THz Control of Complex Oxides

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2012 SwissFEL Photonics „Pump Laser Workshop“

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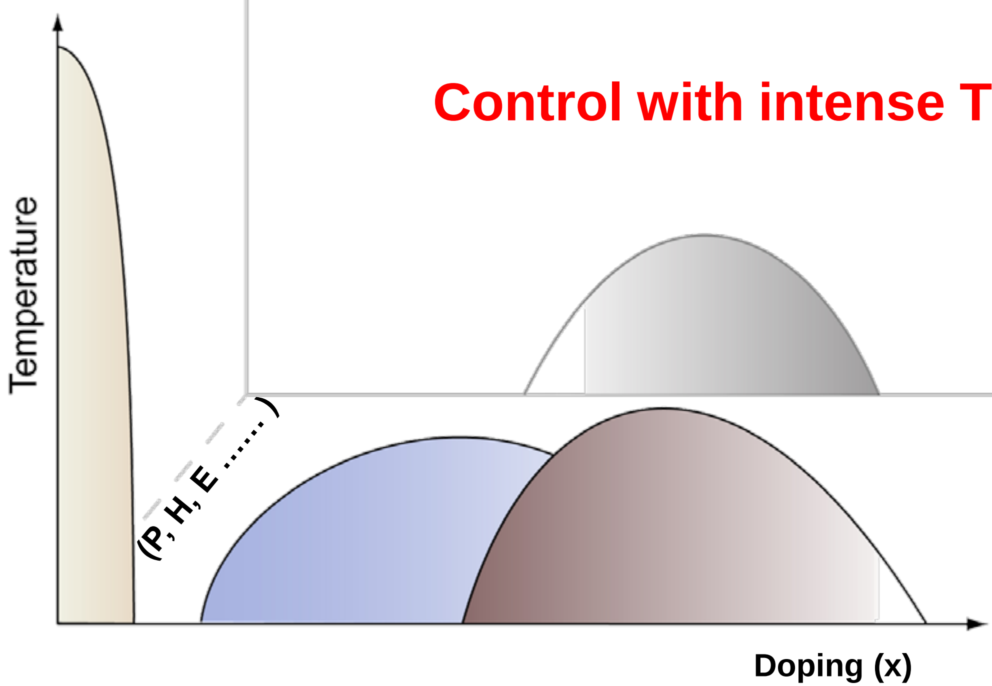
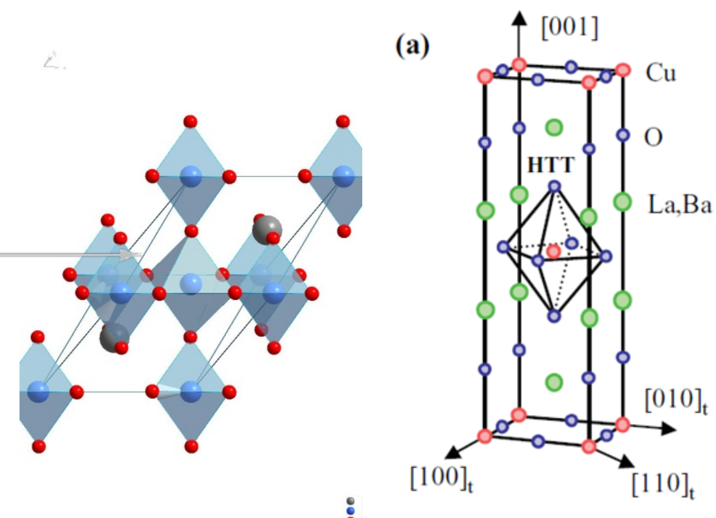
Phase diagram of complex oxides

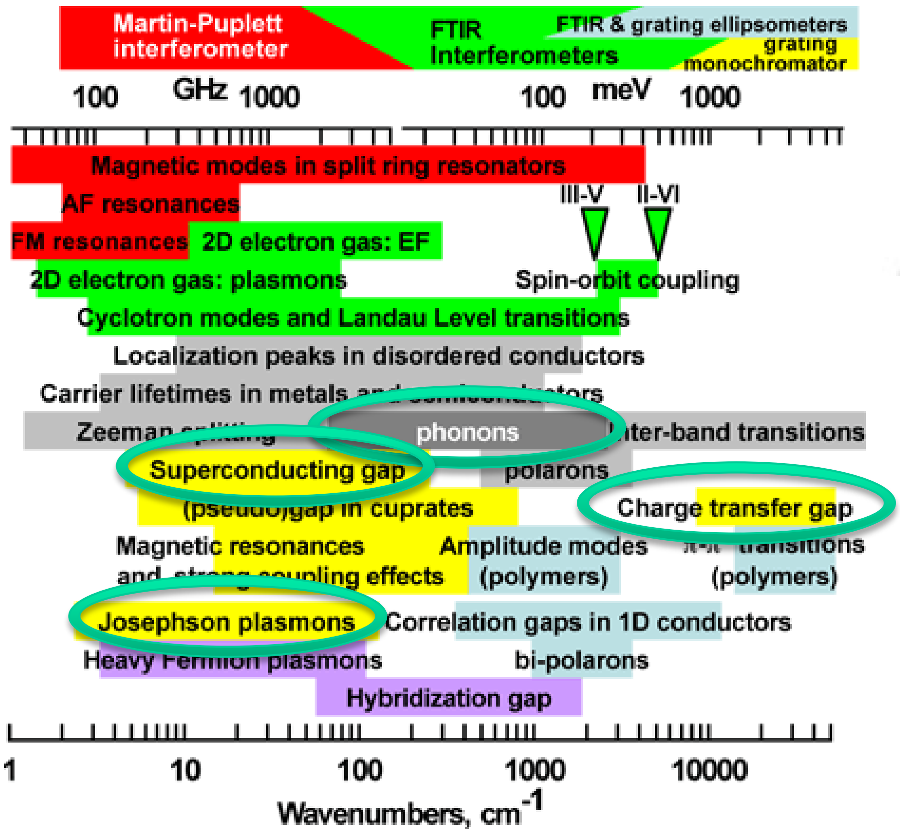
- Competition of different stable phases
- Complex interplay of charge, orbital, spin and lattice degrees of freedom
- Controllable with external parameters (p, H, E, x, …)

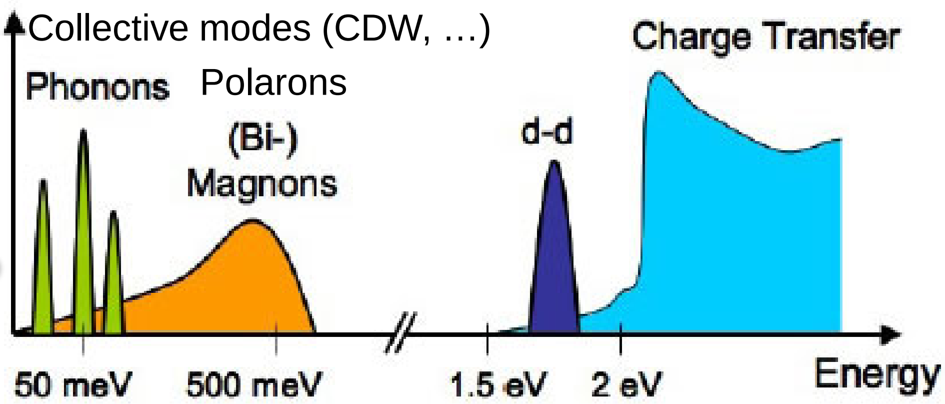
Control with intense THz and mid-IR pulses





Spectrum of collective excitations

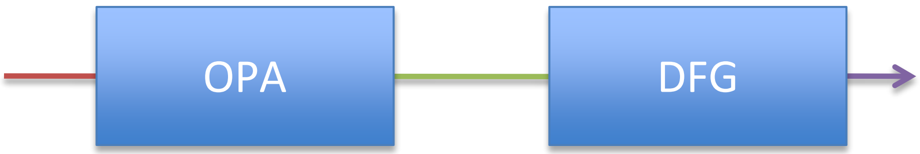




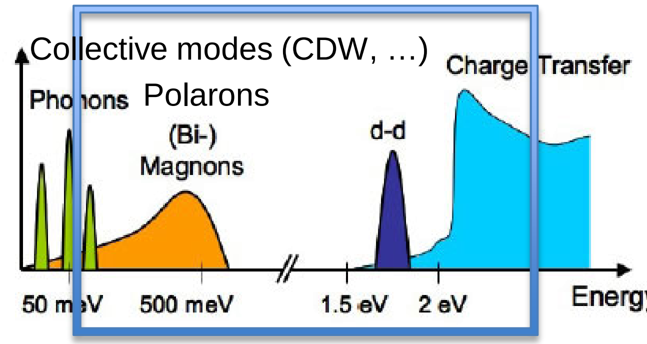
Basov, Rev. Mod. Phys.
Ament, Rev. Mod. Phys.

THz and mid-IR light sources

- **Optical parametric amplifiers: VIS to mid-IR wavelengths**

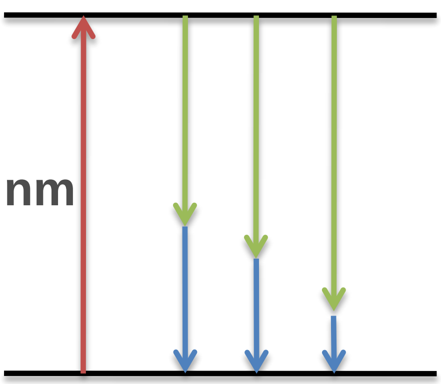


~ 100 fs



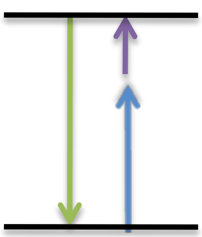
Signal/Idler
1.1-2.3 mm

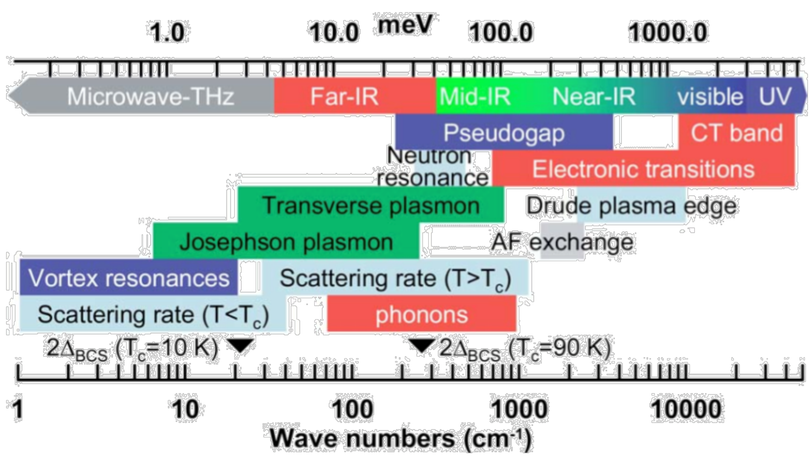
800 nm



$\omega_{DFG}=\omega_s-\omega_i$

3-19 µm
(100-16 THz)





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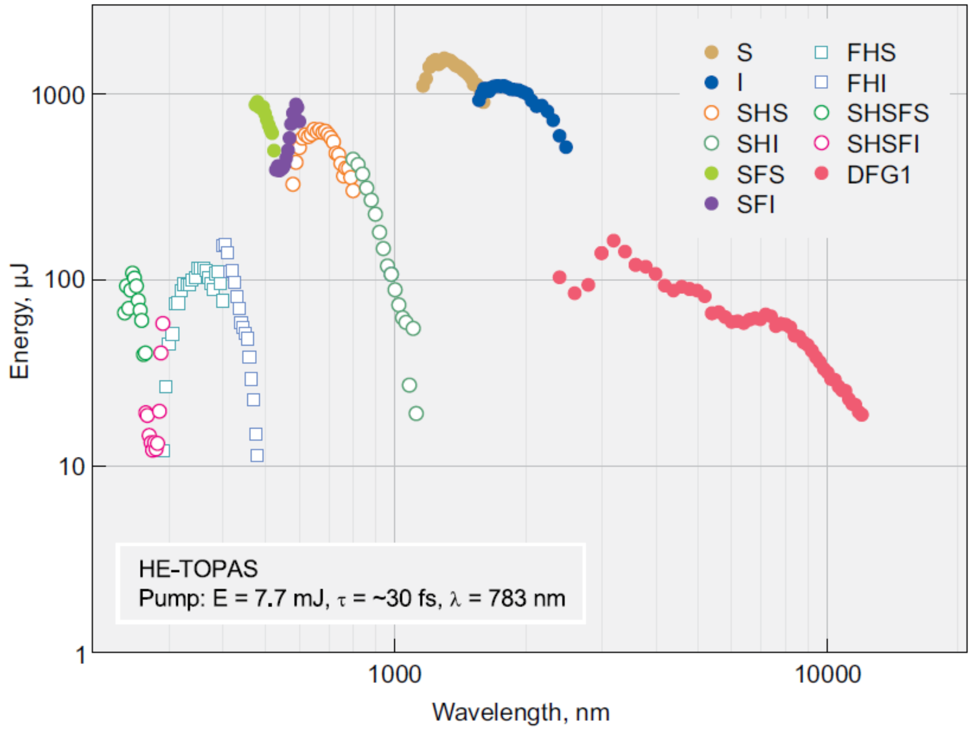
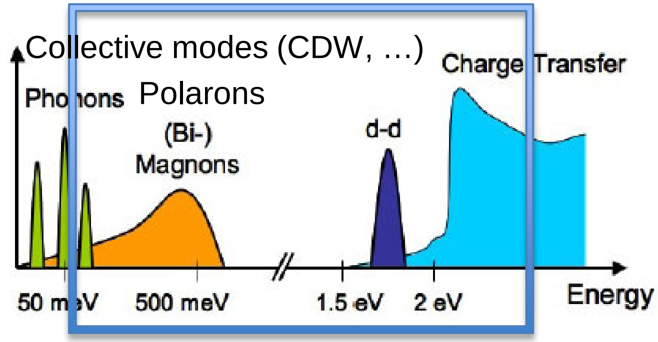
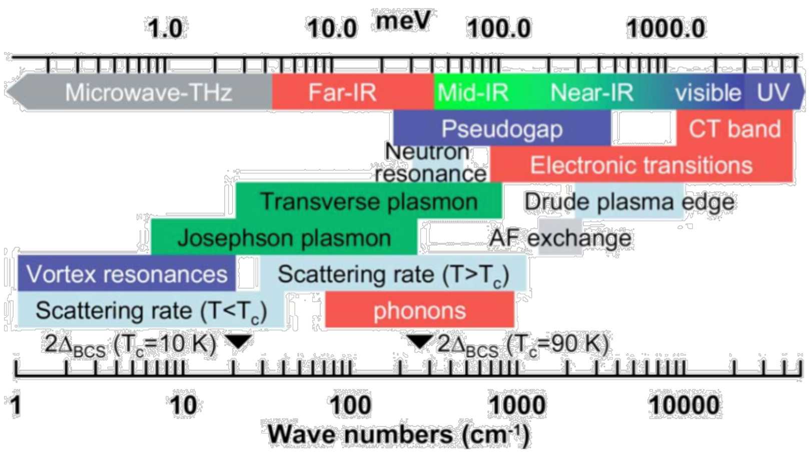
THz and mid-IR light sources

➤ **Optical parametric amplifiers: VIS to mid-IR wavelengths**



TOPAS specification sheet





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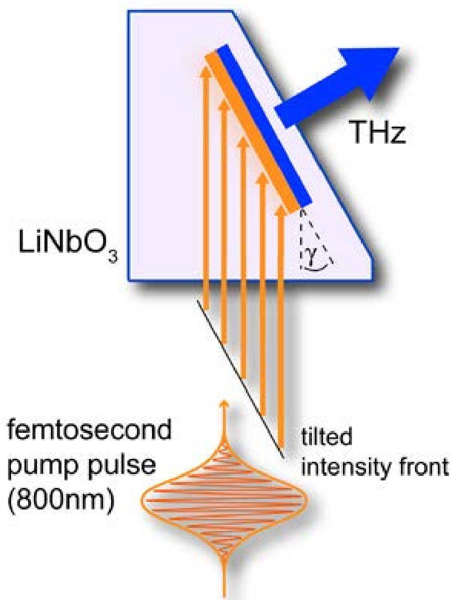
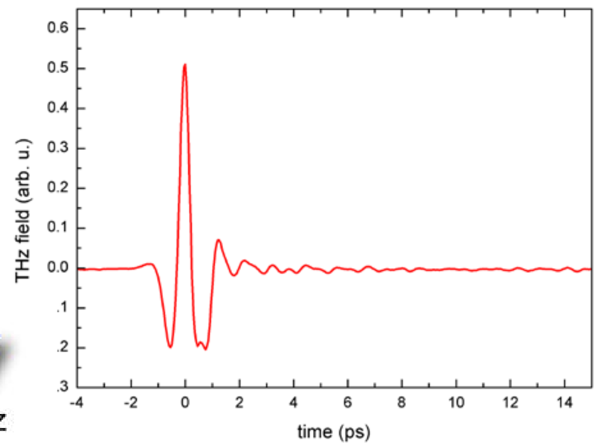
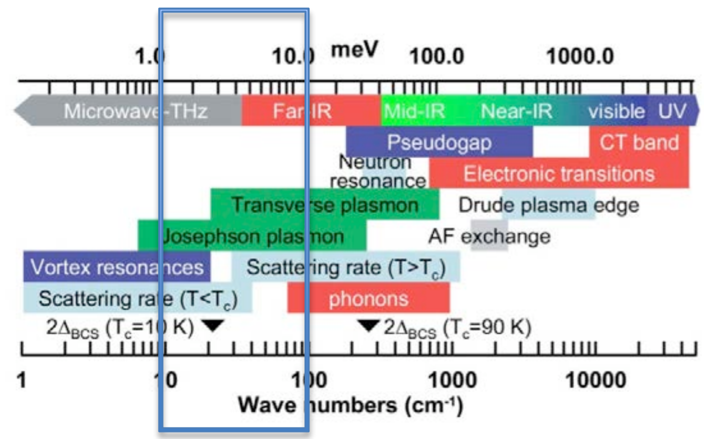
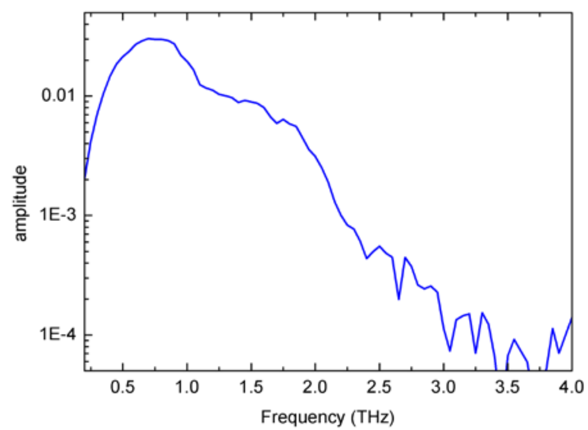
THz and mid-IR light sources

➢ Optical rectification in LiNbO3









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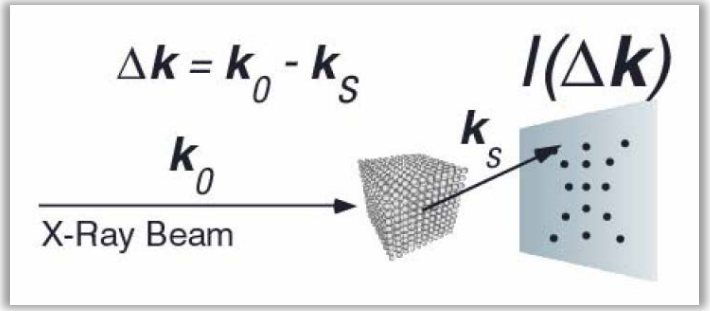
How to probe phase state dynamics?

- **Table-top pump & probe techniques**
  - **Transient optical conductivity from THz to visible spectrum**
  - **Time-resolved magneto-optics (e.g. MOKE)**
  - **Time-resolved photo-emission (e.g., ARPES)**

Structural science

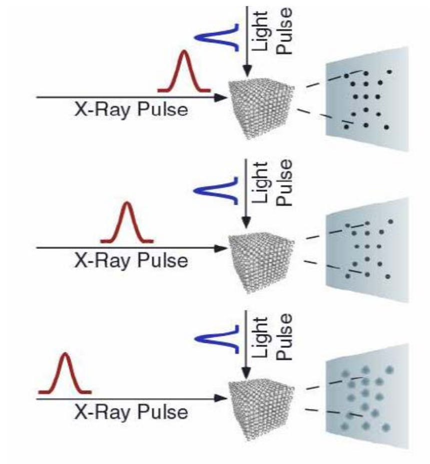
- **Investigate the microscopic arrangement of atoms, charges, orbitals, spins, … within matter**
- **Correlate the structure to the macroscopic physical/chemical (functional) properties**

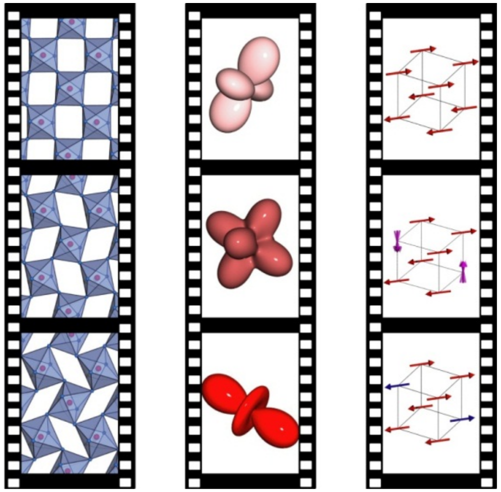


X-ray diffraction

Structural dynamics in condensed matter

- **Explore**
  - **the reaction of a system to external (optical) stimulation**
  - **the relevant mechanisms and time scales**
  - **how to control the functional properties**





Goal: resolve the atomic spatial scale *and* the inherent temporal scale of quantum dynamics

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How to probe phase state dynamics?

➢ **Diffraction techniques at FELs (femtosecond time-resolved)**

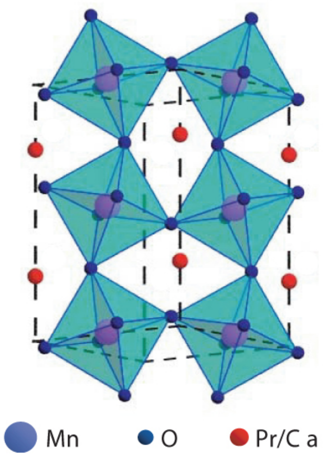
- **Resonant Soft X-ray Diffraction**
- **Hard X-ray Diffraction**

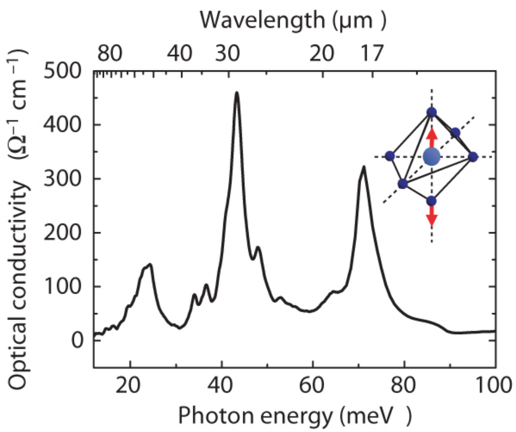
→ **established** (both of the above)

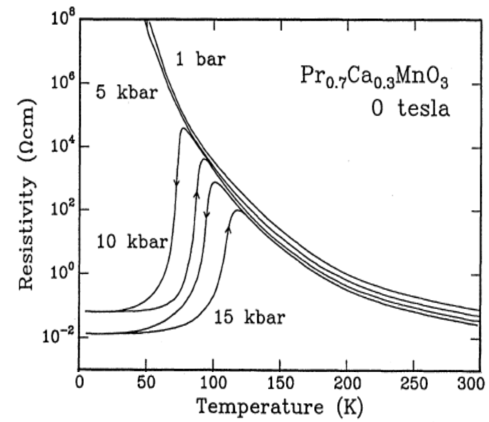
- **Femtosecond Nanocrystallography** → **first experiments done (H. Chapman)**
- **Resonant Inelastic X-ray Scattering** → **future? (J. Hill)**

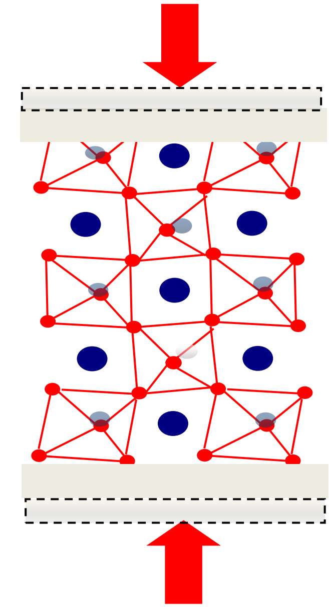
Vibrationally induced phase transitions

➢ **Insulator-metal transition in a manganite**

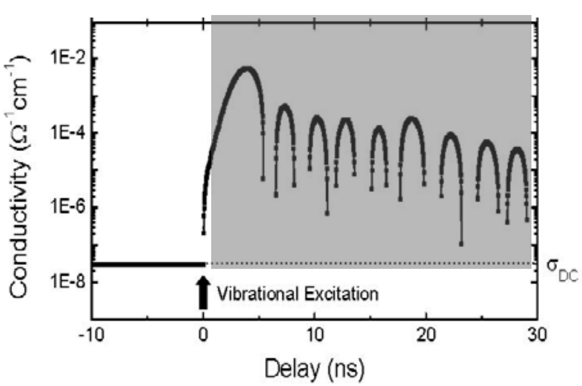




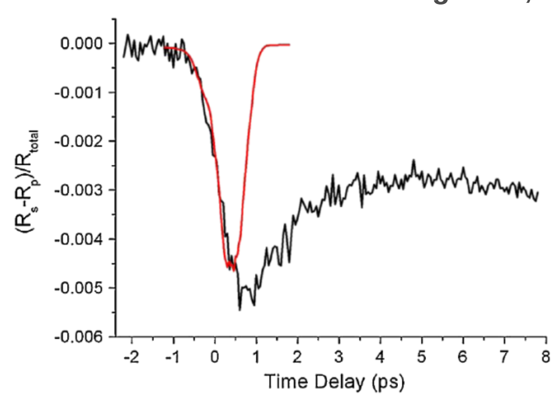




H. Hwang et al., PRL 52, 15046 (1995).



M. Rini et al.,
Nature 449, 72 (2007).



R. Tobey et al.,
PRL 101, 197404 (2009).

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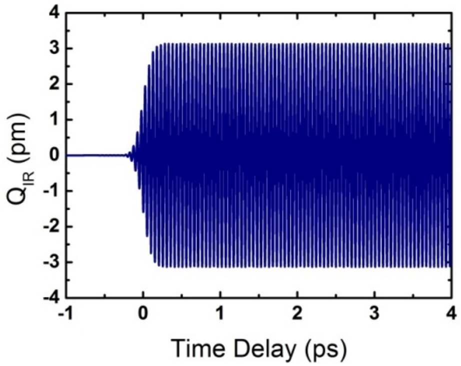
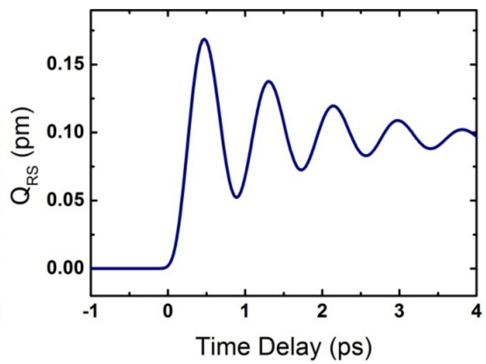
How does the optical pulse drive the lattice?

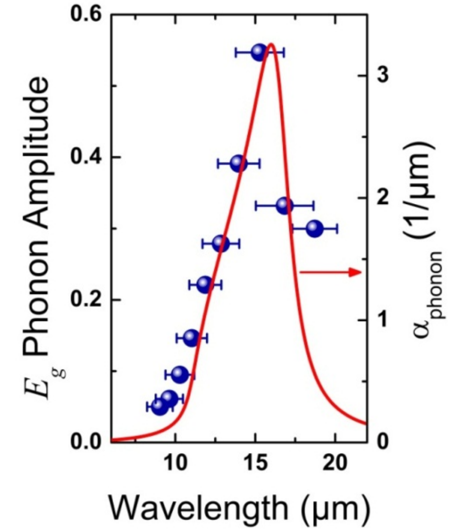
- **Nonlinear phononics:** $H_A = -NAQ_{IR}^2 Q_{RS}$

$$\ddot{Q}_{RS} + \Omega_{RS}^2 Q_{RS} = AQ_{IR}^2$$



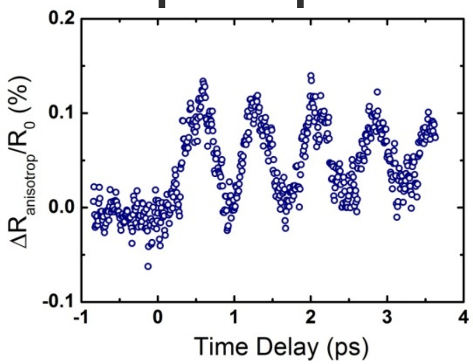


Resonant excitation

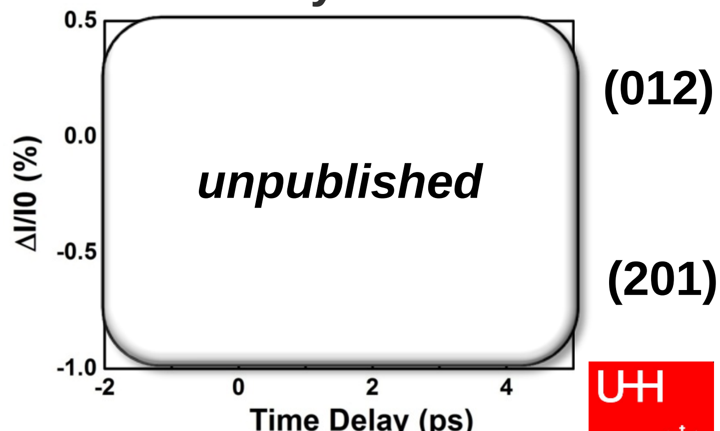


Rectification of the vibrational field

Optical probe



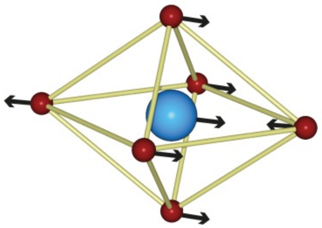
Hard X-ray diffraction

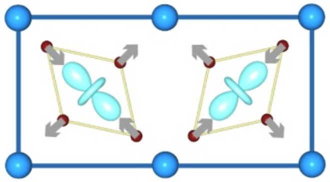


M. Först et al., Nature Physics 7, 854 (2011).
M. Först et al., submitted

Lattice control of magnetism

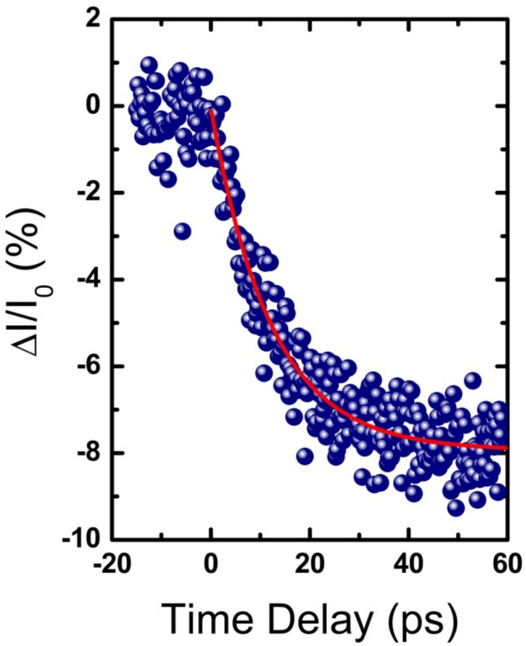
- Resonantly driven IR-active stretching mode couples to Raman-active Jahn-Teller mode

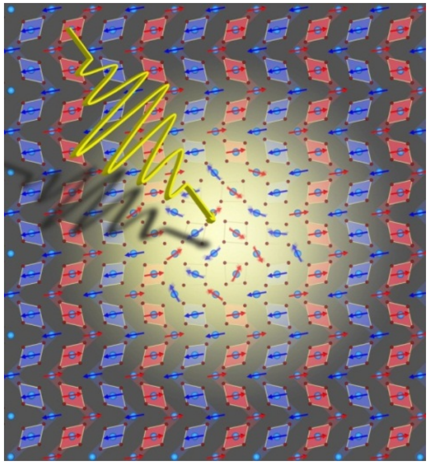




A_g
Jahn-Teller mode

- Melting of AFM order measured via RSXD at LCLS





M. Först et al,
Phys. Rev. B 84, 241104(R) (2011)

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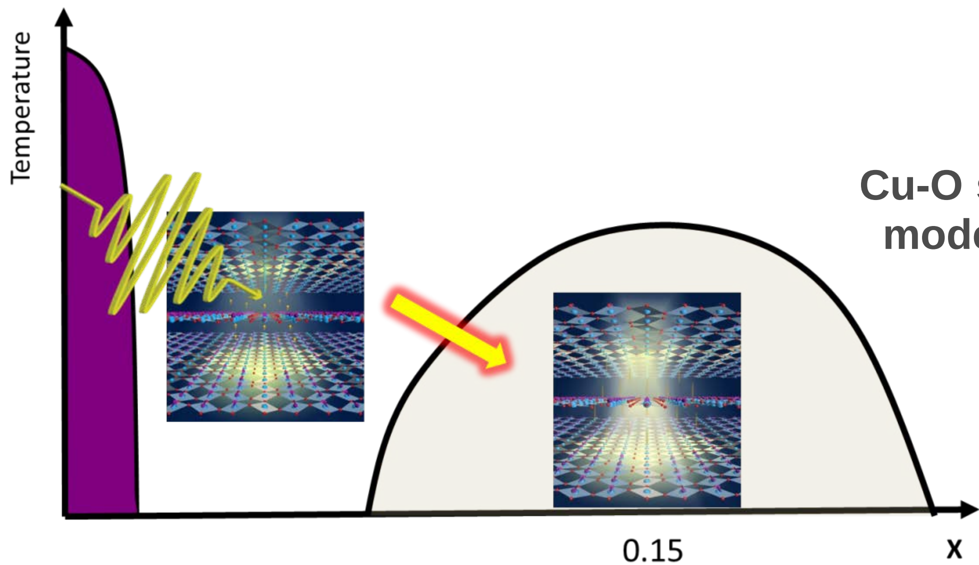
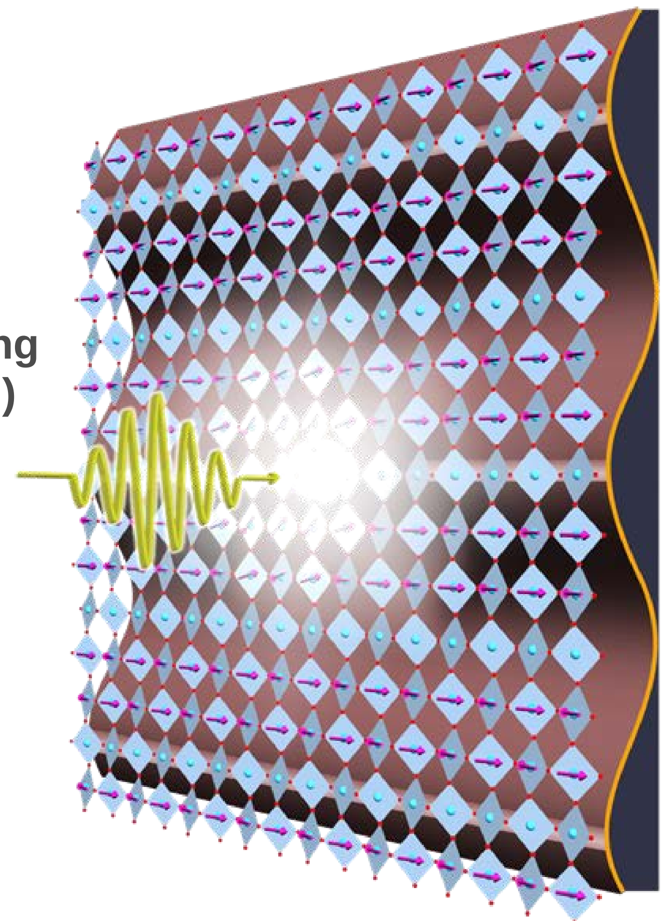
Transient superconductivity in stripe-ordered cuprate

$La_{1.8-x}Eu_{0.2}Sr_xCuO_4$



Cu-O stretching mode (17 μm)



Suppressed superconductivity
Stripe order pinned by LTT distortion

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D. Fausti et al., Science 331, 189 (2011)

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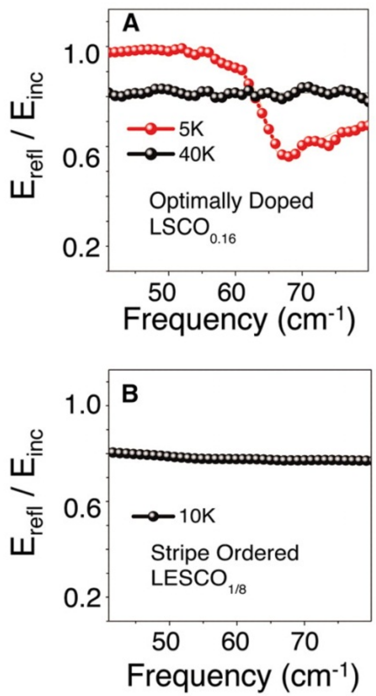
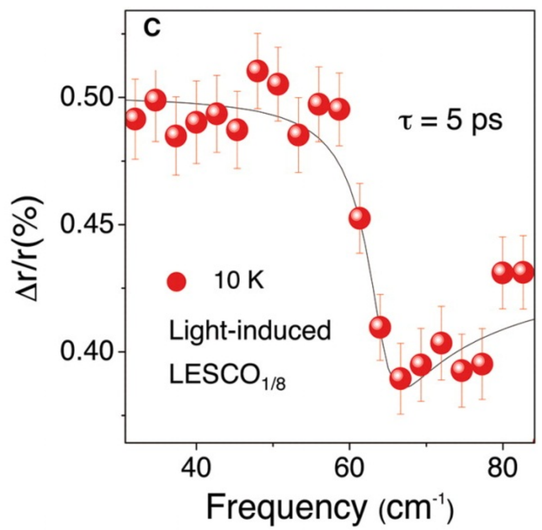
Probing in the THz domain

D. Fausti et al., Science 331, 189 (2011)

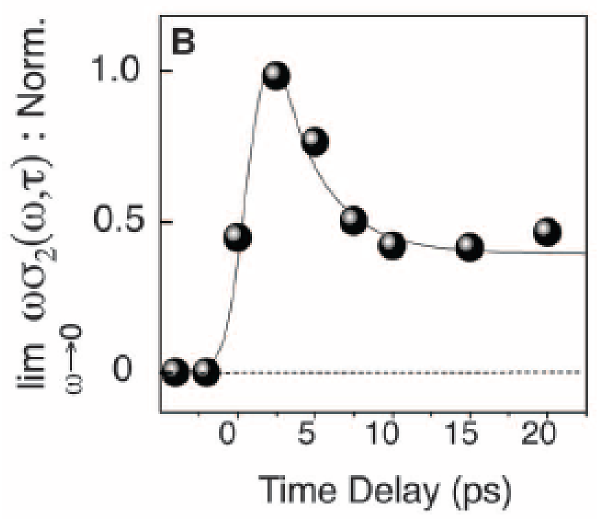
- **Josephson plasma edge**
  → interlayer phase coherence

$La_{1.8-x}Eu_{0.2}Sr_xCuO_4$





condensate density



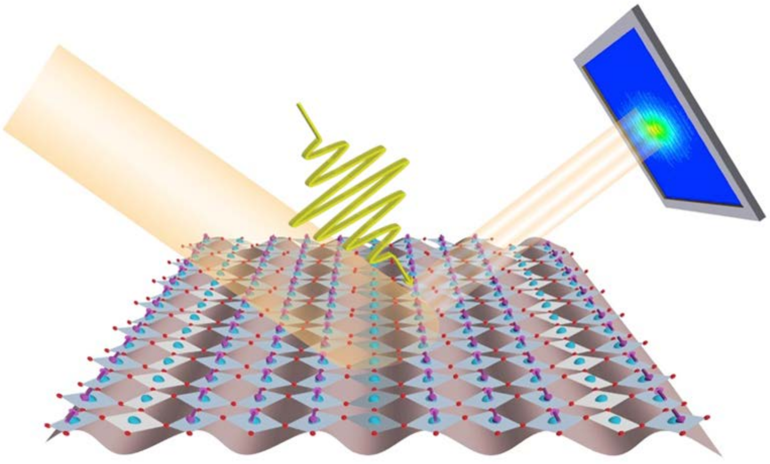
relation to stripe order and LTT distortion?

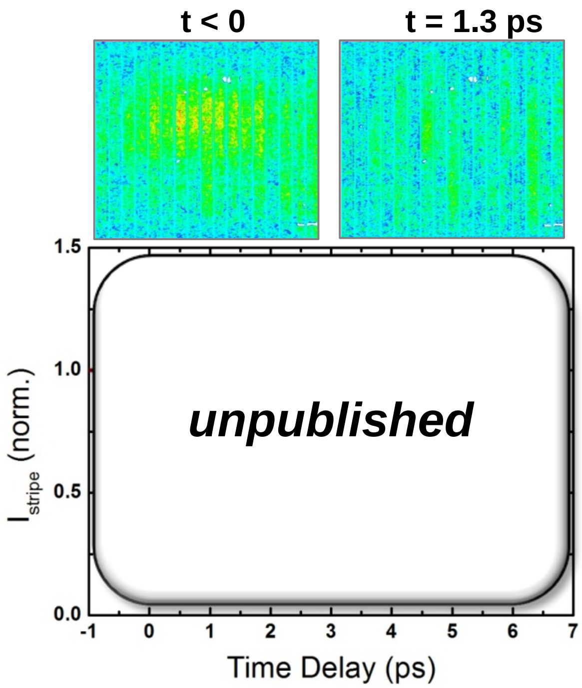
Probing stripe order via resonant soft X-ray diffraction

$La_{1.875}Ba_{0.125}CuO_4$

Charge stripe order at *q* = (0.25 0 0.5)

Oxygen K pre-edge





Complete charge order melting on sub-ps time scale

➔ no coexistence with superconductivity

M. Först, R. Tobey et al., unpublished

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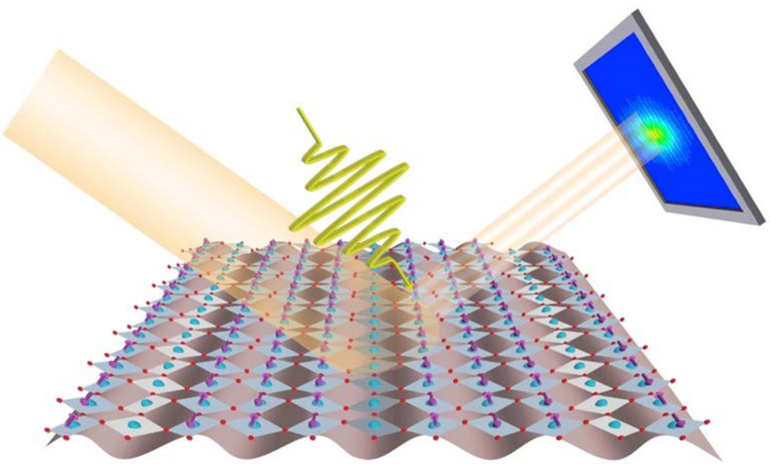
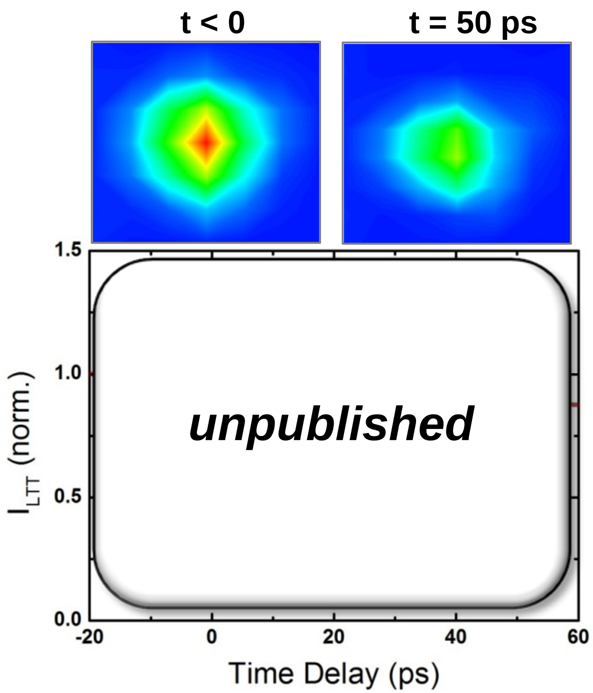
Probing LTT distortion via resonant soft X-ray diffraction

$La_{1.875}Ba_{0.125}CuO_4$



LTT distortion at q = (001)

Oxygen K edge



Weak and slow relaxation

➔ decoupling of LTT distortion and stripe order

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M. Först, R. Tobey et al., unpublished

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Requirements to the pump laser

- **minimum requirements (from experiences at the LCLS)**
  - **>10 μJ energy/pulse**
  - **~ mJ/cm^2 excitation fluences**
  - **reliable tunability and spectral bandwidth**
  - **high power and pointing stability over 1 week of beamtime**
  - **synchronization to the FEL < 250 fs**
  - **collinear alignment with the FEL beam**

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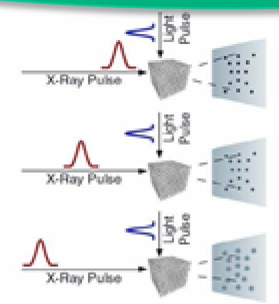
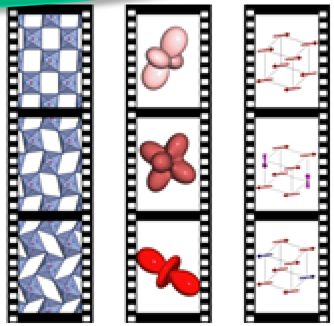
Needs for the future

Structural dynamics in condensed matter

- **Explore**
  - the reaction of a system to external (optical) stimulation
  - the relevant mechanisms and time scales
  - how to control the functional properties





Goal: resolve the atomic spatial scale *and* the inherent temporal scale of quantum dynamics

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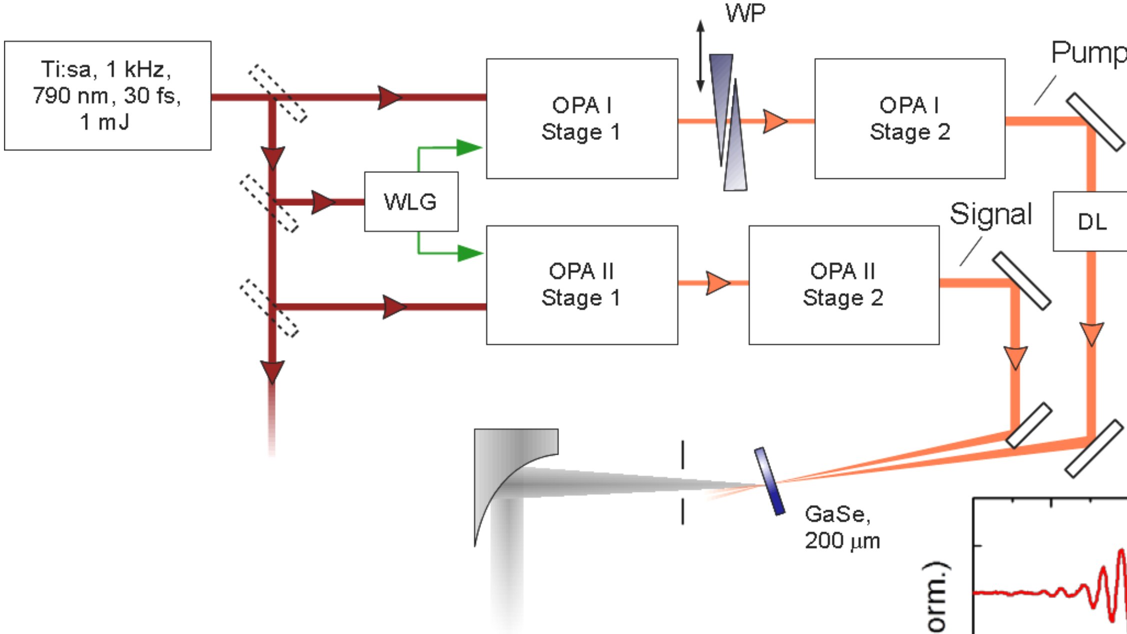
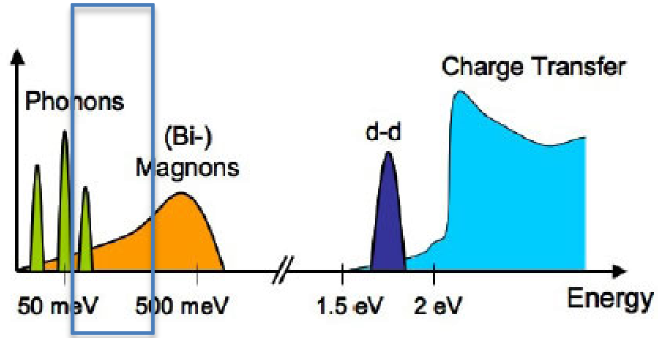
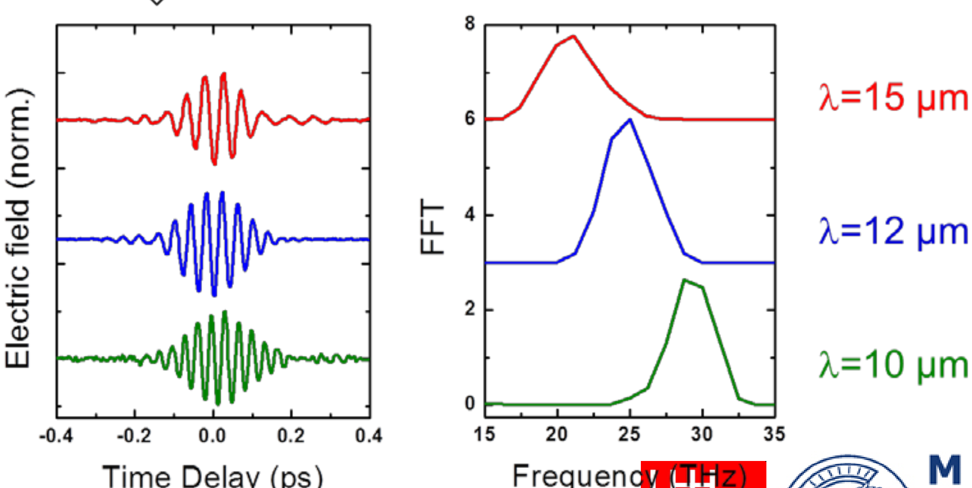
Needs/wishes for the future

- **Phase-stable mid-IR pulses**





requires timing jitter < 30 fs



C. Manzoni, M. Först et al., Opt. Lett. 35, 757 (2010)

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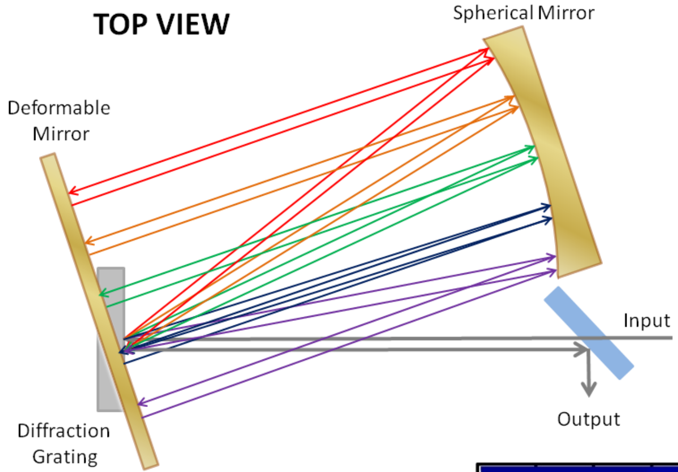
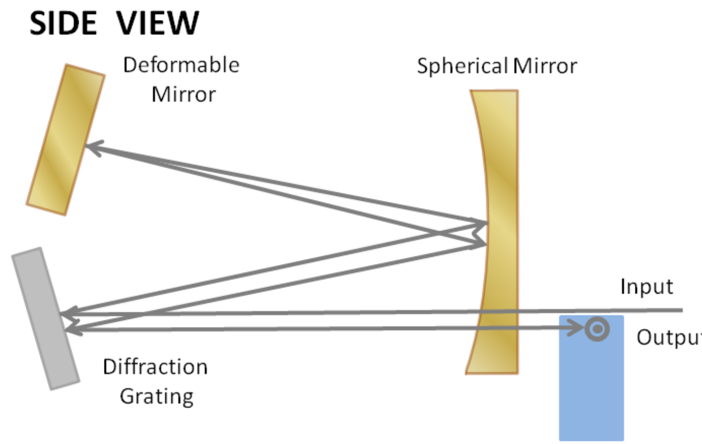
Needs/wishes for the future

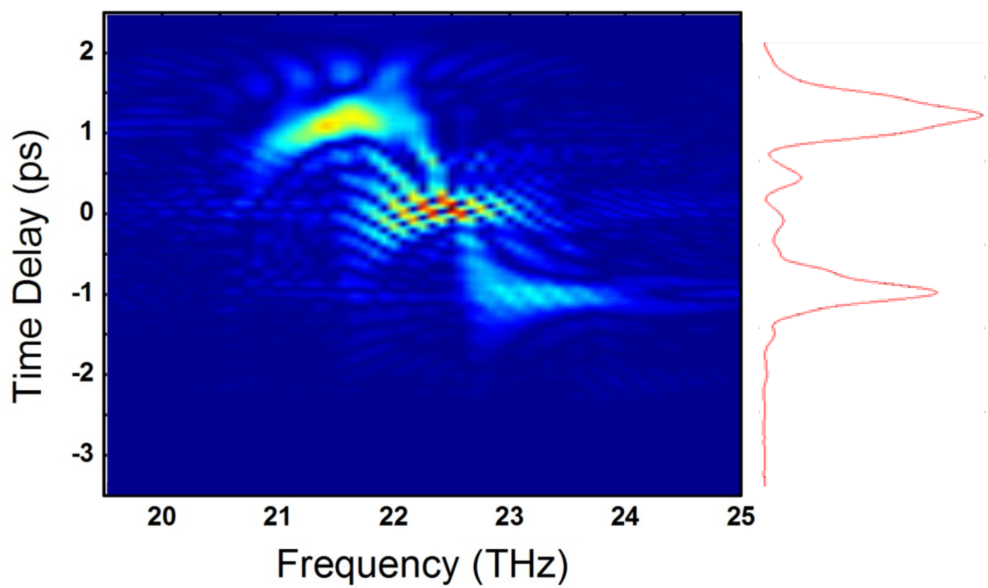
- **Mid-IR pulse shaping**

Deformable mirror:
23 electrodes
± 110µm deformation





Wigner map



C. Manzoni
S. Bonora
A. Cartella

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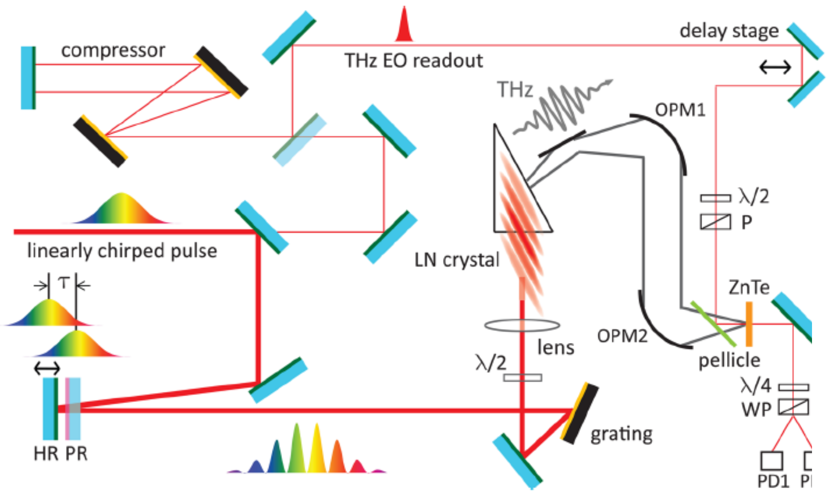
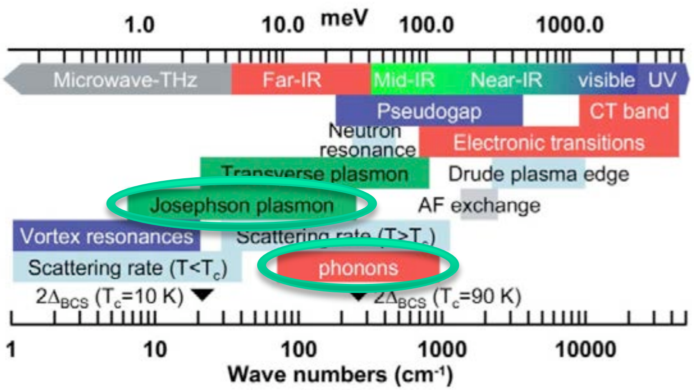
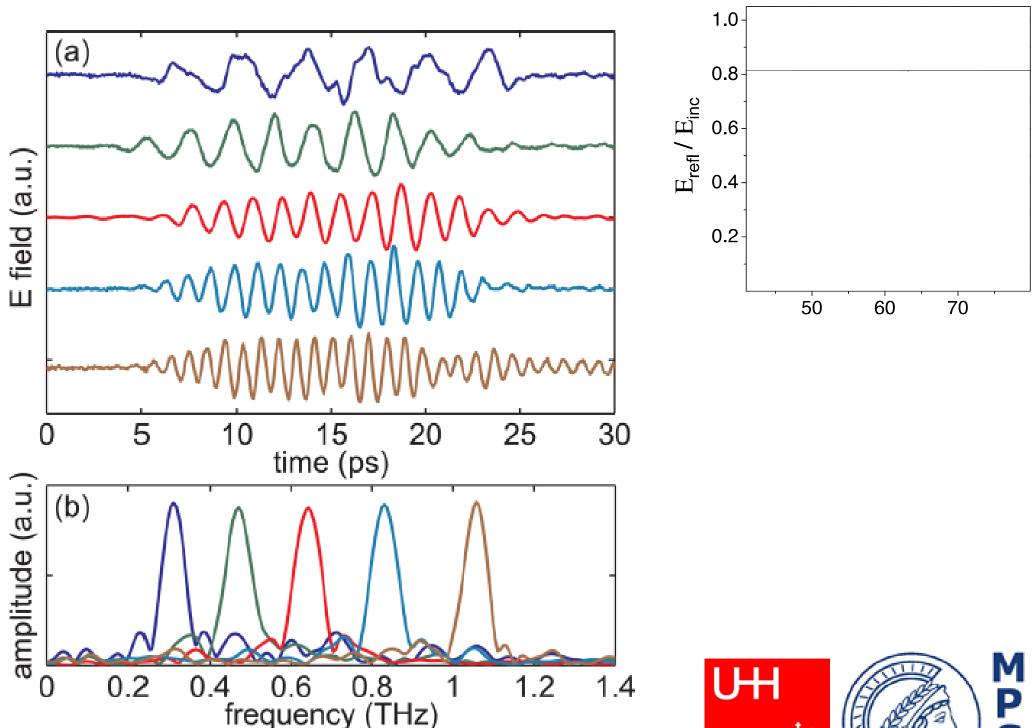
Needs/wishes for the future

➢ **Narrowband low-frequency excitations**





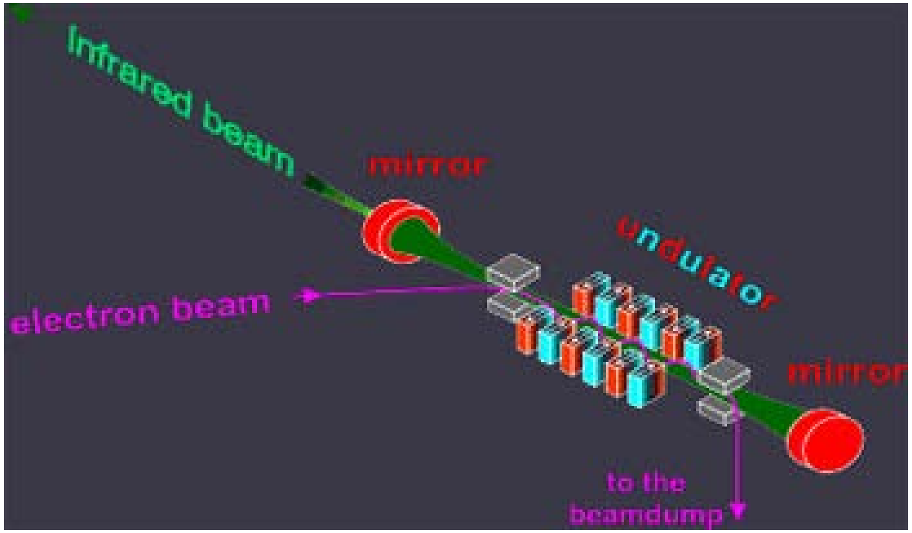


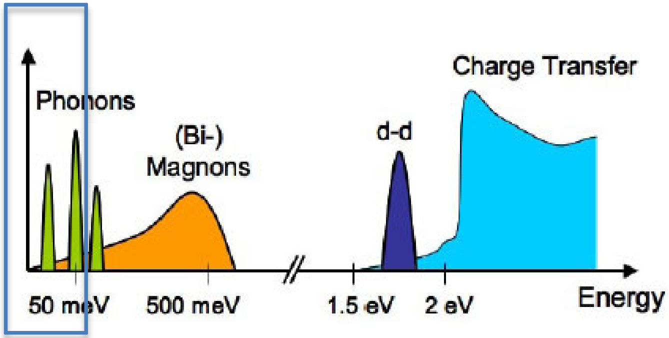
chirp&delay
intensity modulation

Z. Chen et al., Appl. Phys. Lett. 99, 071102 (2011)

Needs/wishes for the future

- **Bridge the 3-15 THz gap**





Access to:

- **further phonon modes in transition metal oxides**
- **Josephson (bi-layer) plasma modes in cuprates**

undulator based

other approaches?

Thanks to...

- H. Bromberger, V. Khanna, R. Mankowsky, C. Manzoni, S. Kaiser, H. Ehrke, A.L. Cavalieri,
  T. Garl, A. Caviglia, A. Cavalleri — *Max-Planck Research Group for Structural Dynamics, University of Hamburg*
- R.I. Tobey, S.B. Wilkins, J.P. Hill — *Physics Division, Brookhaven National Laboratory*
- S. Wall — *ECFO Barcelona*
- M. Gensch — *Helmholtz-Zentrum Dresden Rossendorf*
- S.S. Dhesi, S. Cavill — *Diamond Light Source*
- W.F. Schlotter , J.J. Turner, M.P. Minitti, A.R. Fry, D.M. Fritz, H.T. Lemke, D. Zhu, M. Chollet — *Linac Coherent Light Source, SLAC National Accelerator Laboratory*
- Y.D. Chuang — *Advanced Light Source, Lawrence Berkeley Laboratory*
- W.S. Lee, R. Moore — *SIMES, SLAC National Accelerator Laboratory and Stanford University*
- O. Krupin — *European XFEL GmbH*
- S.L. Johnson — *Eidgenössische Technische Hochschule Zürich*
- R. Merlin — *University of Michigan*
- M. Trigo — *PULSE, SLAC National Accelerator Laboratory*
- J.C. Mitchell — *Materials Science Division, Argonne National Laboratory*
- Y. Tomioka — *Correlated Electron Engineering Group, AIST, Tsukuba,*
- Y. Tokura — *Department of Applied Physics, University of Tokyo*
- R. Scherwitzl, J.-M. Triscone — *University of Geneva*
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