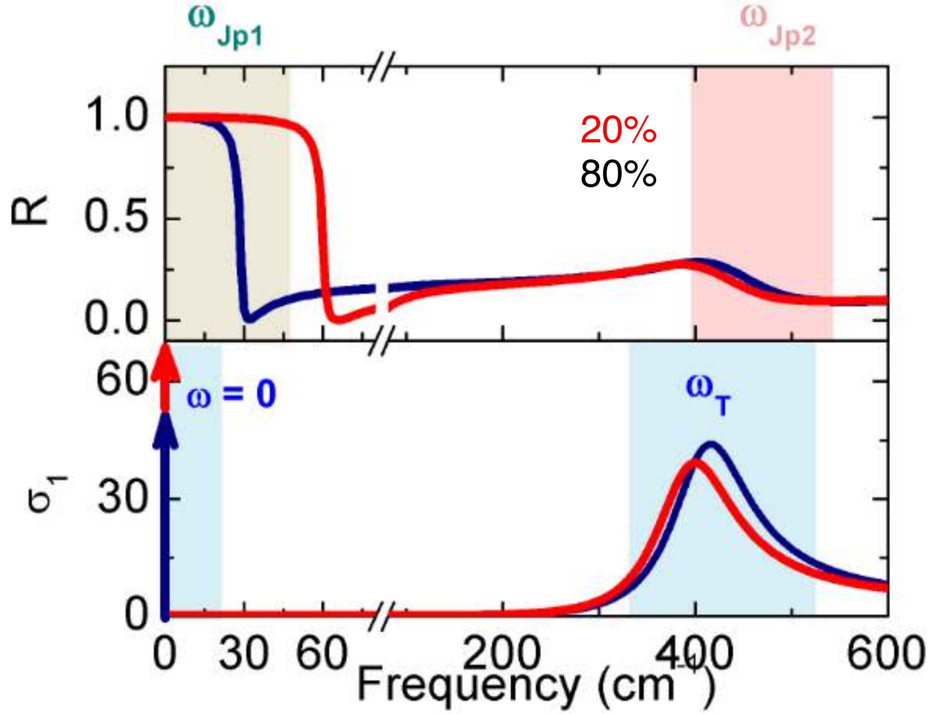
Measure amplitude and phase of the electric field

Obtain complex conductivities ($\sigma_1(\omega)+\iota\sigma_2(\omega)$)

No Kramers Krönig transformation

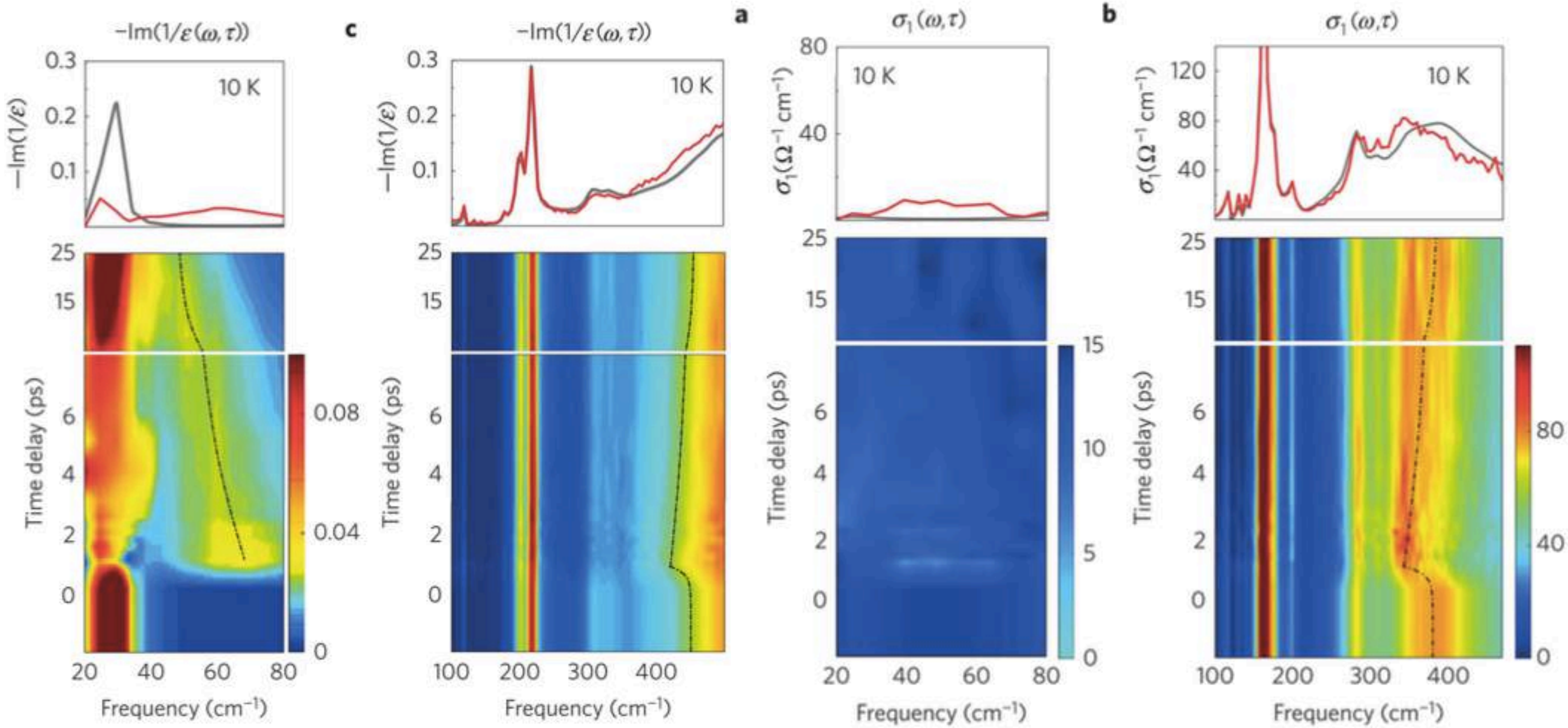
Example: Spectral weight in superconductors





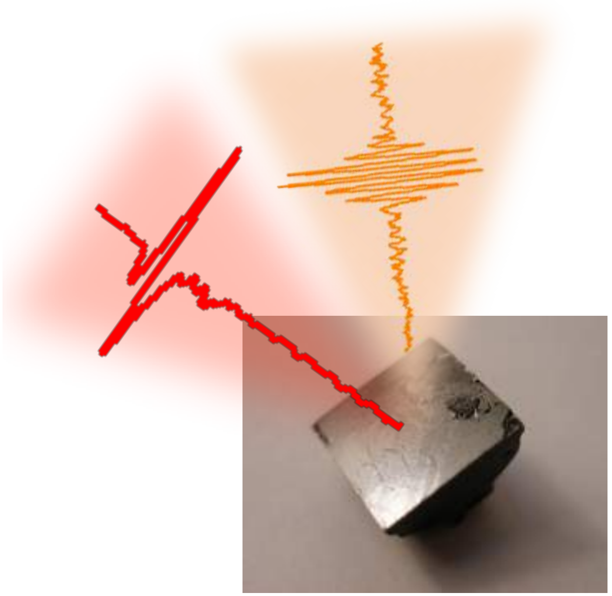
W. Hu *et al.,* **Nature Materials, 13, 705–711 (2014)**

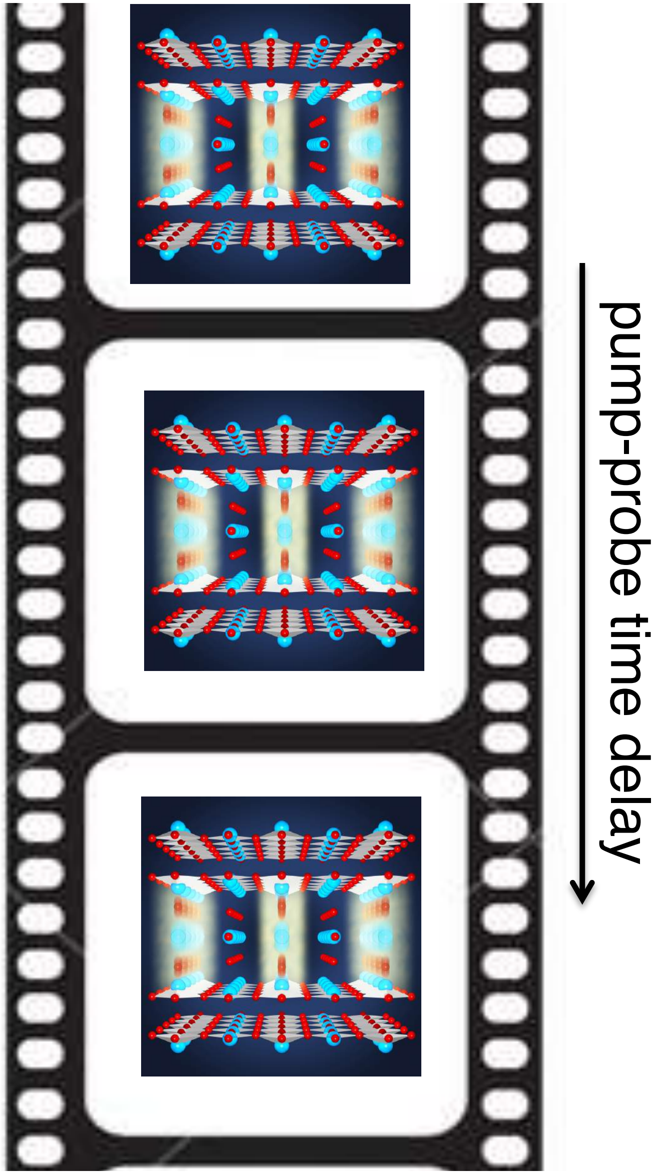
Example: Time Resolved Optics in YBCO



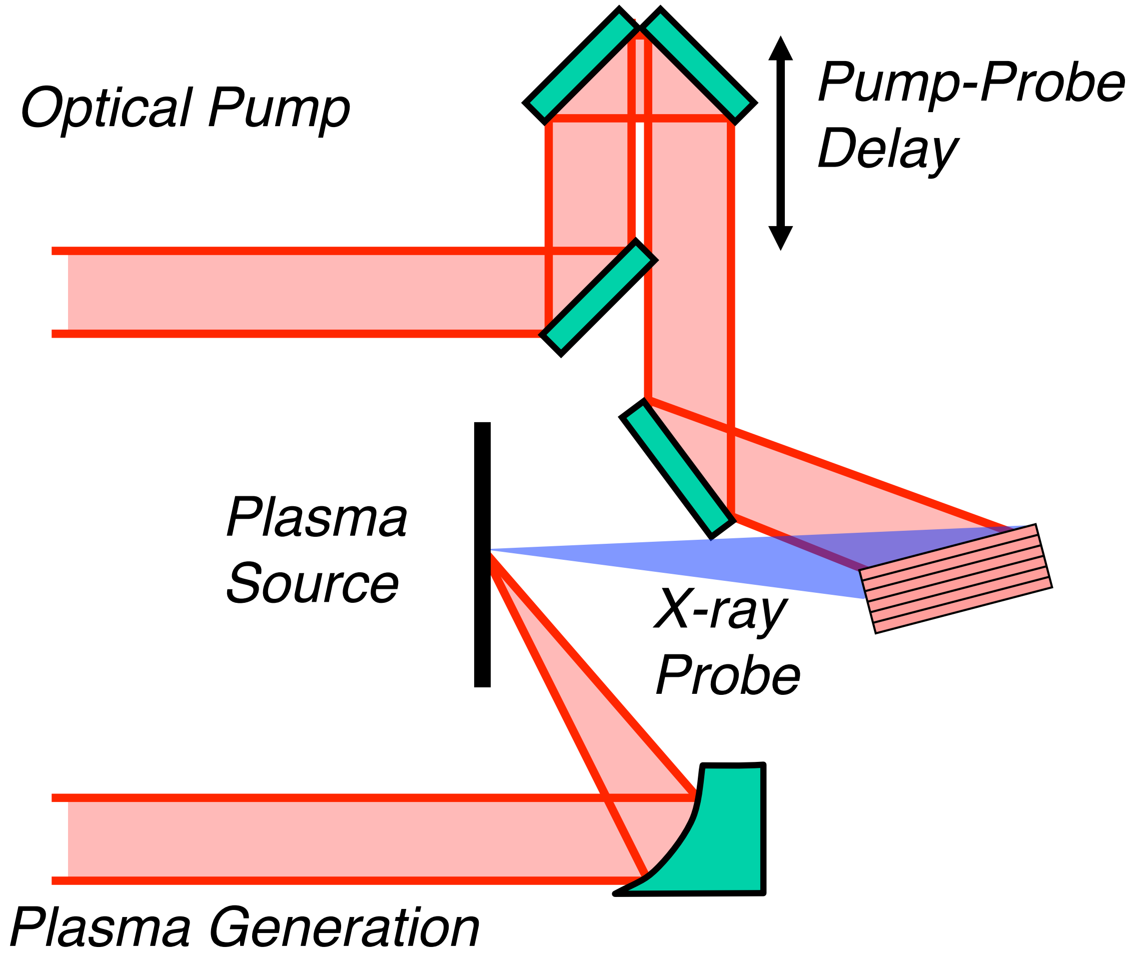
W. Hu *et al.*, Nature Materials, 13, 705–711 (2014)

Measuring atomic rearrangements



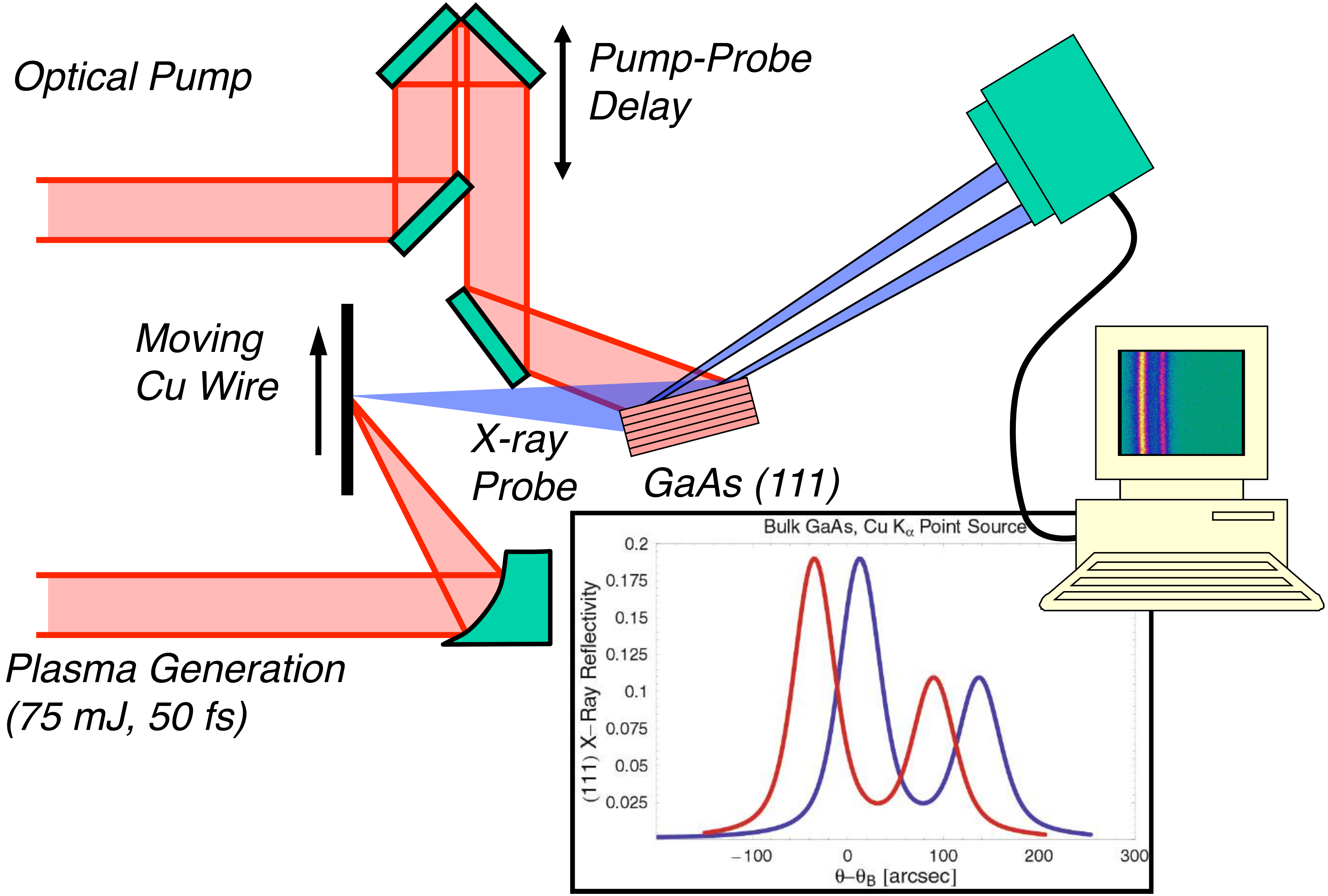


The Stone Age: X-ray Plasma Sources



Ch. Rischel et al. *Nature* <u>390</u>, 490 (1997)

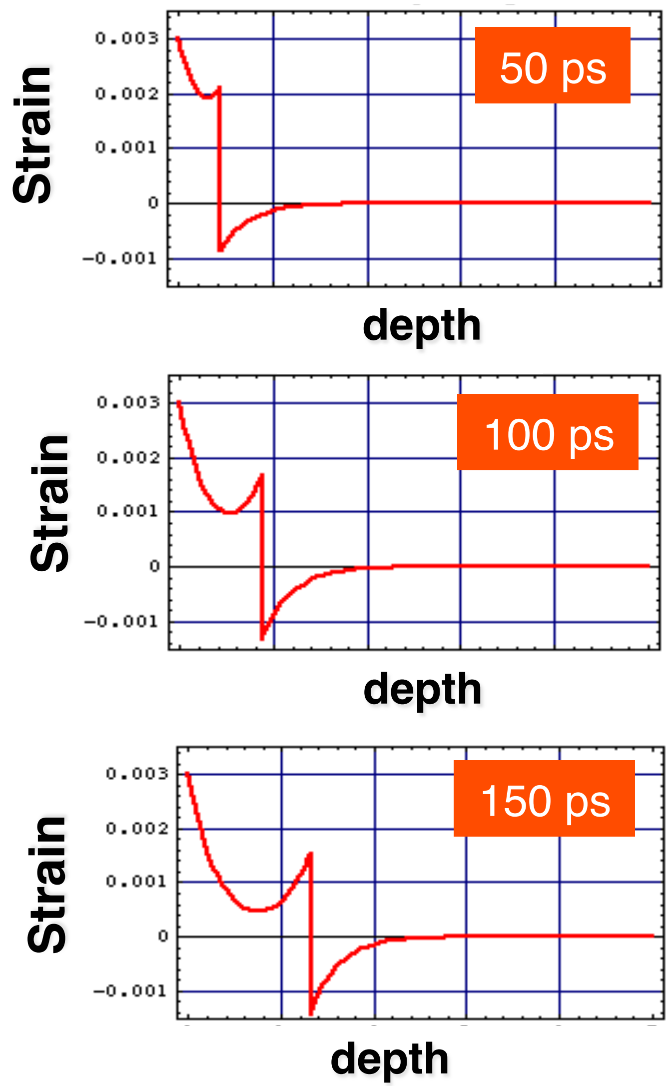
The Stone Age: Measuring Strain Waves



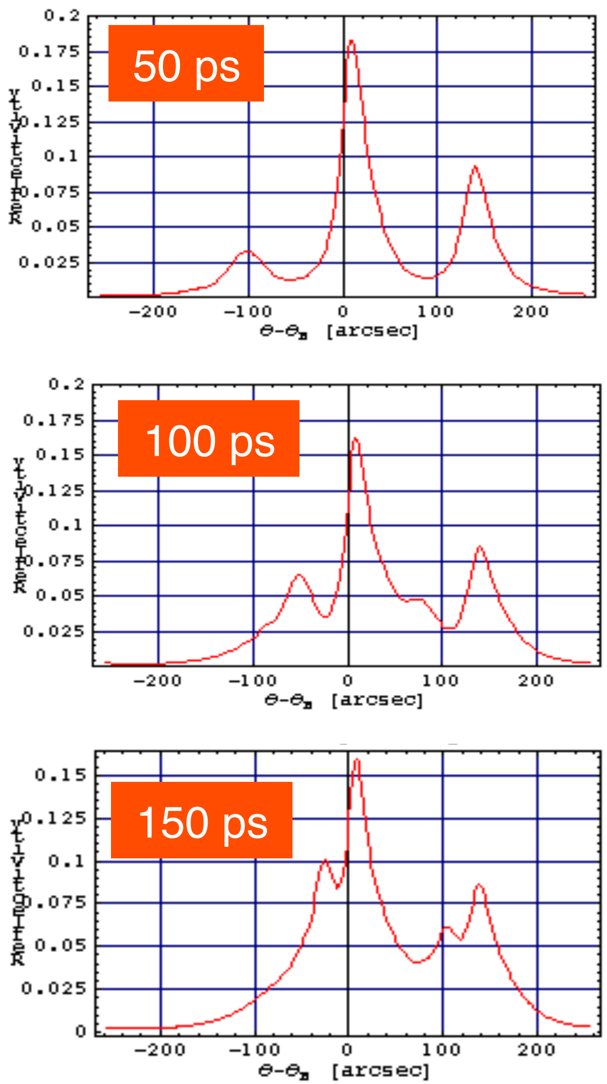
Ch. Rose-Petruck et al. *Nature* 398, 310 (1999)

Strain as a Function of Time

Calculated Strain



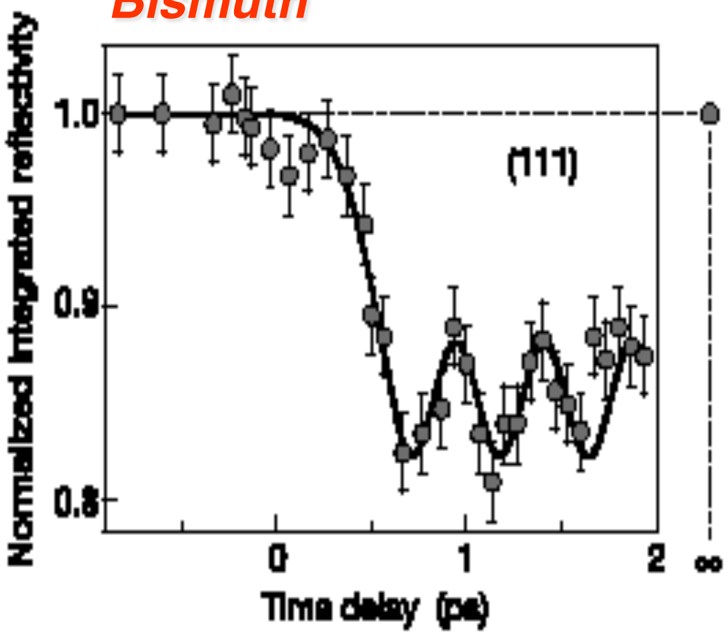
Calculated Diffraction



Ch. Rose-Petruck et al. *Nature* <u>398</u>, 310 (1999)

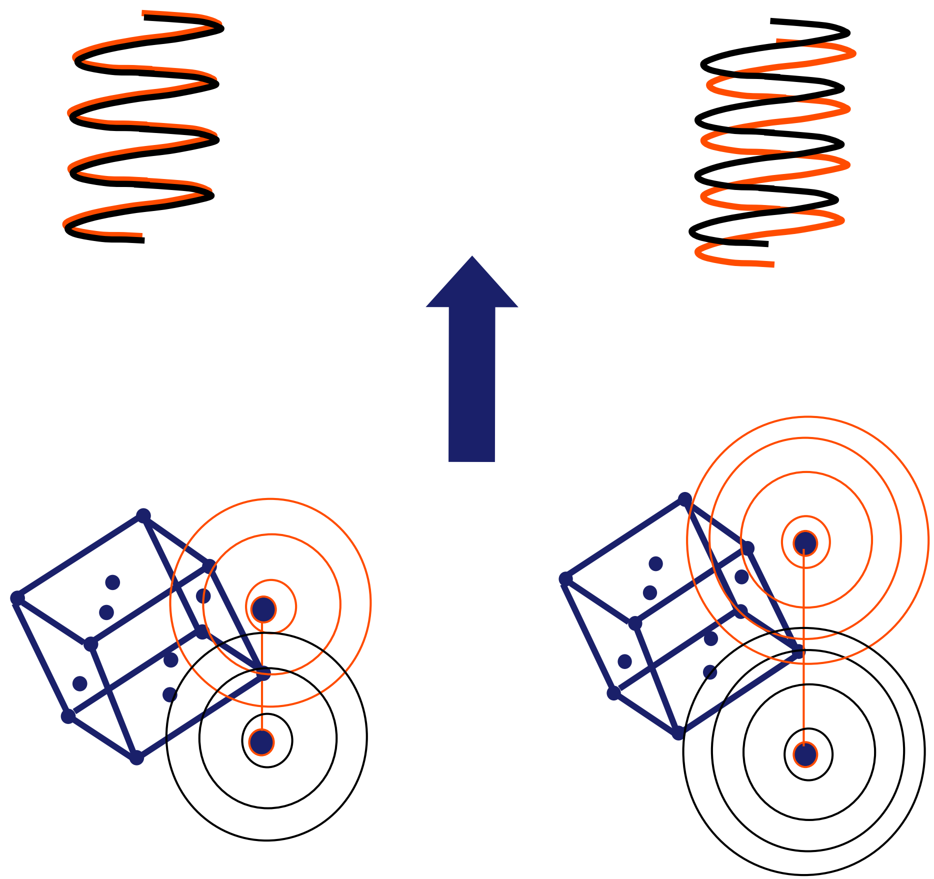
Measurements of Optical Phonons

Bismuth



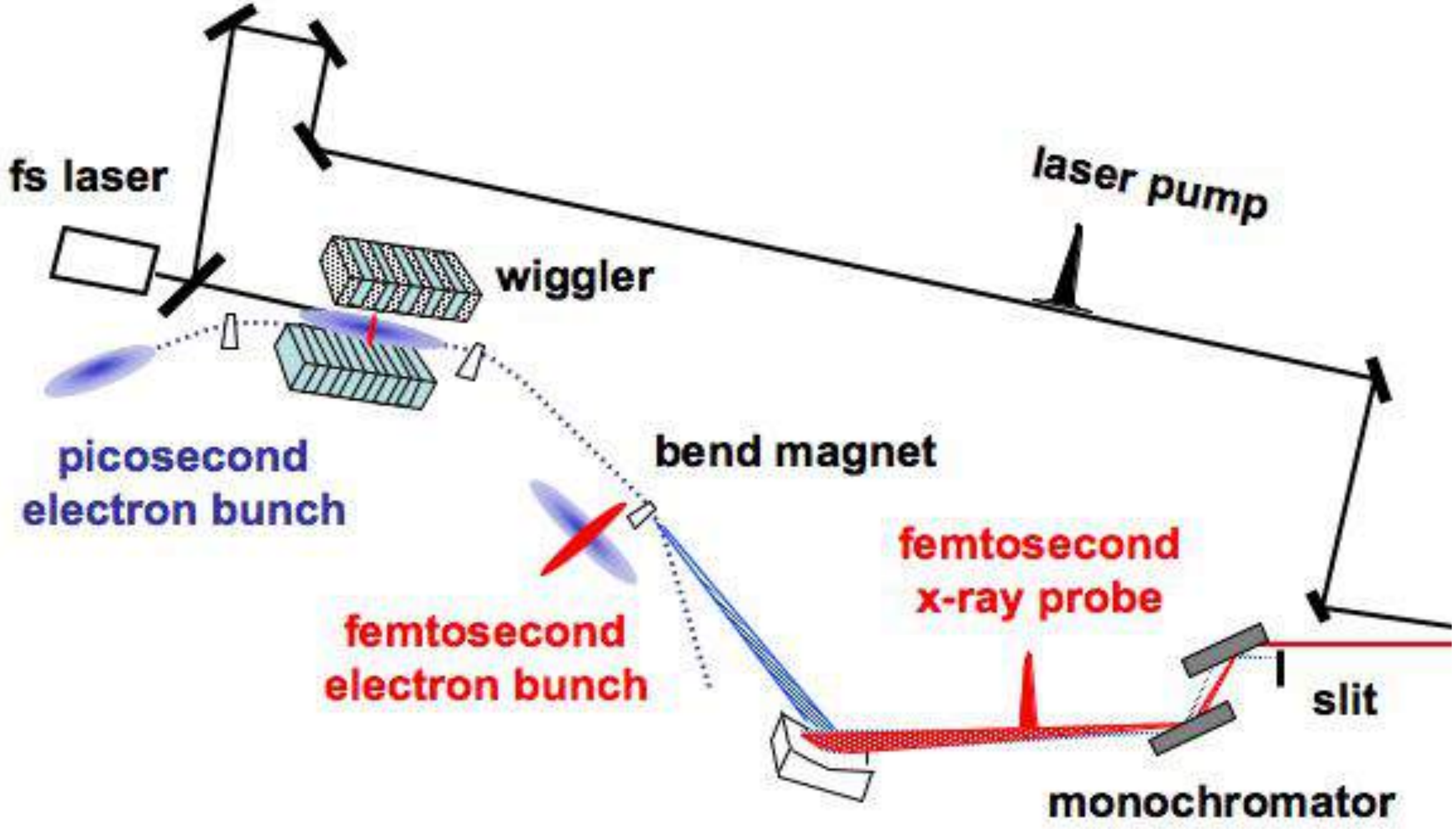
δ = 5% of unit cell

$I\,(111) \propto [1 - \cos(6\,\Pi\,\delta)]$



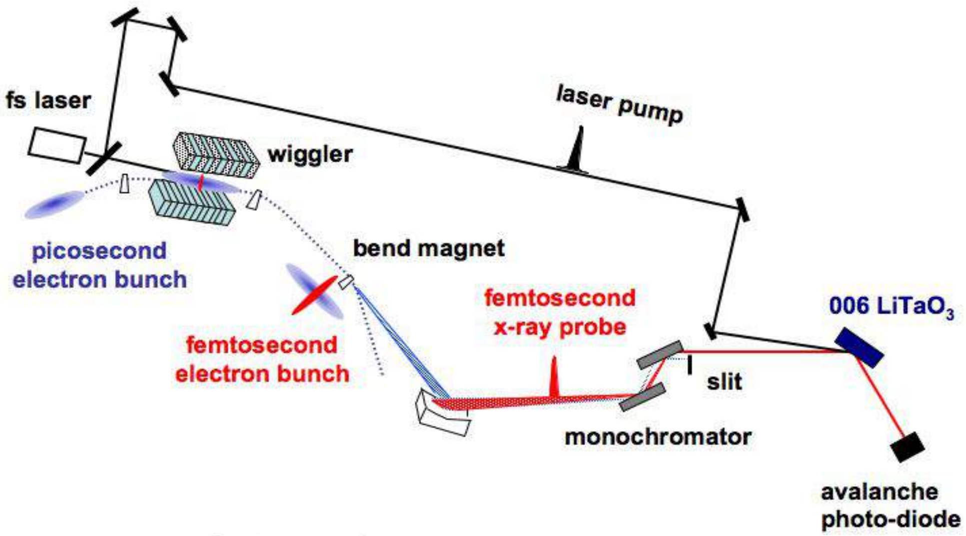
K. Sokolowski-Tinten et al. Nature 422, 287 (2003)

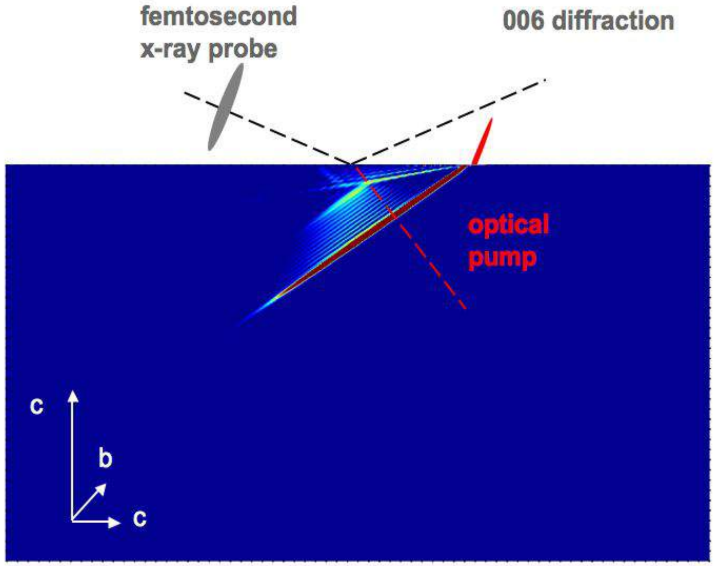
Stone Age 2: Sliced Synchrotron X-rays

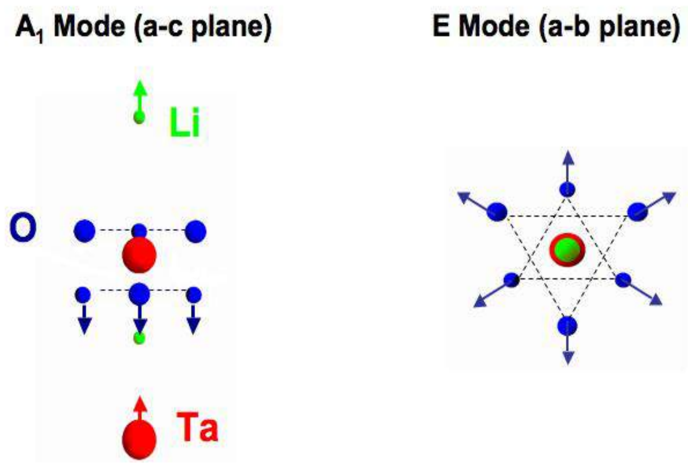


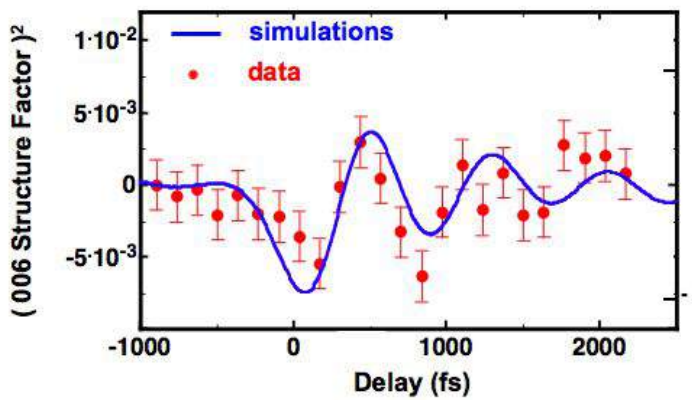
***R.W. Schoenlein et al., Science*, 287, 2237,(2000).**

Stone Age 2: Sliced Synchrotron X-rays



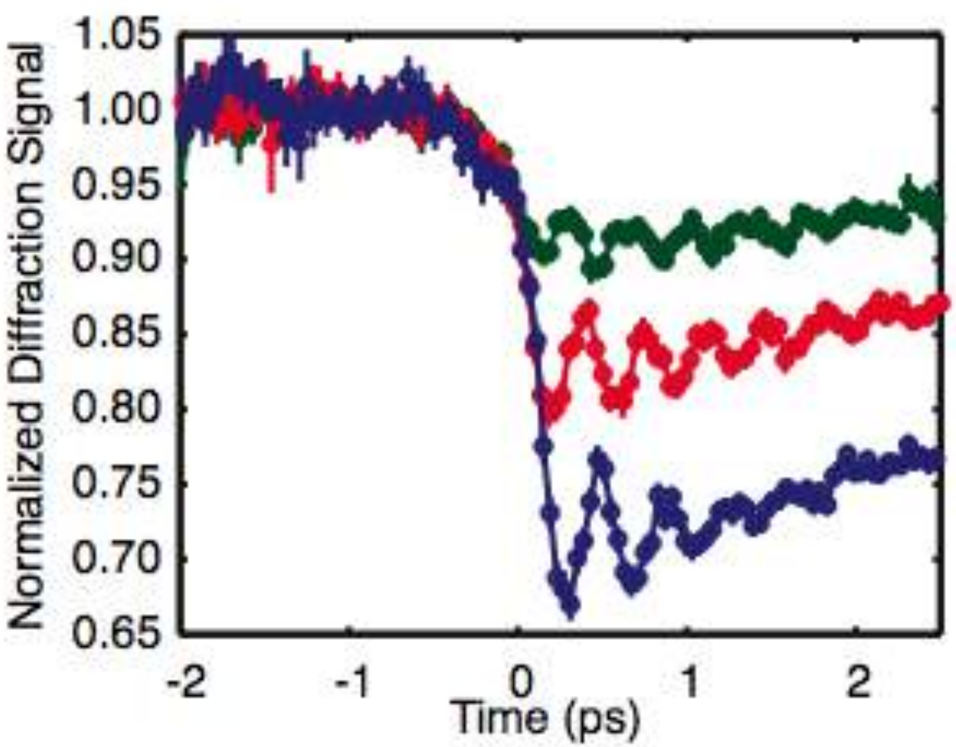


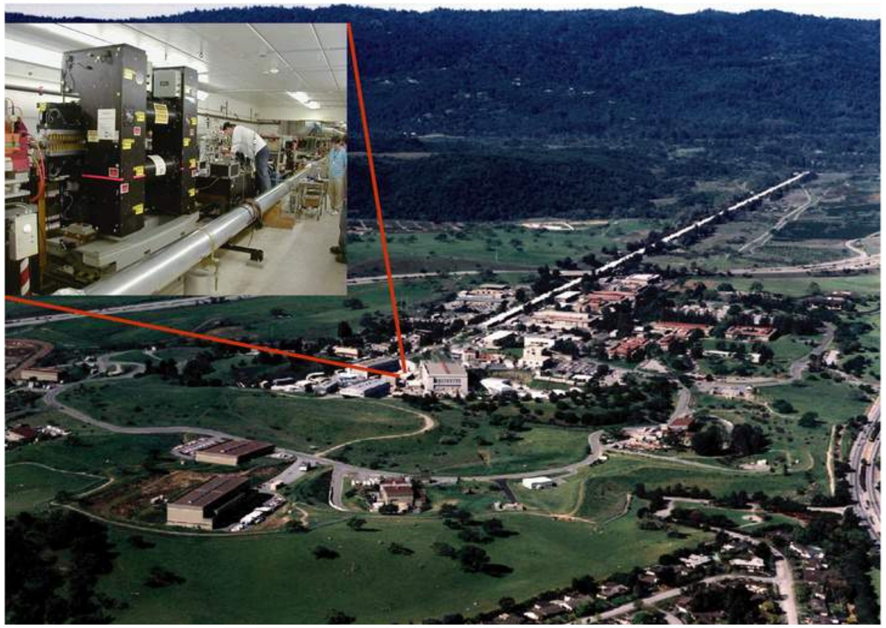




Cavalleri et al., *Nature* 442, 644 (2006).

Bronze Age: Accelerator Based Sources

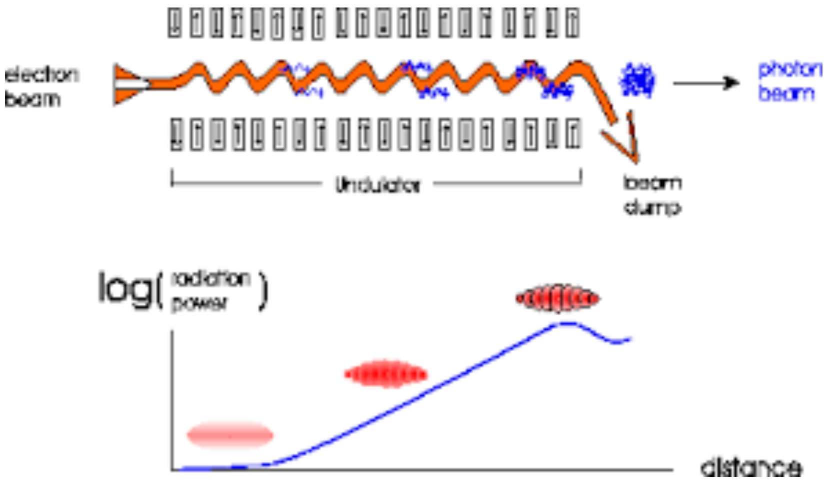


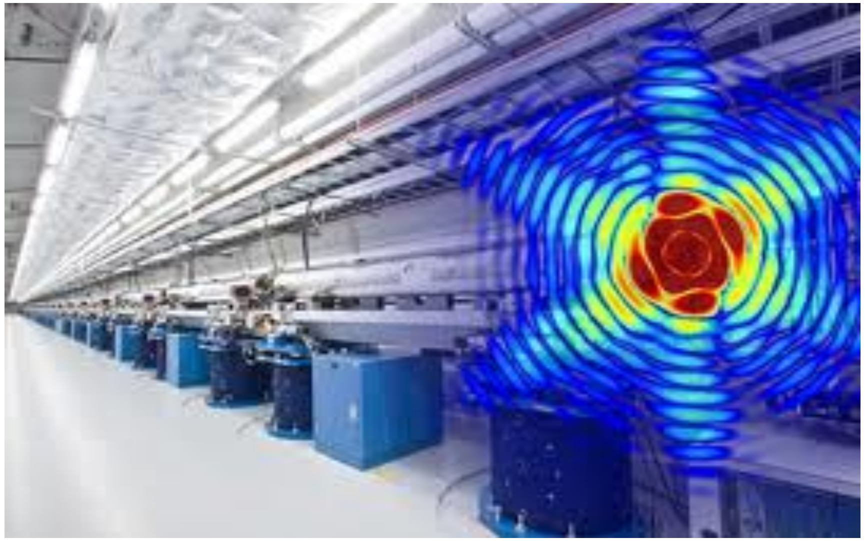


D. M. Fritz et al. Science 315, 633, 2007.

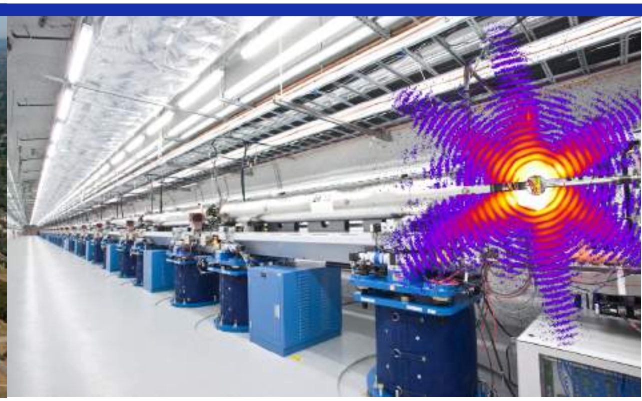
Present time: Free Electron Lasers

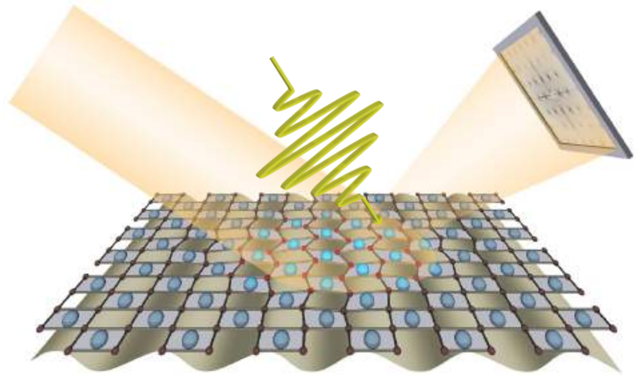
X-ray Lasers

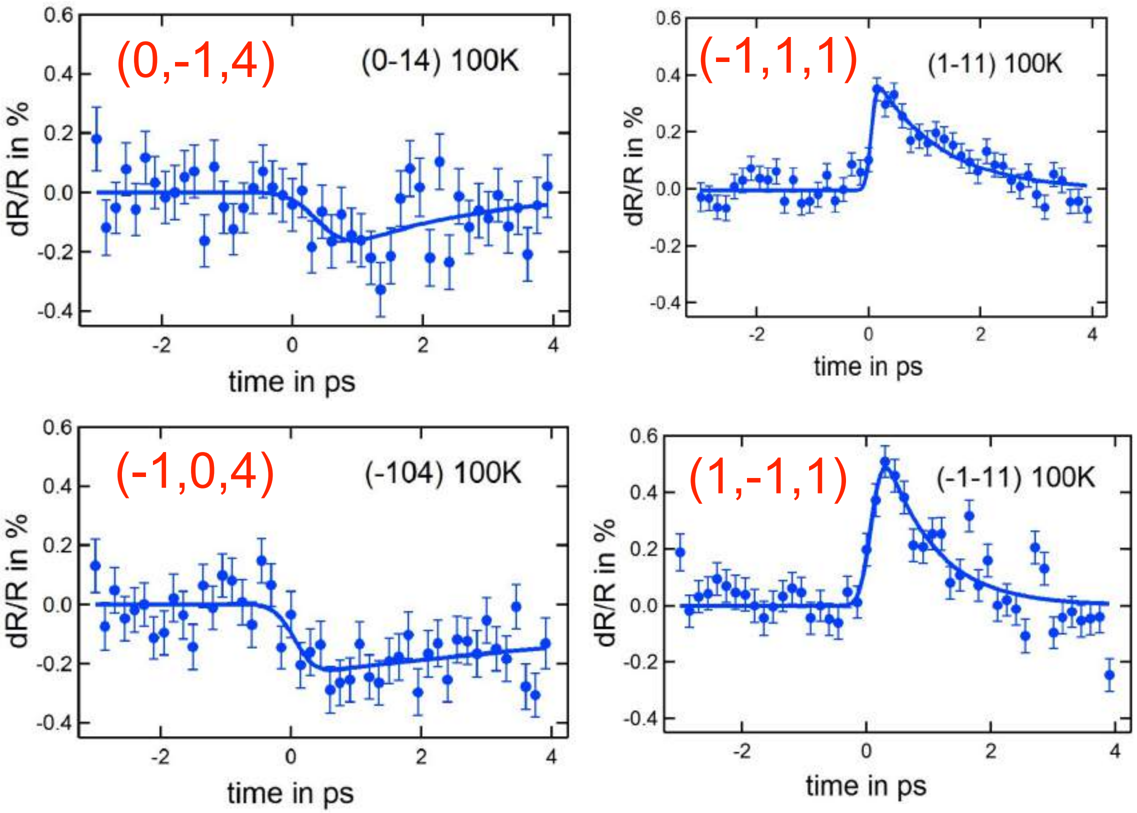




Femtosecond Crystallography at X-ray FELs



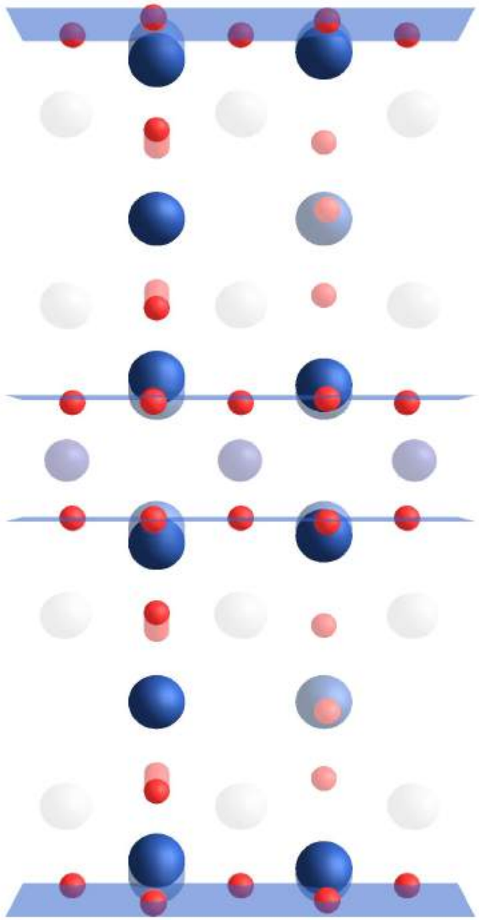


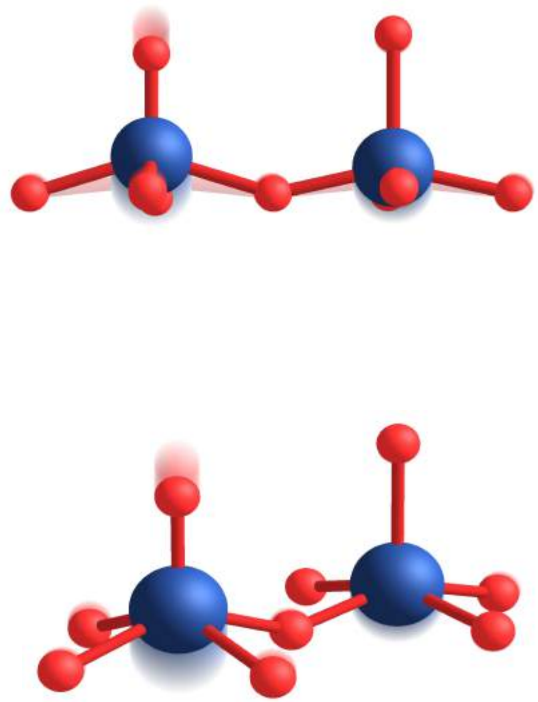


R. Mankowski et al. *Nature 516, 71 (2014)*

Dynamical materials discovery

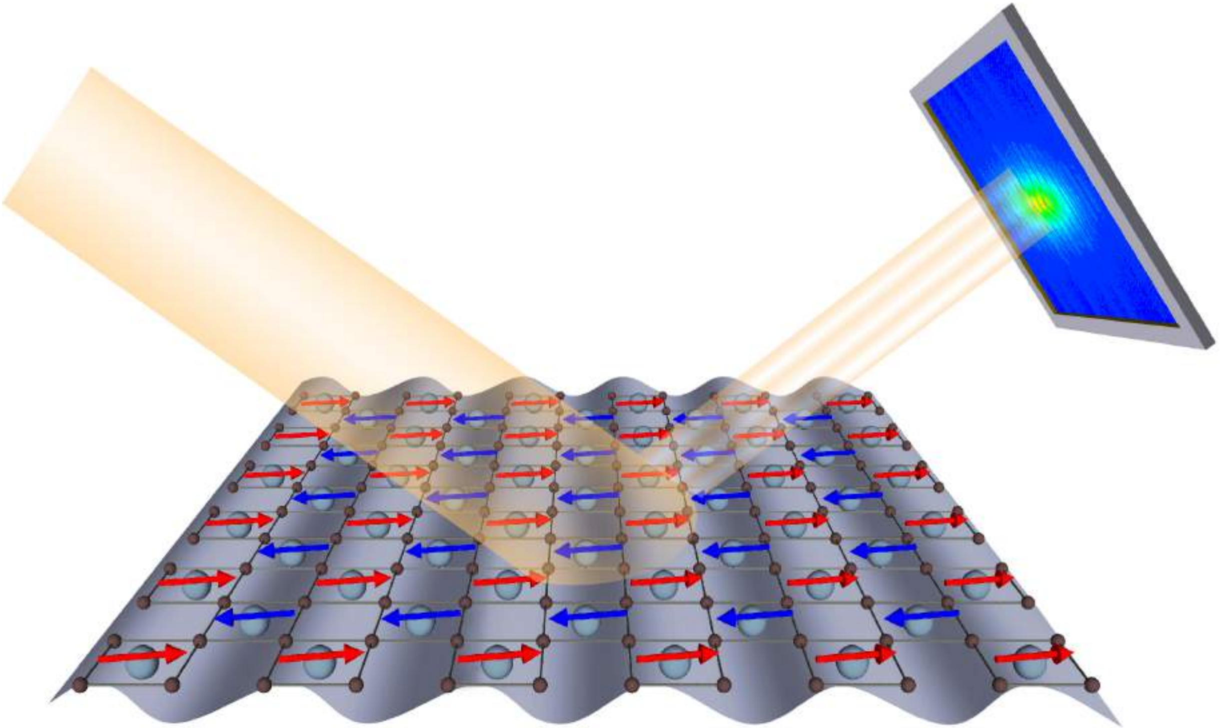
Transient crystal structures with enhanced electronic properties

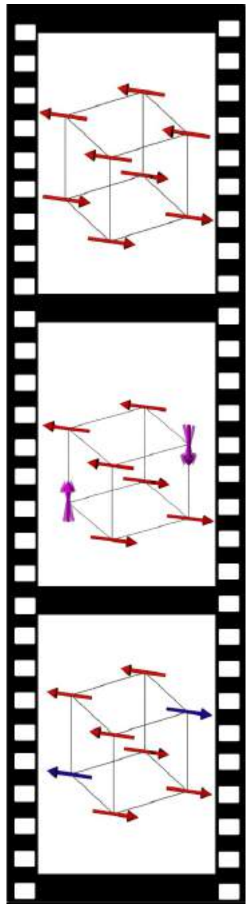




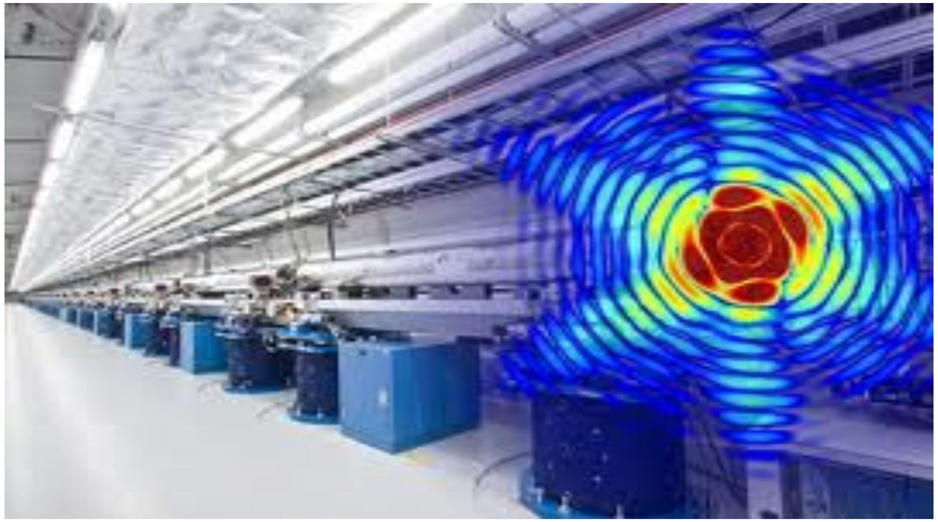
R. Mankowski et al. *Nature* 516, 71 (2014)

Femtosecond soft X-rays: spin dynamics



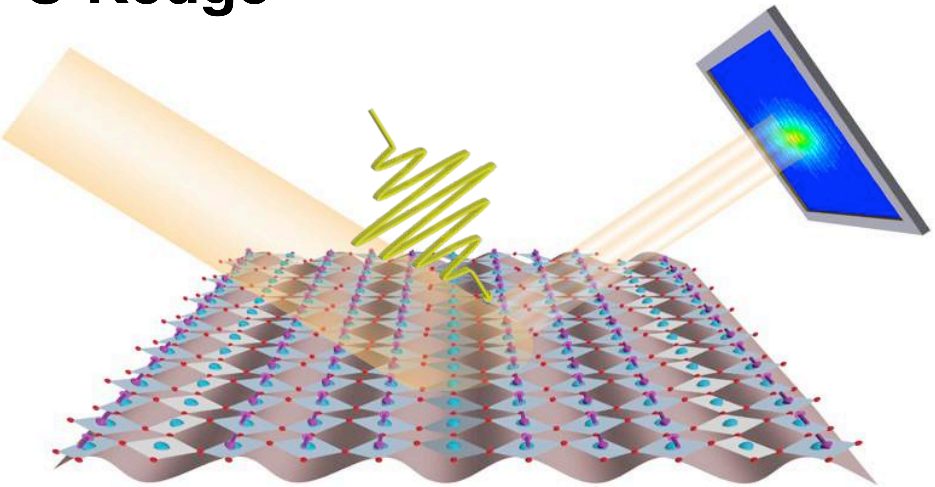


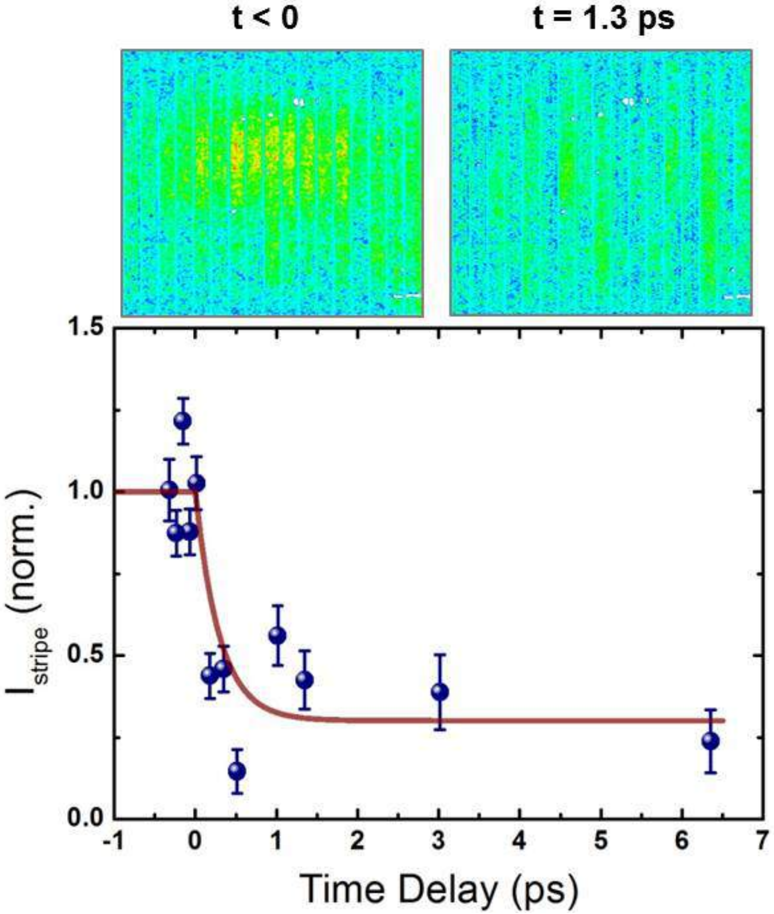
Charge disordering in superconductoprs



O Kedge

(0.25, 0, 0.65)





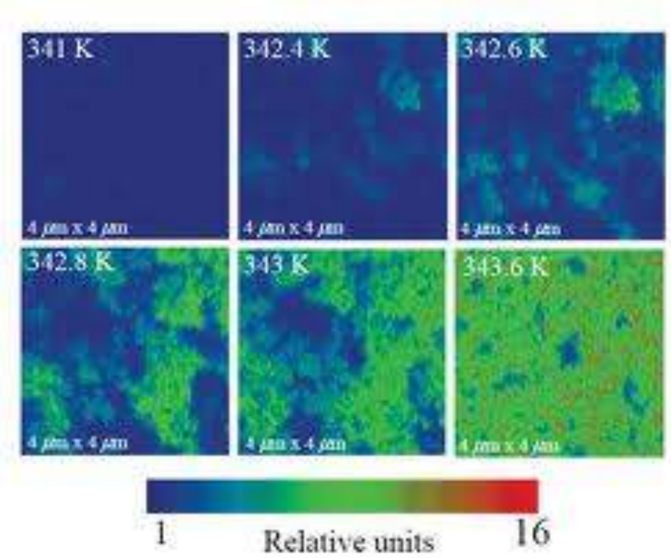
D. Fausti et al., *Science* 331, 6014 (2011)

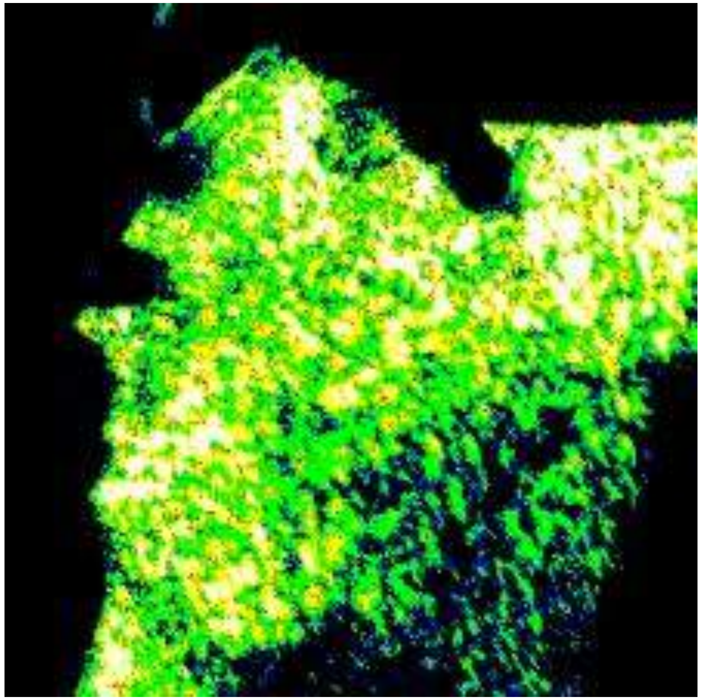
M. Foerst et al., Phys Rev Lett 112, 157002 (2014)

With John Hill, BNL

Challenge: Mesoscopic scale Dynamics

Complex oxides host highly inhomogeneous physics

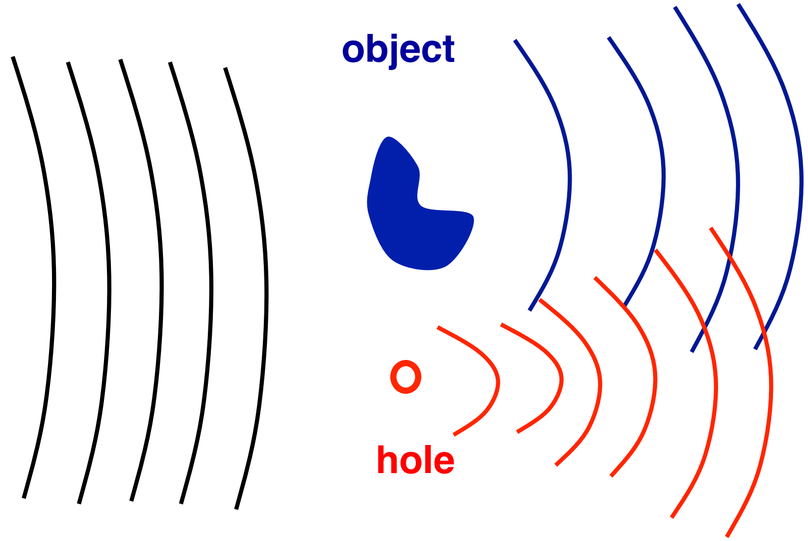


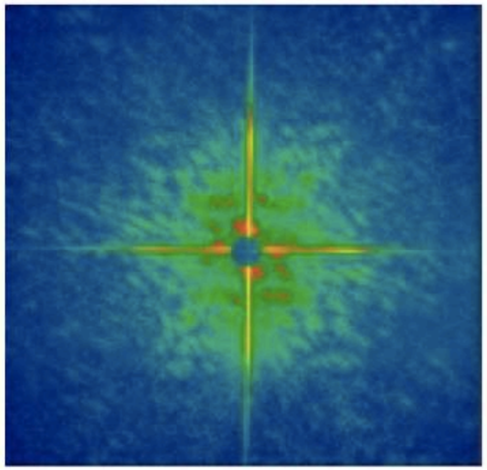


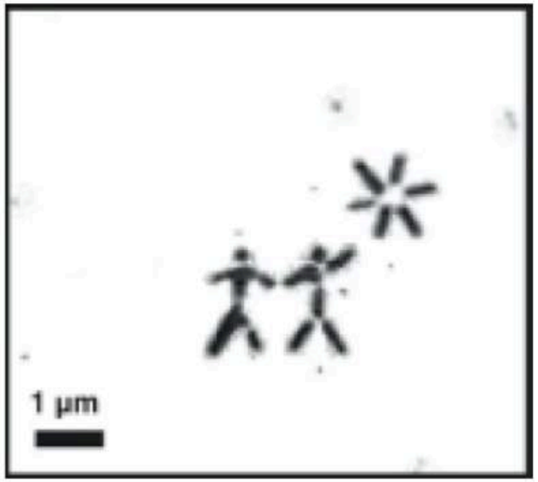
Probing mesoscopic dynamics at Ultrafast time resolutions ?

Exploiting Coherent X-ray Beams at FELs

Free Electron Laser



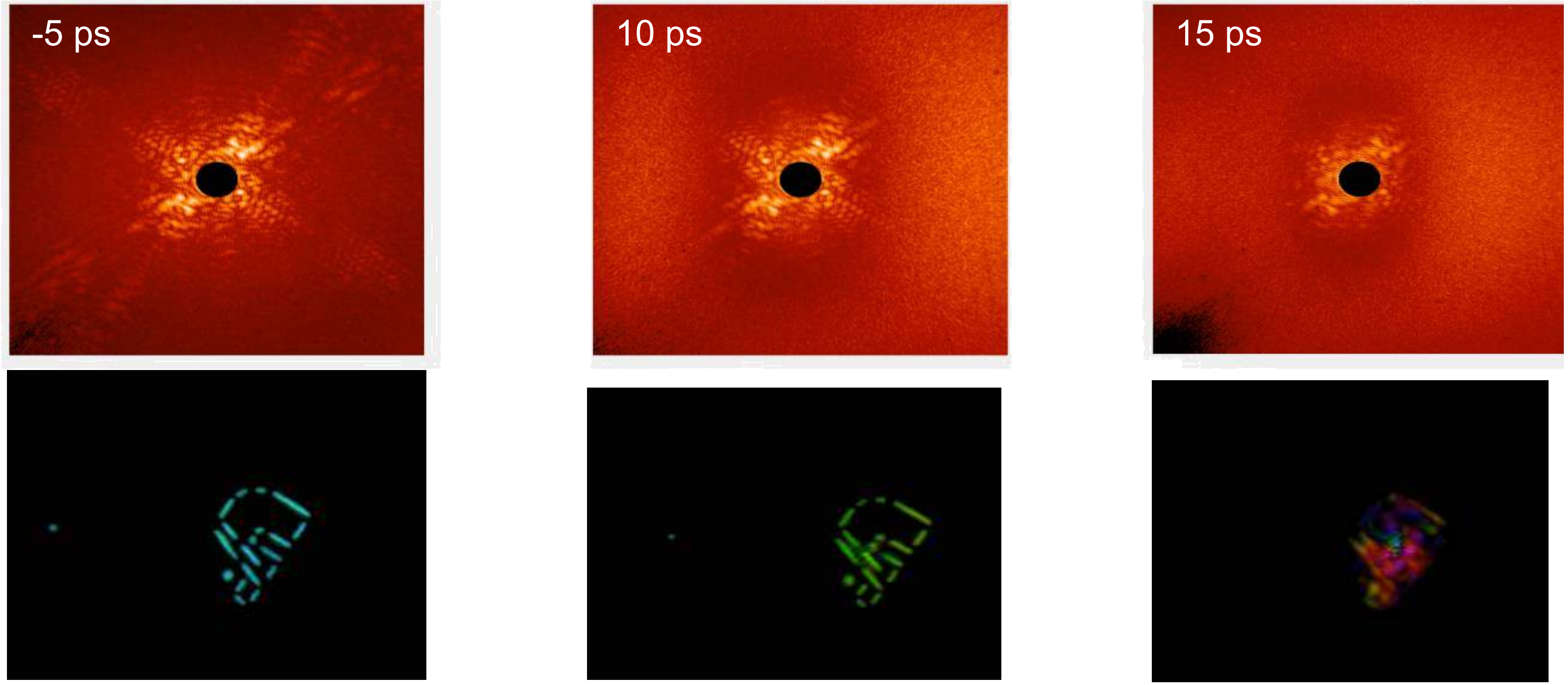


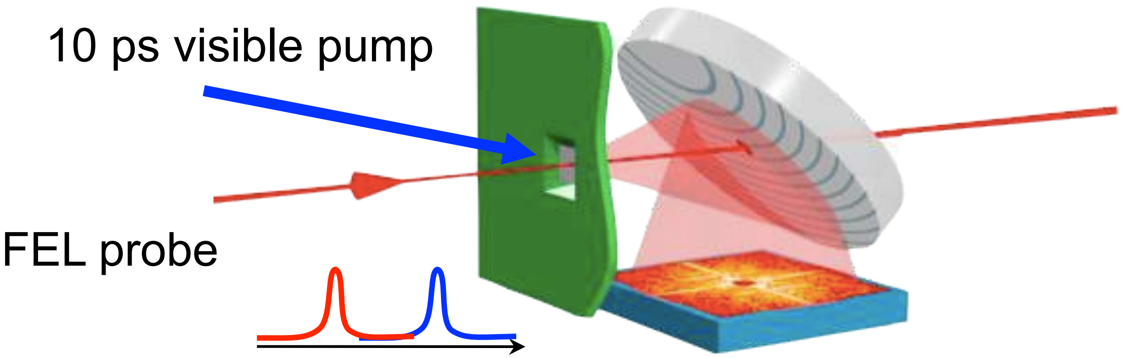


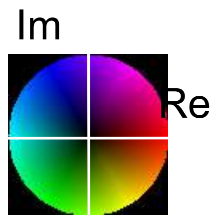
H. Chapman et al. , *Nature Physics* 2, 839 (2006).

S. Eisebitt et al. , *Nature* 432, 885 - 888 (2004).

Time Resolved Imaging of Non-periodic Structures



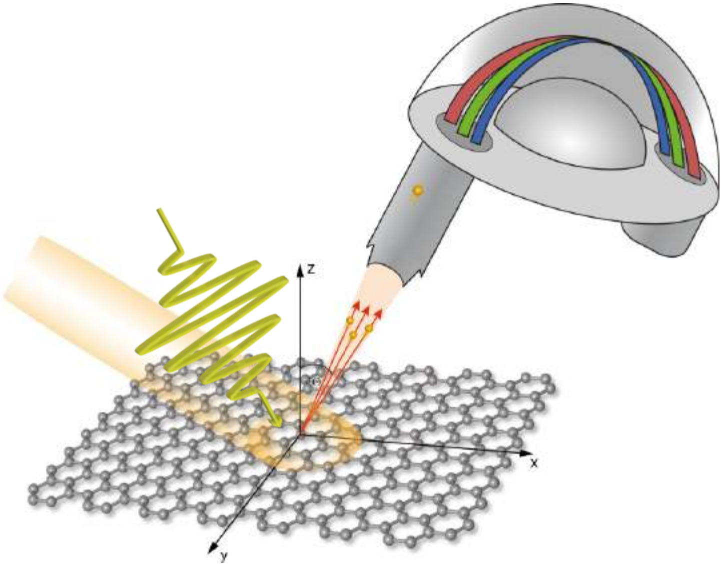




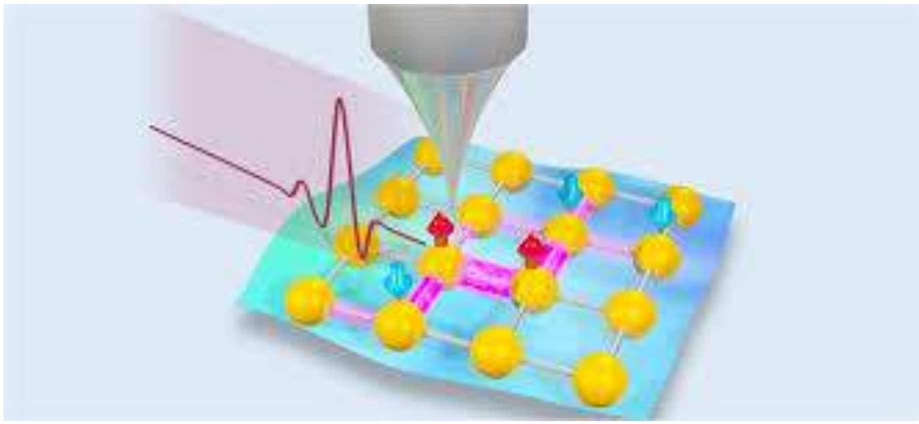
A. Barty et al., Nature Photonics 2 415 (2008)

I have not told you about

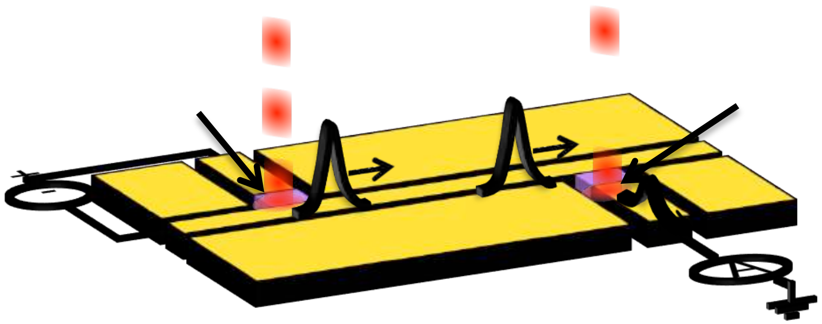
Femtosecond ARPES



Femtosecond STM



Ultrafast Transport



The people who make all this possible

Andrea Cavalleri, *M. Foerst, D. Nicoletti, W. Hu, M. Buzzi, J. McIver, S. Rajasekaran, E. Pomarico, F.U. Stein, M. Budden, A. Cantaluppi, A. Cartella, E. Casandruc. B. Liu, R. Mankowski, T. Nova. A. von Hoegen, Th. Gebert, H. Bromberger*

Summary: Lecture 1

New Methods are revolutionizing measurements of dynamics

We use coherent THz radiation from tabletop sources and Infrared Free Electron Lasers to drive solids away from equilibrium.

We use ultrafast broadband THz optics to measure the electronci properties of materials away from equilibrium

We use ultrafast x-rays from X-ray Free Electron Lasers to characterize lattice, charge, spin and orbital dynamics.

Use tabletop XUV sources for Time and Angle Resolved Photo-emission and Fermi Surface Dynamic Mapping.

Develop techniques to measure transport at picosecond and femtosecond resolution