ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

(Ultrafast) Soft X-ray Spectroscopy of Correlated Electron Materials

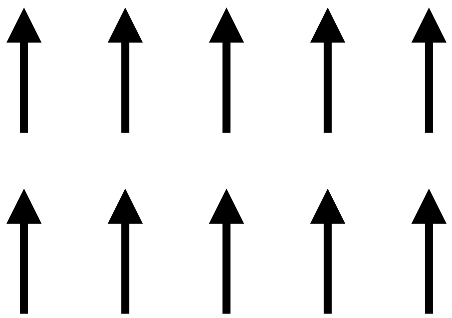
Steve Johnson

ETH Zurich

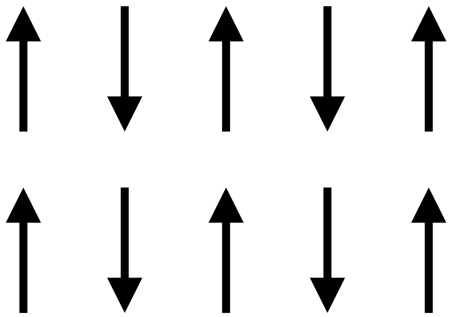
Outline

- Correlated electron systems: why and what is there to learn?
- Role of soft-x-ray spectroscopies at FELs
- Lessons from ultrafast optics
- Frontiers

Correlated Electron Systems

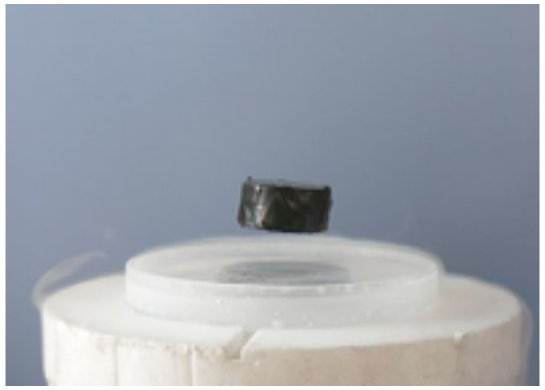


Ferromagnetism



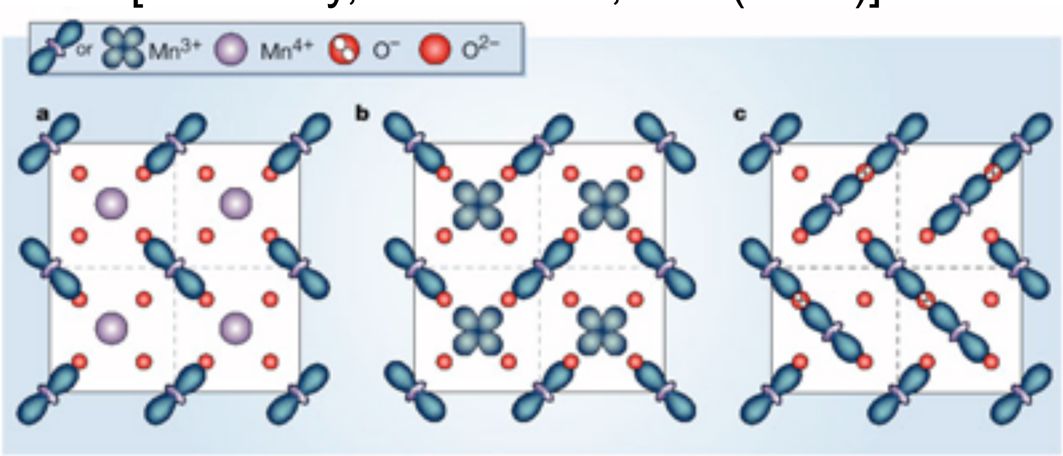
Antiferromagnetism

[Mai-Linh Doan, CC BY-SA 3.0]



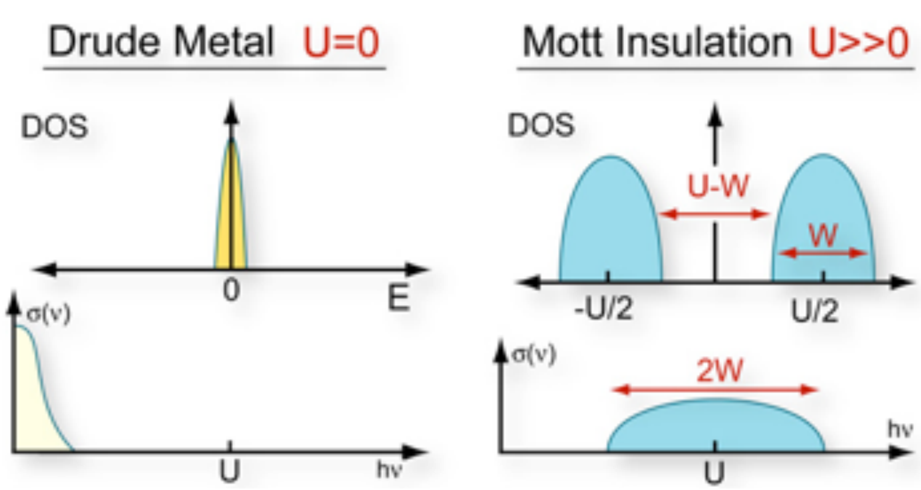
Superconductivity

[M. Cohey, *Nature* **430**, 155 (2004)]



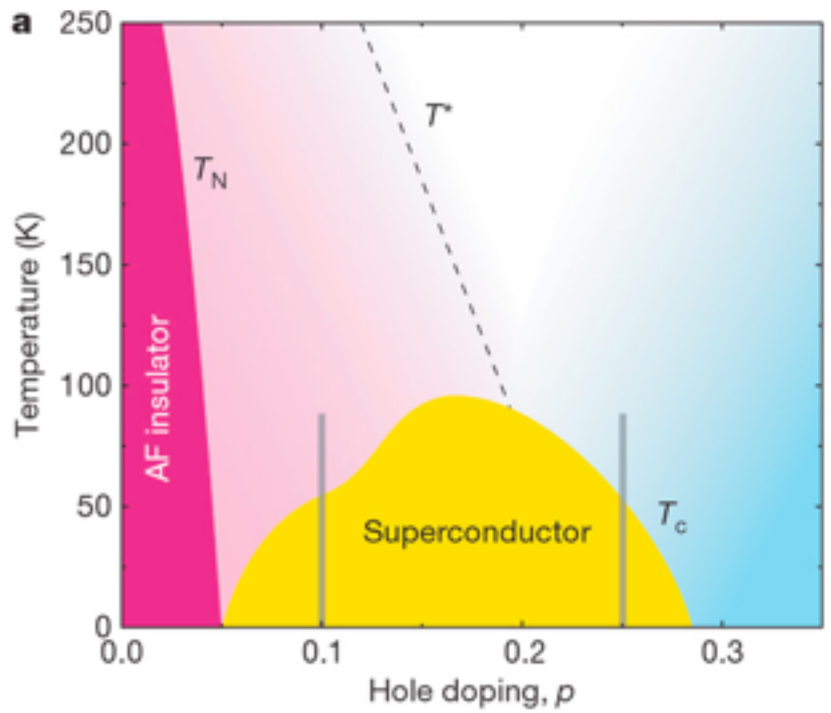
Charge and orbital order

[http://mpsd-cmd.cfel.de/research-scie-motti.html]

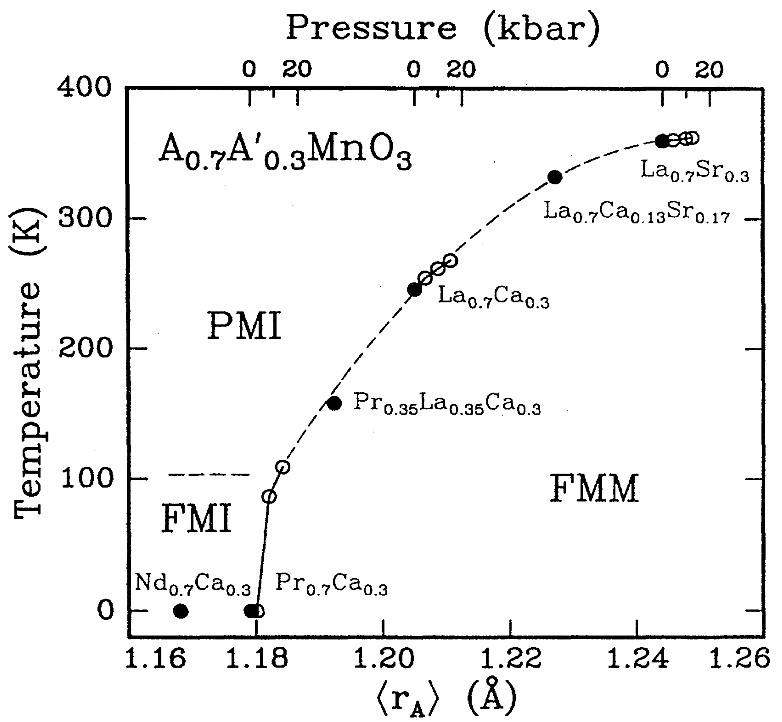


Mott insulation

Correlated Electron Systems



[Doiron-Leyraud et al. Nature 447, 565 (2007)]



[Hwang et al. PRB 52, 15046 (1995)]

- Interactions strongly compete
- Complex phase diagrams

General Questions

- What is the physical origin of the correlations in particular materials?

- Is it possible to manipulate or guide correlations?

General Questions

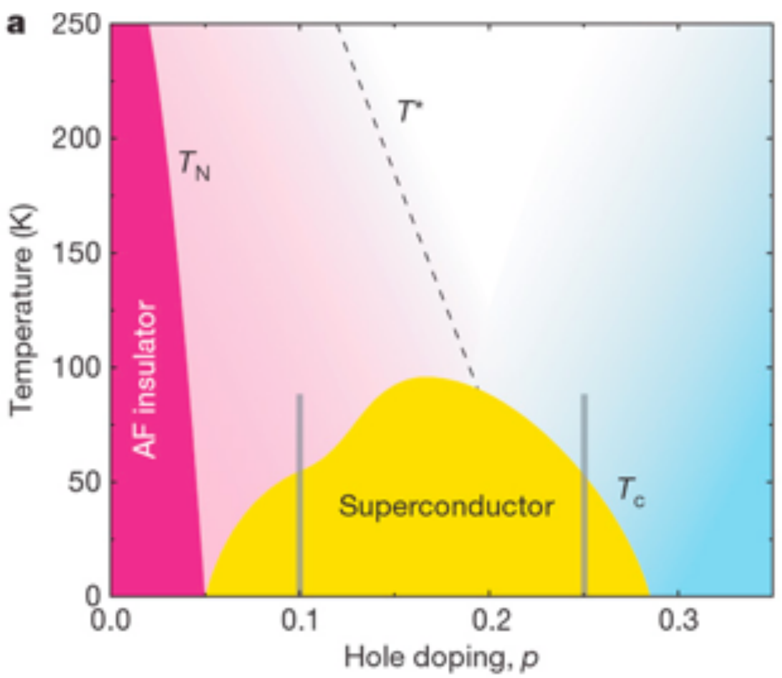
- What is the physical origin of the correlations in particular materials?

- Is it possible to manipulate or guide correlations?

ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

General Questions

- What is the physical origin of the correlations in particular materials?
  - Measure strength of interactions
  - Compare over materials, phases



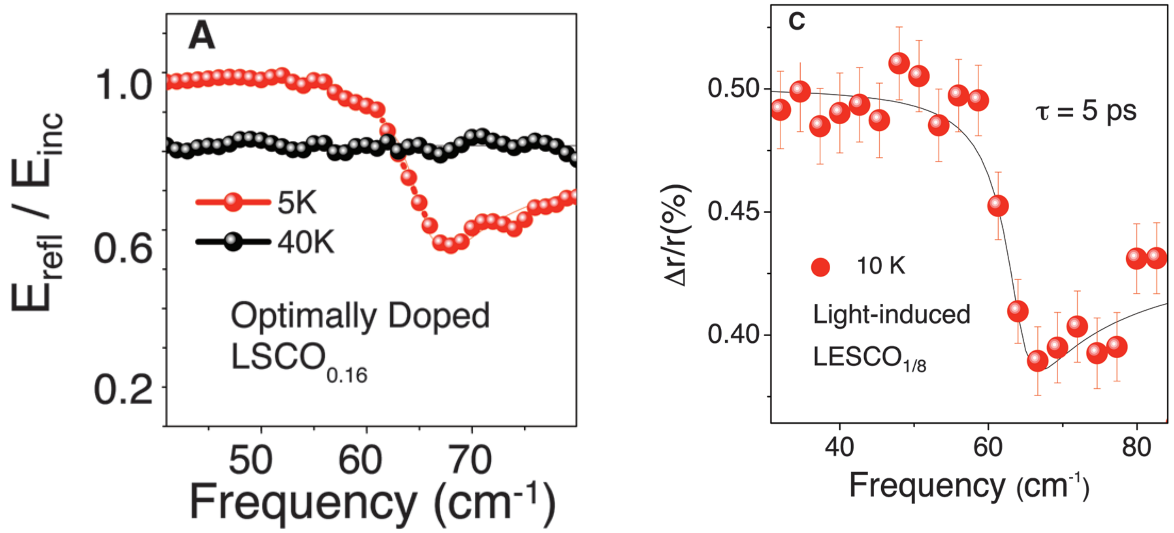
  - Difficulties:
    - Needs theory to directly relate to correlations
    - Comparisons usually change multiple variables, hard to sort out causality
  - Ideal: study interactions vs. multiple single coordinates that switch correlations on and off (quickly)

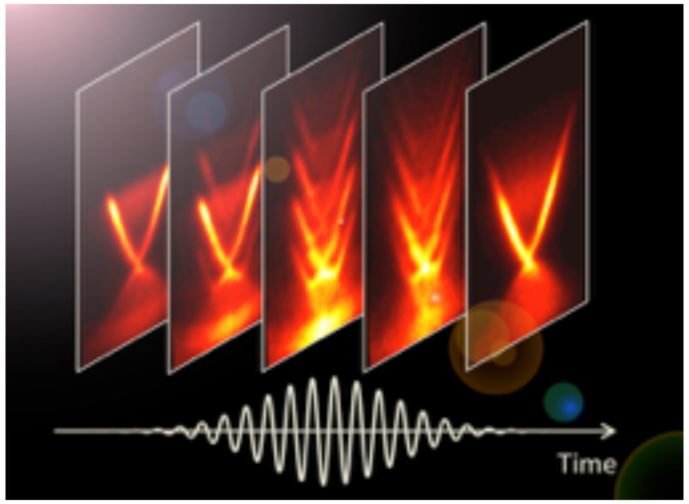
[Doiron-Leyraud et al. Nature 447, 565 (2007)]

General Questions

- Is it possible to manipulate or guide correlations?
  - “Designer” materials
  - Guided search relies on understanding of mechanisms
  - Variety of control parameters
  - Transient vs. persistent

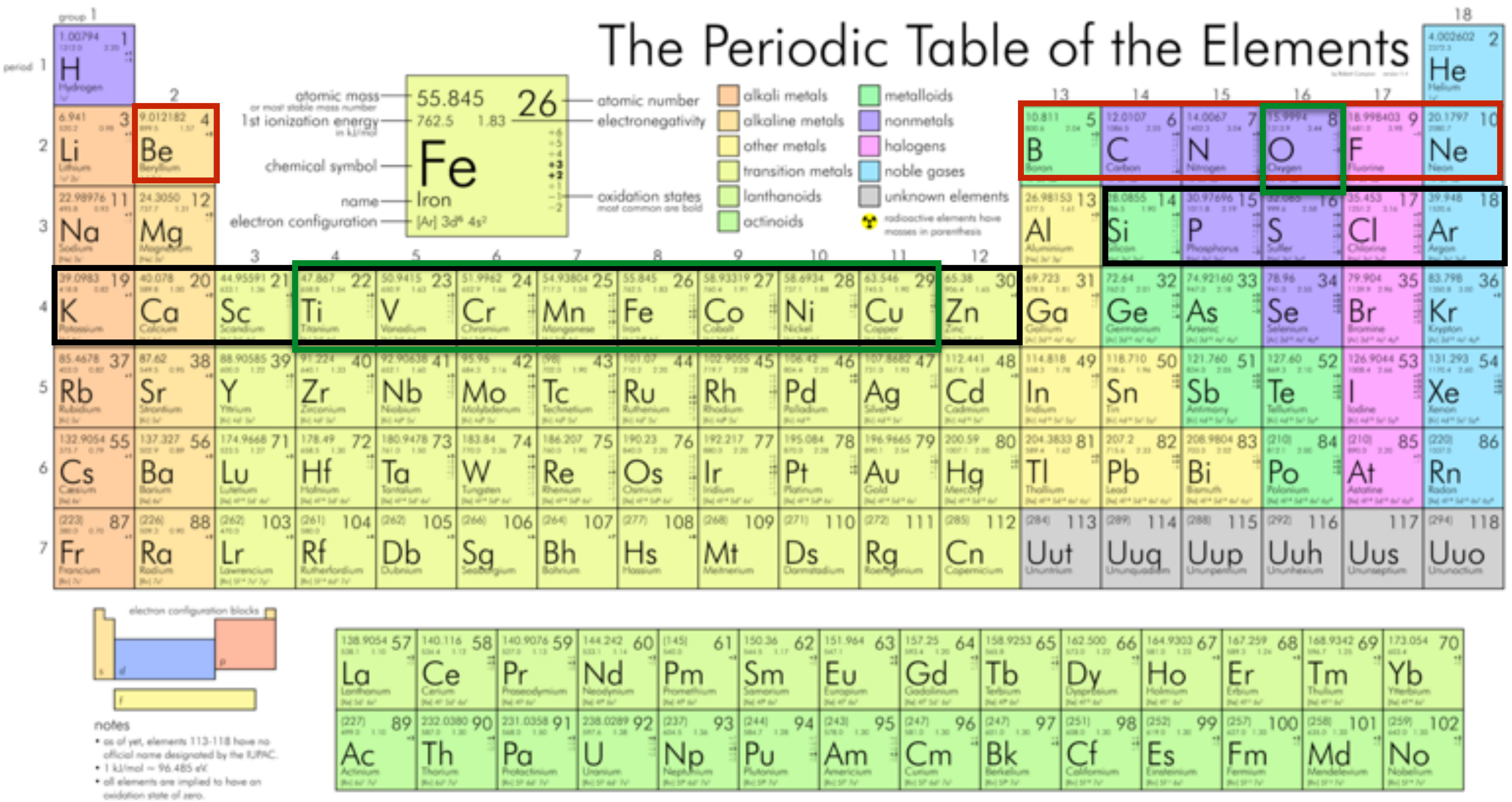
Light-Induced Superconductivity in a Stripe-Ordered Cuprate
D. Fausti *et al.*
Science **331**, 189 (2011);
DOI: 10.1126/science.1197294





[F. Mahmood et al., Nature Physics 12, 306 (2016)]

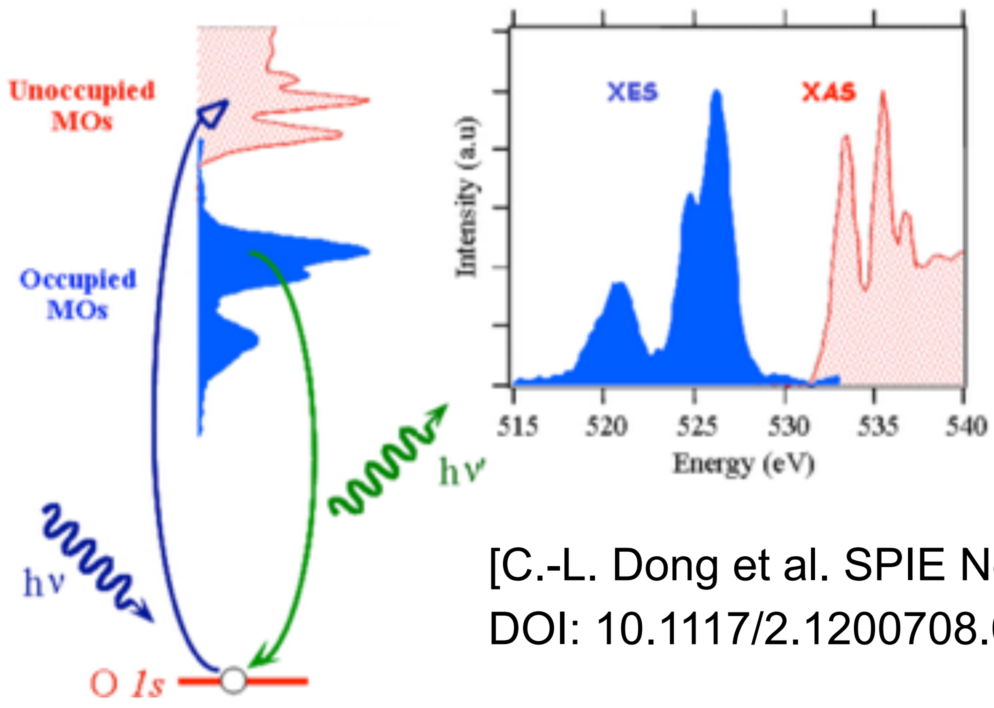
X-ray spectroscopies



- Access to O K-edge, 3d L-edges

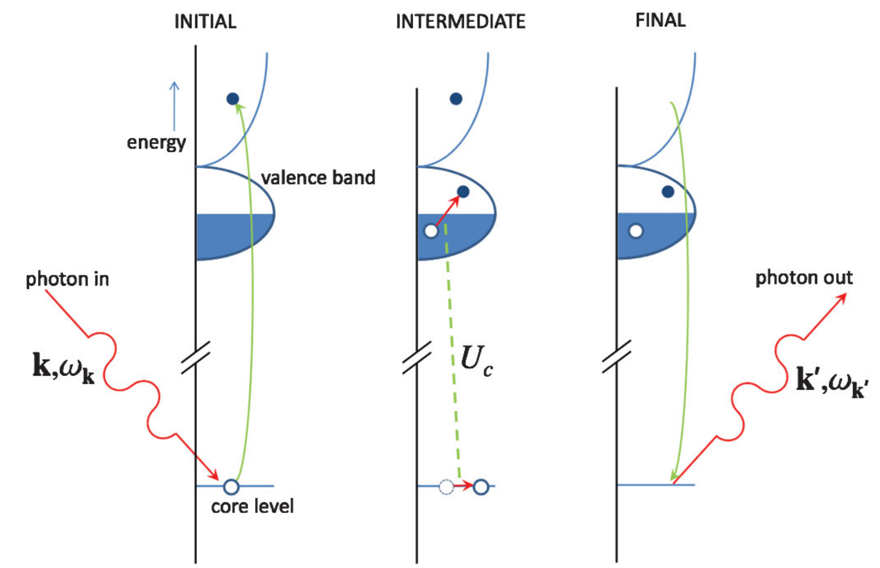
X-ray spectroscopies

X-ray absorption



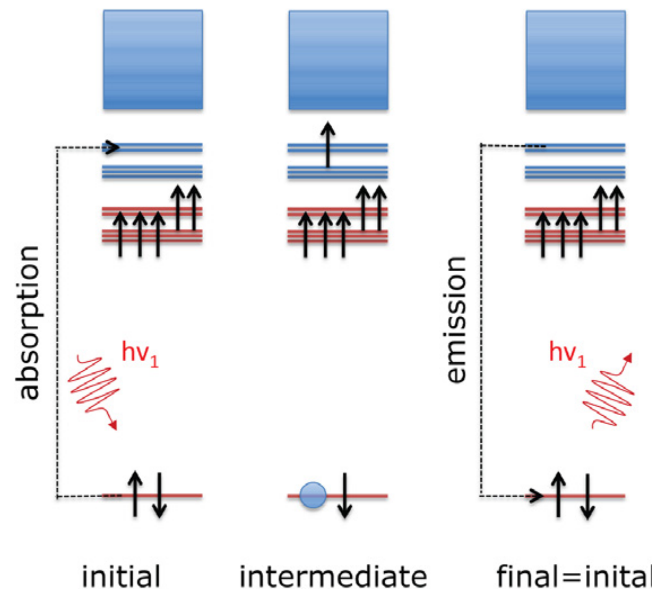
[C.-L. Dong et al. SPIE Newsroom. DOI: 10.1117/2.1200708.0812 (2007)]

- Correlated systems:
  - $L_{2,3}$-edges of transition metals: 3d orbitals
  - K-edge of O: 2p orbitals



[Ament et al. RMP **83**, 709 (2011)]

Resonant inelastic scattering

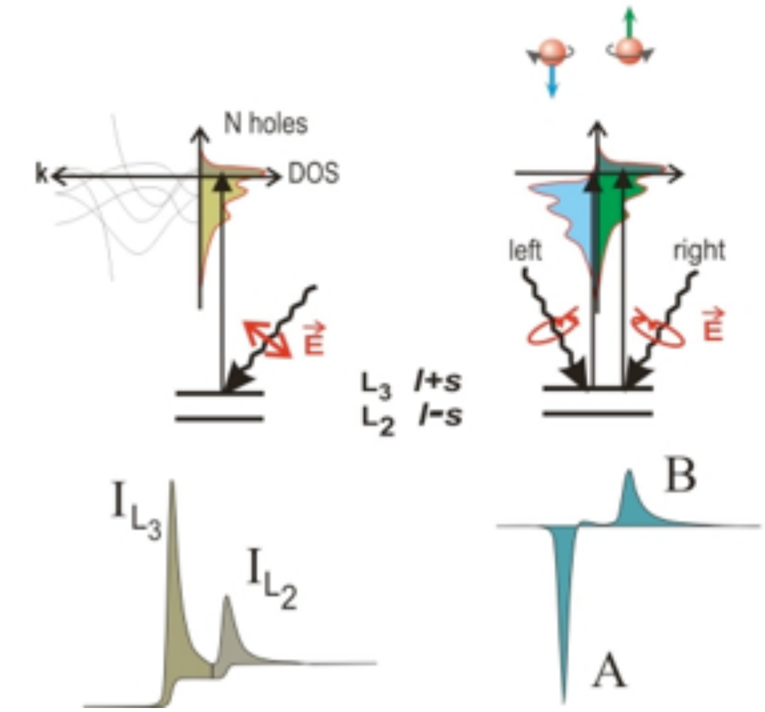


[Fink et al. Rep. Prog. Phys. **76**, 056502 (2013)]

Resonant elastic scattering

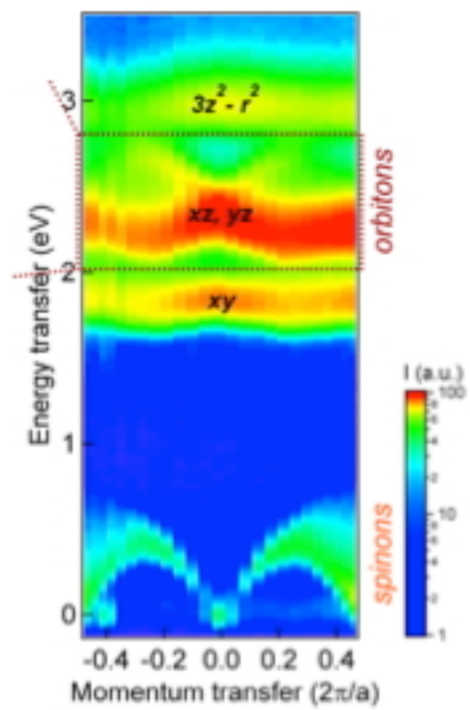
X-ray spectroscopies

- XAS: electronic structure, element-specific magnetic moment (XMCD)



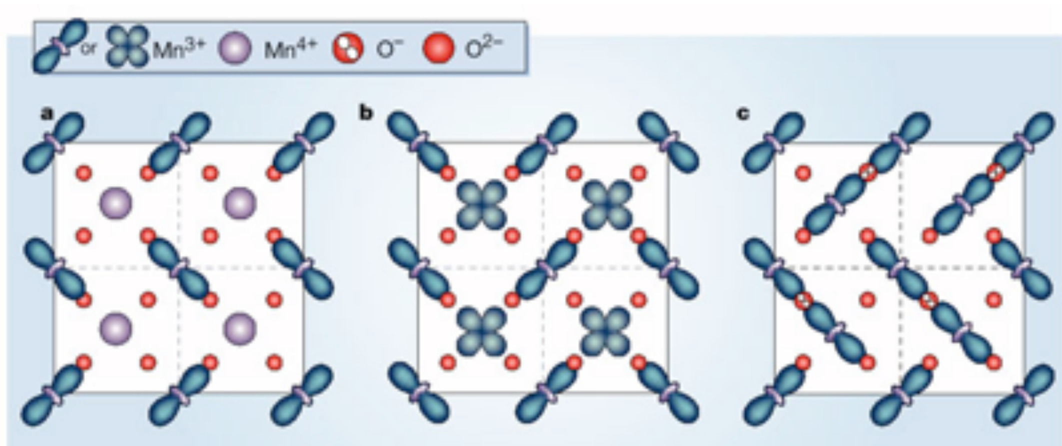
https://www-ssrl.slac.stanford.edu/stohr/xmcd.htr

- RIXS: low-energy excitations, dispersion



[J. Schlappa et al., Nature 485, 82 (2012)]

- REXS: long-range order of valence orders



[M. Cohey, *Nature* **430**, 155 (2004)]

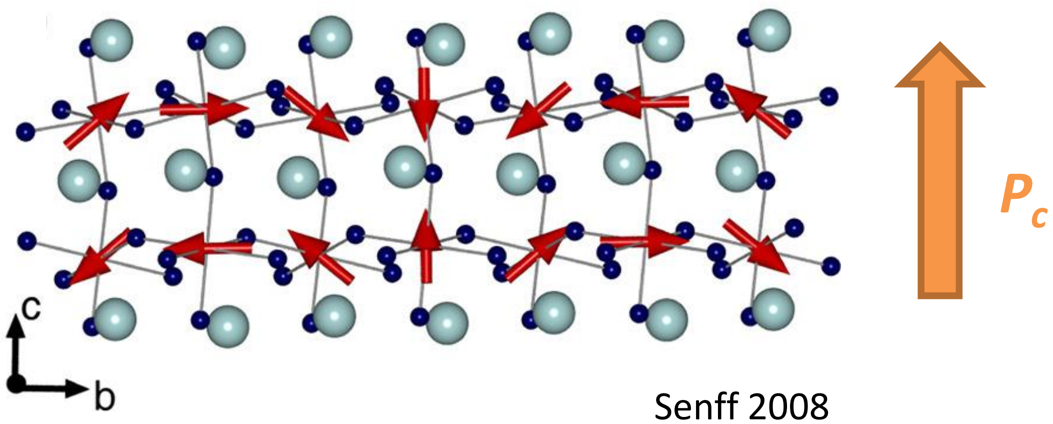
Why FEL?

- Main advantages for time-resolved (pump-probe) measurements
- As probe:
  - “Snapshots” of electronic, magnetic structure (XAS, REXS)
  - Applied to RIXS, gives coupling to low-energy excitations (vs. momentum)
- As pump:
  - Very fast decoherence (~ fs), scattering to many other states (heating)
  - Sample damage issues (esp. for nonlinear cases)

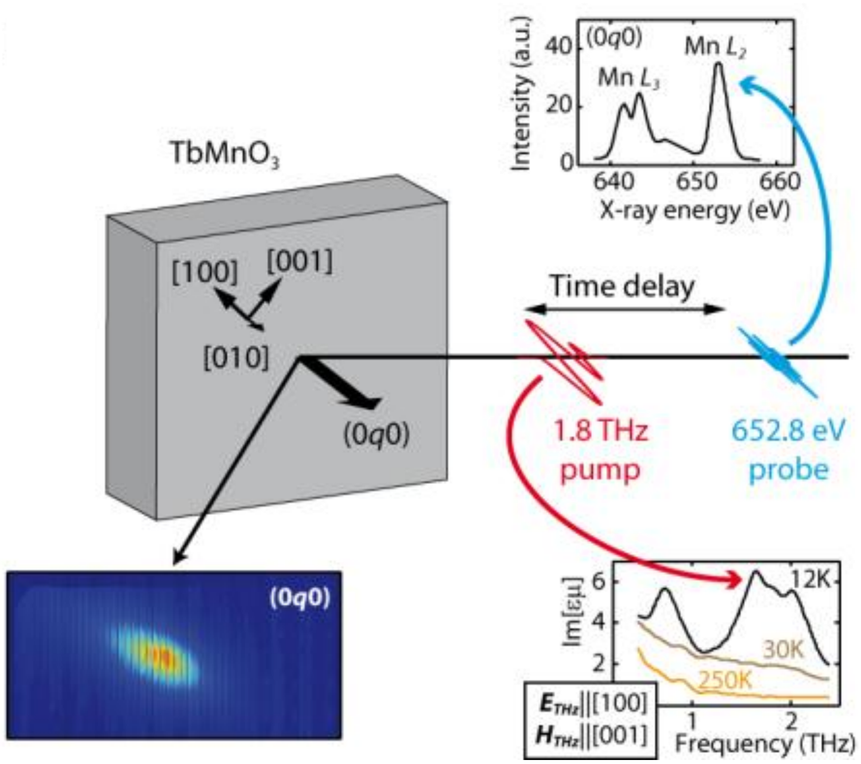
ETH
Eidgenössische Technische Hochschule Zürich
Swiss Federal Institute of Technology Zurich

Example: THz pump, REXS probe

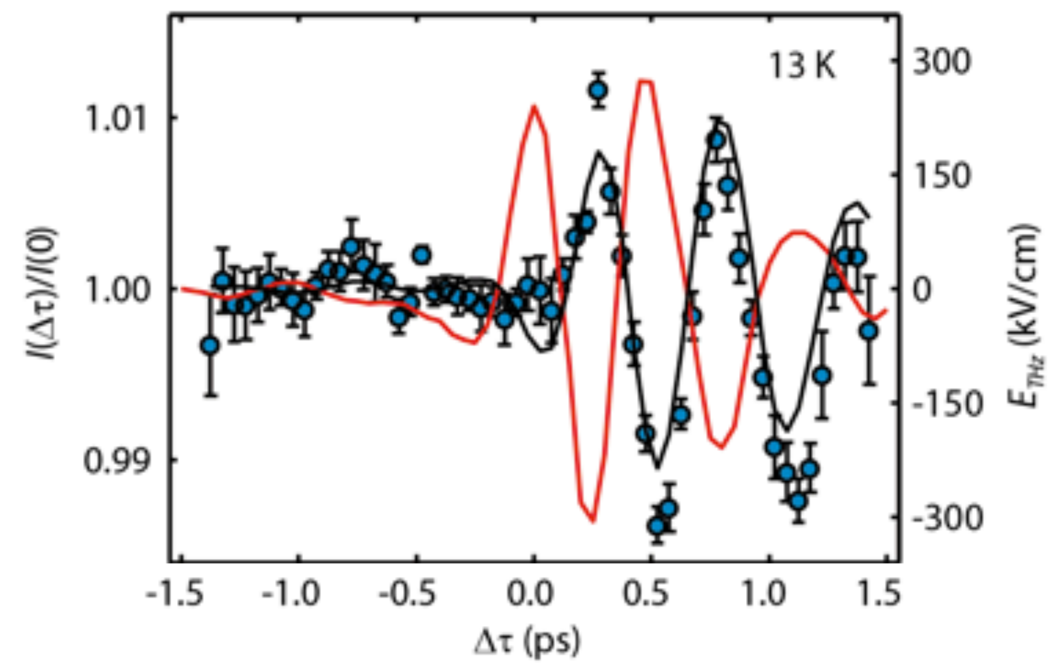
TbMnO3



Senff 2008

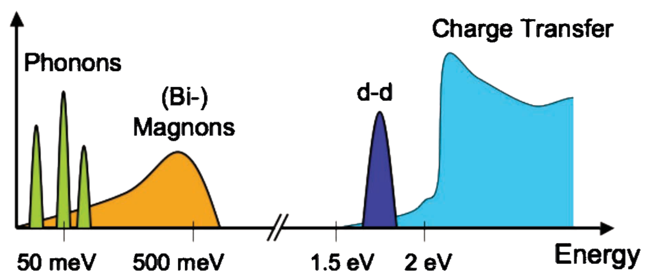


[T. Kubacka et al., Science **343**, 1333 (2014)]



- THz pump of “electromagnon”
  - Hybrid of spin and lattice excitations
  - Very specific excitation: minimal competing channels
- RXES probe of magnetic structure shows spin response

Time vs. Frequency domain

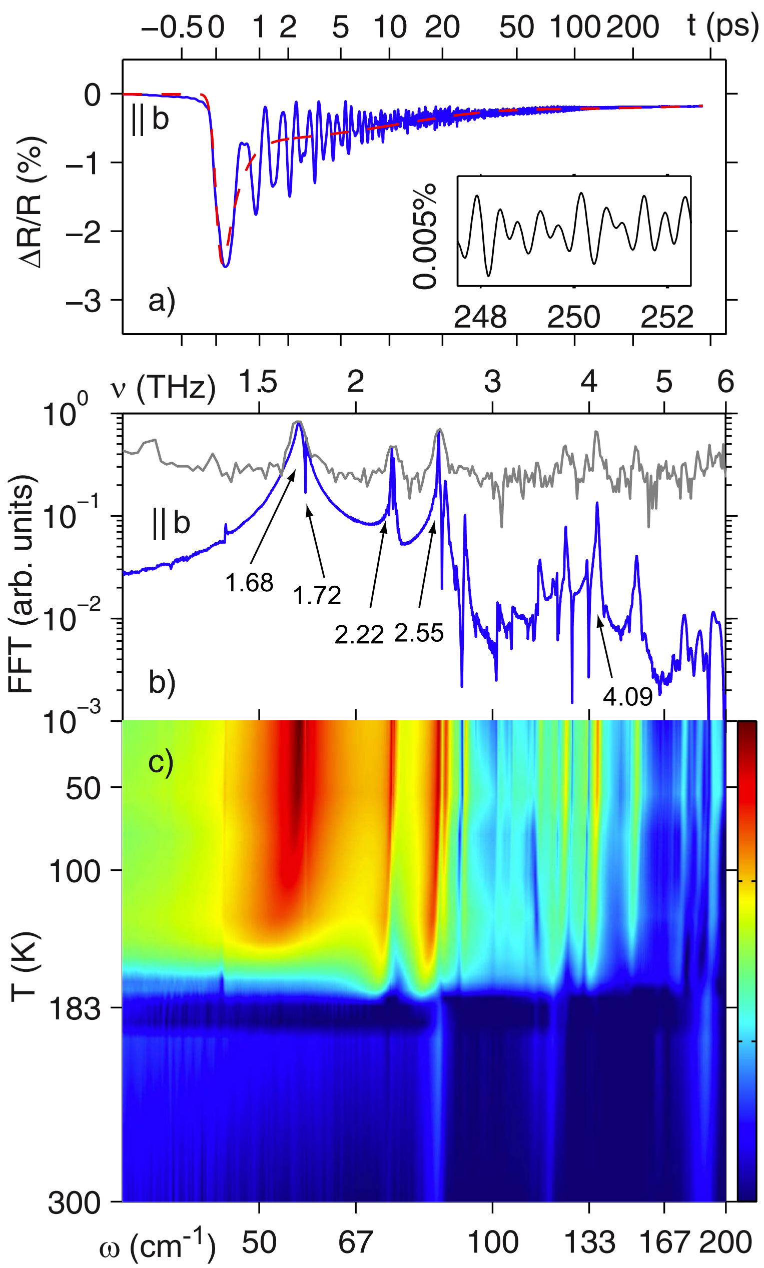


[Ament et al. RMP **83**, 709 (2011)]

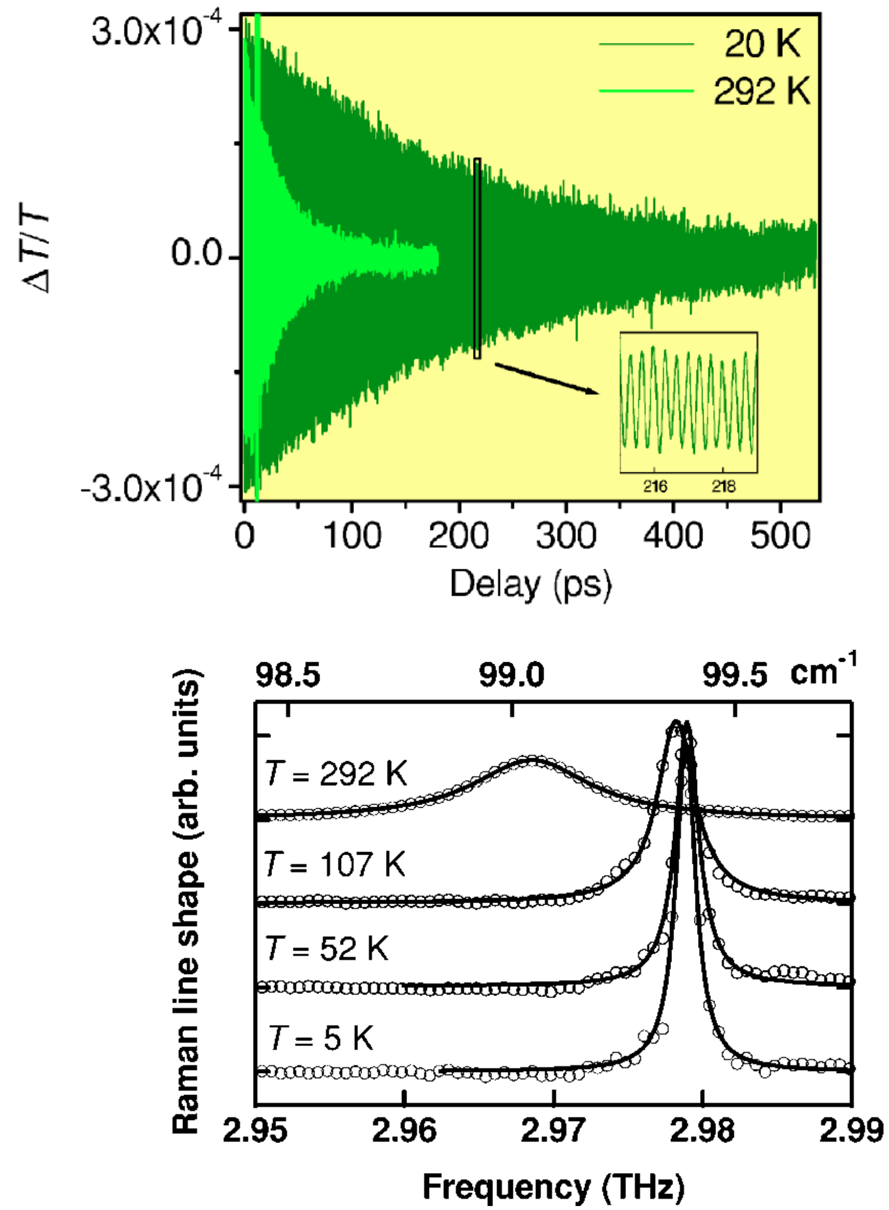
$E = h\nu$	$T = 1/\nu$
10 meV	414 fs
50 meV	83 fs
500 meV	8.3 fs
1.5 eV	2.8 fs
2 eV	2 fs

- High time resolution: unique opportunity to explore new time-domain analogs of conventional spectroscopies

Time vs. Frequency domain



[H. Schäfer et al. PRL **105**, 066402 (2010)]



[C. Aku-Leh et al. PRB **71**, 205211 (2005)]

- Optics: pump-probe in perturbative regime gives information similar to spontaneous Raman scattering
- Requires pulses shorter than period

Time vs. Frequency domain

phonon creation & annihilation operators

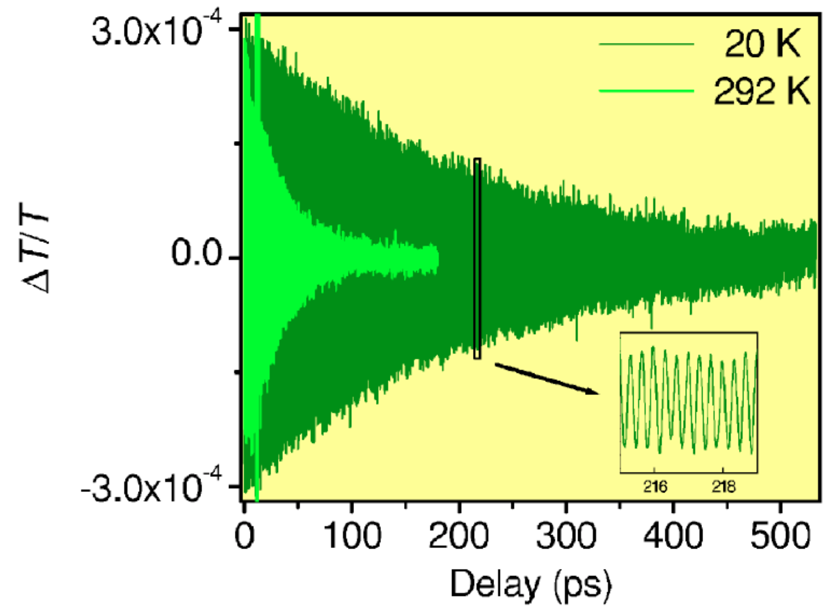
quasiparticle annihilation & creation operators

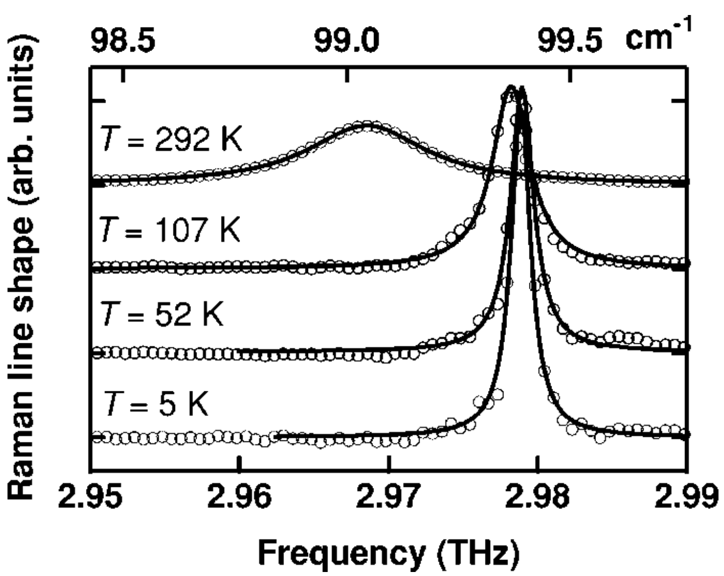
$$H_I \propto \sum_{q,b,b',k,k'} \Xi_{q,b,b',k,k'} \left(a_q^\dagger + a_q\right) c_{bk}^\dagger c_{b'k'}$$

$$\rightarrow F \propto |E(t)|^2$$

(semiclassical approximation

for short-lived states)





[C. Aku-Leh et al. PRB **71**, 205211 (2005)]

- Optics: pump-probe in perturbative regime gives information similar to spontaneous Raman scattering
- Requires pulses shorter than period

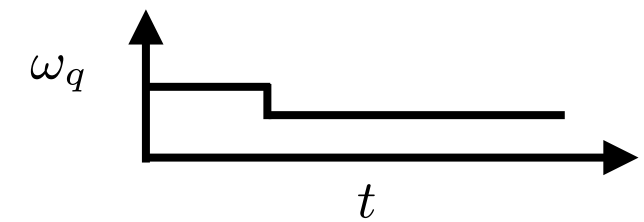
Time vs. Frequency domain

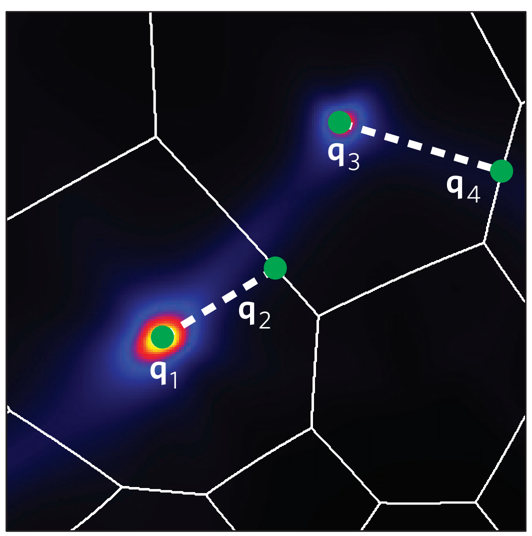
LETTERS
PUBLISHED ONLINE: 27 OCTOBER 2013 | DOI: 10.1038/NPHYS2788
nature physics

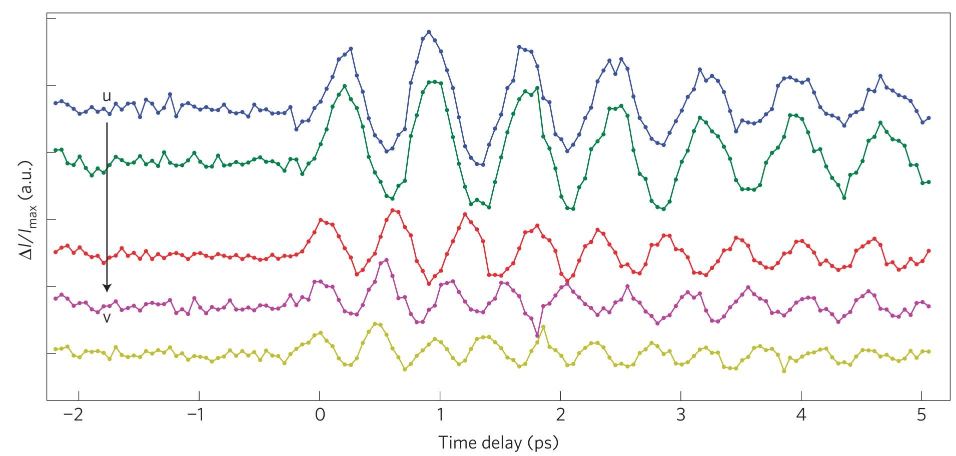
Fourier-transform inelastic X-ray scattering from time- and momentum-dependent phonon–phonon correlations

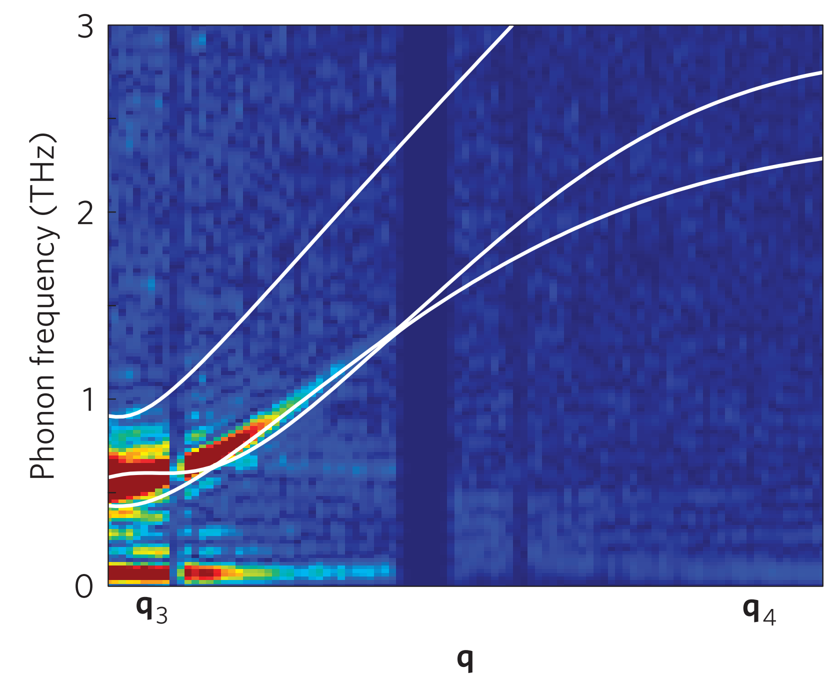
M. Trigo[1,2]*, M. Fuchs[1,2], J. Chen[1,2], M. P. Jiang[1,2], M. Cammarata[3], S. Fahy[4], D. M. Fritz[3], K. Gaffney[2], S. Ghimire[2], A. Higginbotham[5], S. L. Johnson[6], M. E. Kozina[2], J. Larsson[7], H. Lemke[3], A. M. Lindenberg[1,2,8], G. Ndabashimiye[2], F. Quirin[9], K. Sokolowski-Tinten[9], C. Uher[10], G. Wang[10], J. S. Wark[5], D. Zhu[3] and D. A. Reis[1,2,11]*

$$H_I \propto \sum_{q,q',b,b',k,k'} \Pi_{q,q',b,b',k,k'} a_q^\dagger a_{q'} c_{bk}^\dagger c_{b'k'}$$









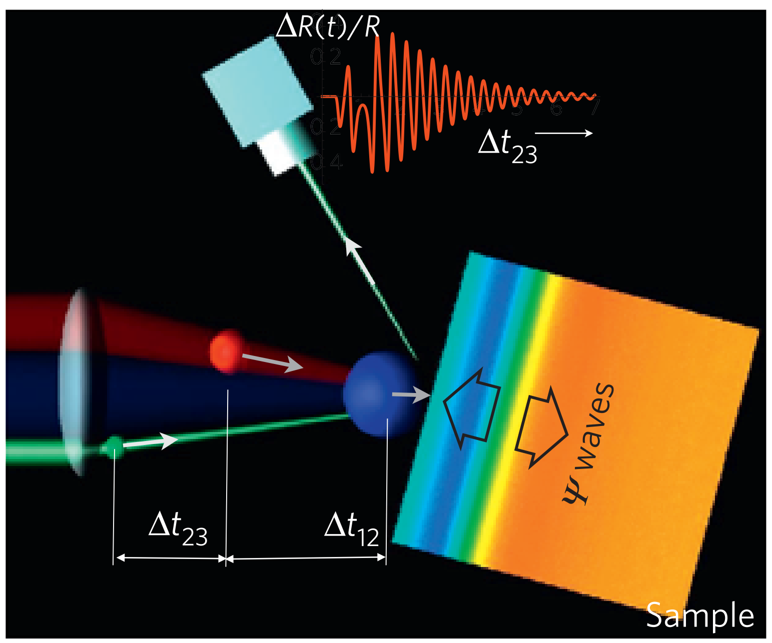
- Sufficiently short pump that couples to excitations induce coherences in time
- Coherences can be higher order, allows momentum coverage

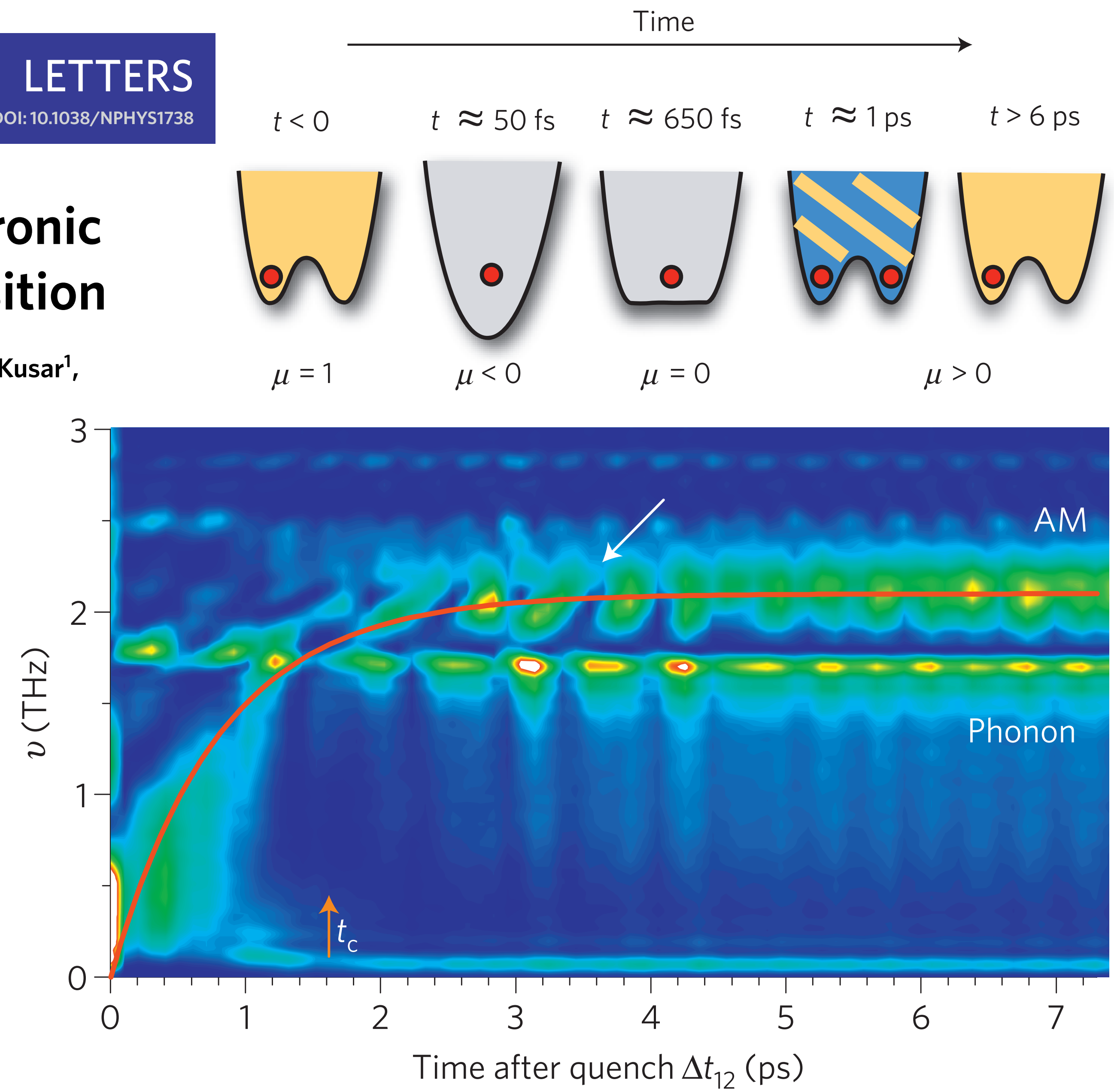
"Double pump" experiments


nature physics
LETTERS
PUBLISHED ONLINE: 22 AUGUST 2010 | DOI: 10.1038/NPHYS1738

Coherent dynamics of macroscopic electronic order through a symmetry breaking transition

Roman Yusupov[1], Tomaz Mertelj[1], Viktor V. Kabanov[1], Serguei Brazovskii[2], Primoz Kusar[1], Jiun-Haw Chu[3], Ian R. Fisher[3] and Dragan Mihailovic[1]*





- Way to use FT spectroscopy to study transient states
- Strong pump followed by weak pump-probe pair
- Subtract out signal without second pump

Limits and Opportunities

- Pulse duration < 10 fs gives enough bandwidth to study < 60 meV excitations via FT methods
  - ~ 0.3 fs may be limit for diffraction (0.3 fs * c = 90 nm)
  - Here pump/synchronization is more of a limit (assuming non-XFEL)
- FT limited pulses: cover higher lying excitations as RIXS via spectral analysis (simultaneously?)
- Ability to smoothly tune pulse duration and bandwidth within FT limit beneficial
- Accurately timed and phased double pulses might be useful for double pump schemes (needs further study)
- Stability essential
- Complete polarization control essential