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Swiss Federal Institute of Technology Zurich

Ultrafast dynamics in strongly correlated systems: from melting to control

Steve Johnson, ETHZ

Outline

- Scientific goal: fast control of technologically relevant material properties
- Thermodynamic vs. dynamic control
- Pump laser wish list

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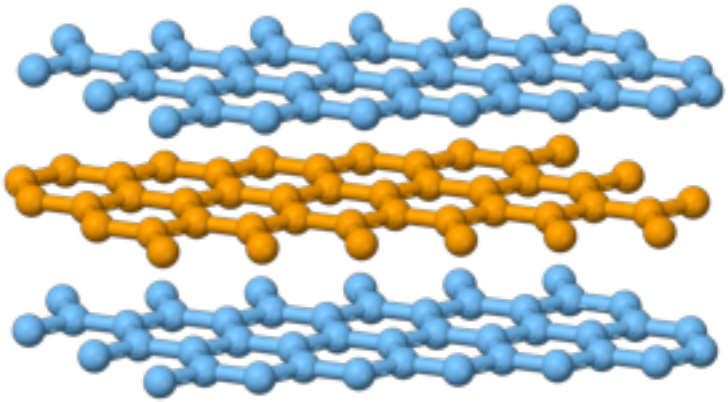
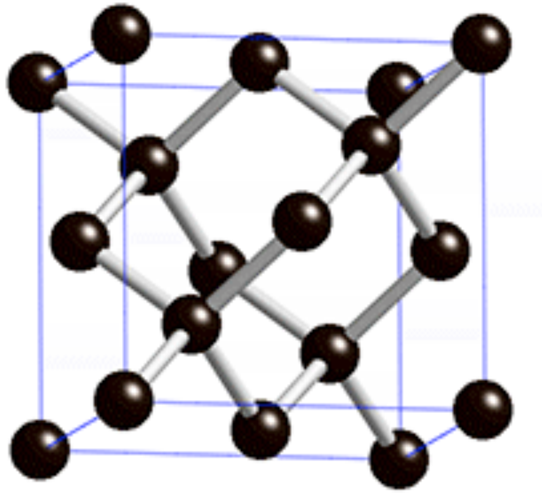
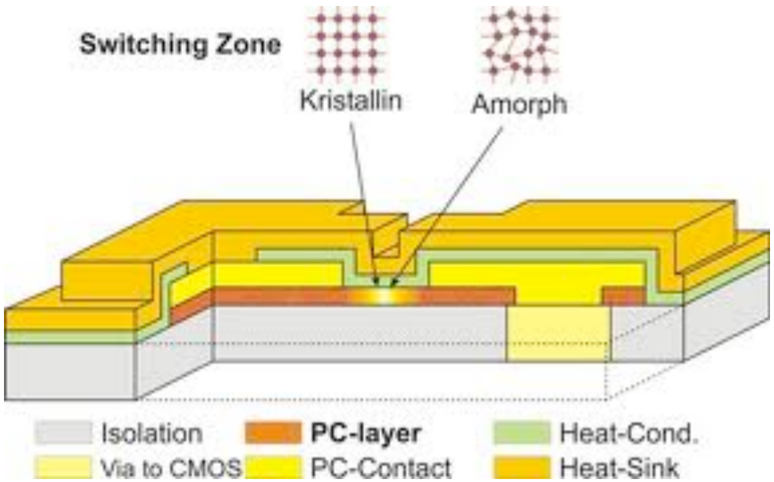
Structure and symmetry



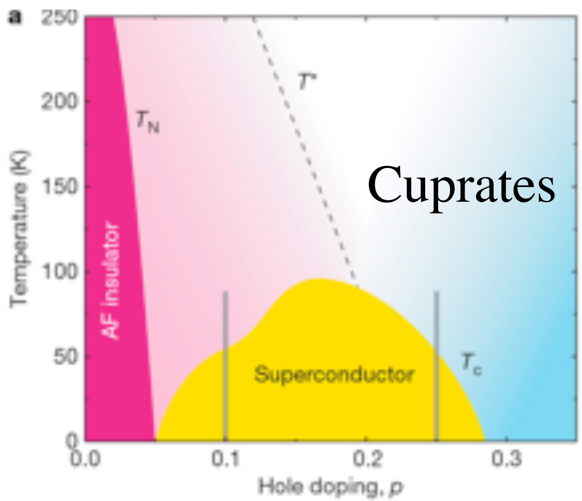


Phase changes

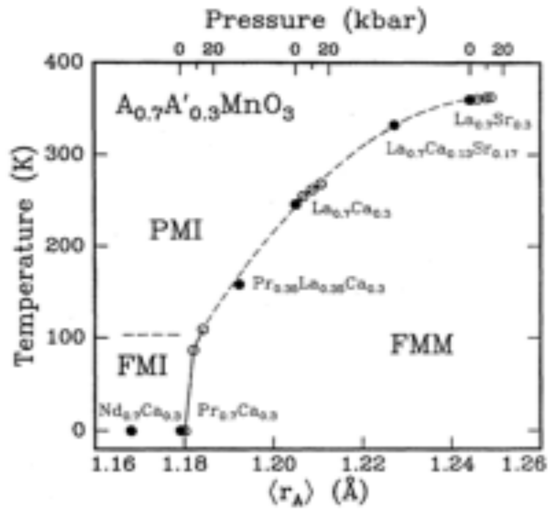
Phase-change RAM



Strongly correlated systems



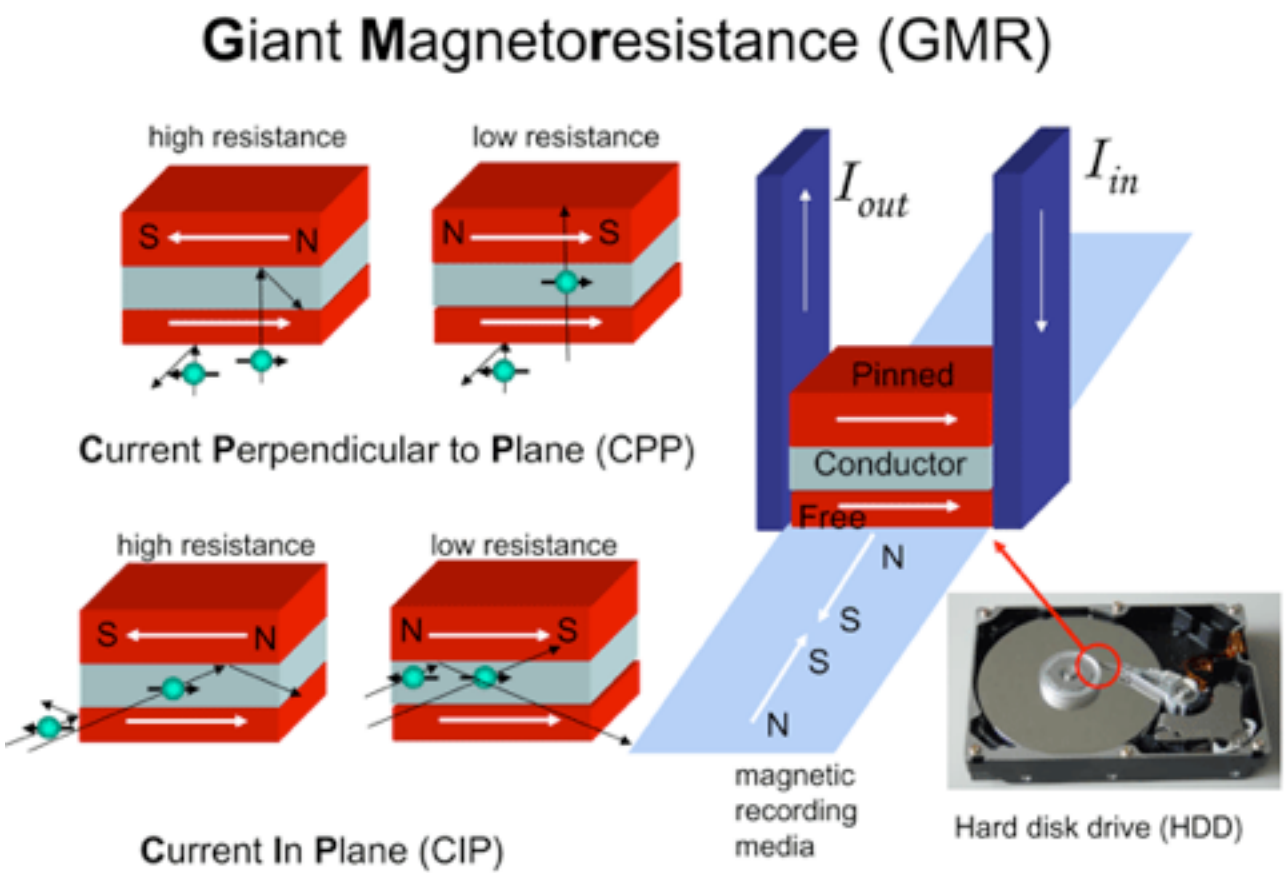
[Doiron-Leyraud et al. Nature 447, 565 (2007)]



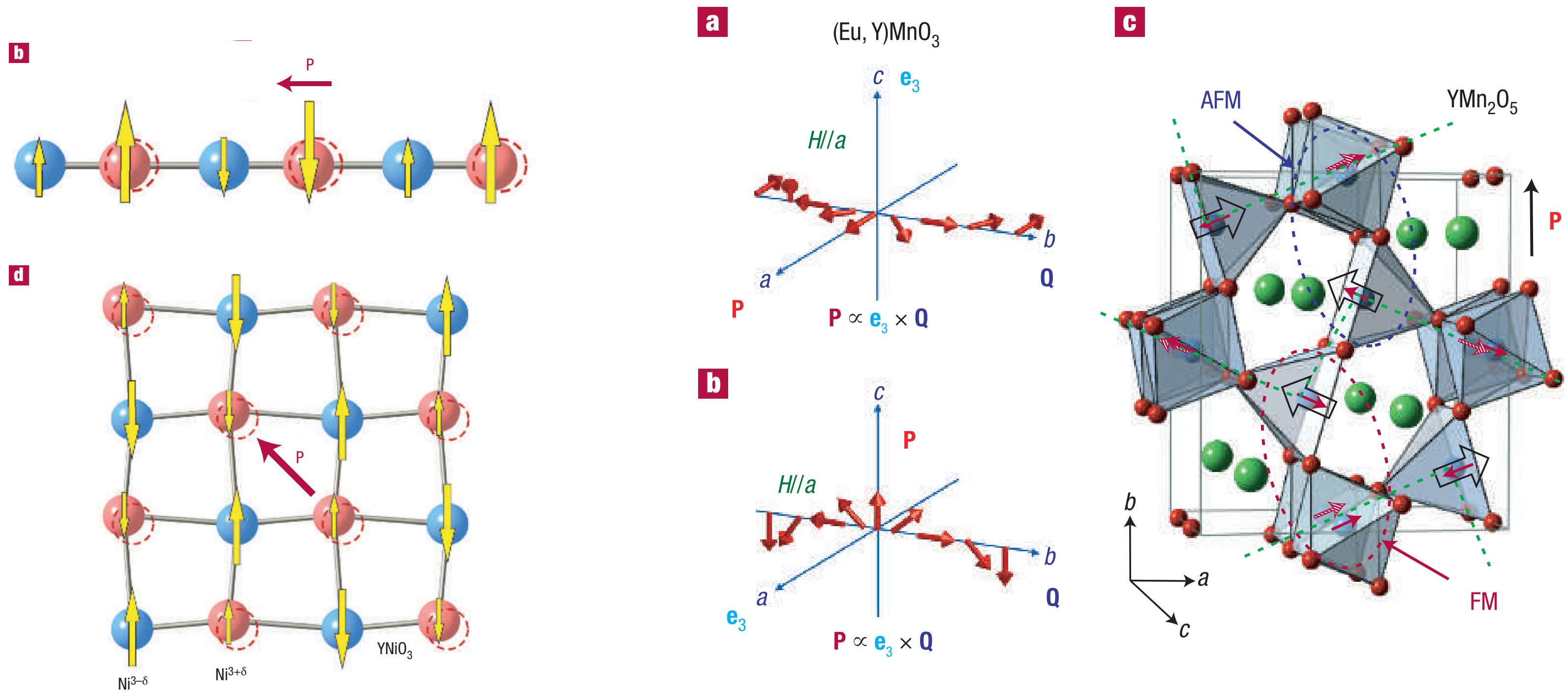
[Hwang et al. PRB 52, 15046 (1995)]

- Electronic correlations important
- Strong phase competition

Domain switching



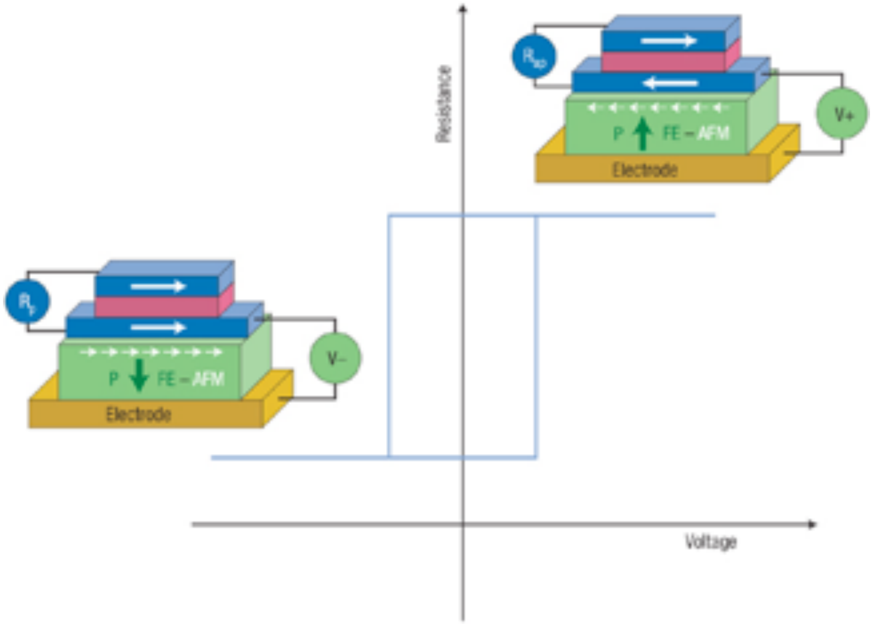
Multiferroics



[S. W. Cheong and M. Mostovoy, Nat. Mat. 6, p. 13 (2007)]

- Coexistence of ferroelectricity with magnetic order
- Magnetic-induced multiferroicity: strong coupling of magnetism and ferroelectricity

MeRAM



[M. Bibes & A. Barthélémy, Nat. Mat. 7, p. 425 (2008)]

- Could lead to devices that combine advantages of magnetic & ferroelectric memory

Basic questions

- How to change phase / domains?
- How fast is this possible?
- Is this reversible?

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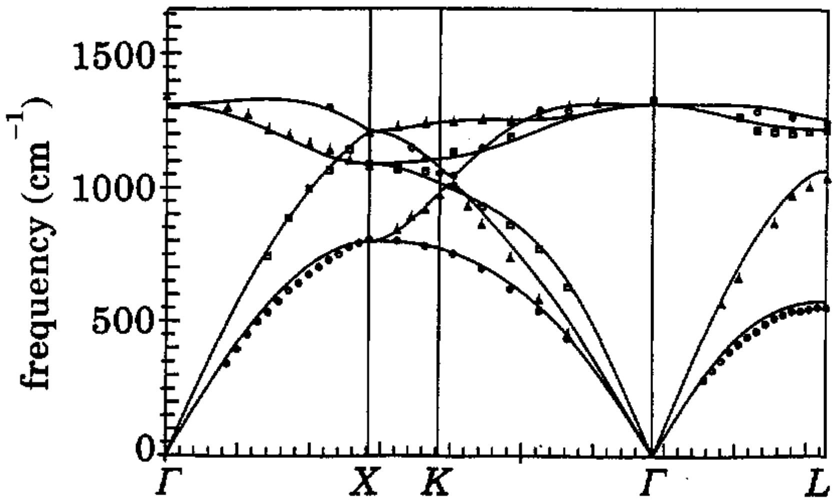
Atomic structure

- How and how fast can we rearrange atoms?
- Small perturbations: phonon frequencies
  - Acoustic: < several THz
  - Optical: 1 - 40 THz (1 ps to 25 fs)

Diamond



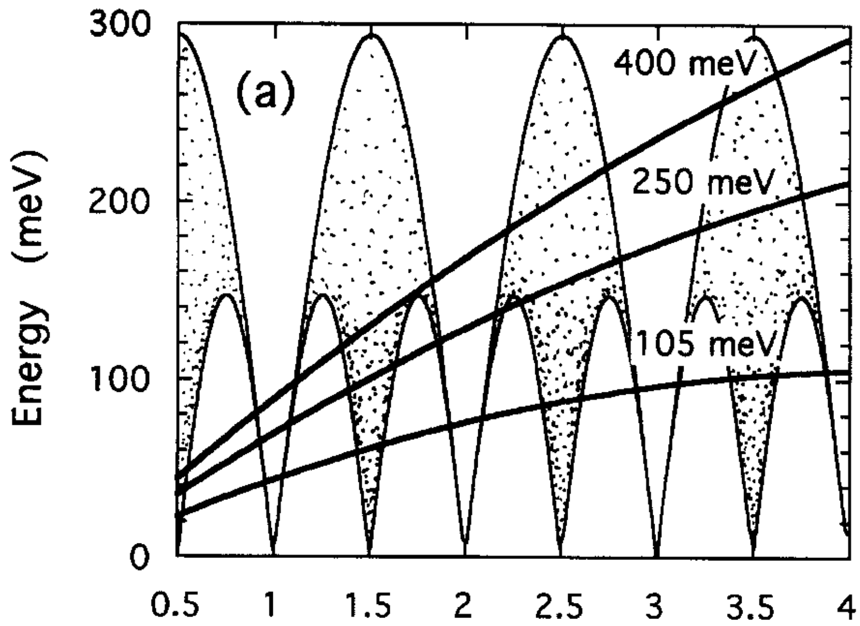
G. KRESSE, J. FURTHMÜLLER and J. HAFNER

Europhys. Lett., **32** (9), pp. 729-734 (1995)

Magnetic structure

- How and how fast can we rearrange spins?
- Small perturbations: magnon/spinon frequencies
  - 0 - 70 THz (14 fs)

CuO



[Boothroyd et al. Physica B 234-236, 731 (1997)]

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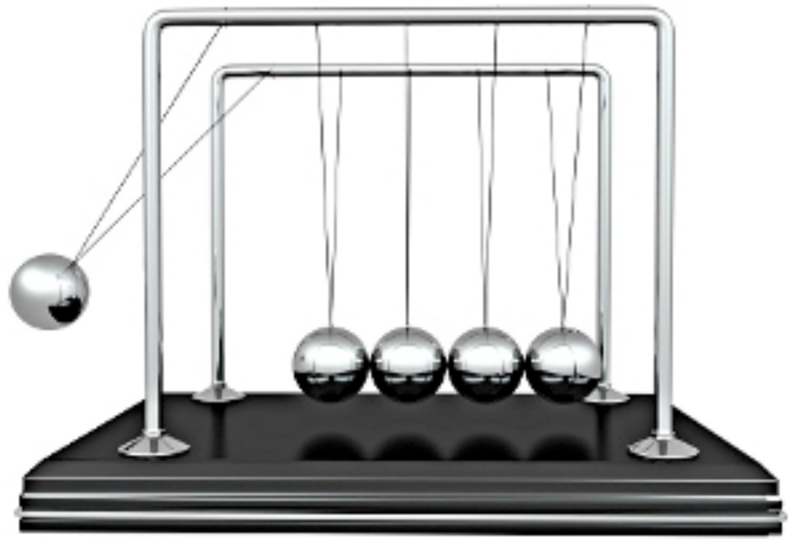
Control: thermodynamics vs. dynamics

- Can access by changing state variables (P,T,V)
- Have to wait for ergodicity
- Can use partial thermalization of subsystems

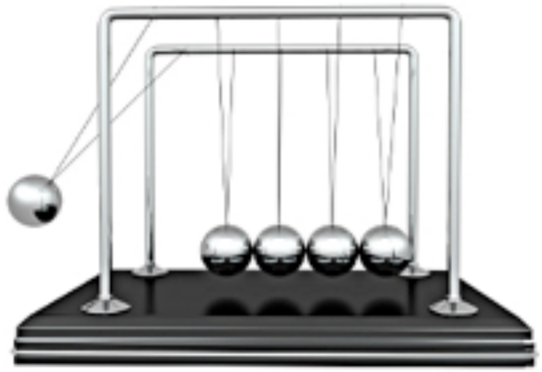
Control: thermodynamics vs. dynamics

- Dynamics: coherent control over specific coordinates
- Nearly reversible
- No need for ergodicity



Control: thermodynamics vs. dynamics

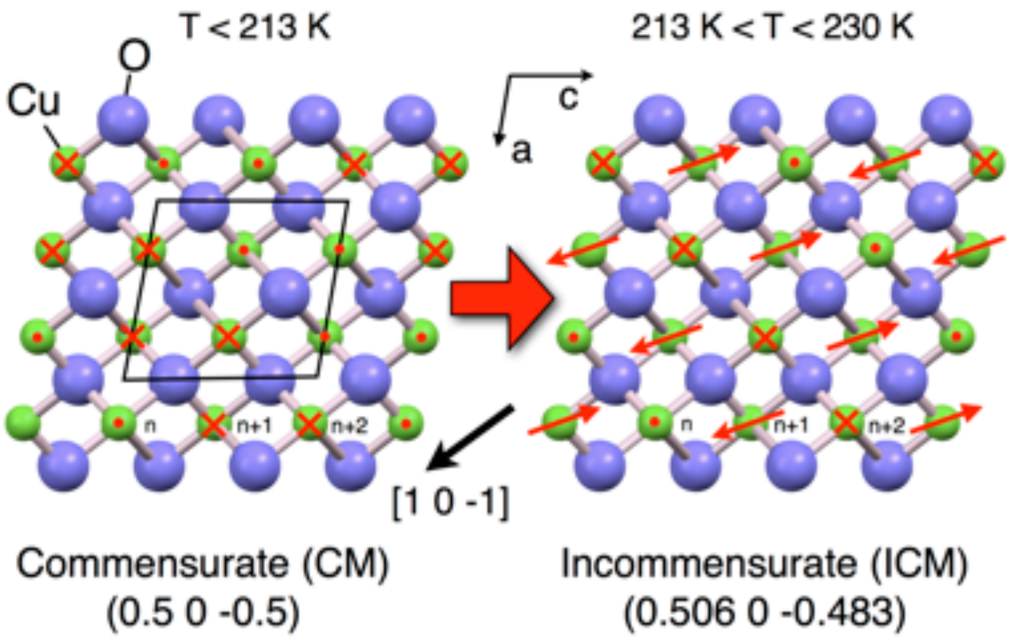
- General pump, must just give energy
- Not very flexible
- Easier to increase entropy than to decrease



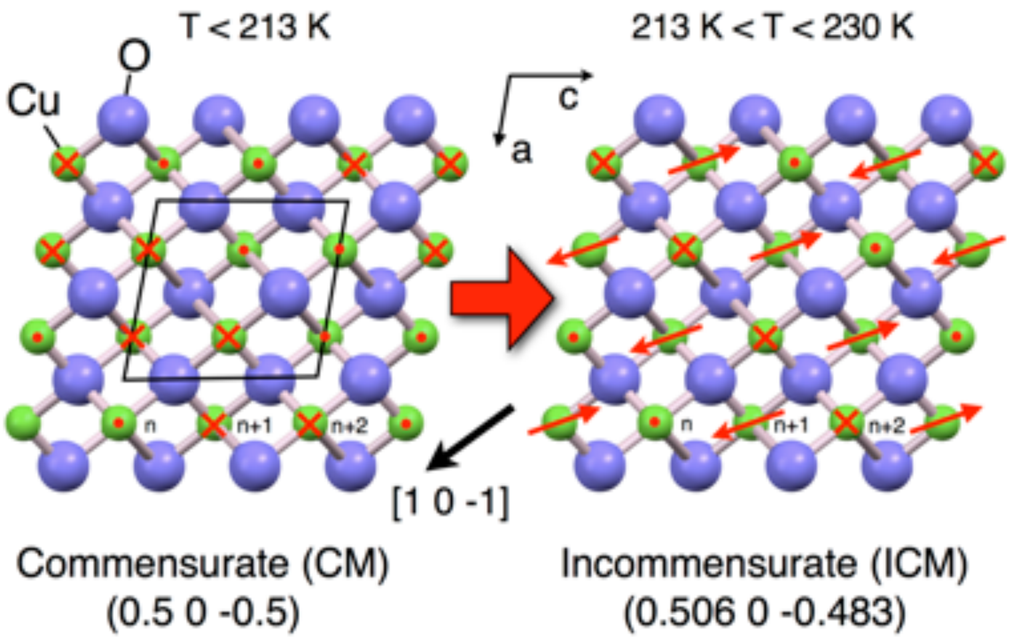
- Requires specific stimulus (pump)
- Not as much wasted energy
- Does not change entropy much

Example: CuO

- Temperature-induced magnetic phase transition
- Long-range reordering of spins on Cu

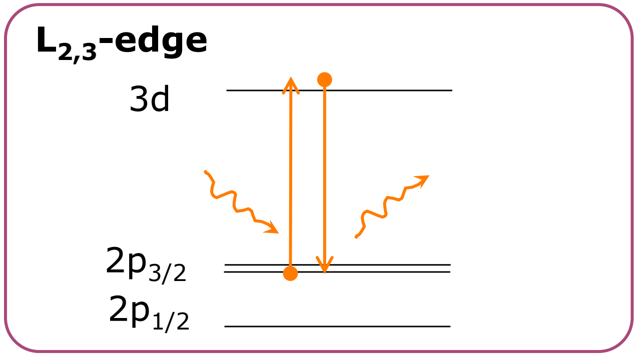


Example: CuO

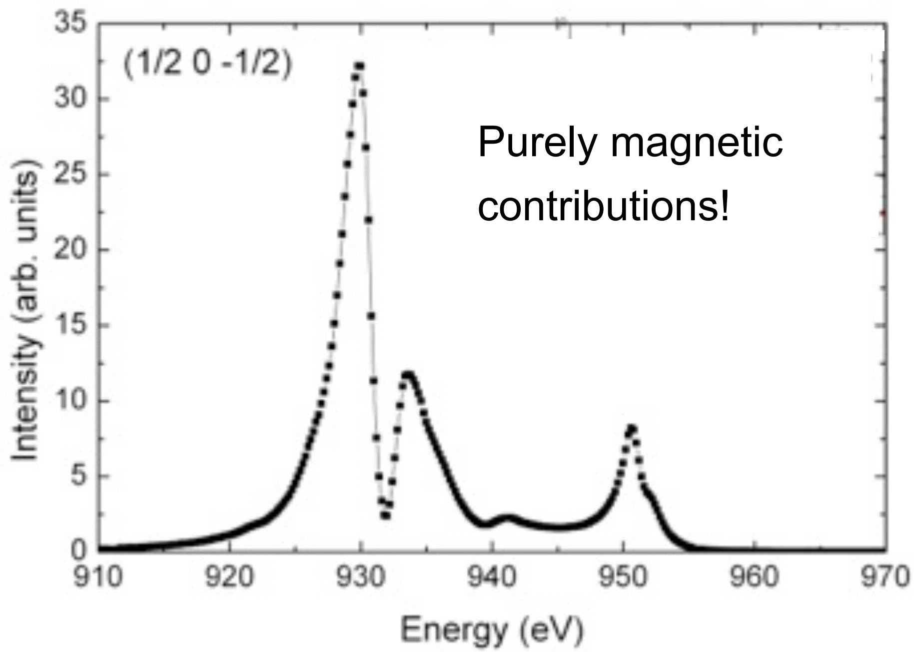

T < 213 K
213 K < T < 230 K
O
Cu
c
a
n
n+1
n+2
[1 0 -1]
Commensurate (CM)
(0.5 0 -0.5)
Incommensurate (ICM)
(0.506 0 -0.483)

Pump-probe experiment

- Near-IR pulse can rapidly "heat" via above bandgap excitation
- Resonant x-ray diffraction as a probe of the magnetic ordering wave-vector

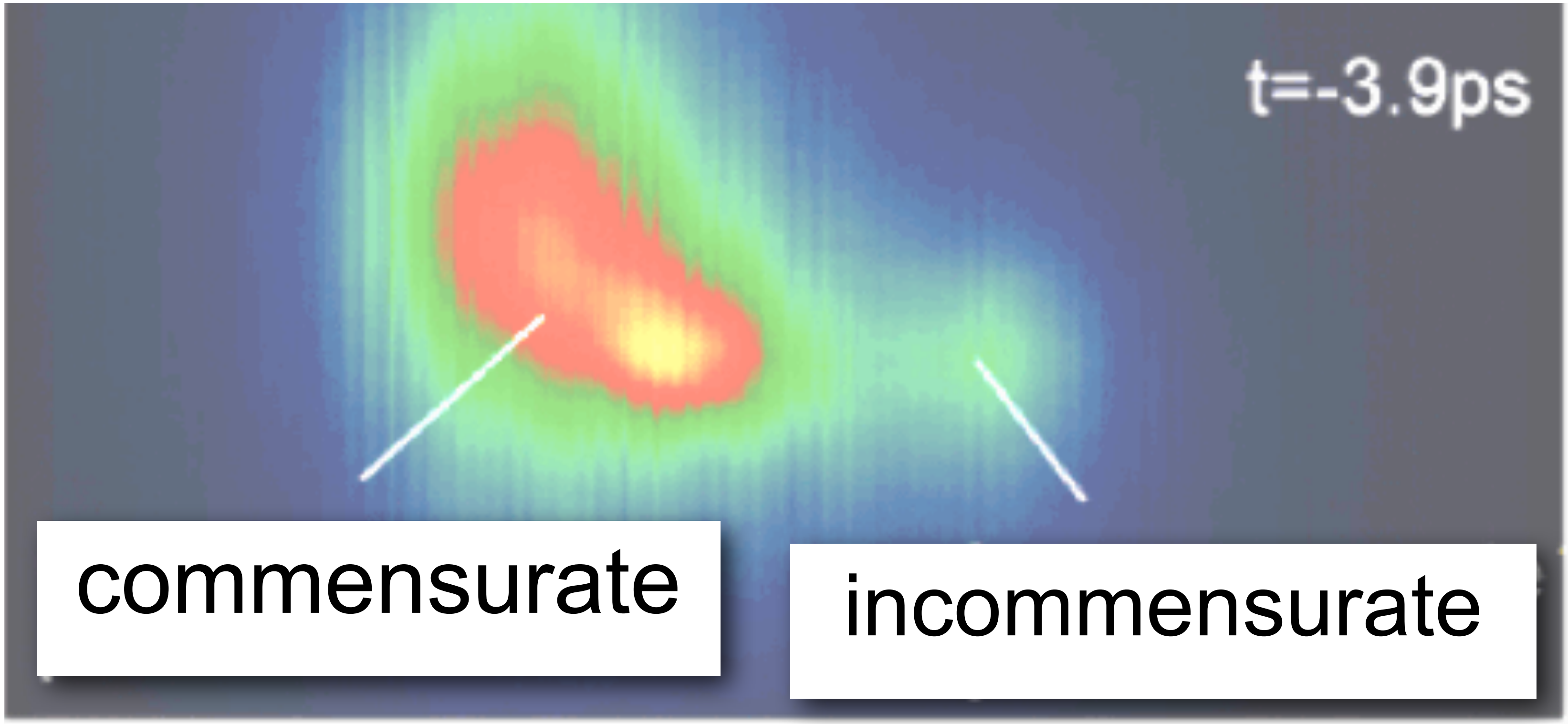


$$\left\langle \mathbf{T}_q^k \right\rangle \propto \sum_n \frac{\langle g| O |n\rangle \langle n| O^* |g\rangle}{E_n - E_g - \hbar\omega + i\Gamma}$$



Phase transition

Before excitation

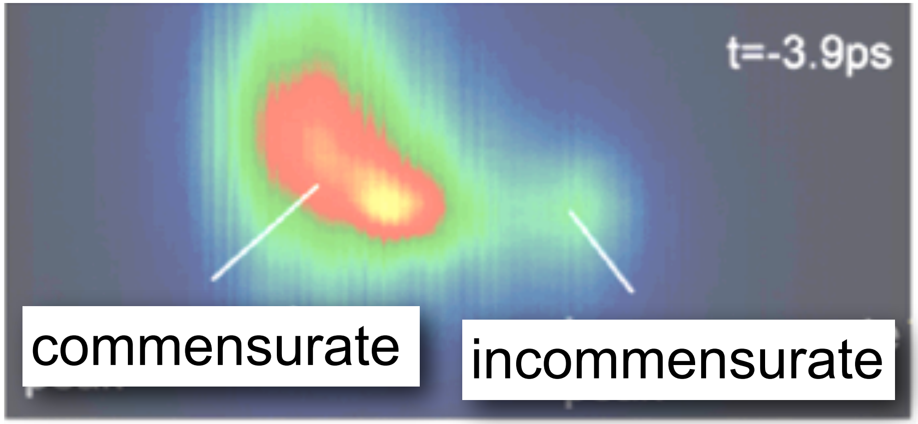


- Optical excitation pushes spins into ICM phase

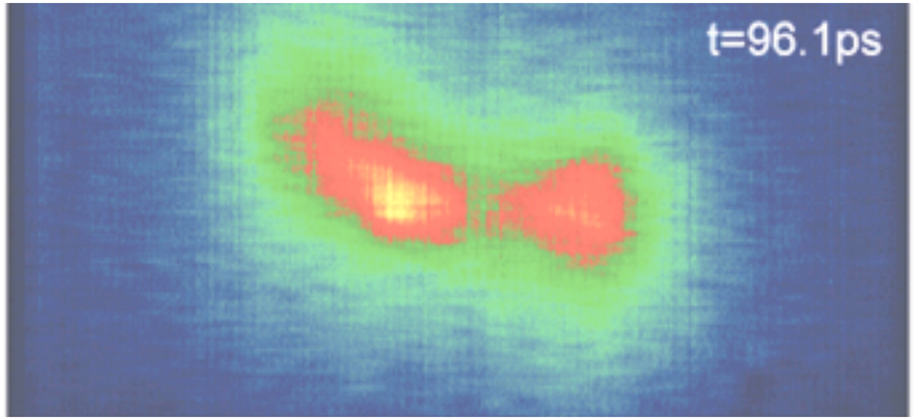
[Johnson et al, PRL 108, 037203 (2012)]

Phase transition

Before excitation



After excitation



- Optical excitation pushes spins into ICM phase

[Johnson et al, PRL 108, 037203 (2012)]

Phase transition onset

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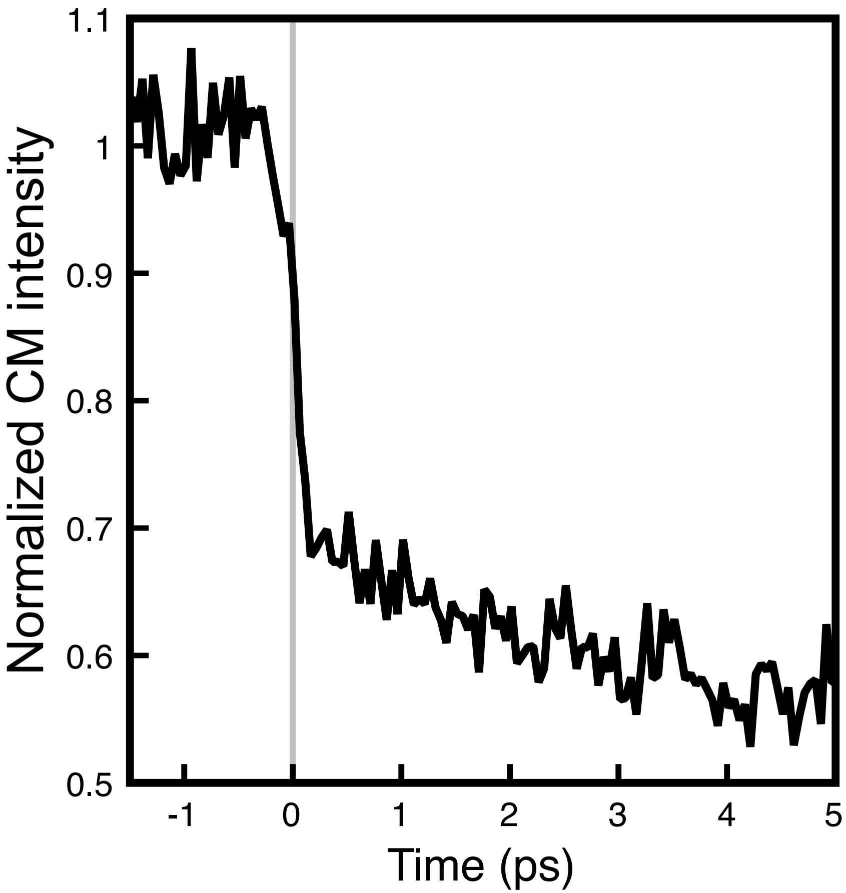
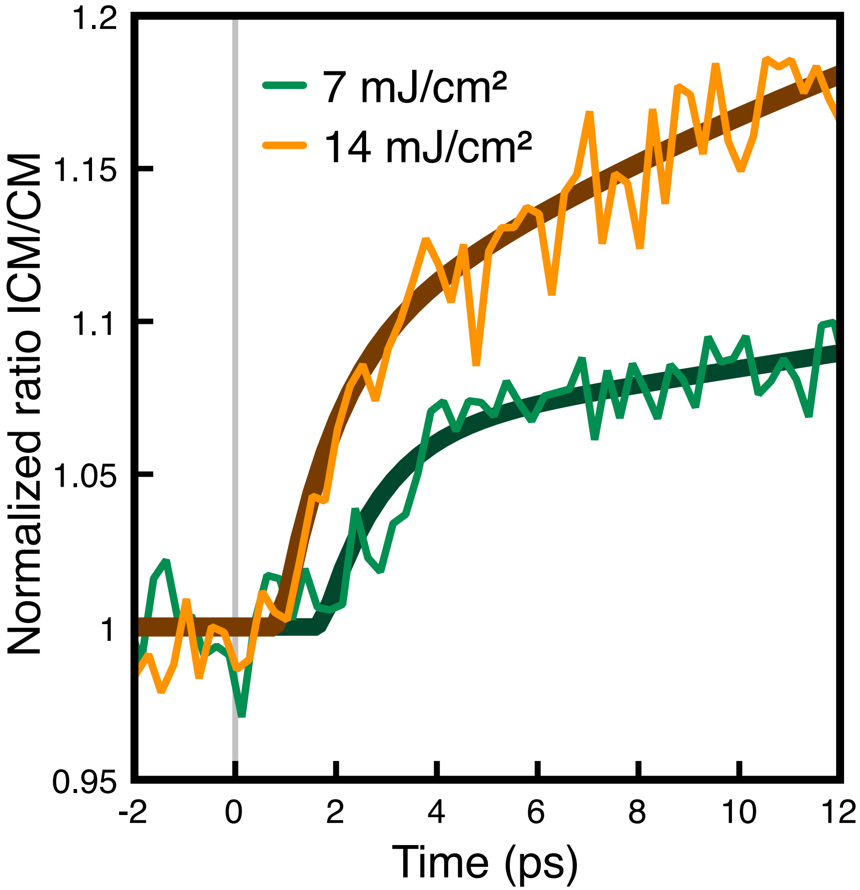


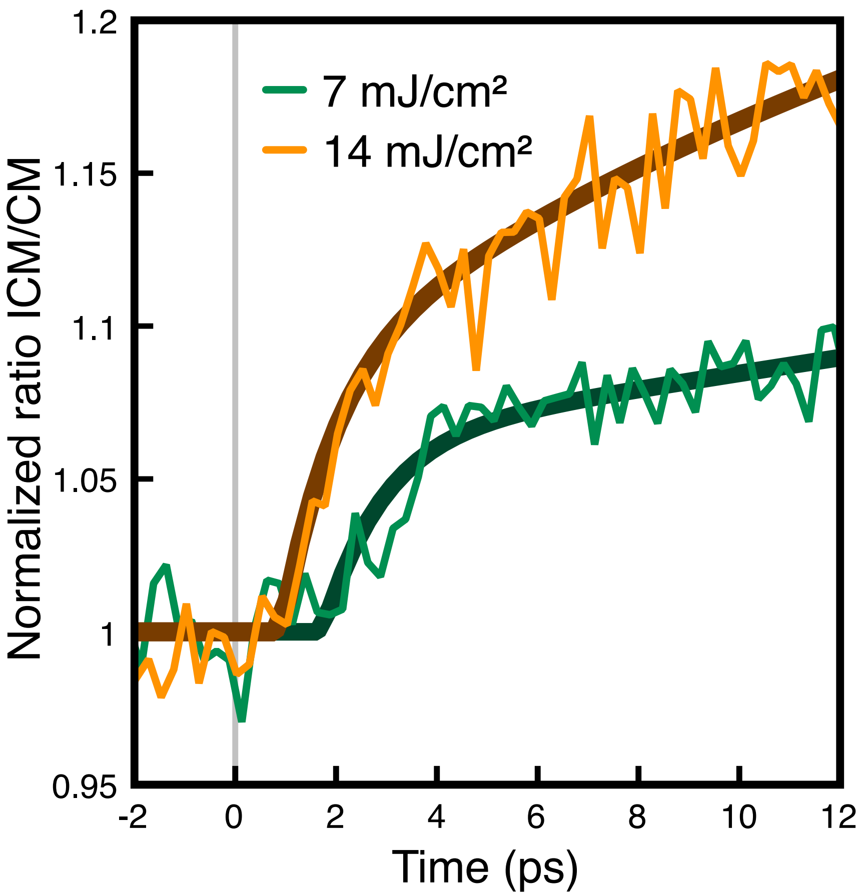
[Johnson et al, PRL 108, 037203 (2012)]

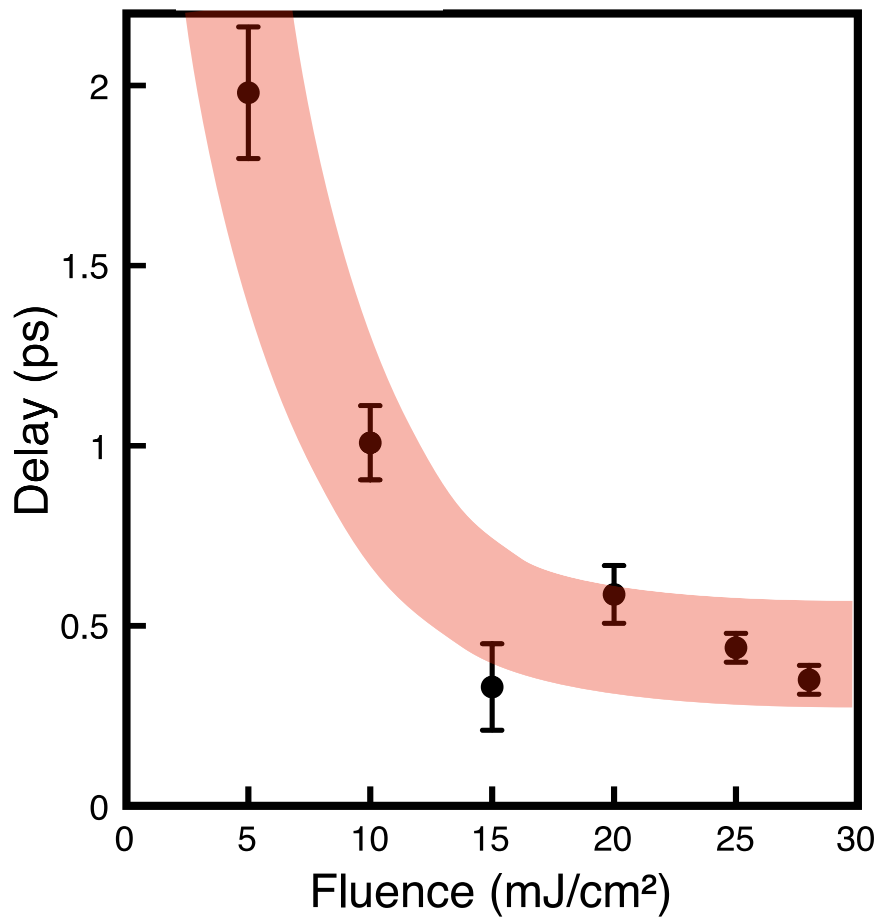
Phase transition onset



[Johnson et al, PRL 108, 037203 (2012)]

Phase transition onset



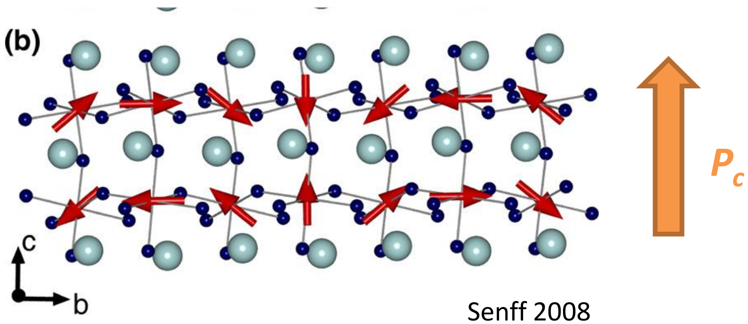


- "Delay" between disorder and onset of phase transition
- Minimum value of ~ 400 fs

[Johnson et al, PRL 108, 037203 (2012)]

Dynamics: THz control?

TbMnO$_3$

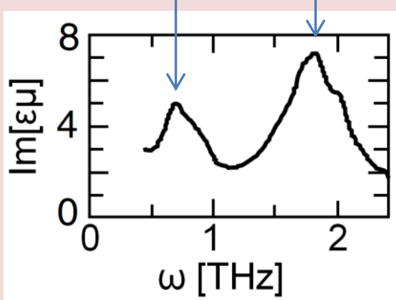


Senff 2008

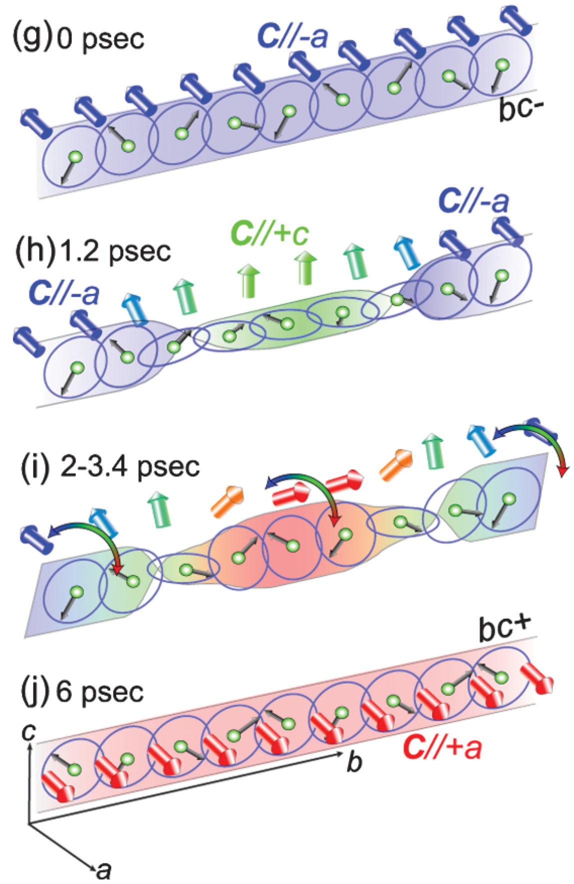
excitations due to the electromagnetic coupling:

higher-harmonic, ellipticity, phonons 0.7 THz

spin-spiral excitation, 1.8 THz



[Y. Takahashi et al., PRL **101**, 187201 (2008)]

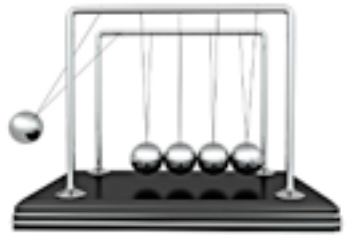


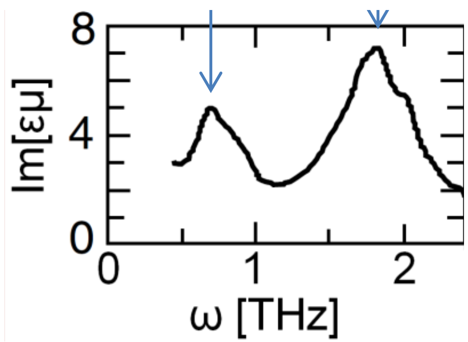
[Mochizuki & Nagaosa, PRL **105**, 147202 (2010)]

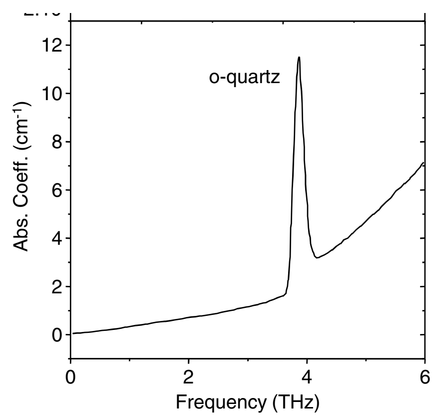
Pump wishes

- In optical and near-IR range, very short pulses (~ 10 fs)
  - electronic heating
  - impulsive Raman excitations

- mid-/far-infrared (5-300 microns), highly tunable, multiple-cycle CEP stable pulses
  - phase stability relevant for time resolution
  - resonant excitation
  - bandwidth material dependent (1% to 30%)







Pump wishes

- Critical parameters:
  - Synchronization/time arrival
    - < 10 fs (FWHM)
  - Fluence/field strengths
    - For THz “few cycle,” > 5 MV/cm
    - 20 mJ/cm^2 for higher frequencies
  - Flexible focusing, steering options (collinear, non-collinear, tilting)
  - Stability
    - Noise of FEL makes long integration needed for small signals

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J. Turner

W. Schlotter

G. Dakovski

M. Minitti

M. Trigo

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M. Yi

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Z. X. Shen

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