Proposed pump-probe experimental station for FLASH II

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Motivation I

We like to **get answers** to **important questions** such as:

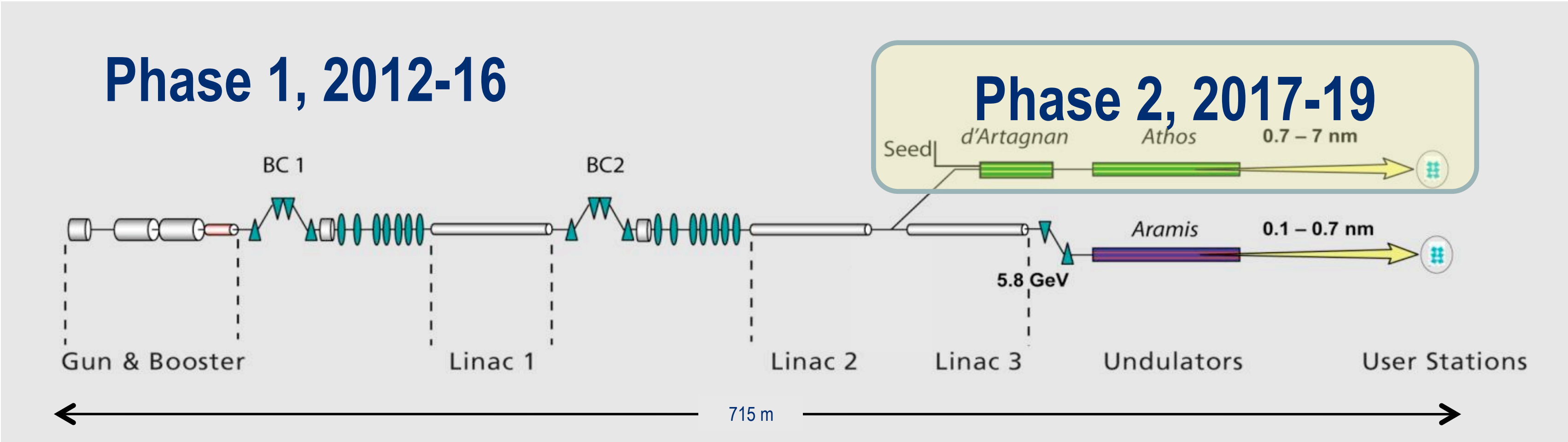
How fast can a phase transition involving a change in symmetry occur?

What is the *driving force* behind phase transitions in interesting (strongly correlated electron) materials?

How can we manipulate the structure of solids via *non-thermodynamic pathways*?

How is *thermal equilibrium established* after a perturbation?

SwissFEL (to be built at PSI)



Aramis: 1-7 Å hard X-ray SASE FEL,
In-vacuum , planar undulators with variable gap.
User operation from mid 2017

Athos : (2nd phase) 7-70 Å soft X-ray FEL for SASE & Seeded operation .
APPLE II undulators with variable gap and full polarization control.
User operation end 2019?

SwissFEL parameters

Wavelength from	1 Å - 70 Å
Photon energy	0.2-12 keV
Pulse duration	1 fs - 20 fs
e^- Energy	5.8 GeV
e^- Bunch charge	10-200 pC
Repetition rate	100 Hz

Focus

Physics:
Strongly correlated electron systems

Focus on pump-probe experiments

Pump:
Optical, IR/THz radiation, x-ray (two color)

Probe:
Resonant x-ray scattering
X-ray absorption

Other **stimuli**:
Magnetic fields, electric fields, (low) temperature

Resonant X-ray diffraction

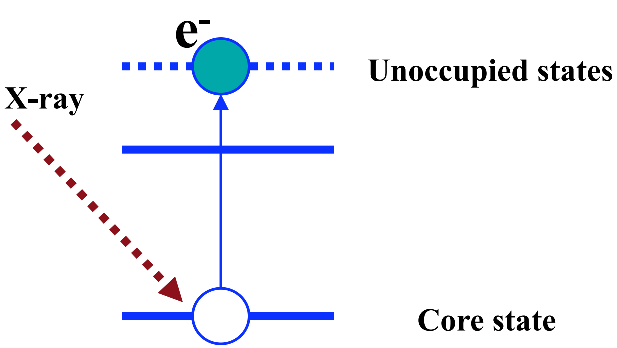
Electronic states

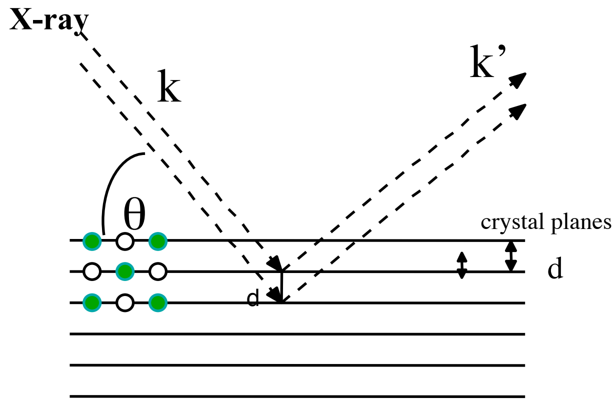
Spectroscopy (XAS)

Energy dependence of Bragg reflections

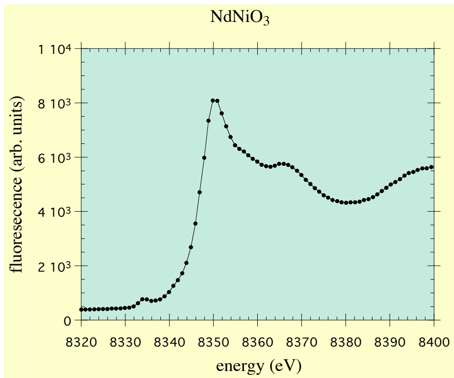
Structure

Diffraction





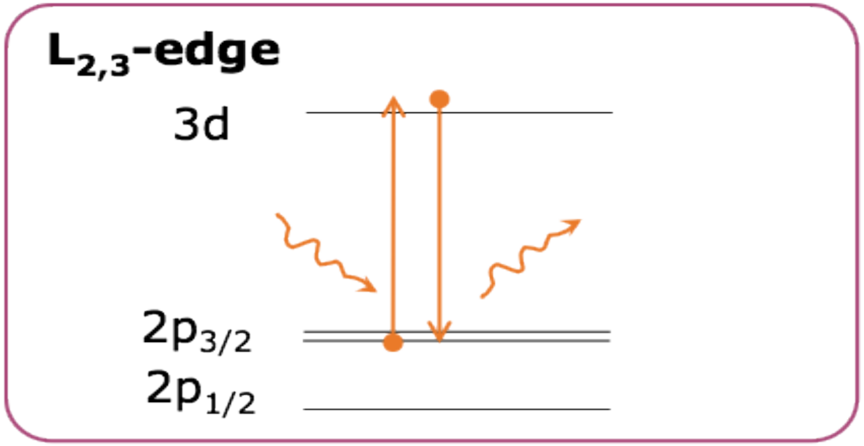
$2d \sin(\theta)=\lambda$ positive Interference

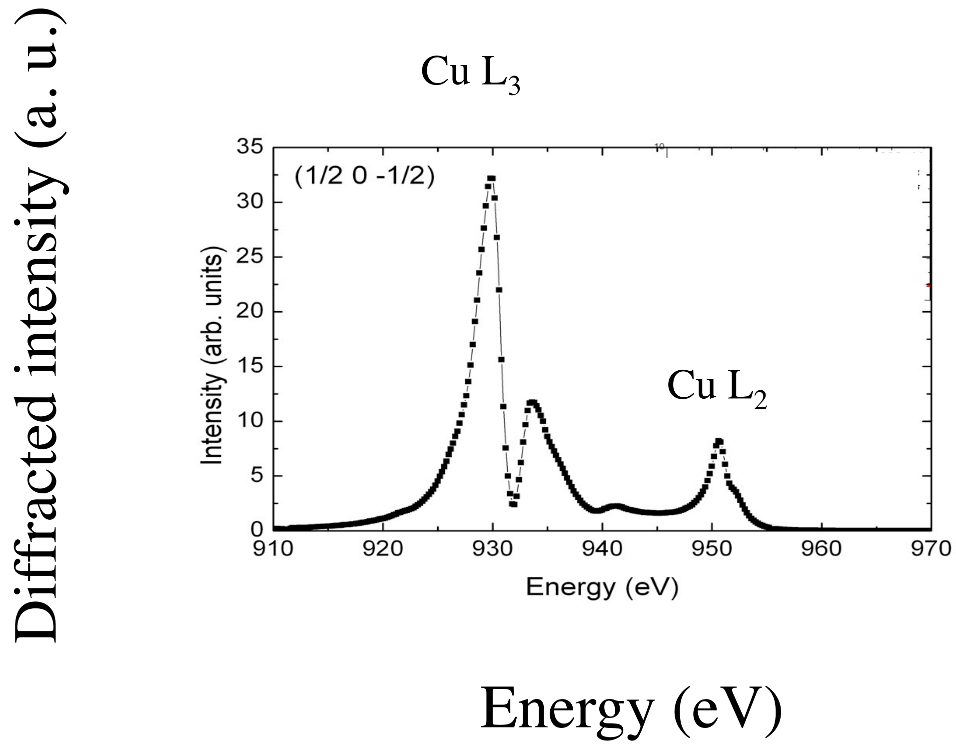


Spectroscopic and spatial information ==> site selectivity
requires ordering

enhanced magnetic, charge and orbital sensitivity

Methods: Resonant XRD



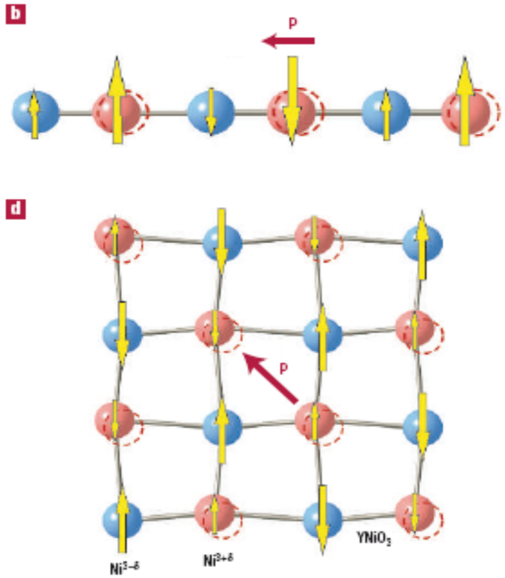


$$\left\langle \mathbf{T}_q^k \right\rangle \propto \sum_n \frac{\langle g| O |n\rangle \langle n| O^* |g\rangle}{E_n - E_g - \hbar\omega + i\Gamma}$$

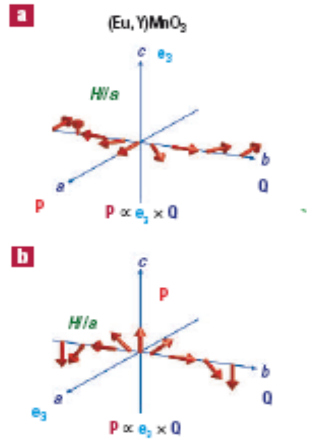
- At resonance: direct probe of orbital, charge or magnetic structure

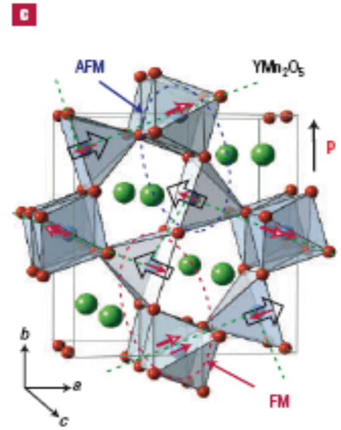
Multiferroics

Exchange striction



Spin current
DM interaction





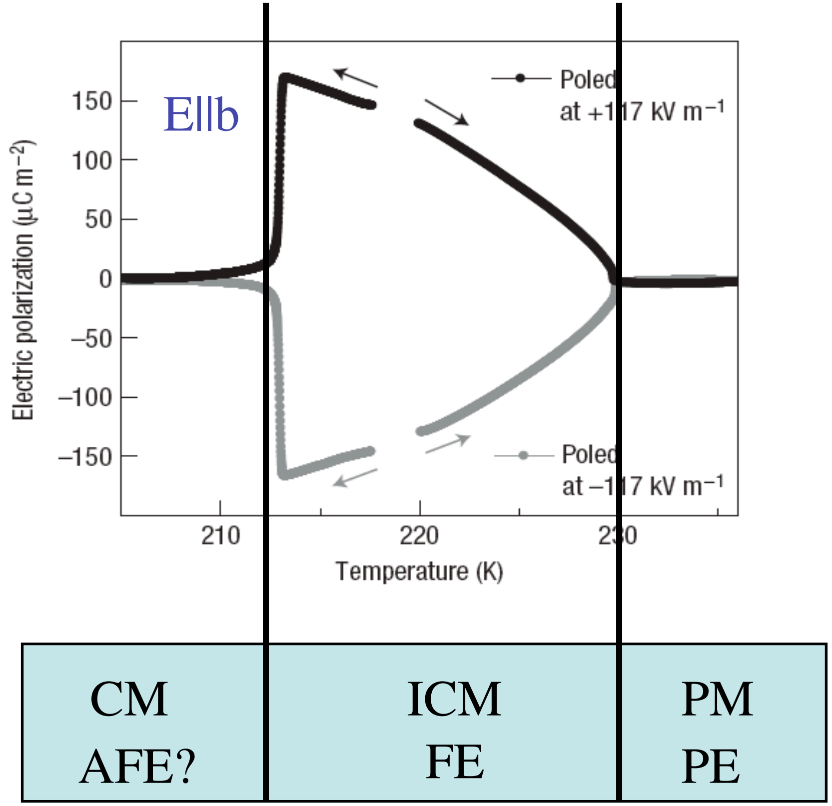
$\mathbf{j} = \mathbf{s}_1 \mathrm{x} \mathbf{s}_2, \mathbf{P} = \mathbf{j} \mathrm{x} \mathbf{r}$

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

- Magnetic-induced multiferroicity: gigantic coupling of magnetism & ferroelectricity

CuO: a high-T induced multiferroic

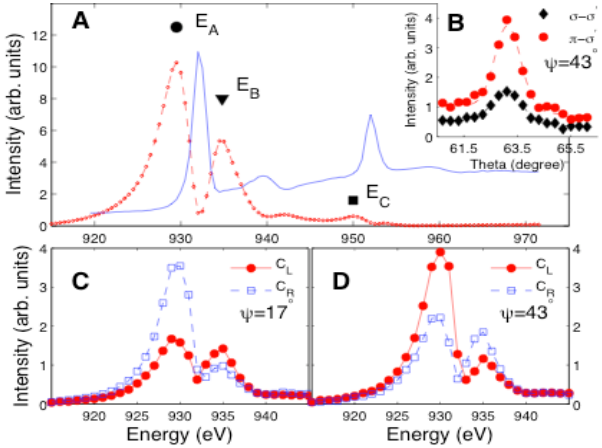
Spontaneous electric polarization

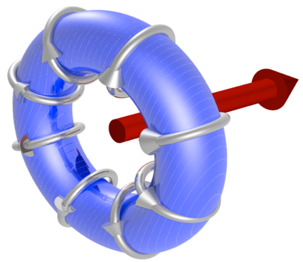


[Kimura et al. Nat. Mater. 7, p. 291 (2008)]

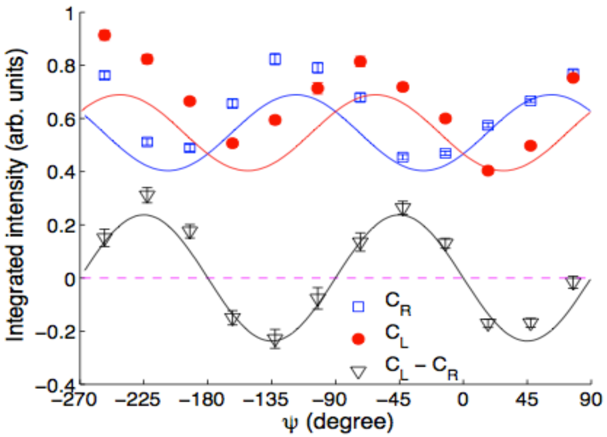
Orbital currents in cupric oxide (CuO)

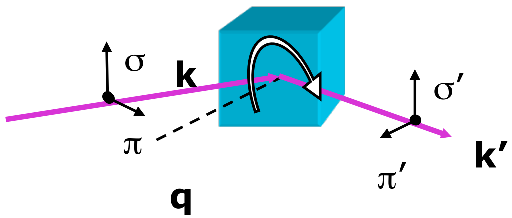
(1/2 0 -1/2) “magnetic” reflection



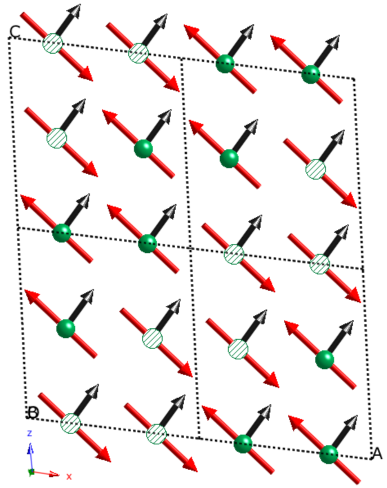


Discovery of orbital current in a CuO plaquette





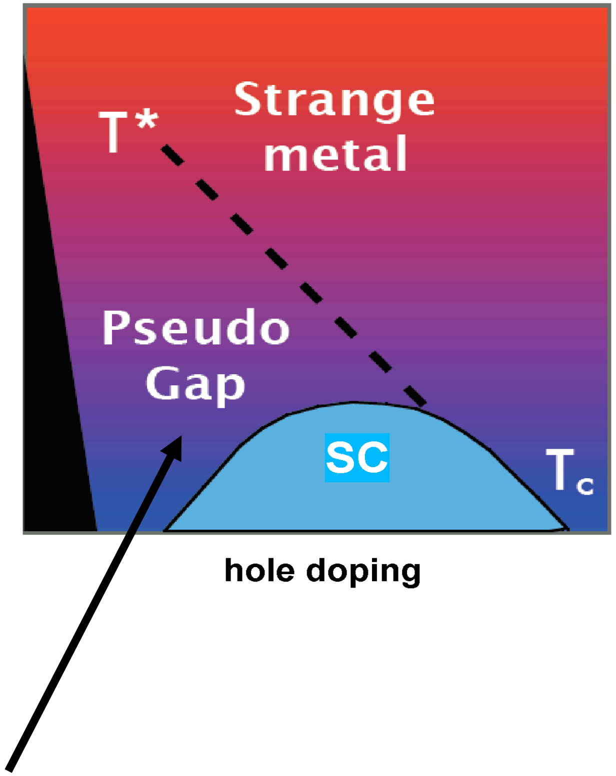
Anti-ferro order



V. Scagnoli, U. Staub et al. Science **332**, 696 (2011)

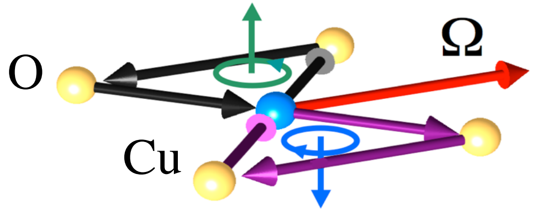
Orbital currents

proposed order parameter of the pseudo-gap phase in high-T_c superconductors

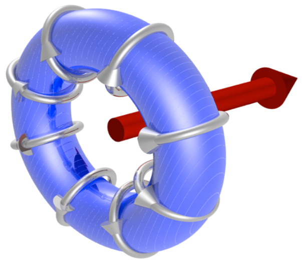


Varma PRB **55**, 14554 (1997)

Orbital current



Toroidal moment



anapole moment

$$\vec{\Omega} = \vec{r} \times \vec{\mu}$$

CP invariant

How fast can we induced a magnetic order-order transition?

Example I

CuO

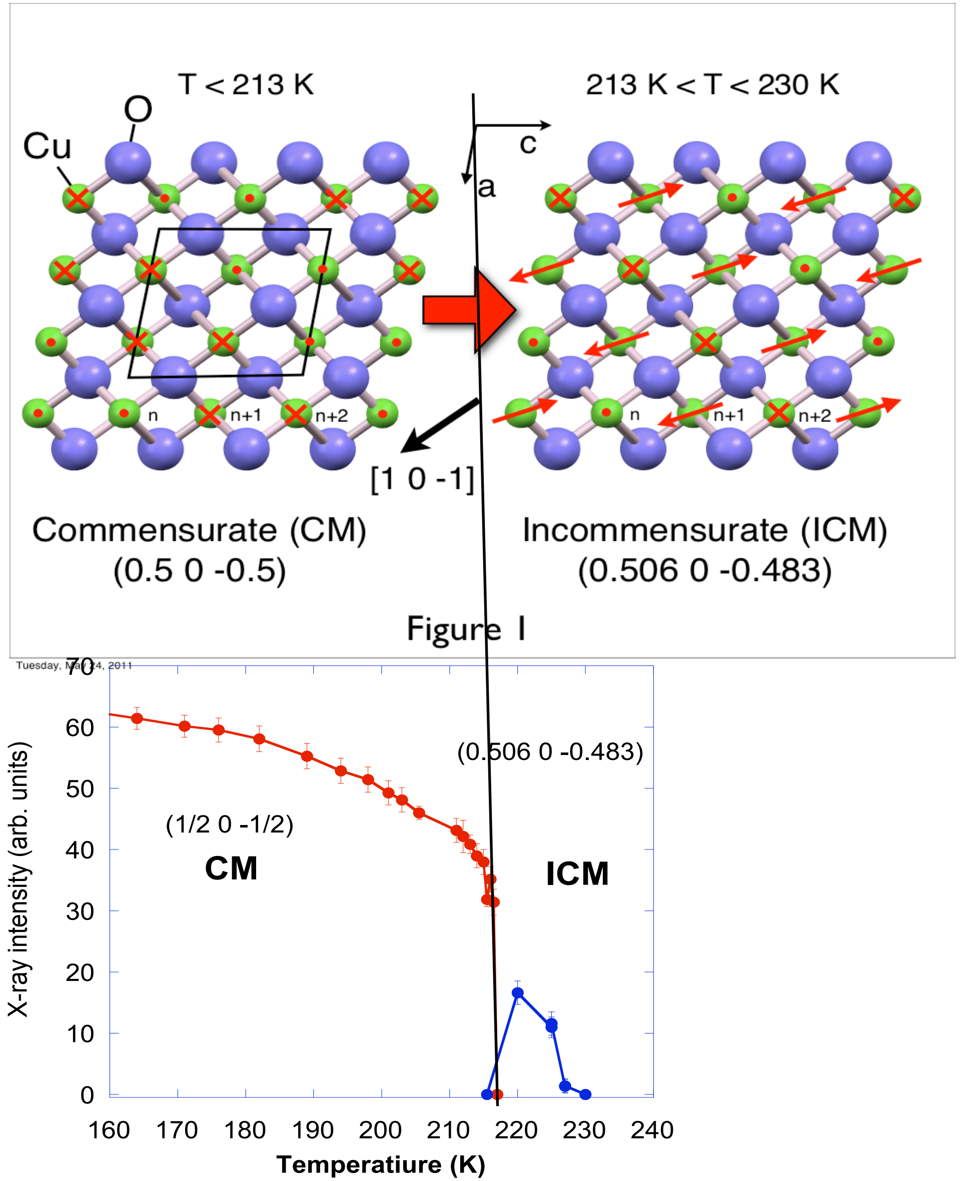
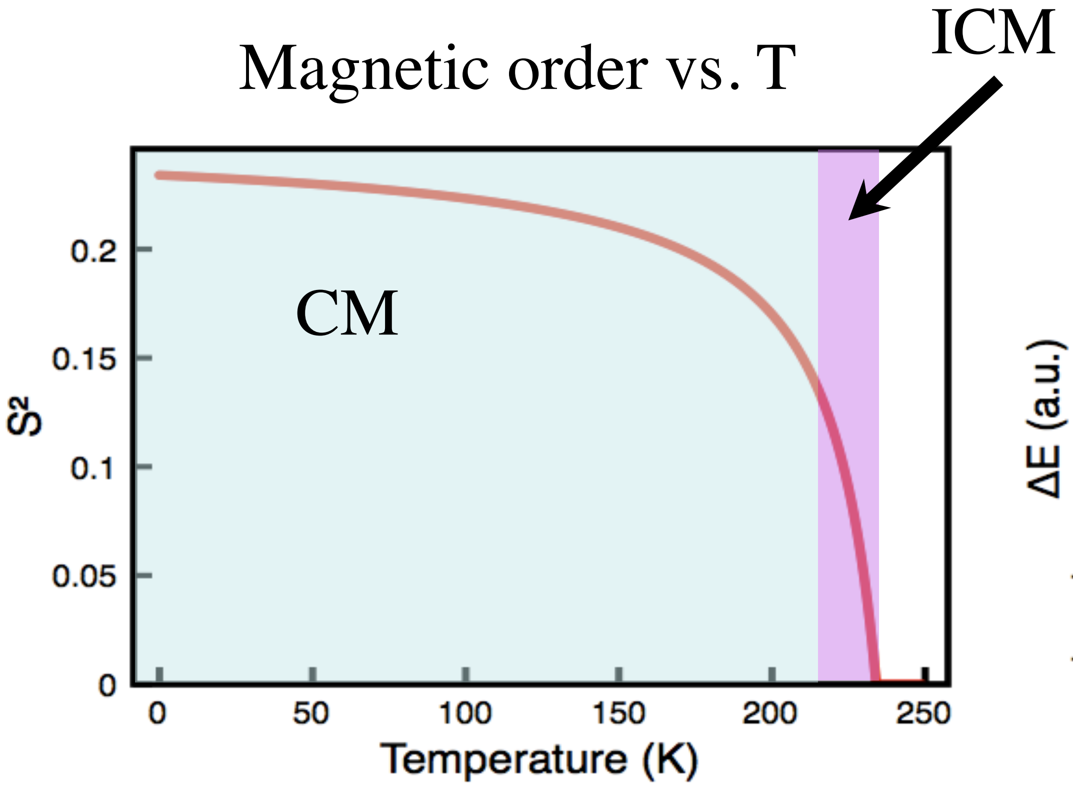


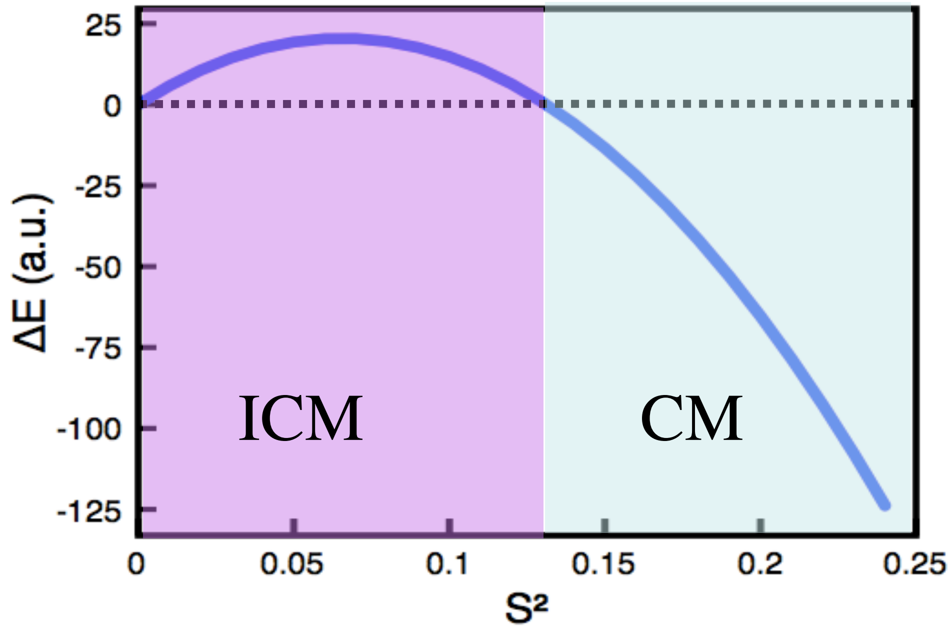
Figure I

CM / ICM Phase transition: driven by disorder?

Magnetic order vs. T



$E_{CM} - E_{ICM}$ vs. S^2

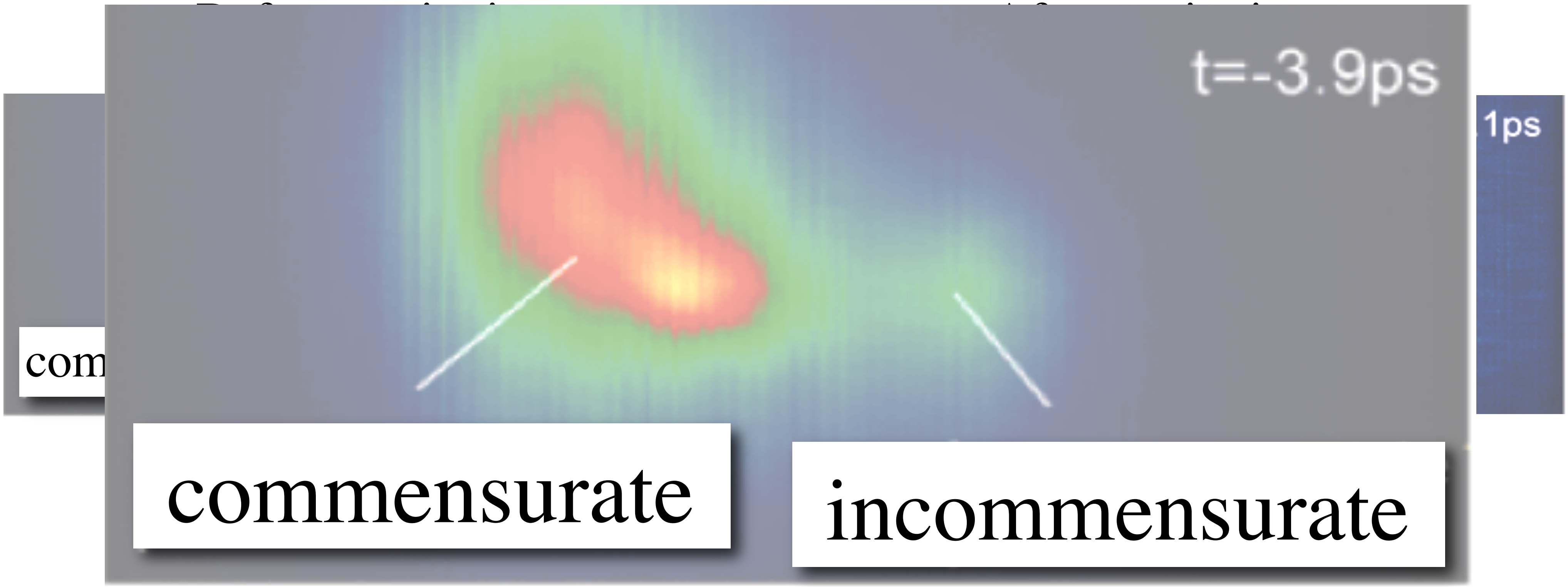


$$E = [J_1 \cos\phi + J_2 \cos(2\phi)]S^2 + IS^4 \cos^2\phi$$

[D. A. Yablonskii, Physica C 171, 454 (1990)]

- Phase stability depends on temperature via S^2

Phase transition



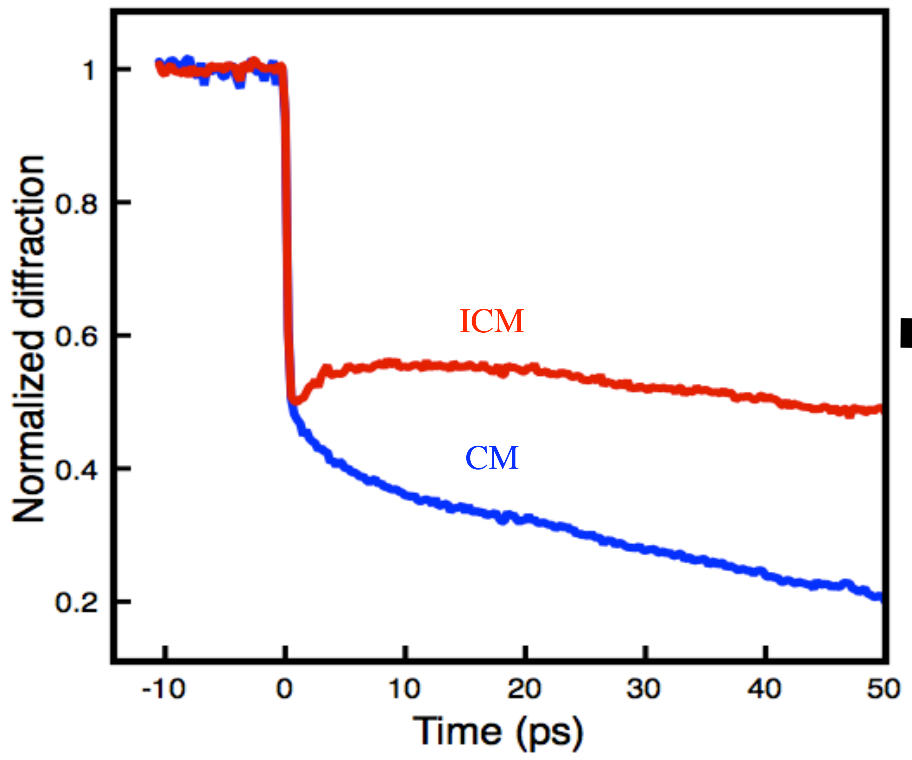
- Optical excitation pushes spins into ICM phase

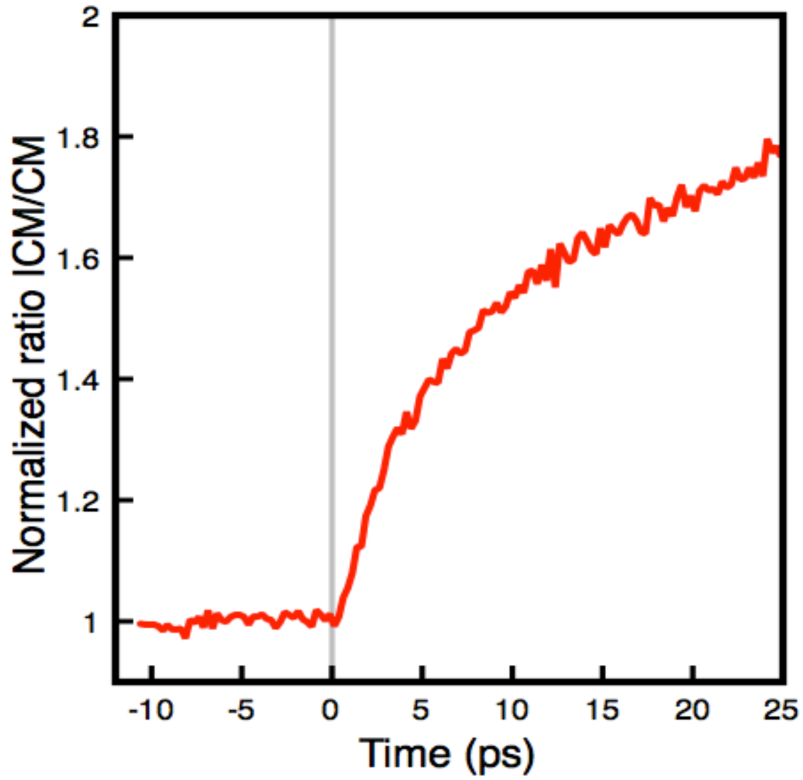
[Johnson et al, arXiv:1106.6128]

Analysis

t = 0: onset of spin disorder, relative drop identical

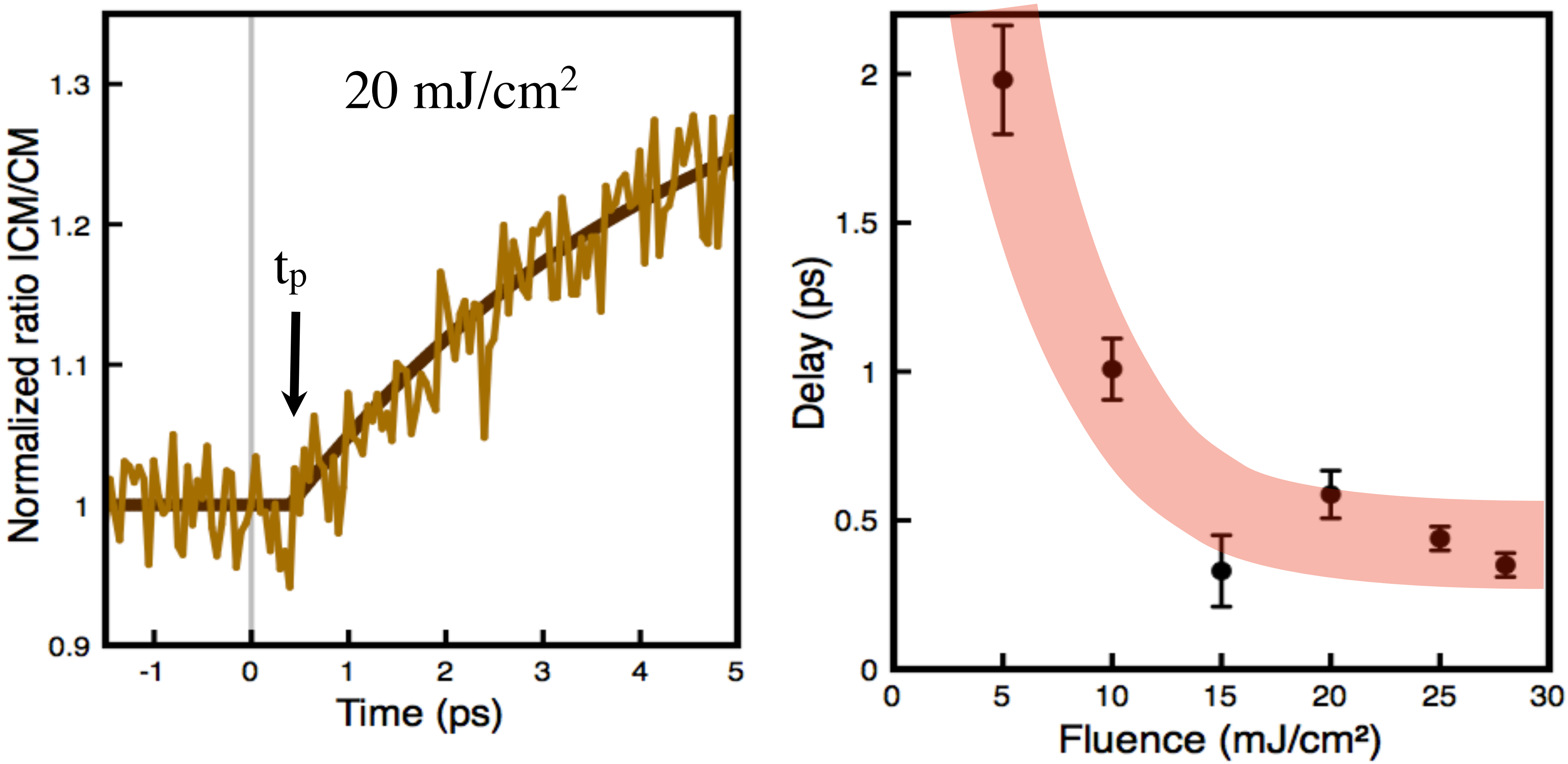
Ratio: relative phase population





[Johnson et al, arXiv:1106.6128]

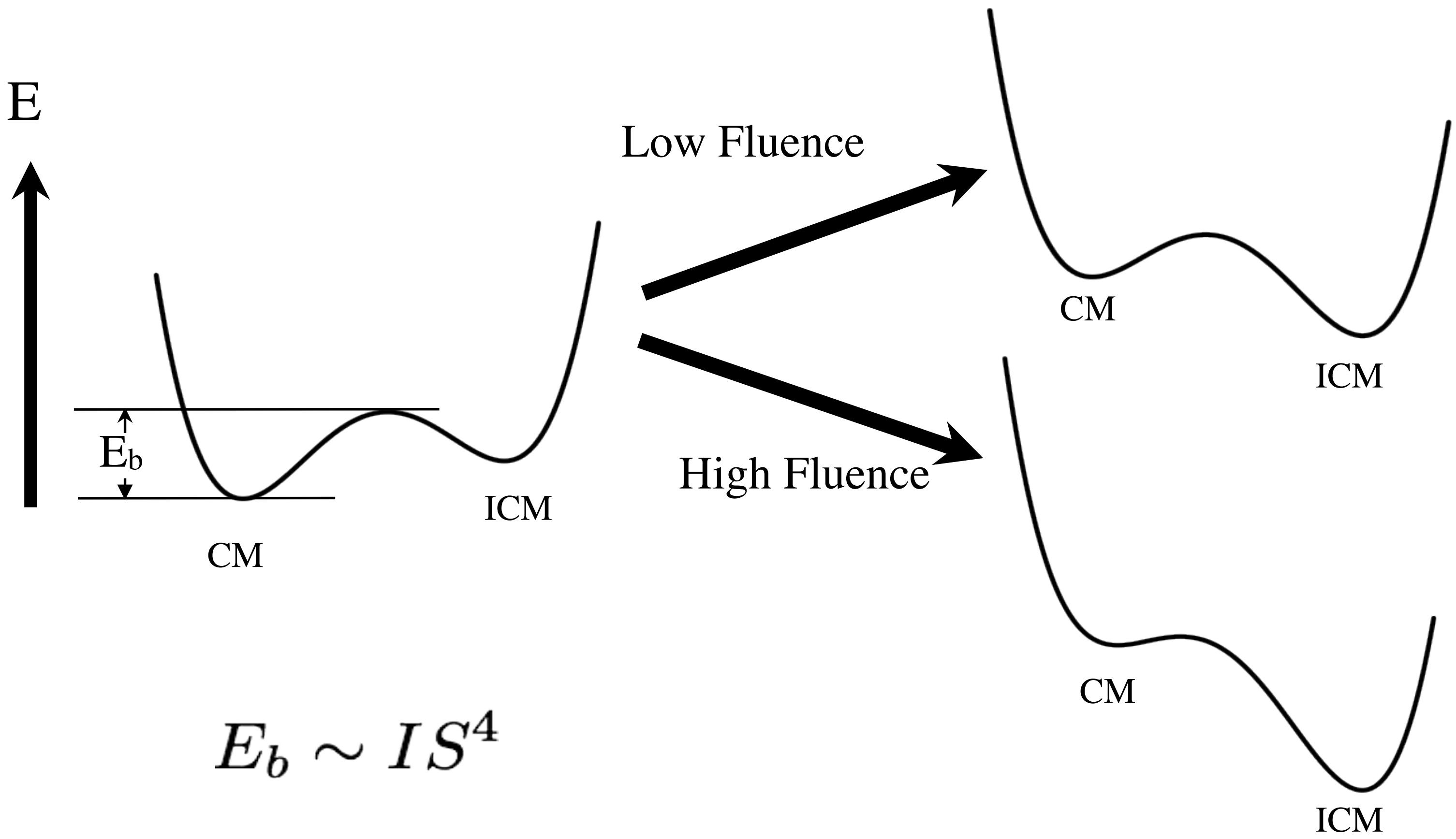
Phase transition onset



- Delay between generation of disorder and onset of phase transition

[Johnson et al, arXiv:1106.6128]

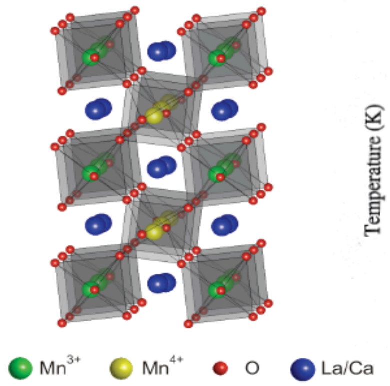
Onset time: theory expectations

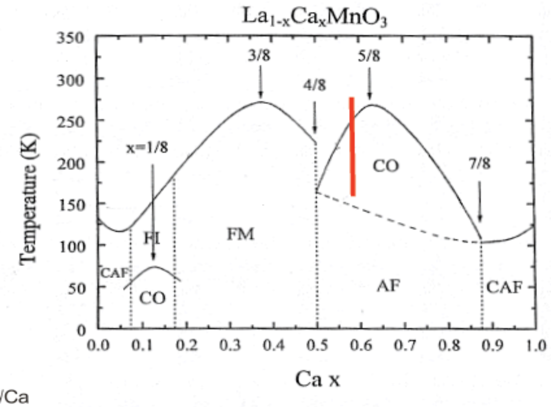


$$E_b \sim I S^4$$

- Excitation → fast S^2 drop, changes relative energies
- ... lowers height of energy barrier (~ biquadratic term)
- Minimum delay roughly 1/4 period of spin wave gap (1.6 ps)

Manganites

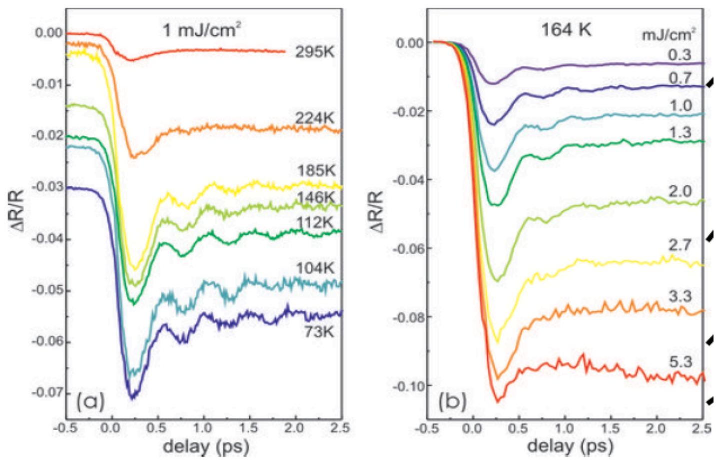




[Cheong & Hwang (1999)]

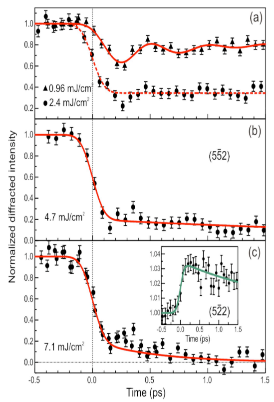
Can we learn how charge & orbital order couples to structure?

Optical pump-probe



Hard x-ray pump-probe

$La_{0.42}Ca_{0.58}MnO_3$



PRL **103** 155702 (2009).

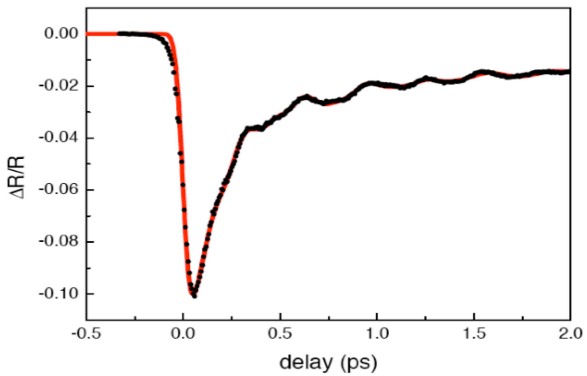
Structural phase transition < 140 fs

Manganites II

LSMO 1/8 doped

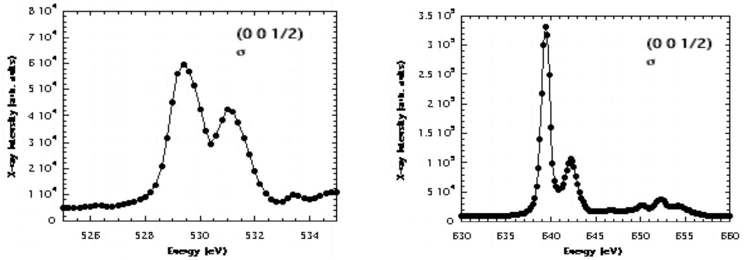
Optical measurement

Fig. 2: Optical reflectivity of $La_{7/8}Sr_{1/8}MnO_3$ at 100K



Static resonant scattering

O K edge Mn $L_{2,3}$ edge



Resonant soft x-ray diffraction (0 0 1/2) reflection at Mn L2,3 and O K edge of $La_{7/8}Sr_{1/8}MnO_3$ taken at the SLS. (PRL **99**, 206402 (2007) ref. 1)

Creation of coherent optical phonon

Is sensitive to charge order!

Parameters for the Source/Station (Wish list)

Ideal characteristics:

Temperature control: 5-400 K

2D detector, one image / pulse X-ray energies: < 950 eV monochromatic scanable

Polarization of source adjustable circular, linear

Polarization analysis of diffracted beam could be implemented

Pump options: optical (excite charge / spin), IR, THz (excite phonon modes directly), use first harmonic (x-rays) to pump (reduce time jiter)

Time jitter as small as possible

Conclusion

PSI Proposes to built/bring and operate an experimental station for FLASHII
Open for users and suggestions

Physics:
Strongly correlated electron systems, Focus on pump-probe experiments

Variable Pump schemes

Probe:
Resonant x-ray scattering (large angle coverage)
X-ray absorption

Other **stimuli**:
Magnetic fields, electric fields, (low) temperature

Acknowledgments

LCLS CuO Experiment

PSI
E. Vorobeva
P. Beaud
A. Caviezel
G. Ingold
R. De Souza
V. Scagnoli
L. Patthey

S. L. Johnson (ETHZ)

W. Schlotter (SLAC)
J. Turner (SLAC)
M. Trigo (SLAC)

A. T. Boothroyd (Oxford)

Y.-D. Chuang (LBNL)
P. Denes (LBNL)
D. Doering (LBNL)
Z. Hussain (LBNL)

W.-S. Lee (Stanford)
R. G. Moore (Stanford)
D. Lu (Stanford)
M. Yi (Stanford)
P. Kirchmann (Stanford)
Z. X. Shen (Stanford)

Manganites

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C. Schuessler-Langeheine

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