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Ultrafast X-ray scattering in correlated materials

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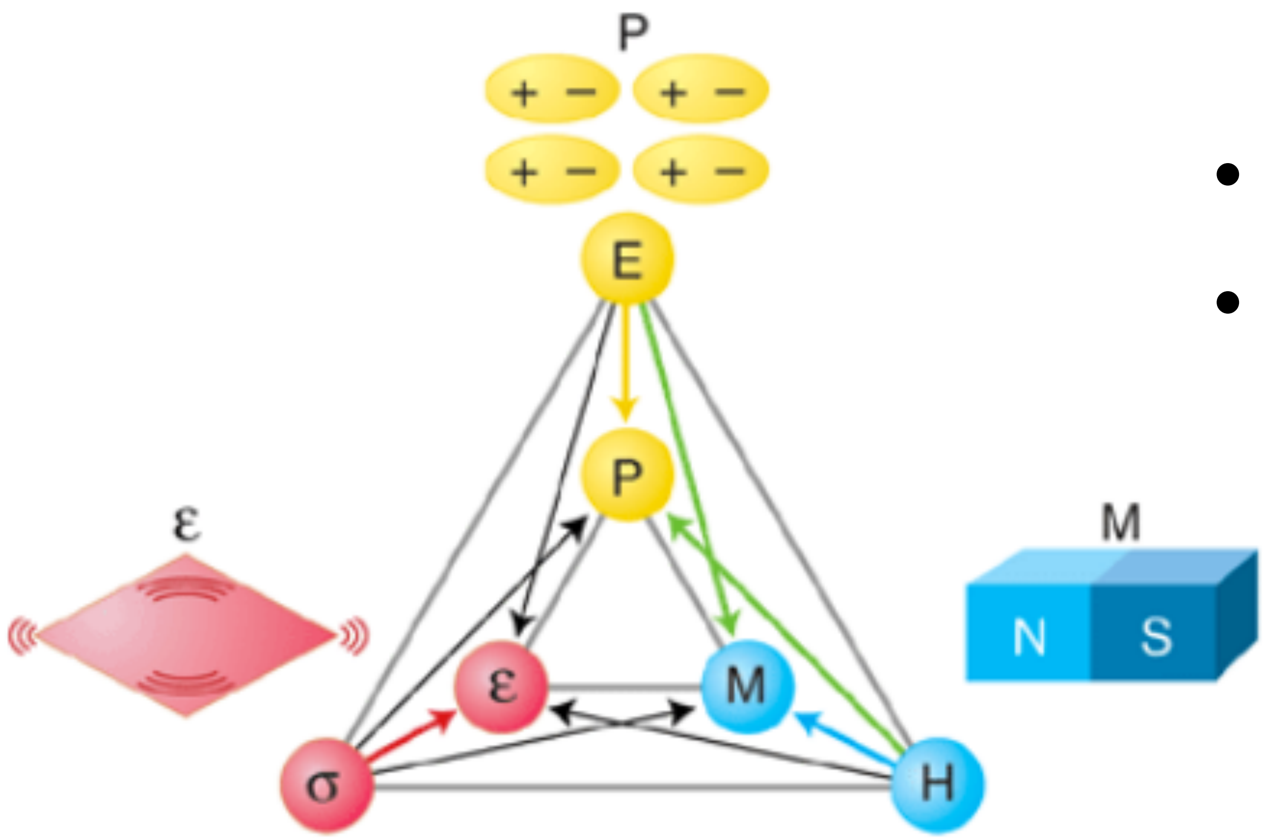
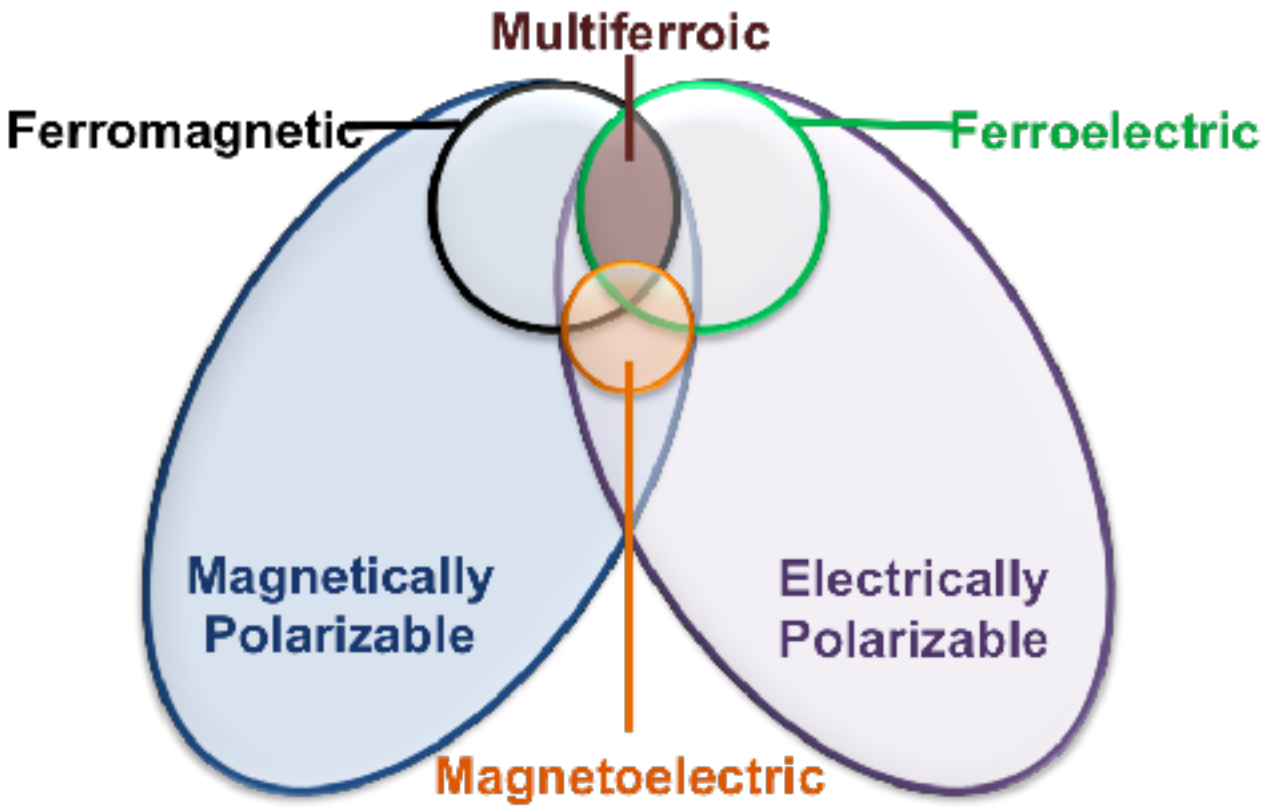
Correlated materials and ferroic order



- Interactions strongly compete
- Complex phase diagrams



- We study nonlinear dynamics up to the point of ferroic order reversal

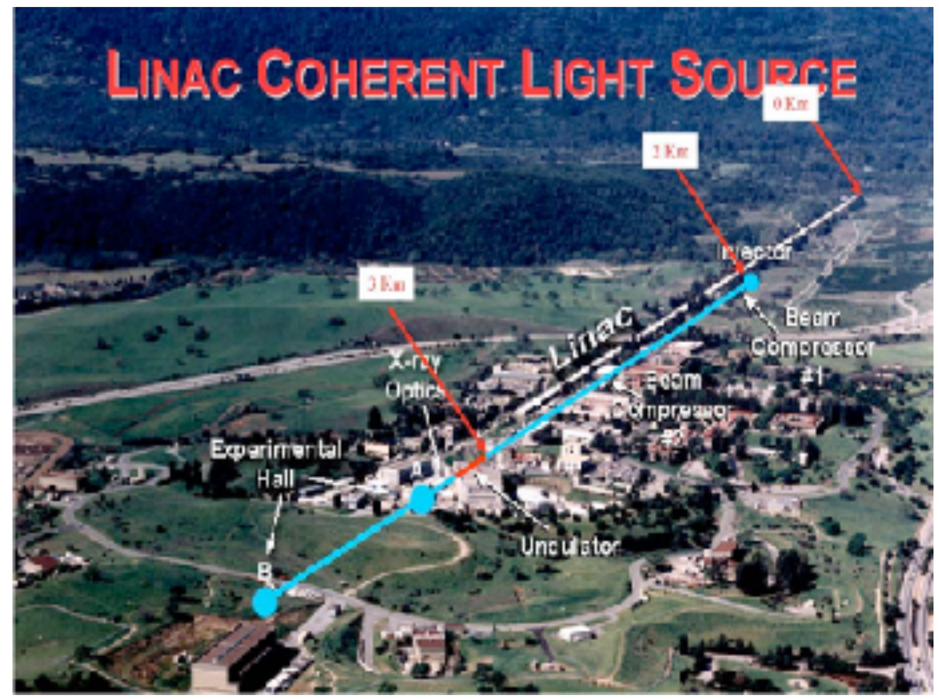
Spaldin, Fiebig, Science 309, 391 (2005)

Zhou *et al*. Energy Harvesting and Systems, 3 (2015)

Accelerator based x-ray sources

"Third" generation

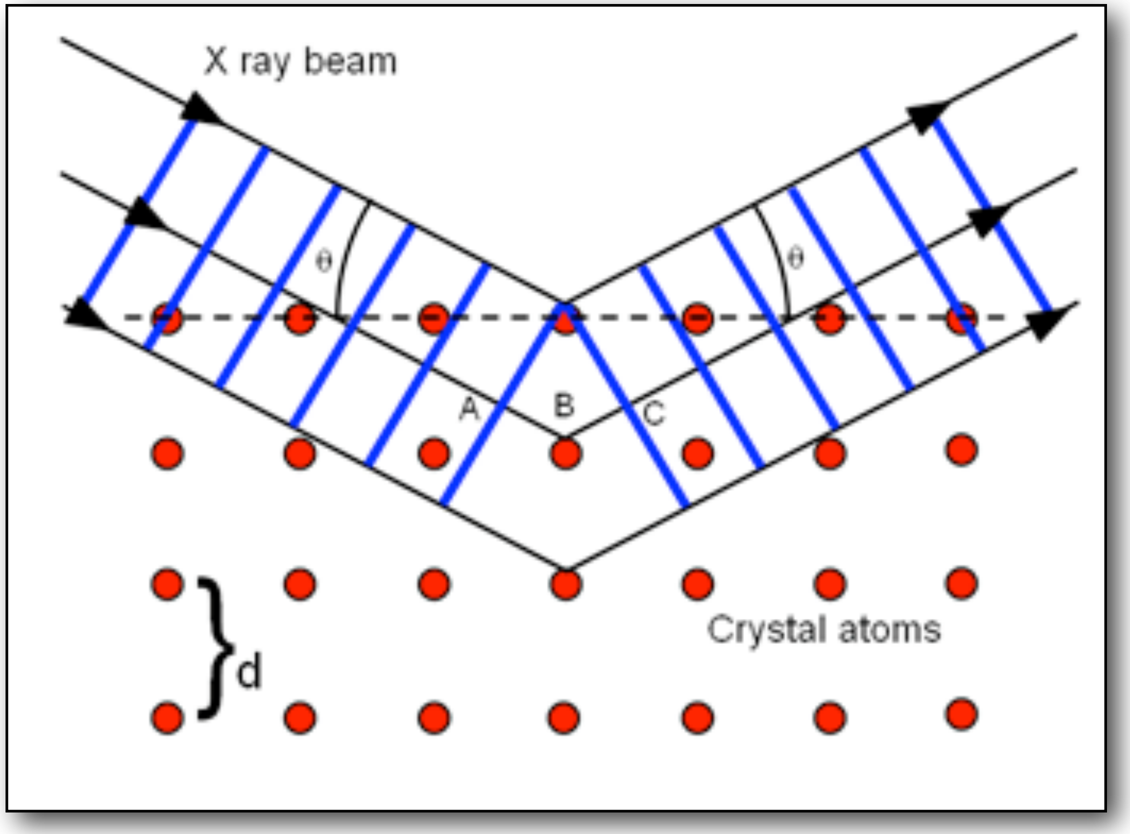
"Fourth" generation

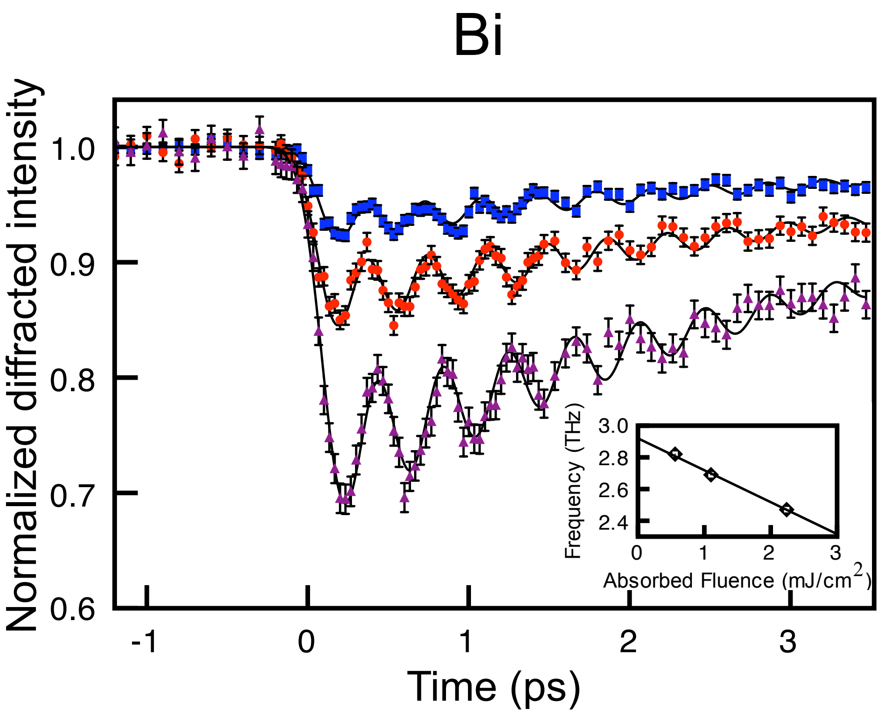


"Fifth" generation

~10^{10} more photons in one pulse!

X-ray diffraction



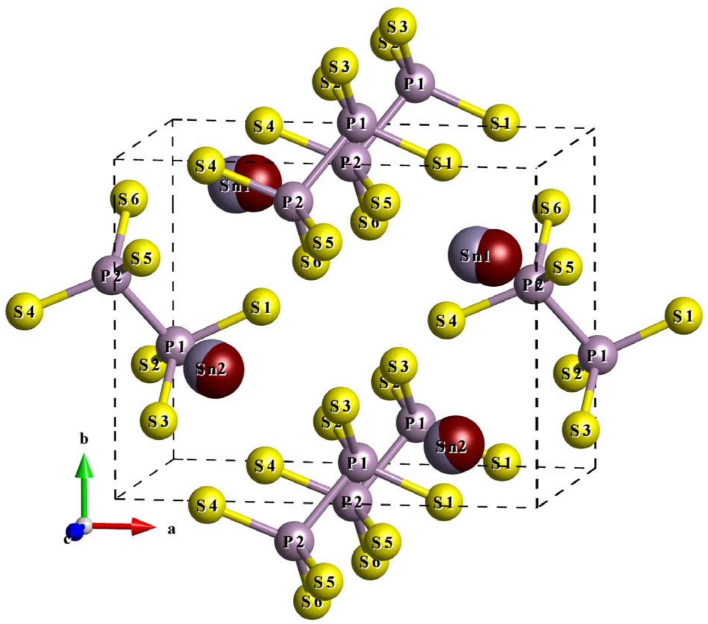


[Johnson et al. PRL 100, 155501 (2008)]

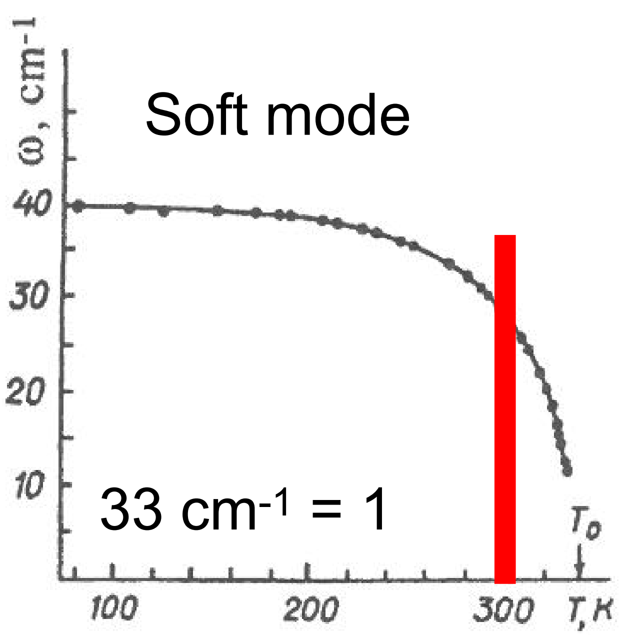
$$I(Q) \propto |F(Q)|^2 = \left| \sum_j f_j e^{i\mathbf{r}_j \cdot \mathbf{Q}} \right|^2$$

Intensity as function of photon momentum transfer directly related to Fourier Transform of electron density

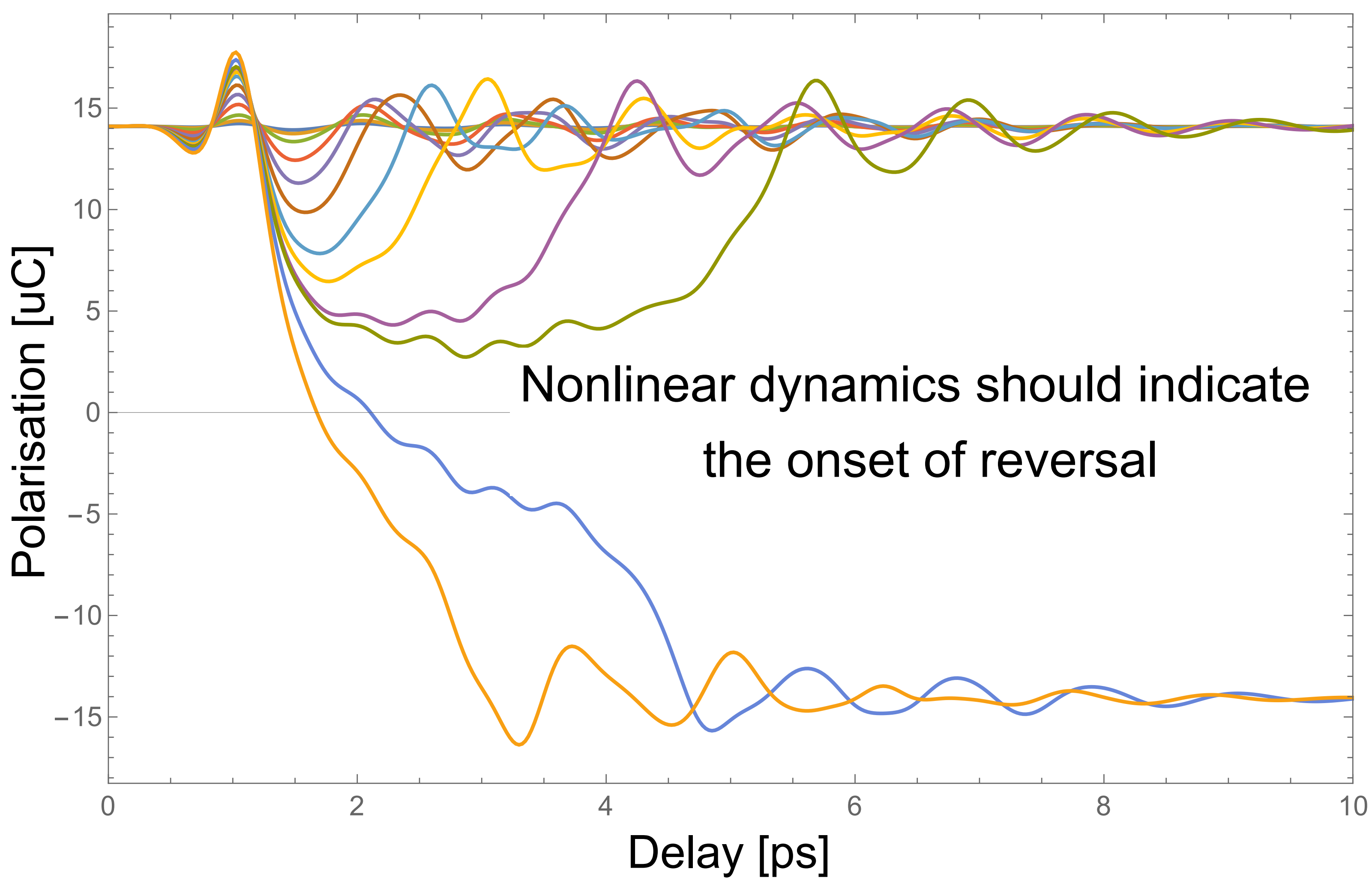
Case Study # 1: $Sn_2P_2S_6$ a soft mode ferroelectric



[Glukhov et al. Int. J. Mol. Sci. 2012, 13, 14356]



[Vysochanskii et al. Fiz. Tv. Tela 20, 90 (1978)]



Sn-atoms position determine the ferroelectric properties.

We want to resonantly excite the soft phonon mode to induce reversal in the ferroelectric polarisation

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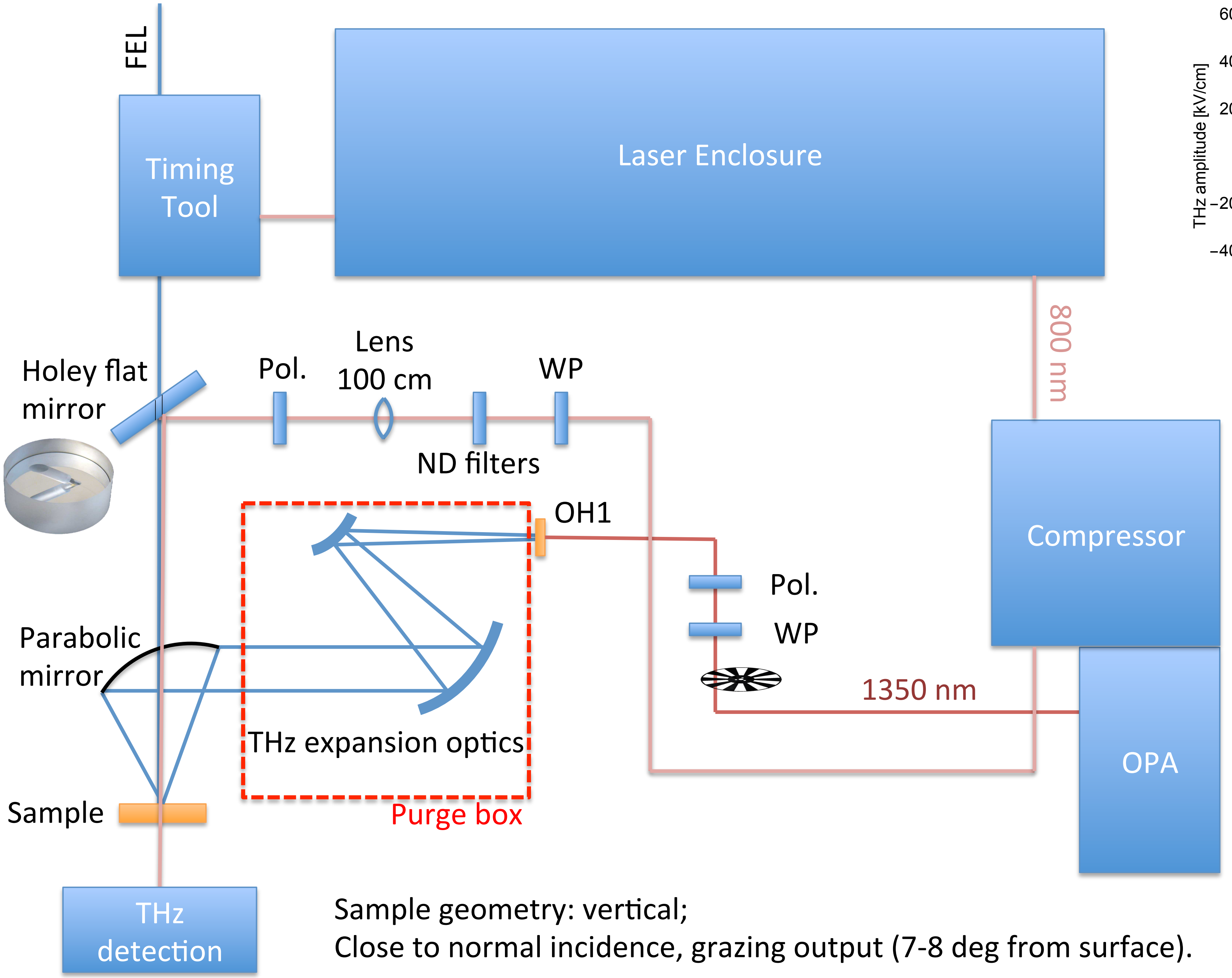
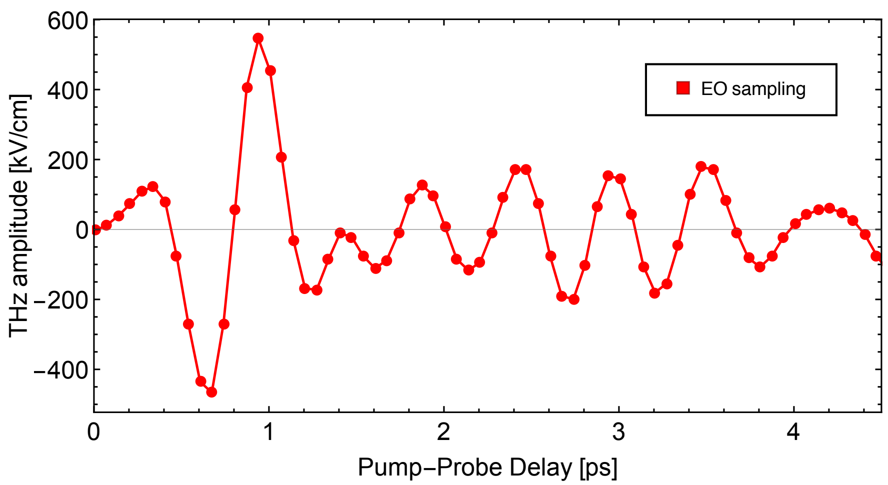
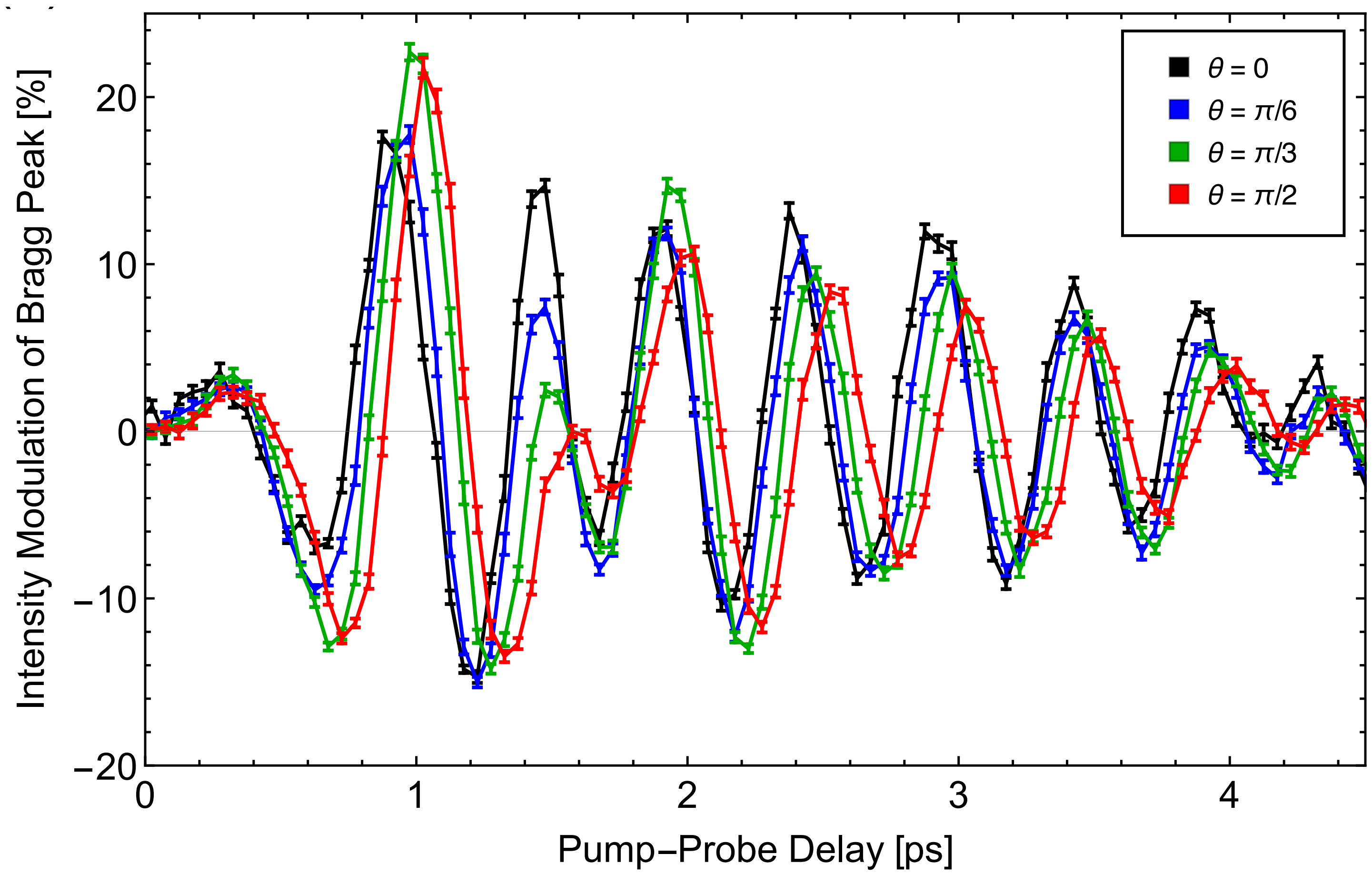
$Sn_2P_2S_6$: THz pump - x-ray probe





Sample geometry: vertical;
Close to normal incidence, grazing output (7-8 deg from surface).

$Sn_2P_2S_6$ THz pumping at LCLS, XPP



Savoini et al. *in preparation*

We can excite large variations in the diffracted intensity (>20%)

We quantify the corresponding change in FE polarisation to > 5% of the full reversal

We can track the phase transition by measuring the induced effect at different temperatures

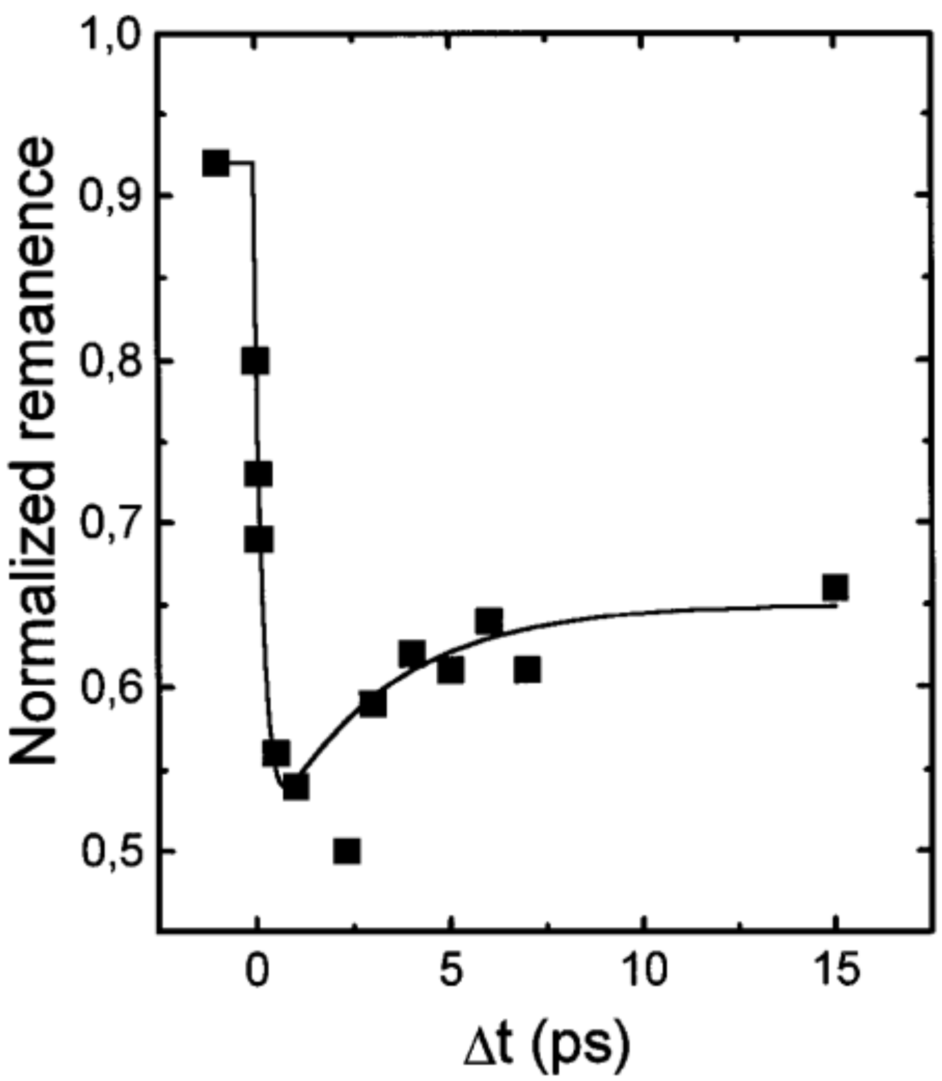
Case Study # 2: ultrafast demagnetisation

- Unexpectedly fast drop in a Ni sample magnetization upon laser pulse illumination

Beaurepaire, PRL (1996)

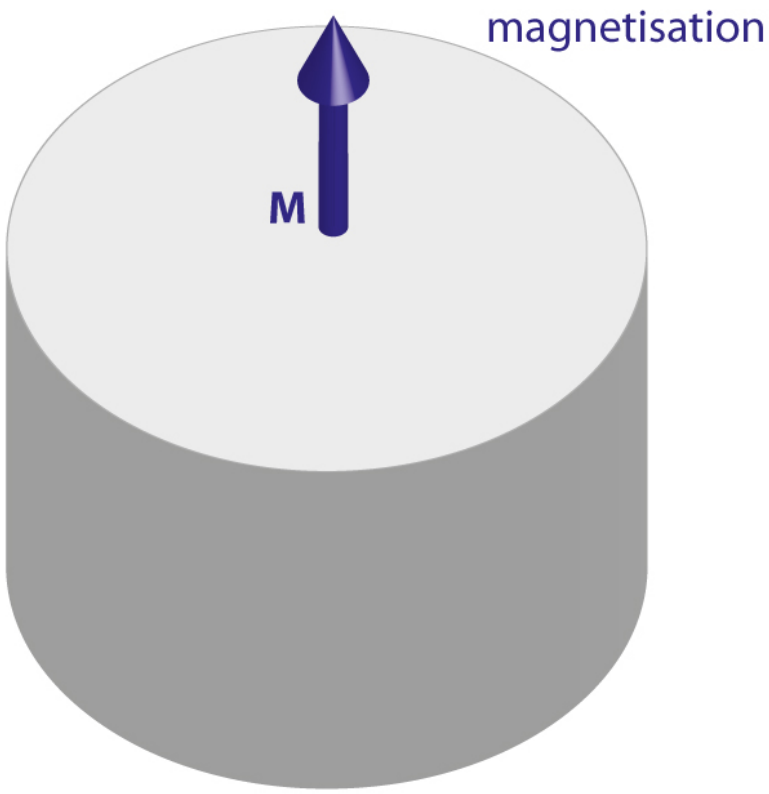
- Not well understood how angular momentum is conserved

- In 2016 we designed a possible test experiment.

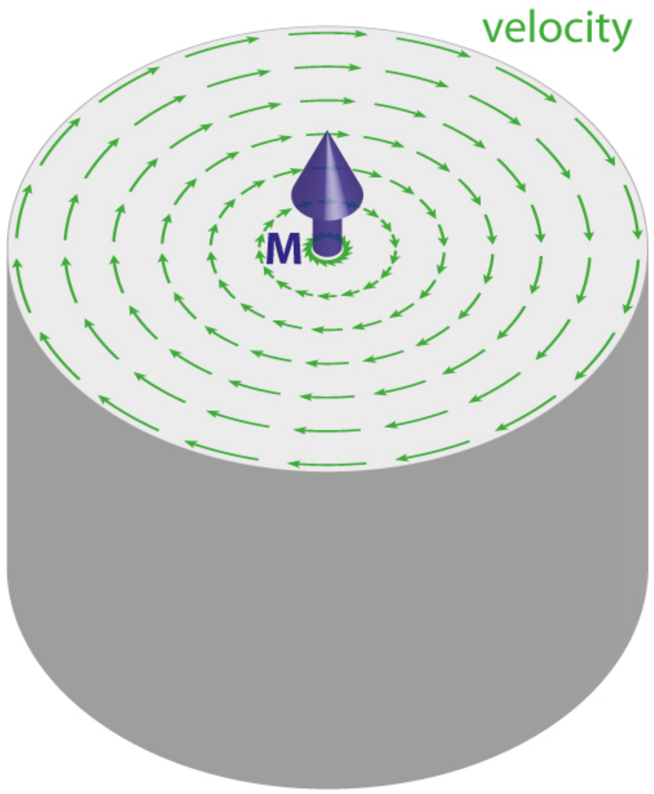


Our idea: ultrafast Einstein - de Haas effect

rod at rest, magnetisation
M along the z-axis
t < 0



macroscopically
rotating rod
on long timescales

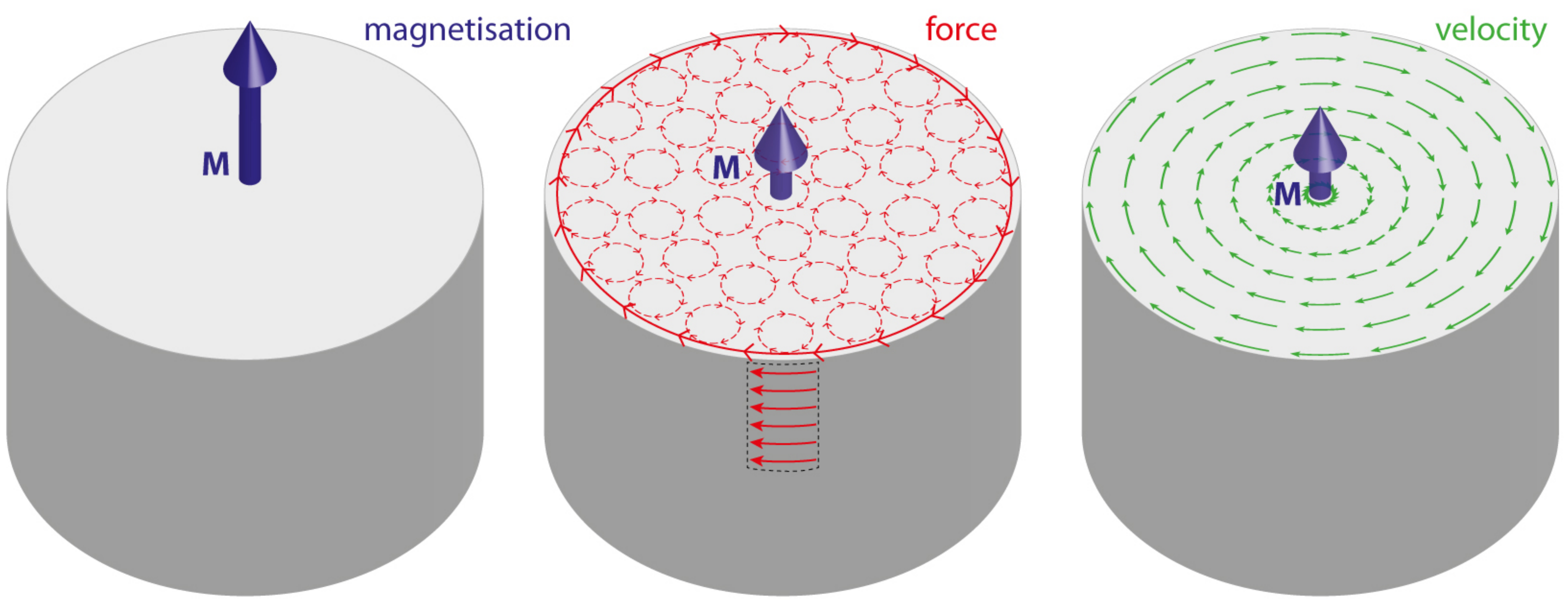


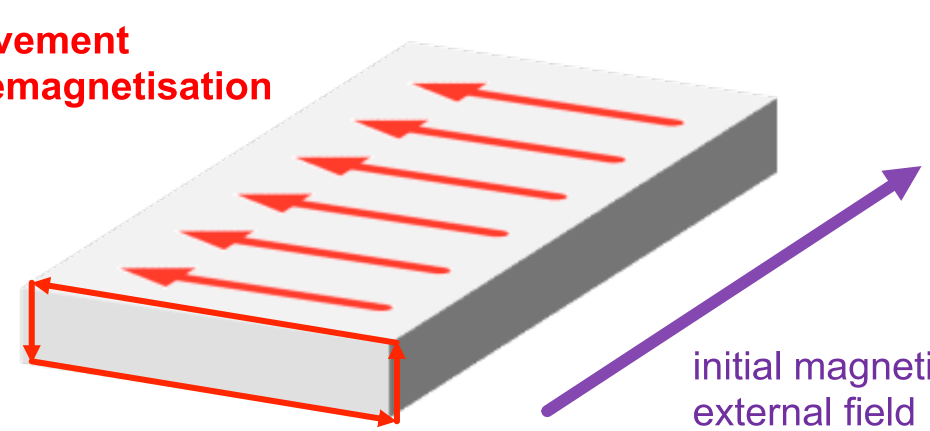
Our idea: ultrafast Einstein - de Haas effect

rod at rest, magnetisation
M along the z-axis
$t < 0$

instantly after ultrafast
demagnetisation;
the forces cancel except at
the surface parallel to M
$t \approx 100$ fs

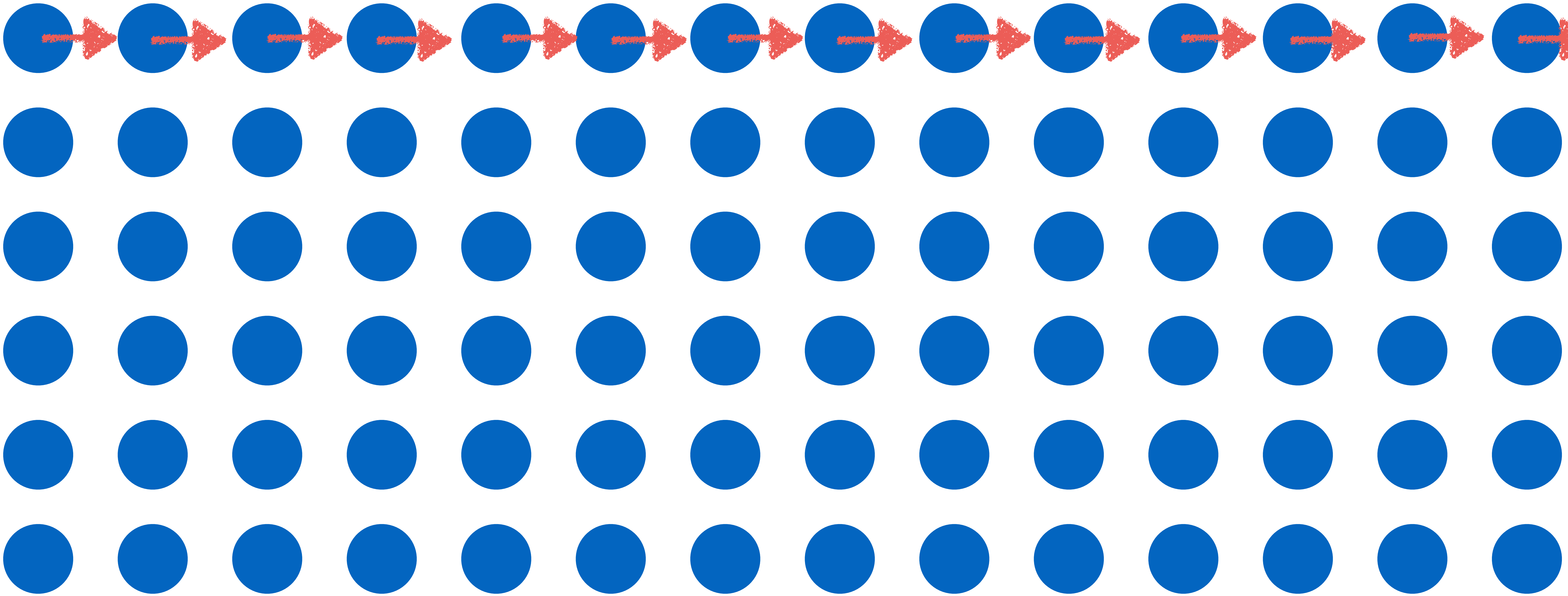
macroscopically
rotating rod
on long timescales



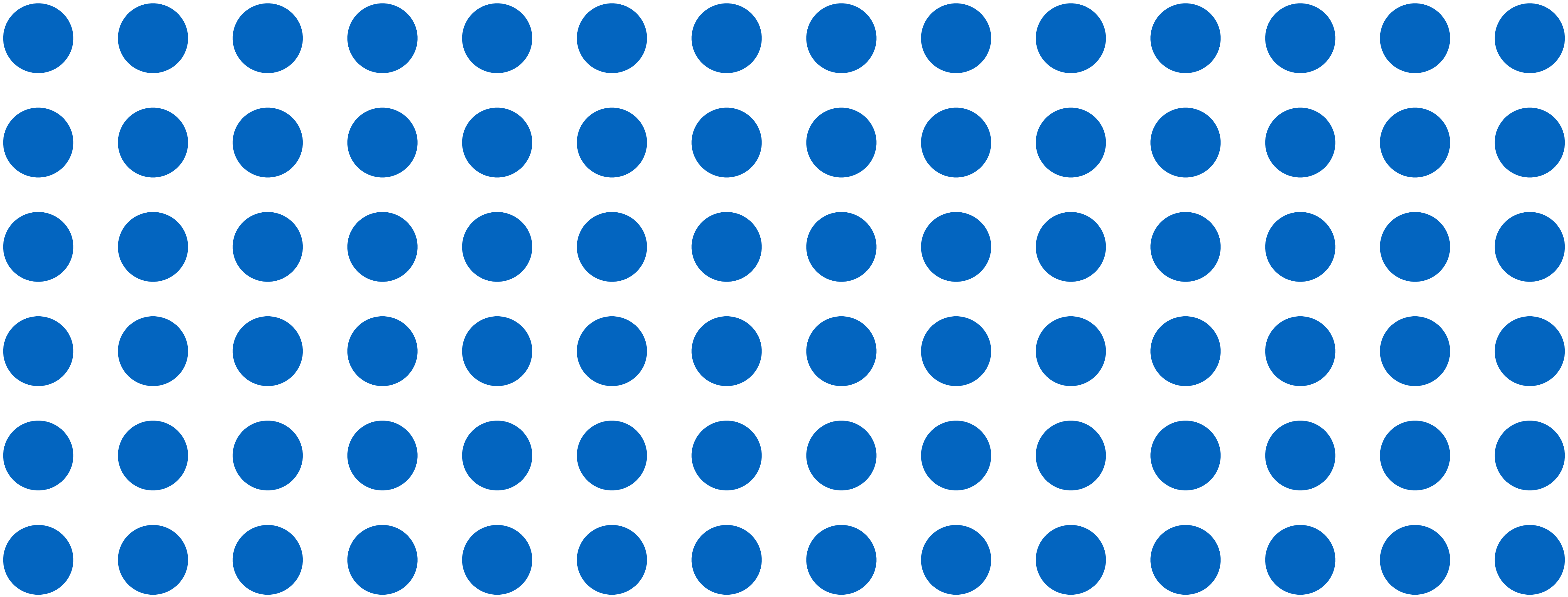


$$\vec{f} = \nabla \cdot \sigma_M$$

$\Rightarrow$ force density non-zero only at surfaces!!

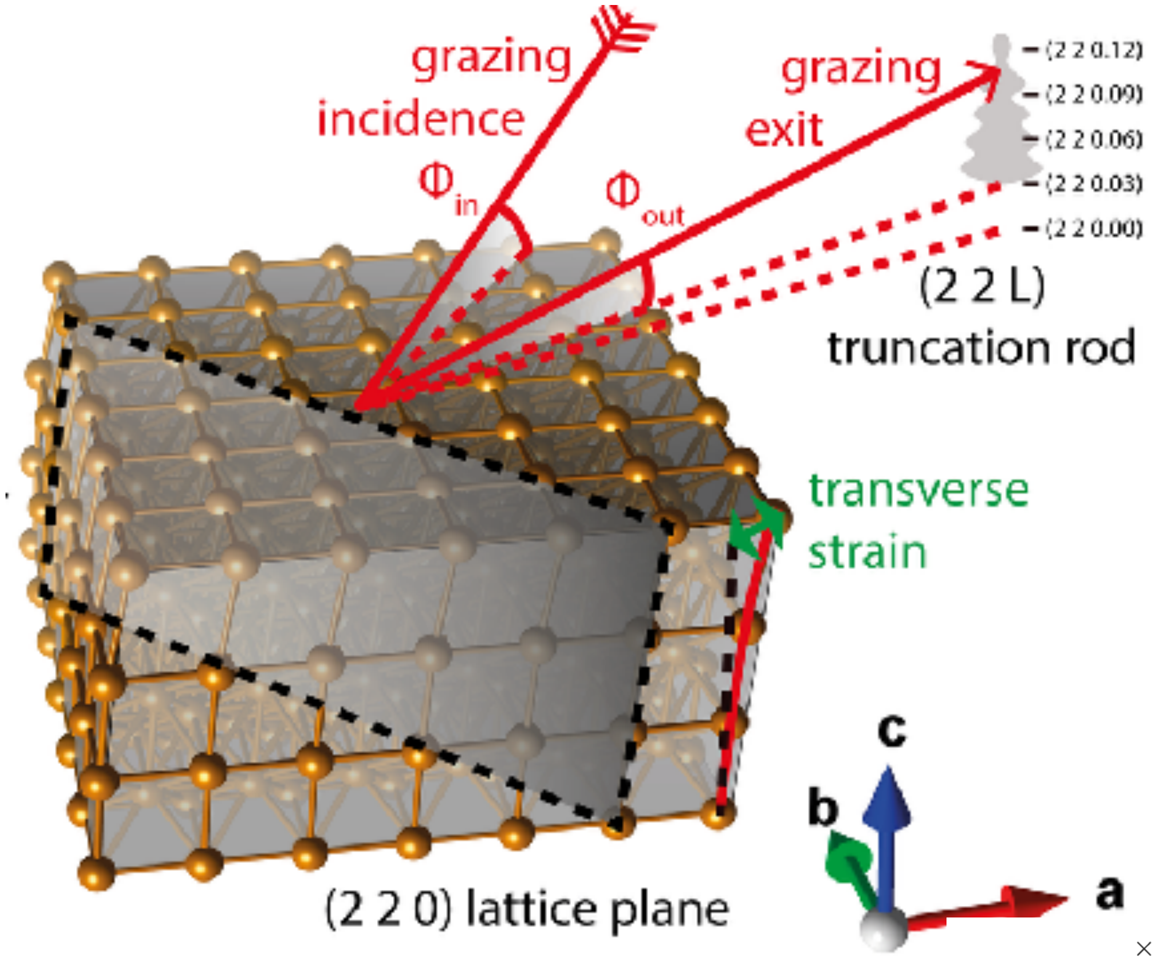


Transverse displacement wave

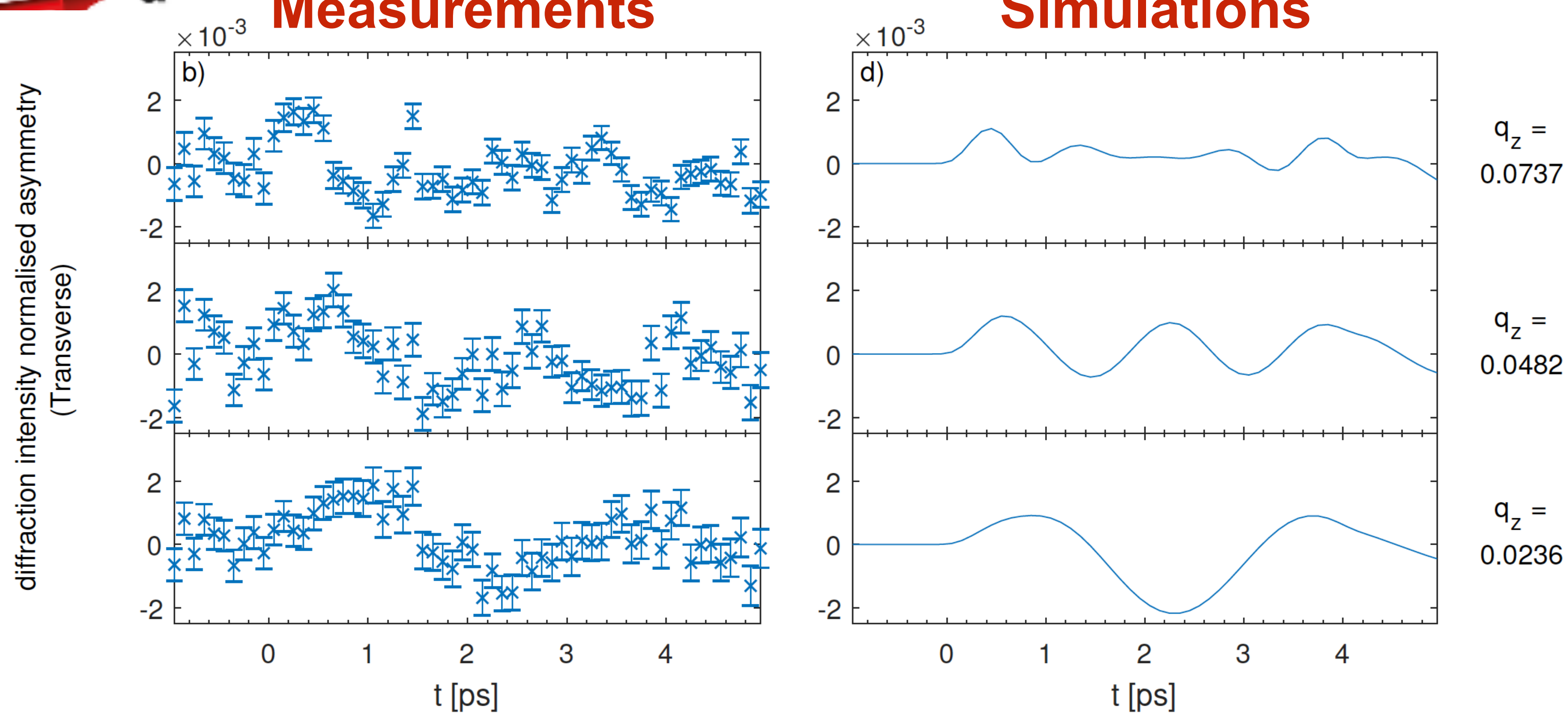


Experiment & results

- X-ray diffraction
- In-plane geometry with grazing incidence and exit
- Crystal Truncation Rod
- Recover lattice dynamics by comparing with simulation



What we measured



From our measurements we estimate that the lattice is taking up to 80% of the lost magnetic moment within the first 200 fs.

Dornes et al., Nature *(in publication)*

What we dream about the next generation?

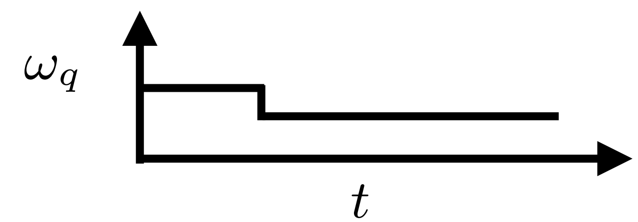
Manipulating correlations

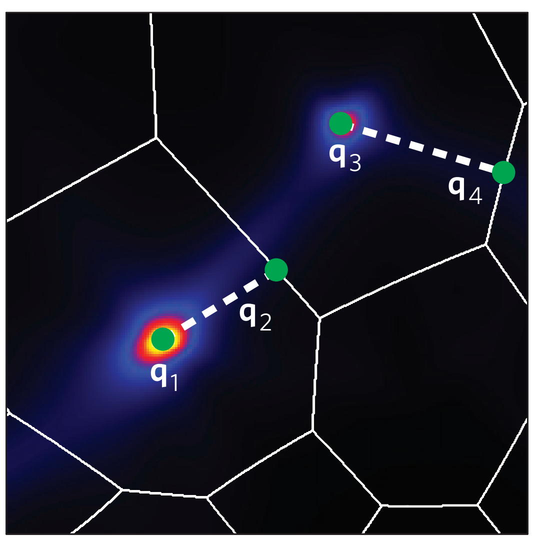
LETTERS
PUBLISHED ONLINE: 27 OCTOBER 2013 | DOI: 10.1038/NPHYS2788
nature physics

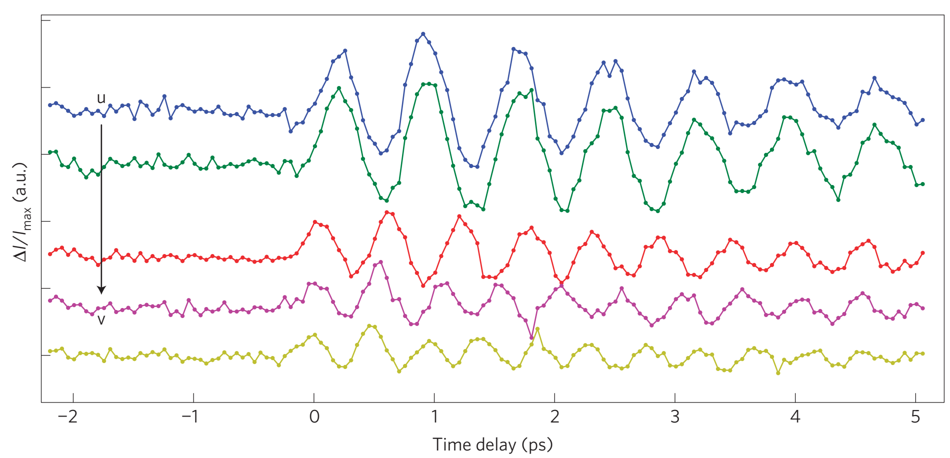
Fourier-transform inelastic X-ray scattering from time- and momentum-dependent phonon–phonon correlations

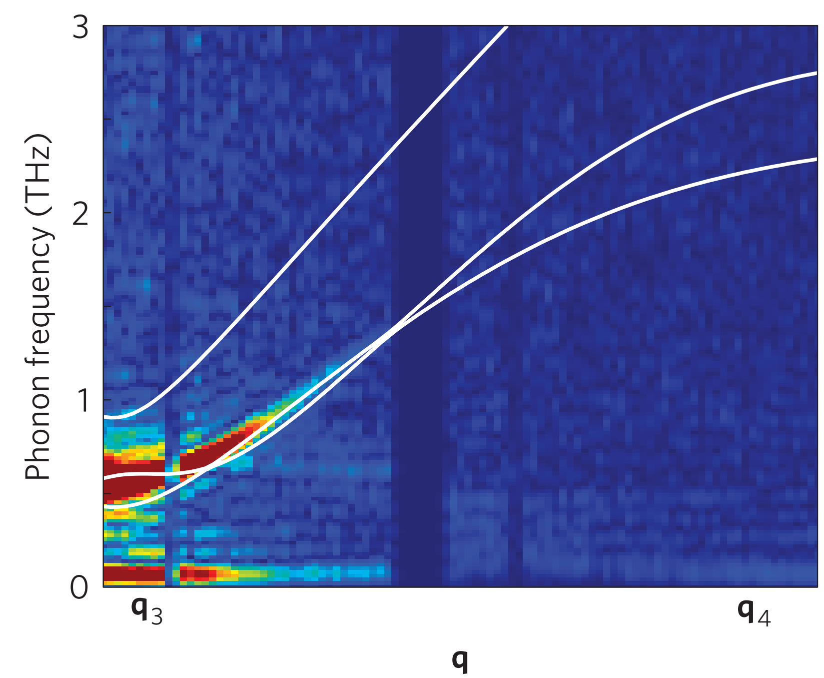
M. Trigo[1,2]*, M. Fuchs[1,2], J. Chen[1,2], M. P. Jiang[1,2], M. Cammarata[3], S. Fahy[4], D. M. Fritz[3], K. Gaffney[2], S. Ghimire[2], A. Higginbotham[5], S. L. Johnson[6], M. E. Kozina[2], J. Larsson[7], H. Lemke[3], A. M. Lindenberg[1,2,8], G. Ndabashimiye[2], F. Quirin[9], K. Sokolowski-Tinten[9], 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
- Xray photons have reasonably large momentum, allows momentum coverage

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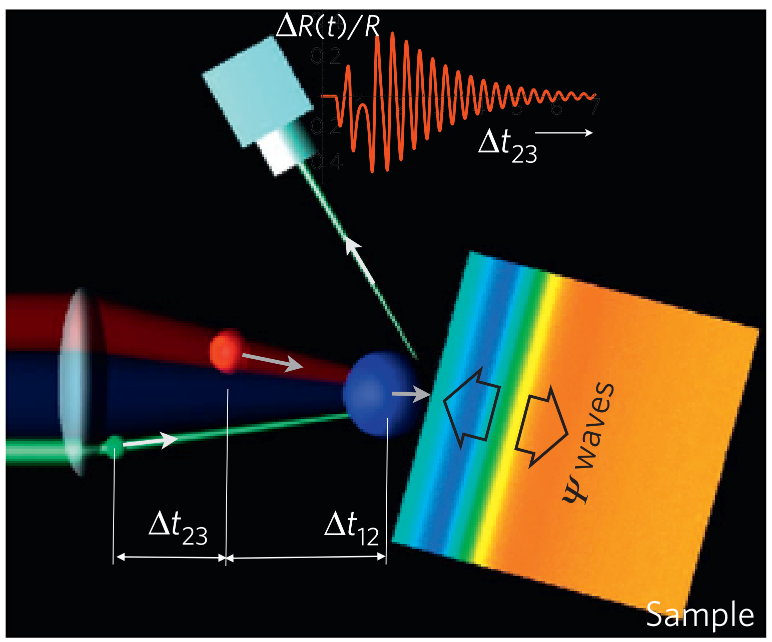
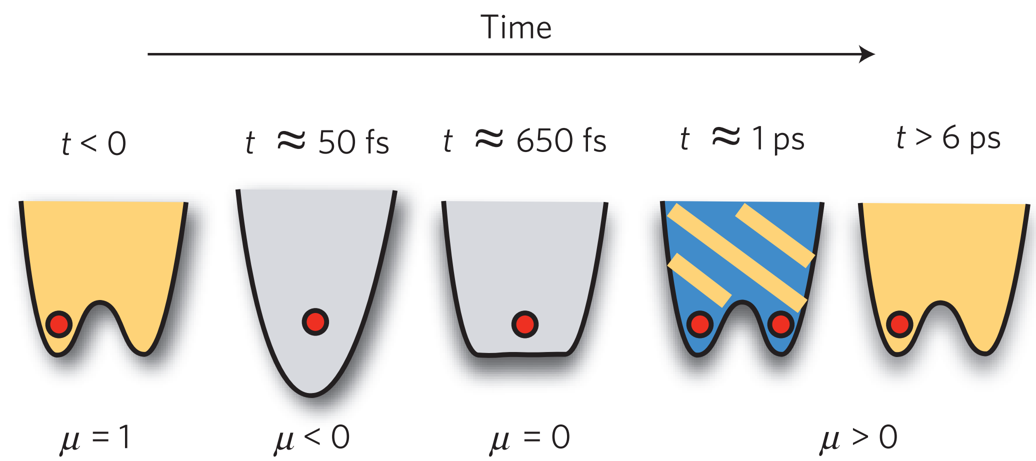
“Double pump” experiments

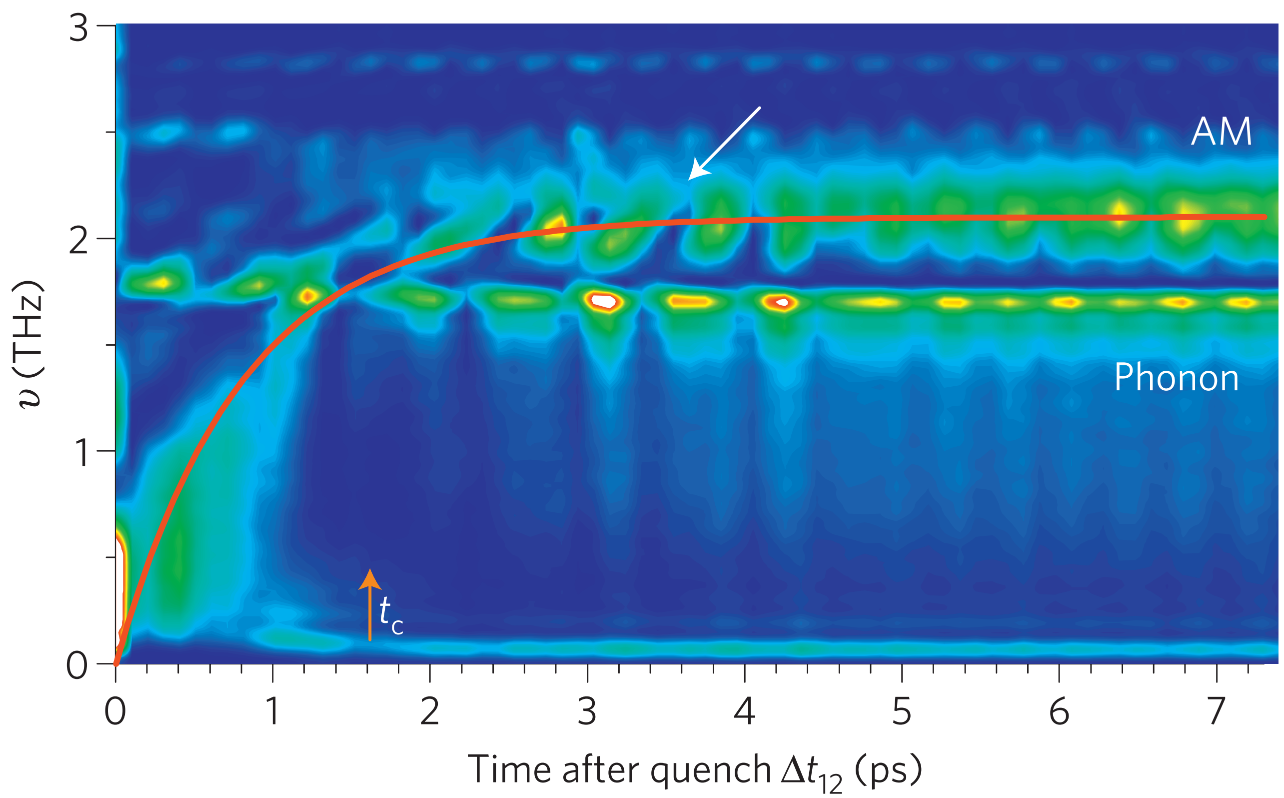
nature physics
LETTERS
PUBLISHED ONLINE: 22 AUGUST 2010 | DOI: 10.1038/NPHYS1738

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
- Initially optical - optical - X-ray, afterwards optical - X-ray - X-ray
- Subtract out signal without second pump

Limits and Opportunities

- Pulse duration < 10 fs gives enough bandwidth to study < 60 meV excitations via FT methods
  - going to sub-fs is not a strong requirement
- Transform limited pulses desirable
- Ability to smoothly tune pulse duration and bandwidth within FT limit beneficial
- Extra lasers synchronised for double pump experiments
- High rep rate not critical, unless coupled with THz pulses directly from the machine
- Accurately timed and phased double pulses might be useful for double pump schemes (needs further study)
- Stability essential
- Complete polarization control essential (for E < 1 keV)

Acknowledgments

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Questions??