Shaken, not stirred:
a recipe for ultrafast magnetization reversal

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https://www.ru.nl/en/hfml-felix

big toys for great science

Acknowledgements

The Netherlands

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Italy

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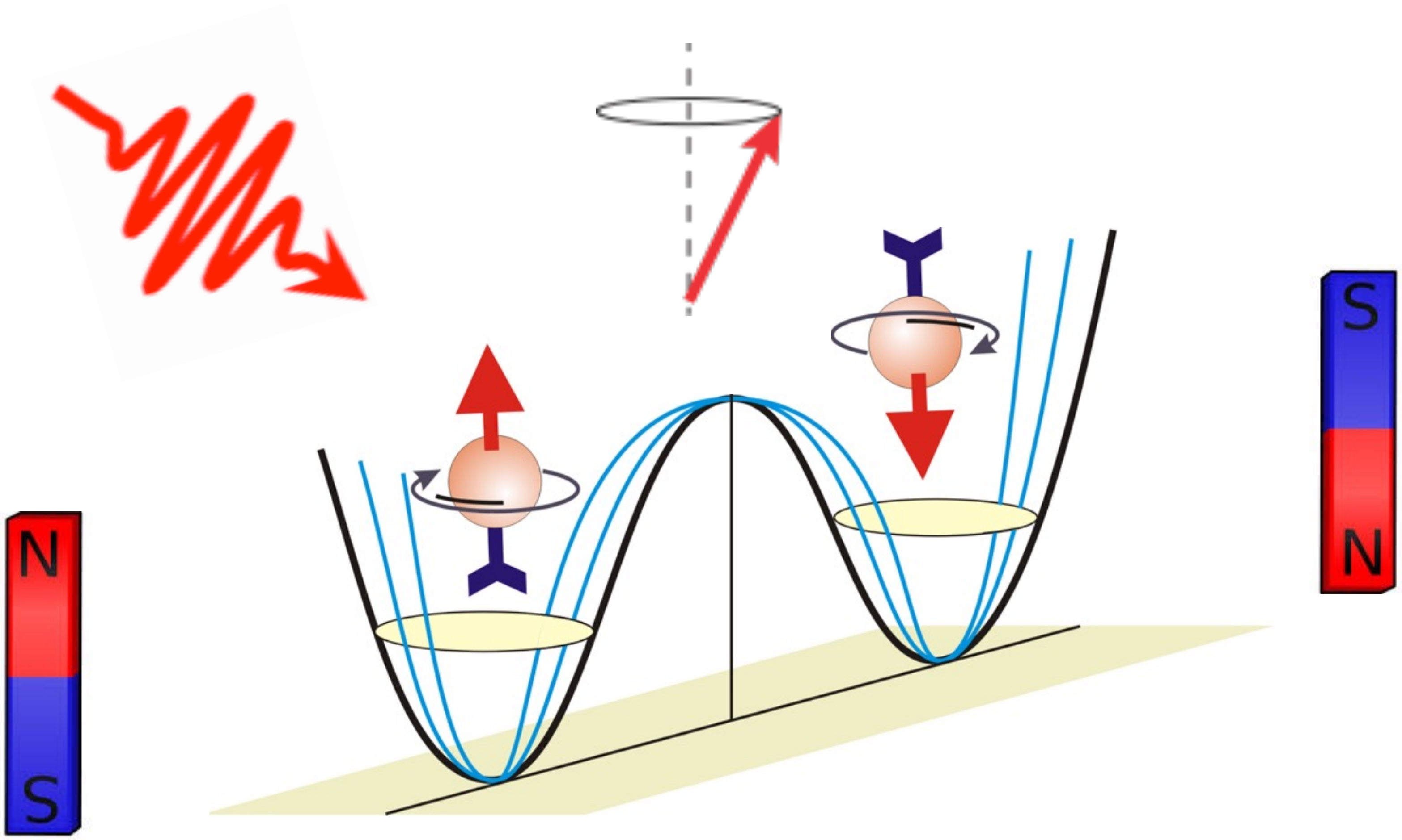
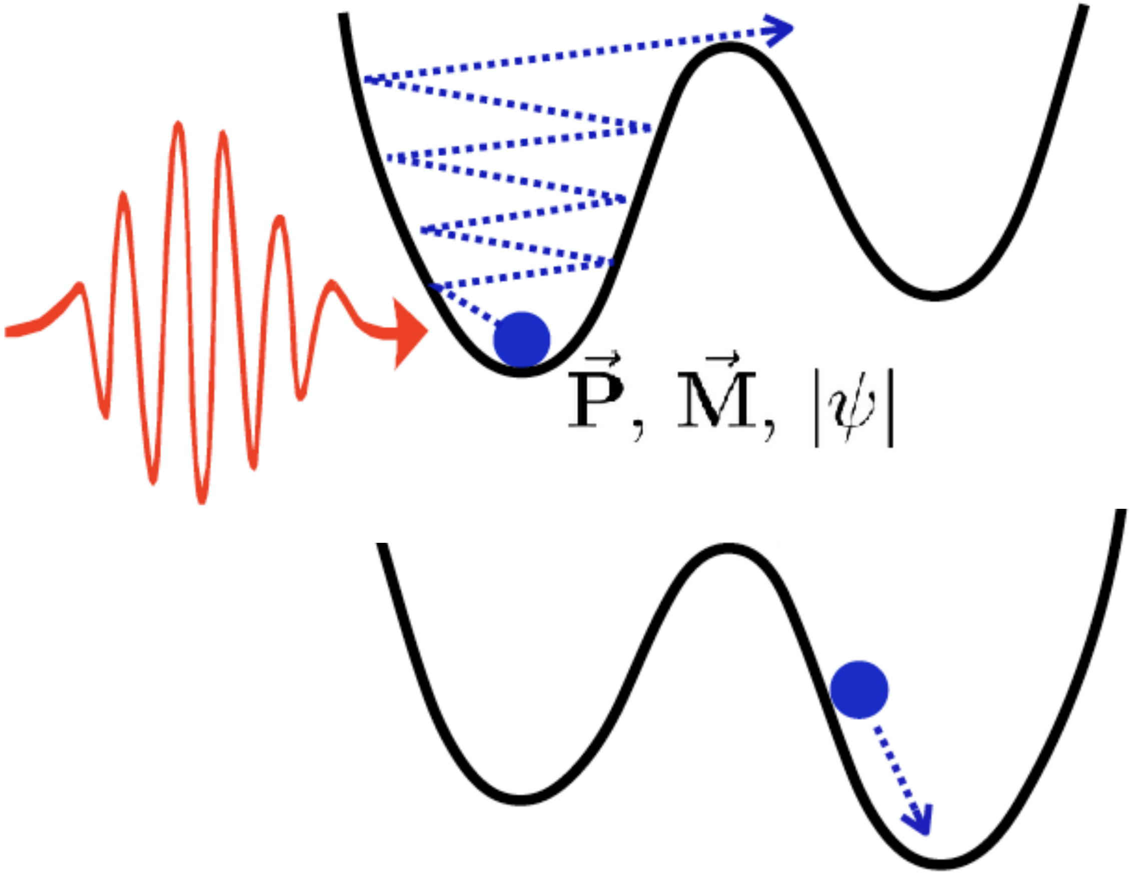
Giovanni Finocchio
Anna Giordano
Riccardo Tomasello

Magnetization dynamics: precession and reversal



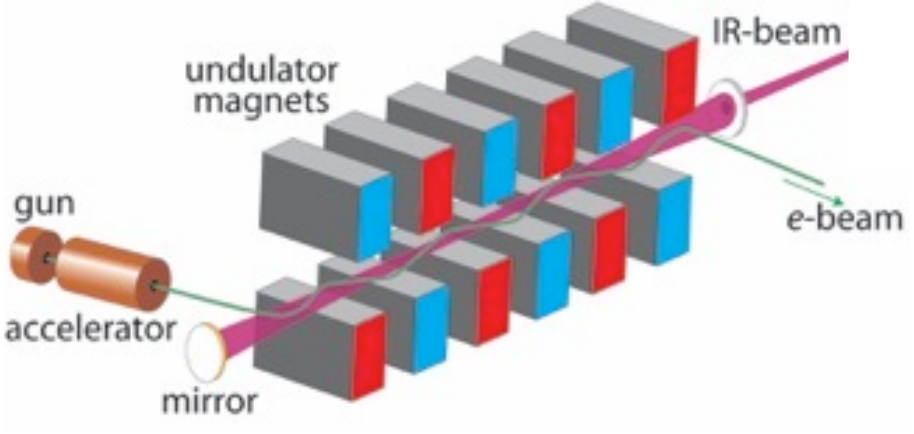
General idea: Coherent Control of (magnetic) Matter

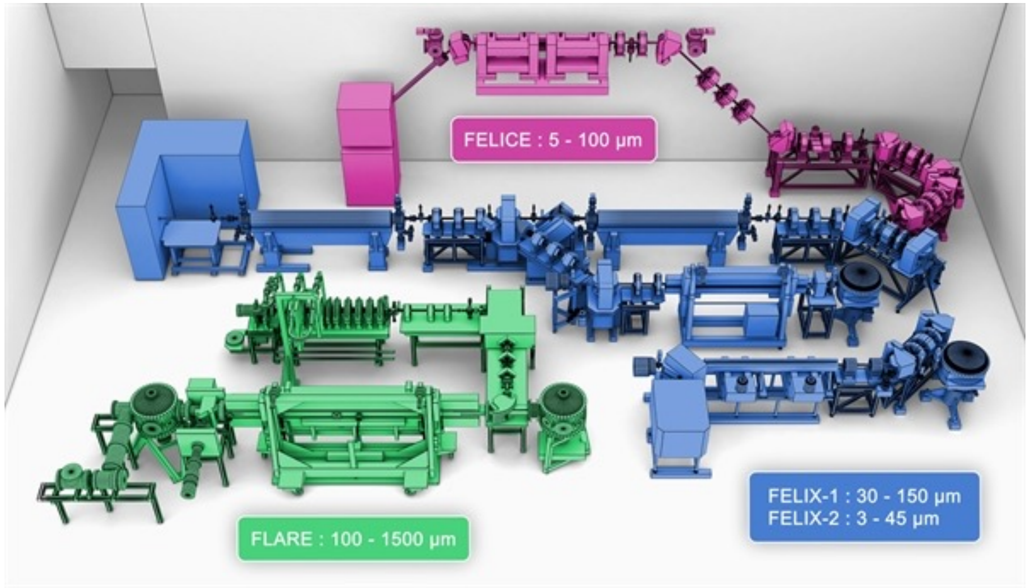


might need to simultaneously modify the potential, ideally by the same excitation and for the time of switching

Infrared / THz lasers at FELIX Laboratory

https://www.ru.nl/felix/facility/apply-beam-time/





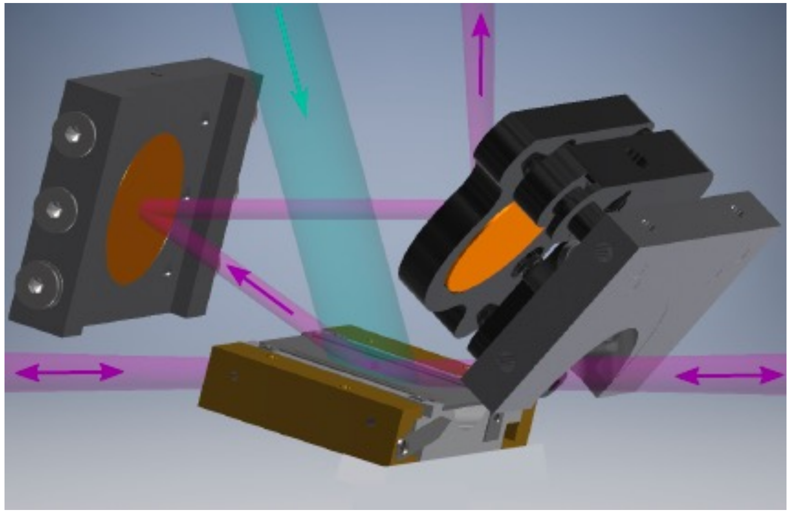
- λ tunable between 2.7µm and 1.5mm or between 0.2 – 90 THz
- <10 µJ's per (sub)ps pulse
- 0.4-4 % bandwidth, Fourier-transform limited

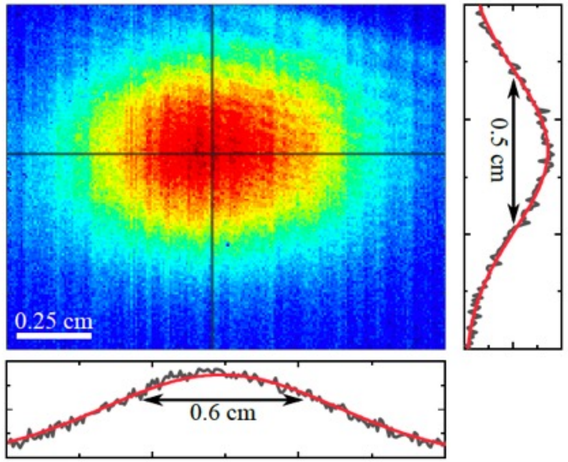
Oepts et al, *The free-electron-laser user facility FELIX*, Infrared Phys. Technol. **36**, 297 (1995)

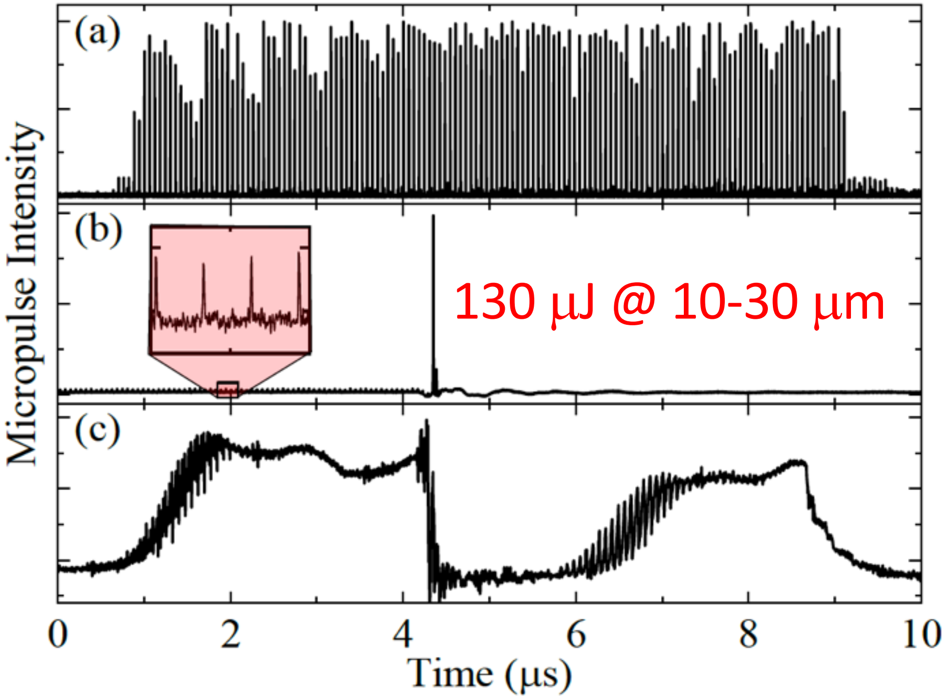
Infrared / THz range for dummies

3 μm	=	100 THz	=	3333 cm^{-1}	=	413 meV
10 μm	=	30 THz	=	1000 cm^{-1}	=	124 meV
30 μm	=	10 THz	=	333 cm^{-1}	=	41 meV
100 μm	=	3 THz	=	100 cm^{-1}	=	12 meV

Strong single picosecond pulses from cavity-dump







130 μJ @ 10-30 μm

to compare: outside the cavity, FELIX typically delivers pulse energies of <10 μJ

T. Janssen et al, Rev. Sci. Instrum. **93**, 043007 (2022)

Interaction between magnetism and lattice (phonons)

The ultrafast Einstein–de Haas effect

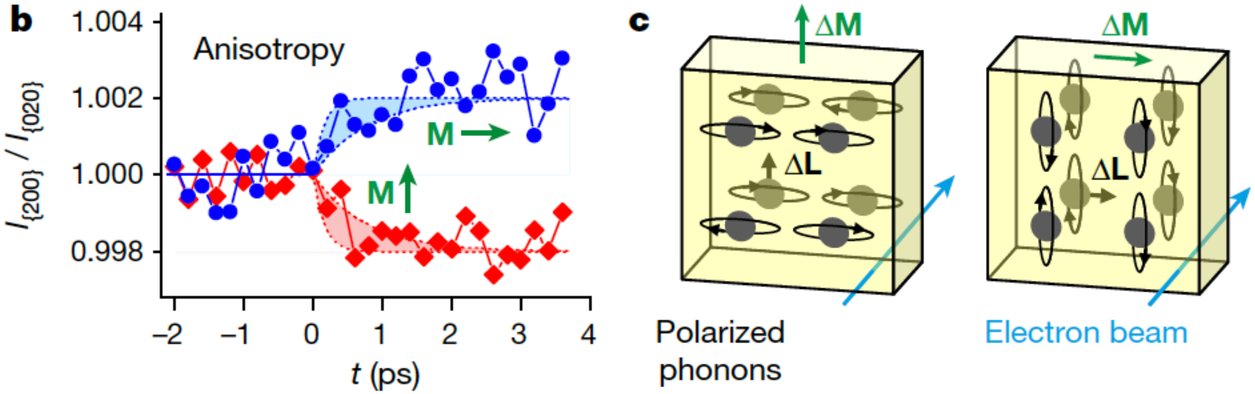
C. Dornes[1]*, Y. Acremann[2], M. Savoini[1], M. Kubli[1], M. J. Neugebauer[1], E. Abreu[1], L. Huber[1], G. Lantz[1], C. A. F. Vaz[3], H. Lemke[4], E. M. Bothschafter[3], M. Porer[3], V. Esposito[3], L. Rettig[3,5], M. Buzzi[3,6], A. Alberca[3], Y. W. Windsor[3,5], P. Beaud[4], U. Staub[3], Diling Zhu[7], Sanghoon Song[7], J. M. Glownia[7] & S. L. Johnson[1,4]*

Nature **565**, 209 (2019).

Polarized phonons carry angular momentum in ultrafast demagnetization

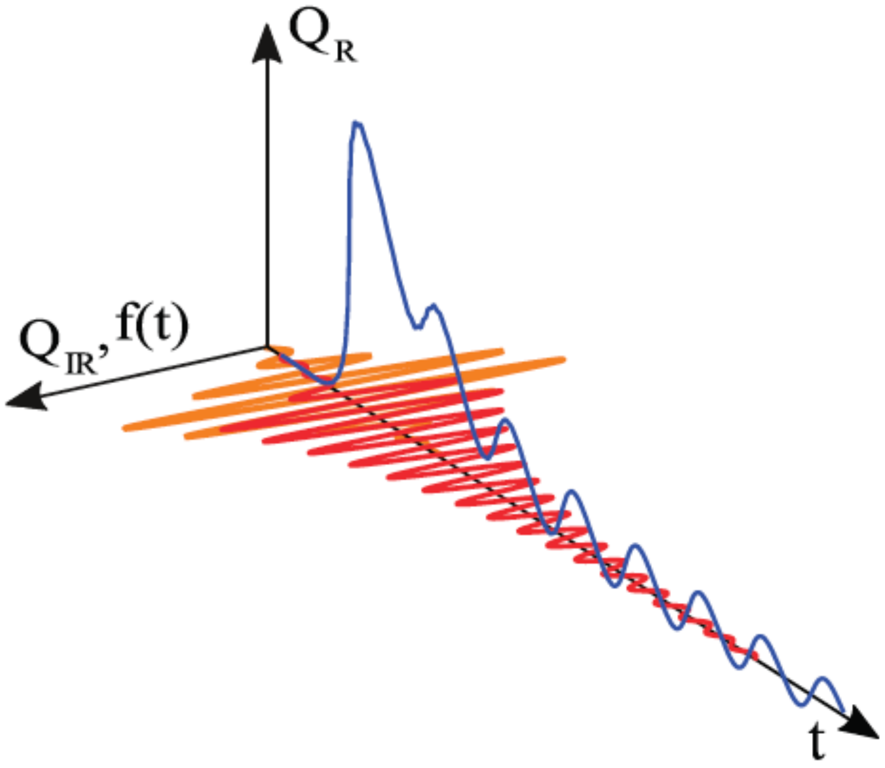
S. R. Tauchert[1,2], M. Volkov[1,2], D. Ehberger[2], D. Kazenwadel[1], M. Evers[1], H. Lange[1], A. Donges[1], A. Book[3], W. Kreuzpaintner[3,4,5], U. Nowak[1] & P. Baum[1,2]✉

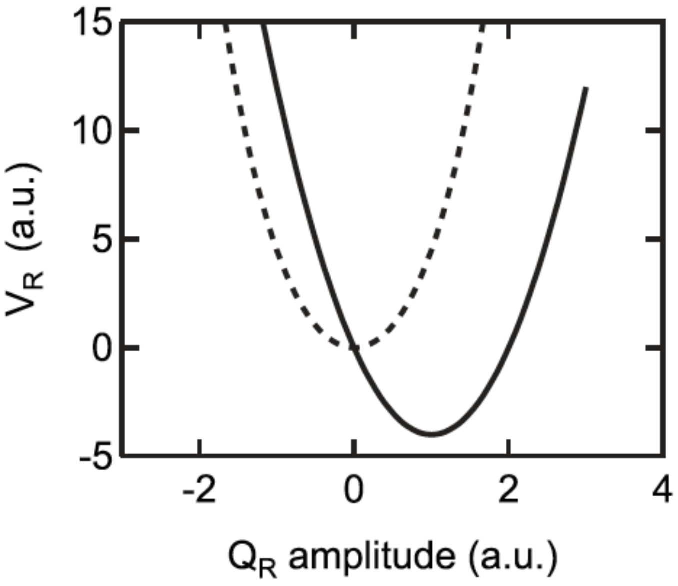
Nature **602**, 73 (2022).



Controlling the lattice (statics and dynamics) can provide full control of magnetism

The idea of 'nonlinear phononics'





Mankowsky et al, Rep. Prog. Phys. **79**, 064503 (2016)
Subedy et al, Phys. Rev. B 89, 220301(R) (2014).

$$\ddot{Q}_{\mathrm{IR}} + 2\gamma_{\mathrm{IR}}\dot{Q}_{\mathrm{IR}} + \omega_{\mathrm{IR}}^2 Q_{\mathrm{IR}} = f(t)$$

$$V_{\mathrm{NL}} = a_{21} Q_{\mathrm{IR}}^2 Q_{\mathrm{R}}$$

shift of Raman mode coordinate breaks symmetry and induces anisotropy

Phono-magnetism

1. “Dynamical multiferroicity”

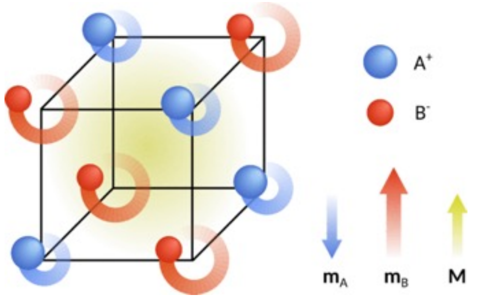
Featured in Physics

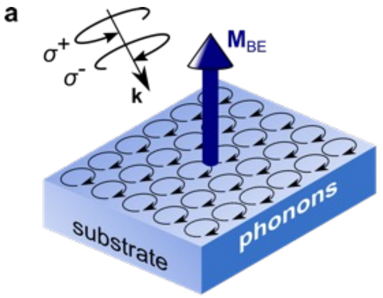
Editors' Suggestion

Dynamical multiferroicity

Dominik M. Juraschek, Michael Fechner, Alexander V. Balatsky, and Nicola A. Spaldin
Phys. Rev. Materials **1**, 014401 – Published 19 June 2017

$$\boldsymbol{M} \propto \boldsymbol{P} \times \frac{\partial \boldsymbol{P}}{\partial t}$$





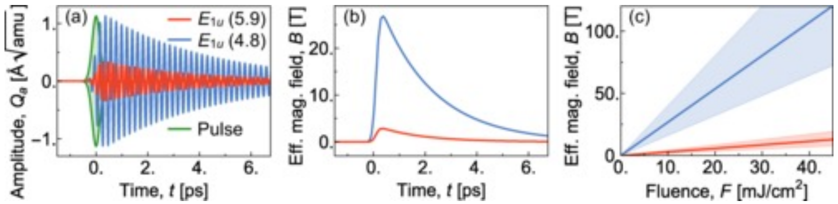
2. Modify crystal electric field → “spin-orbit”-like interaction

Open Access

Giant effective magnetic fields from optically driven chiral phonons in 4f paramagnets

Dominik M. Juraschek, Tomáš Neuman, and Prineha Narang
Phys. Rev. Research **4**, 013129 – Published 17 February 2022

$$H^{ph-sp} = K\boldsymbol{m} \cdot \boldsymbol{Q} \times \frac{\partial \boldsymbol{Q}}{\partial t}$$

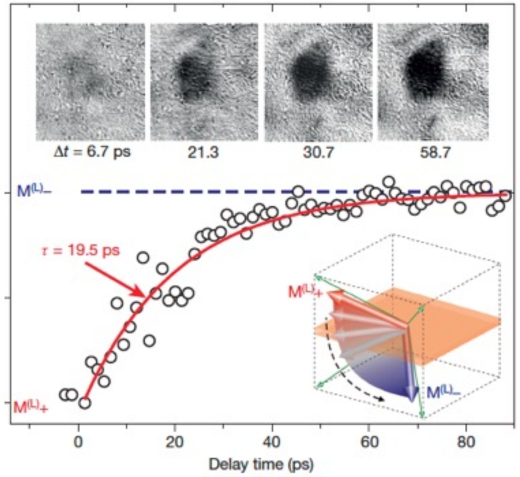


$$B = \frac{\partial H^{ph-sp}}{\partial \boldsymbol{m}}$$

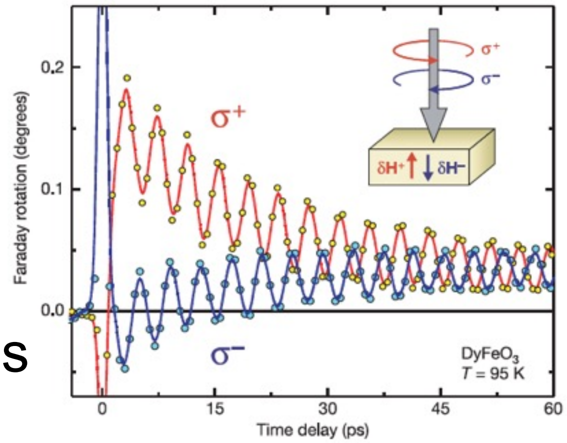
Nonlinear phononics vs phono-magnetism

- **Nonlinear phononics:** stimulus changes the equilibrium, magnetization follows: **displacive effect**
  - for example, light-induced magnetic anisotropy
  - limited by the life-time of the excited states

Stupakiewicz *et al*, Nature (2017)



- **Phono-magnetism**: stimulus changes the magnetization directly: **impulsive effect**
  - for example, inverse Faraday effect*
  - limited by the pulse width & coherence of excited states



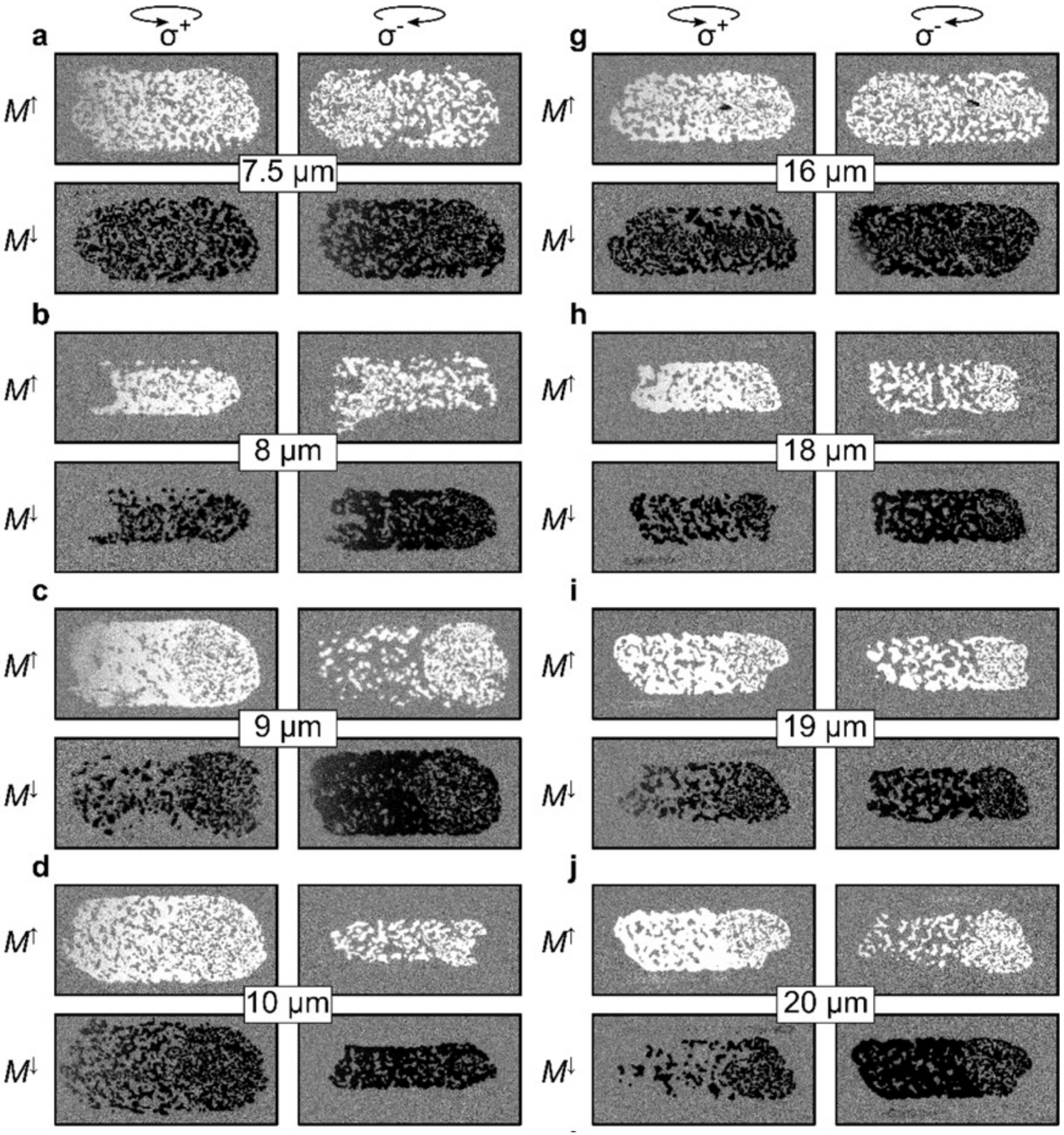
*circular-polarized light: $\mathbf{H}(0) = \frac{\varepsilon_0}{\mu_0}\alpha\left[\mathbf{E}(\omega) \times \mathbf{E}^*(\omega)\right]$

Kimel *et al*, Nature (2005)

GdFeCo/SiO_2

Samples:

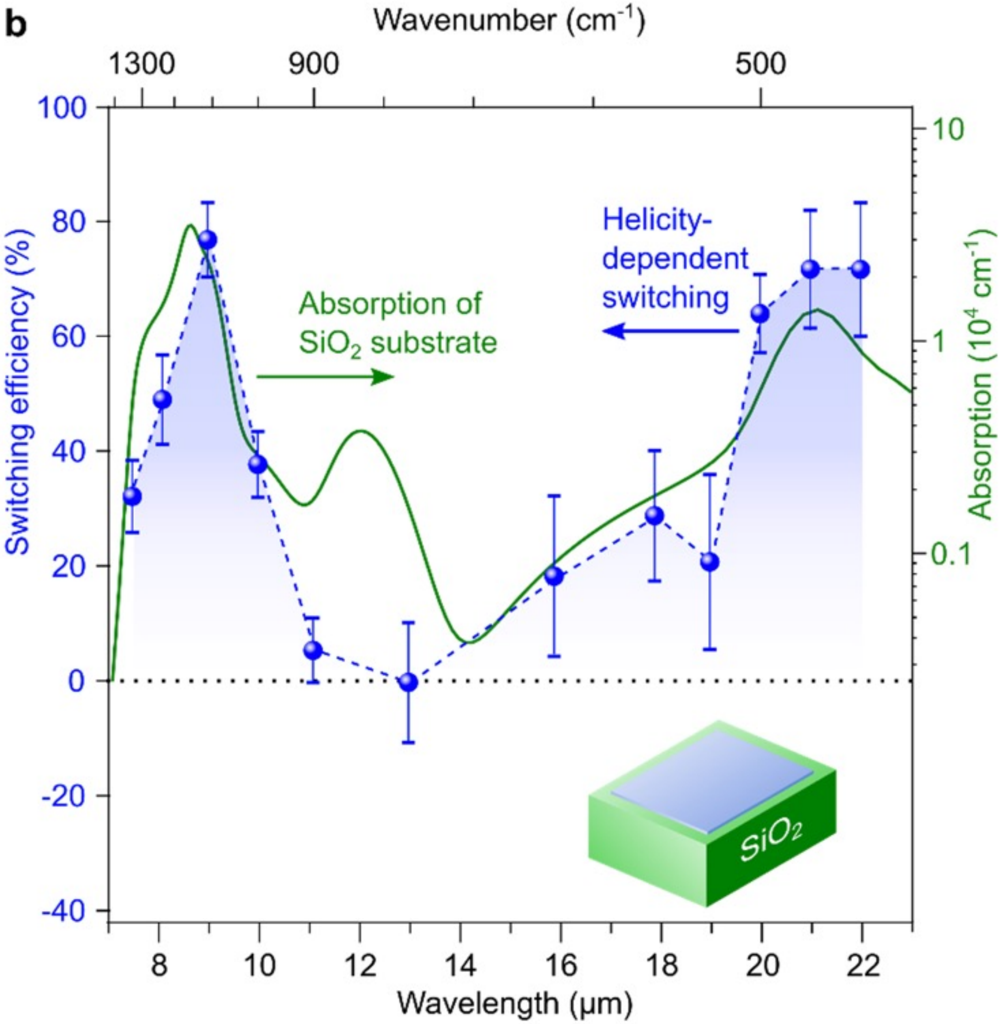
Si_3N_4 (60nm)
$Gd_{24}(FeCo)_{76}$ (20nm)
Si_3N_4 (5nm)
Fused silica (170µm)



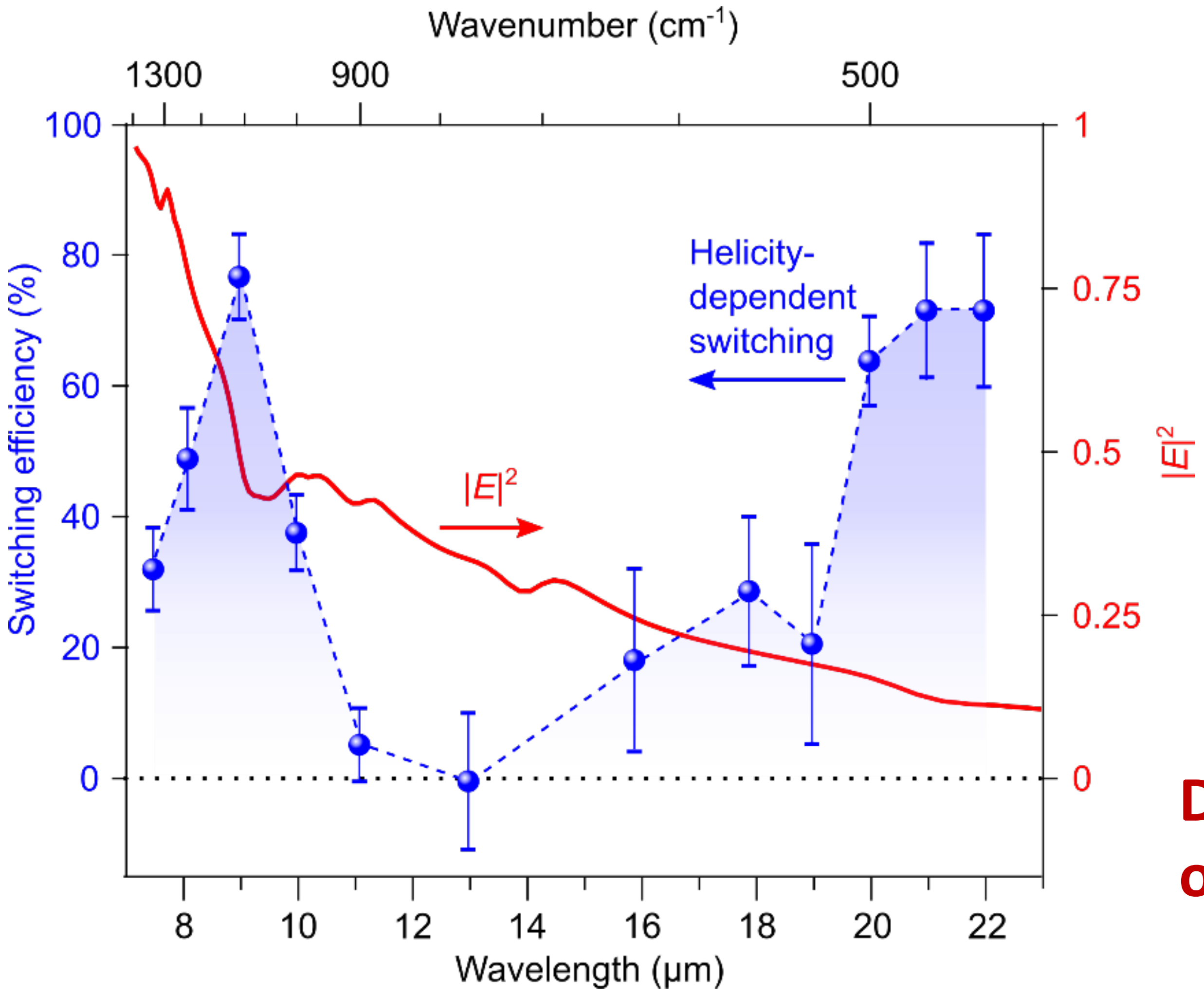
HD-AOS in GdFeCo

C.S. Davies et al, arXiv:2305.11551 (2023).

switching matches TO phonon modes



Absorption in the GdFeCo layer?

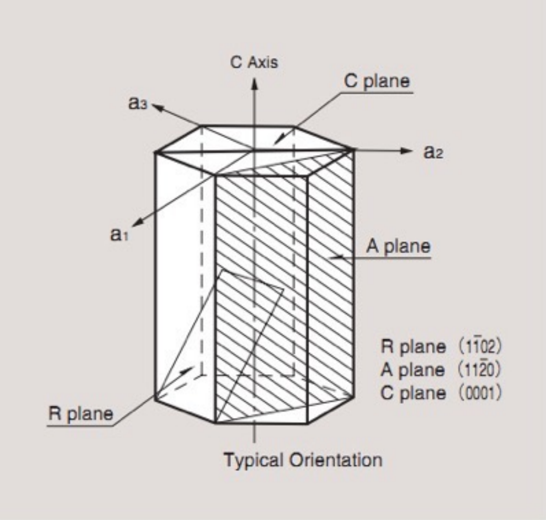


Does this actually depend on the substrate?

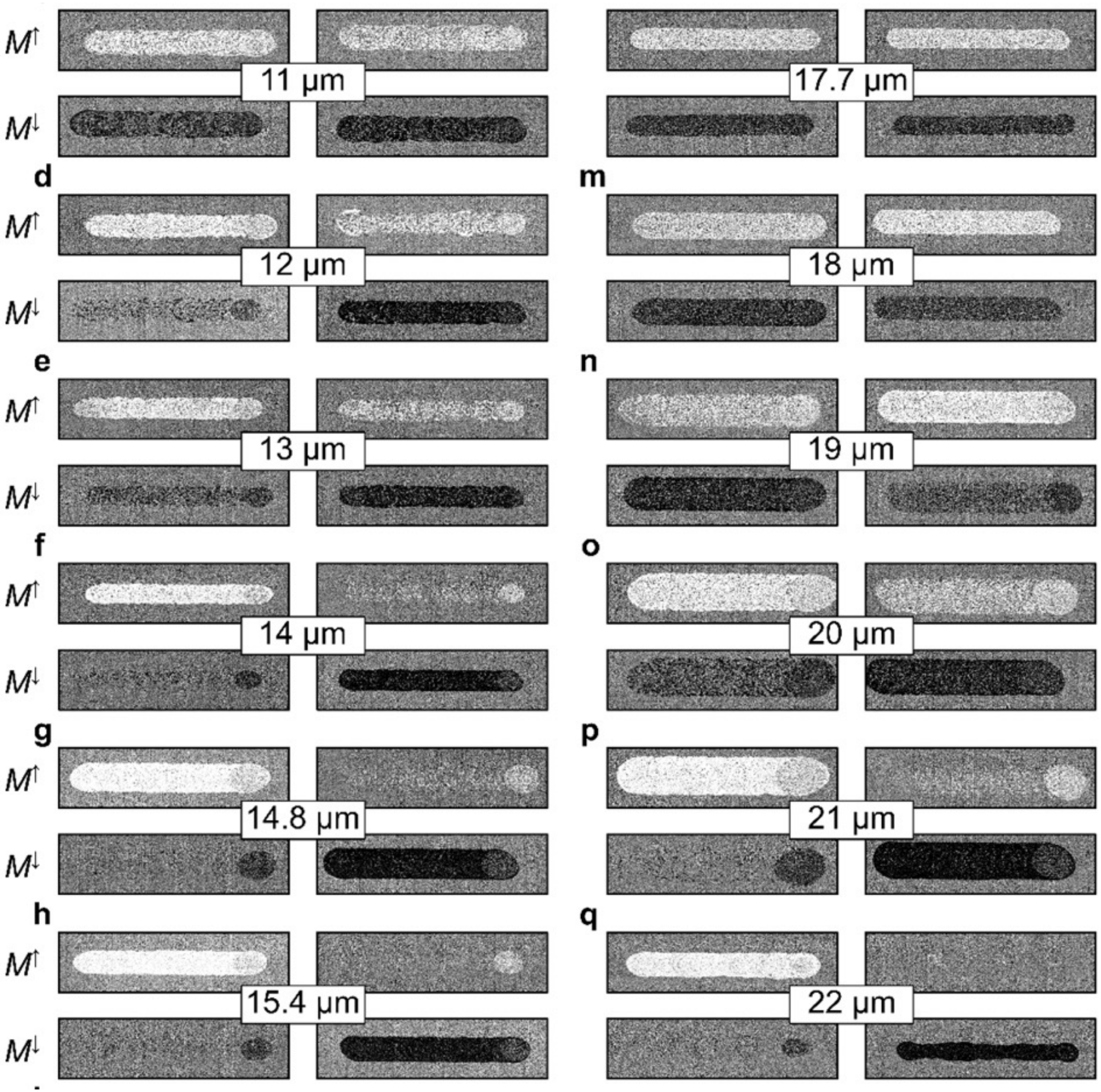
GdFeCo/Al_2O_3

Samples:

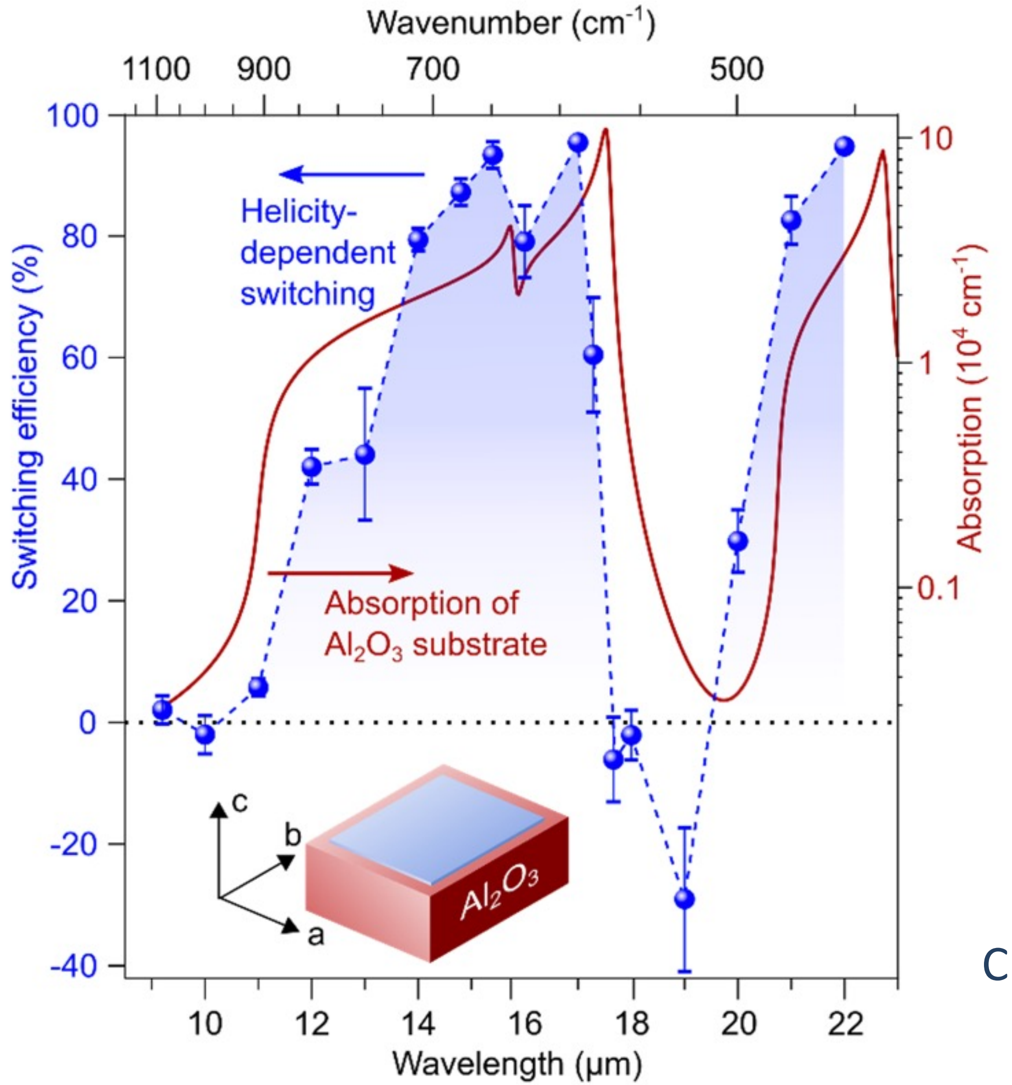
Si_3N_4 (60nm)
$Gd_{24}(FeCo)_{76}$ (20nm)
Si_3N_4 (5nm)
Sapphire (c-cut)



Unit Cell of Sapphire



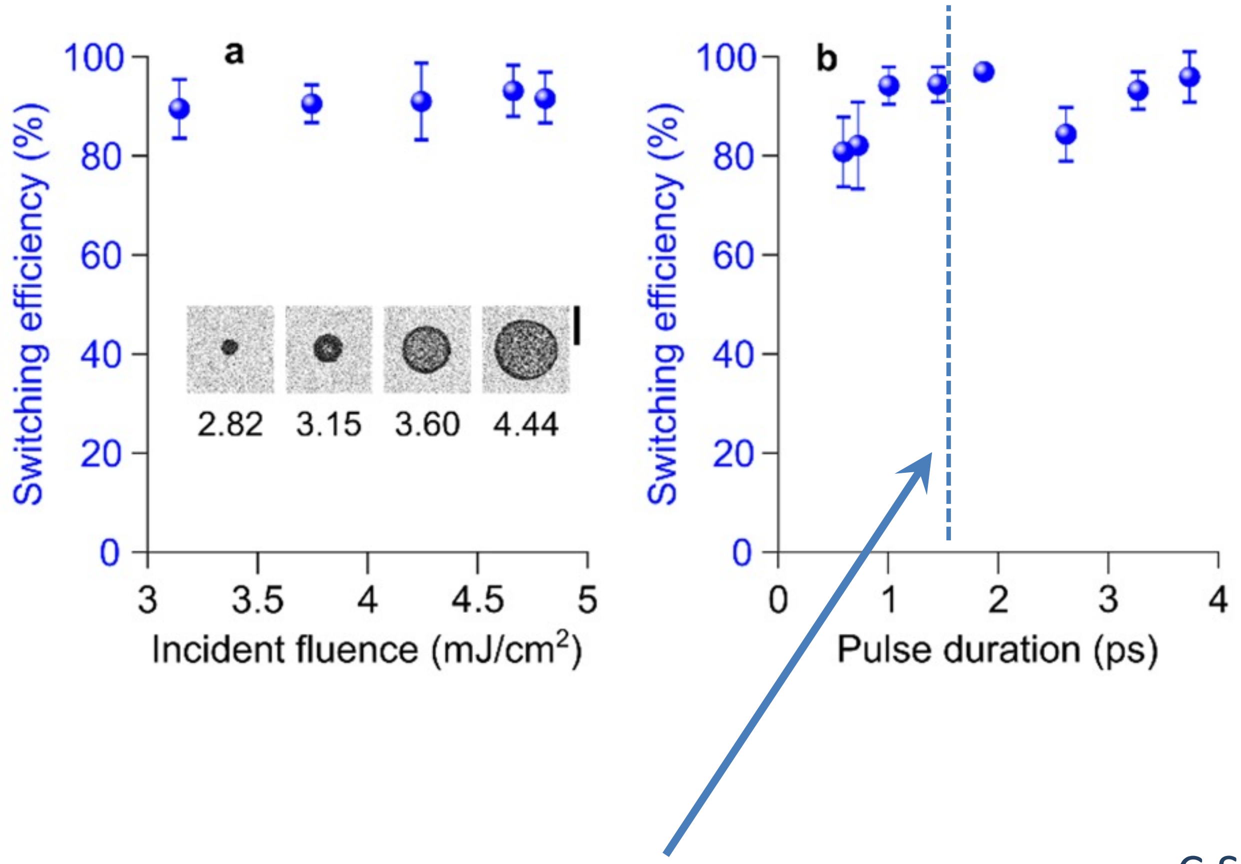
Also here a good match with TO phonons of the substrate!



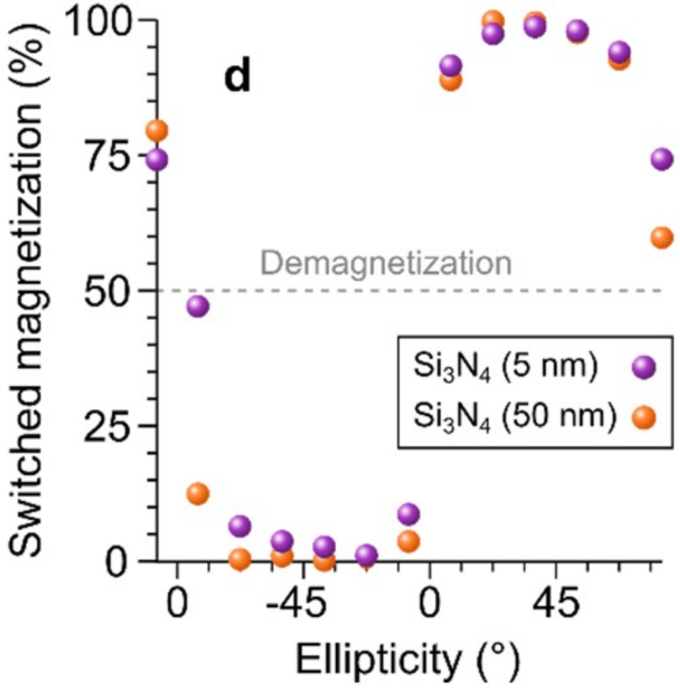
Excitation of circularly polarized phonons in ionic substrate induces magneetic fields that drive the reversal.

C.S. Davies et al, arXiv:2305.11551 (2023).

Switching is very robust (in particular on Al_2O_3 substrate)



limit toggle switching



(angle of λ/4 plate)

C.S. Davies et al, arXiv:2305.11551 (2023).

The interaction mechanism?

Still an open question… Two partial answers may already be here

1. “Dynamical multiferroicity”

Featured in Physics Editors' Suggestion

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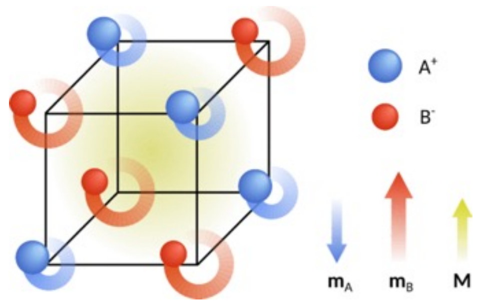

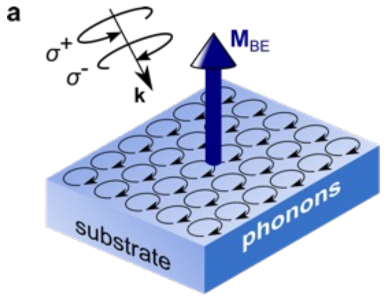

Probably a B-field!

2. Modify crystal electric field → “spin-orbit”-like interaction

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Giant effective magnetic fields from optically driven chiral phonons in $4f$ paramagnets

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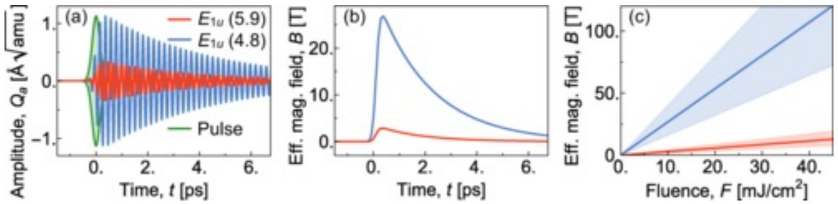
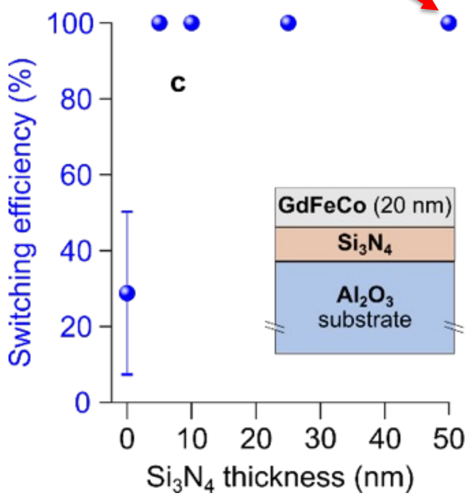
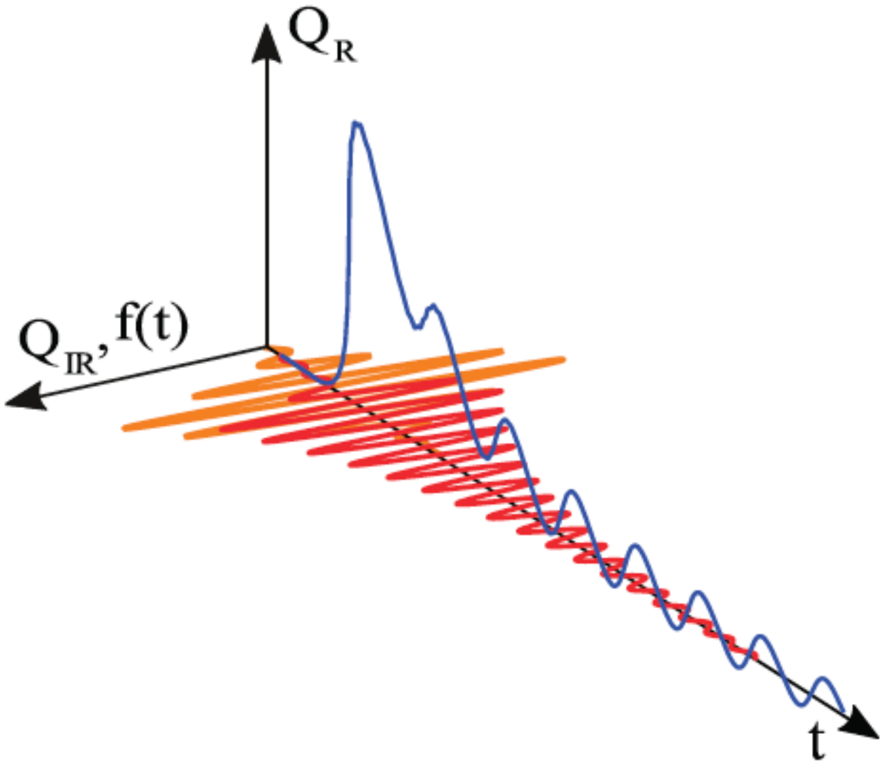
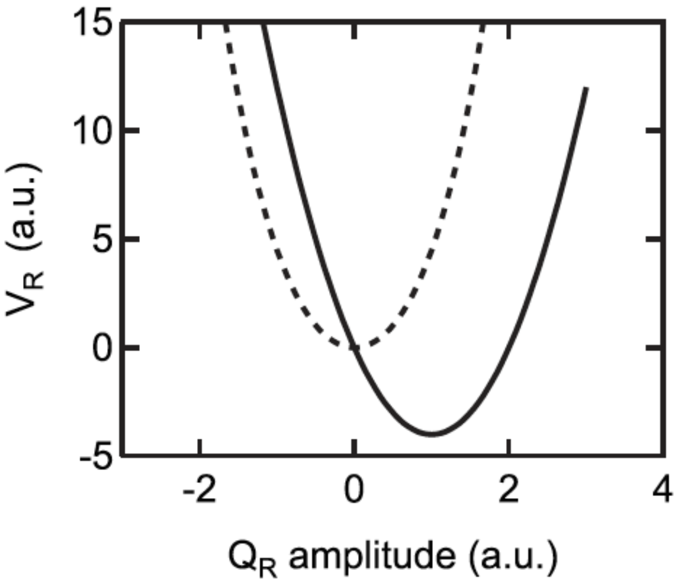


$$B = \frac{\partial H^{ph-sp}}{\partial \boldsymbol{m}}$$



Back to ‘nonlinear phononics’





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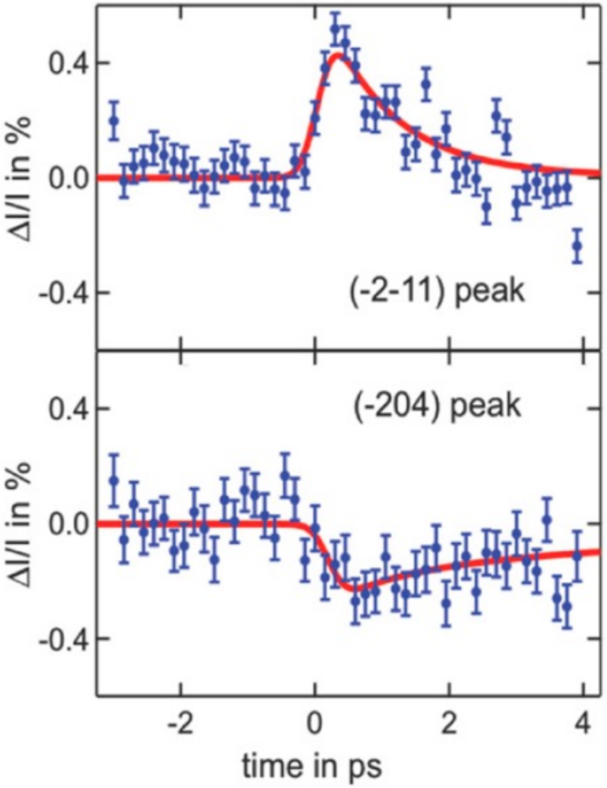
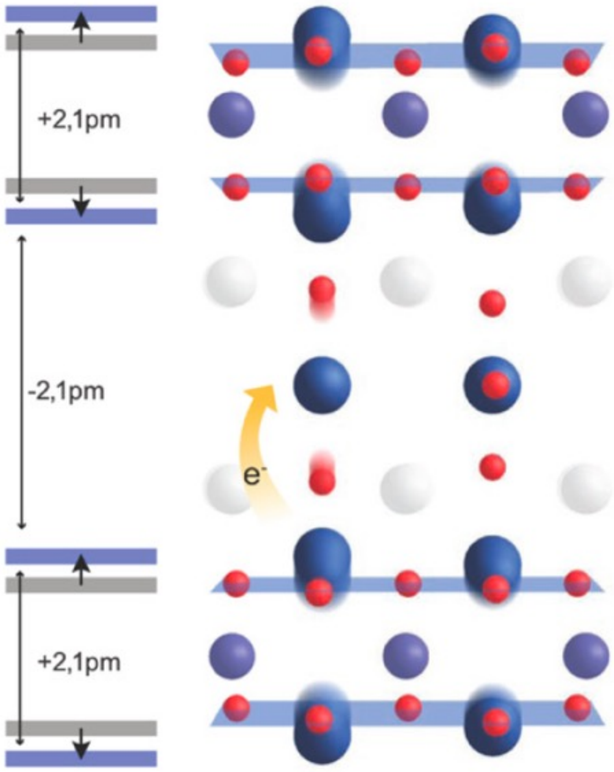
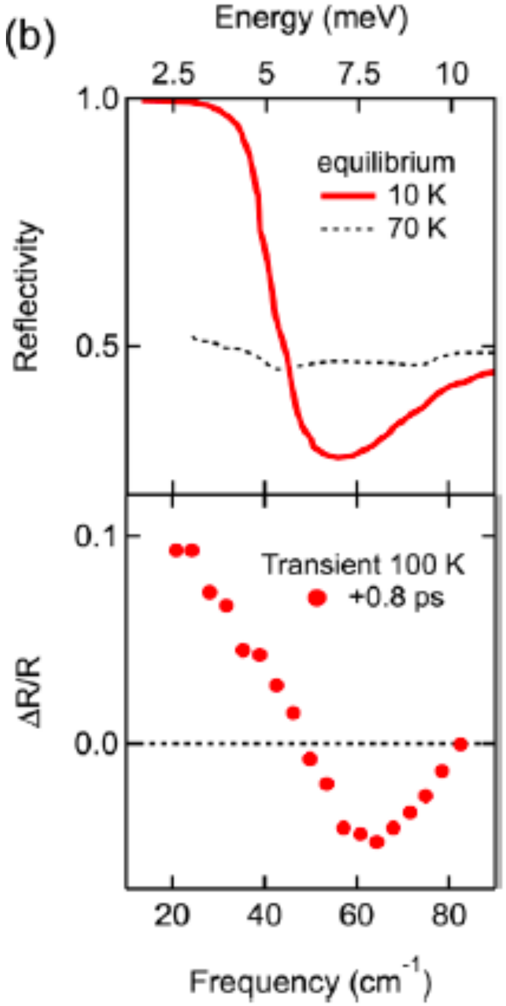
$$V_{NL} = a_{21} Q_{IR}^2 Q_R$$

shift of Raman mode coordinate breaks symmetry and induces anisotropy

Change of equilibrium: enhanced superconductivity in $YBa_2Cu_3O_{6.5}$





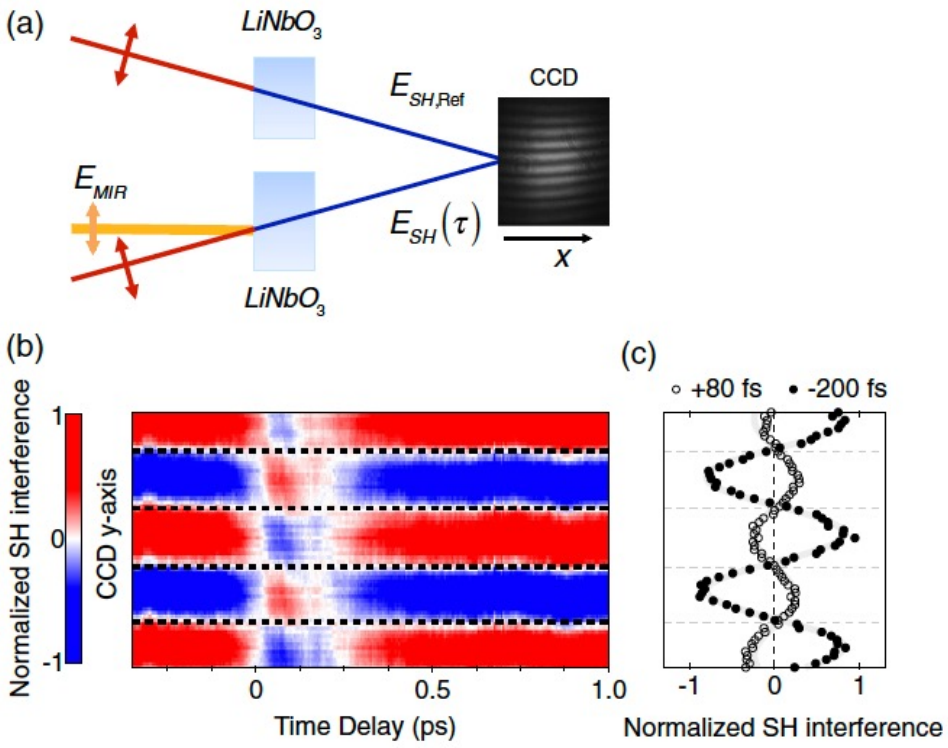


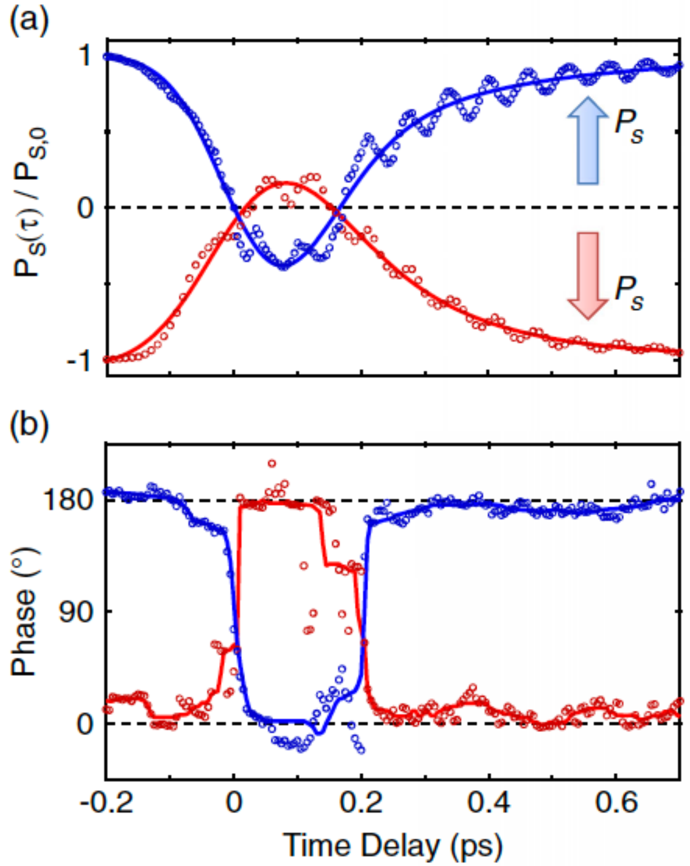
Mankowsky et al, *Nonlinear lattice dynamics as a basis for enhanced superconductivity in* $YBa_2Cu_3O_{6.5}$, Nature **516**, 71 (2014).

PRL **118,** 197601 (2017) PHYSICAL REVIEW LETTERS week ending 12 MAY 2017

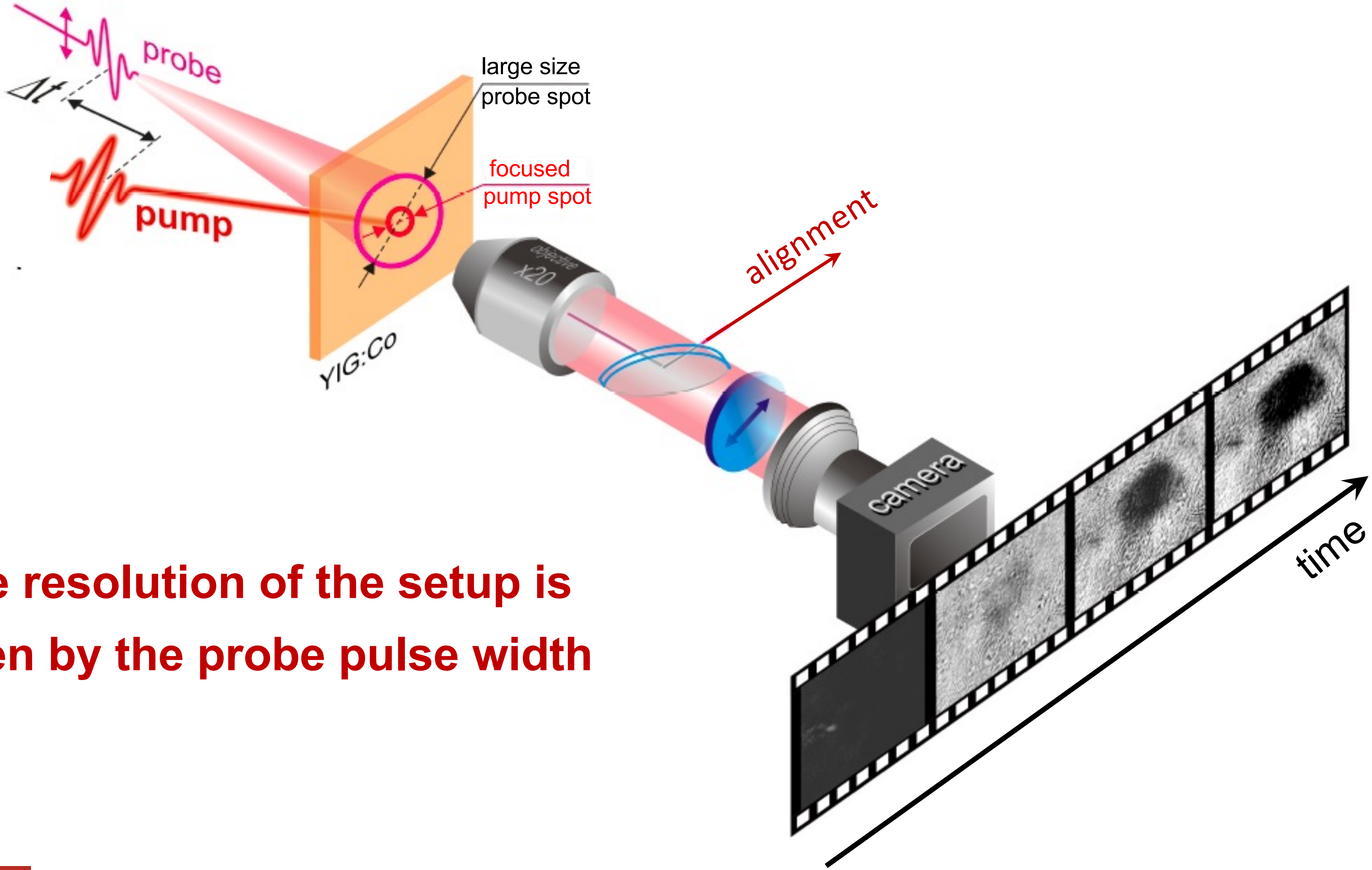
Ultrafast Reversal of the Ferroelectric Polarization

R. Mankowsky,[1] A. von Hoegen,[1] M. Först,[1] and A. Cavalleri[1,2]





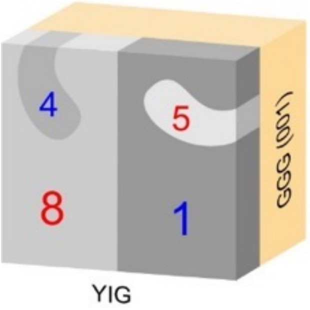
Our experimental approach: single shot imaging for the detection

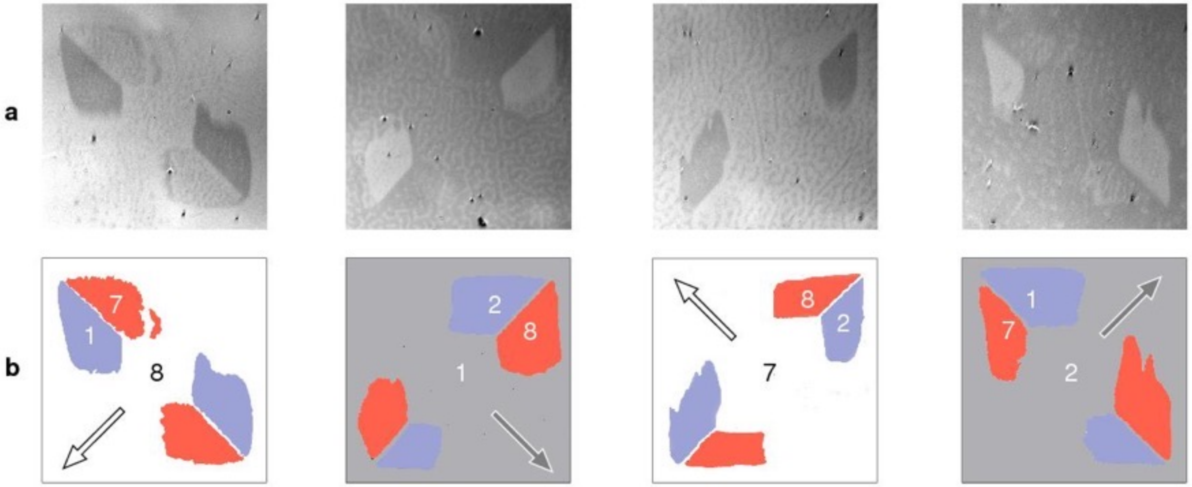


time resolution of the setup is given by the probe pulse width

IR-pulse induced switching in magnetic garnet (Co-doped)

- Polarizing microscope





- Single micropulse
  ($\lambda = 14\ \mu m$, $\tau \approx 1$ ps)

1 pulse, 10 pulses, 100's of pulses…

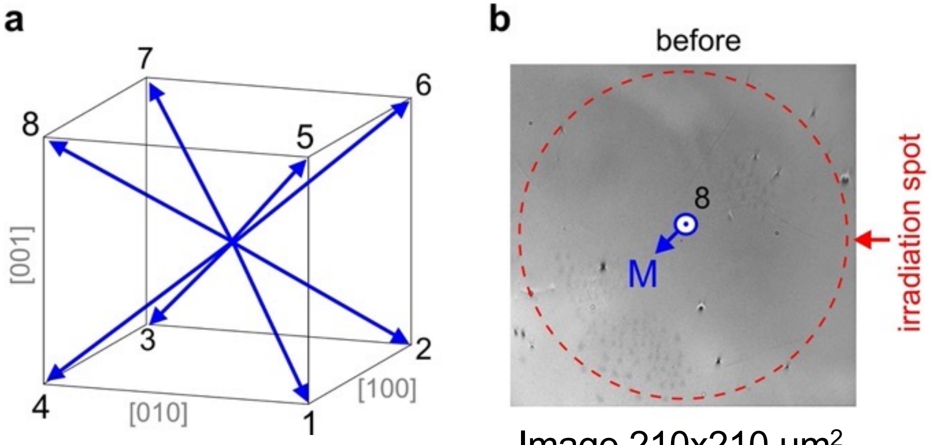


Image 210x210 μm^2

A. Stupakiewicz et al, Nature Physics **17**, 489 (2021)

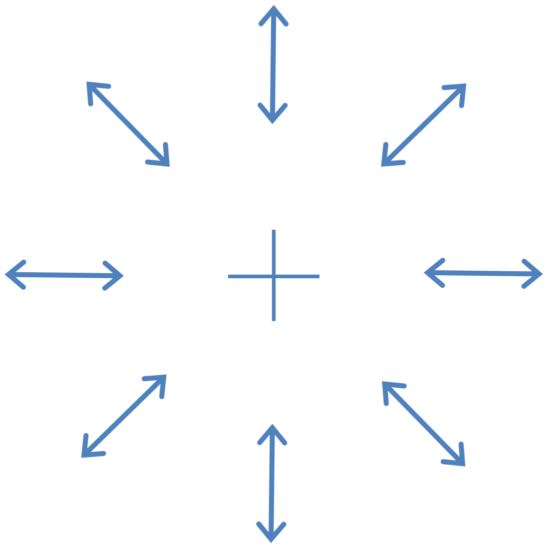
Spatial distribution of strain in a Gaussian beam profile

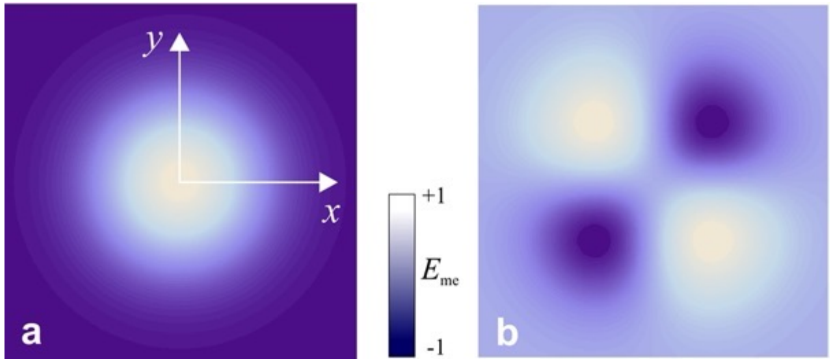
Assume the induced in-plane shift of potential ***u*** has a 2D Gaussian profile

$$u = \frac{Aqa^2}{3}\frac{1+\sigma}{1-\sigma}\left[\frac{1}{r}\left(1-e^{-\frac{r^2}{2a^2}}\right)+(1-2\sigma)\frac{r}{R^2}e^{-\frac{R^2}{2a^2}}\right]$$

Induced strain → $\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial \boldsymbol{u}_i}{\partial x_j}+\frac{\partial \boldsymbol{u}_j}{\partial x_i}\right)$

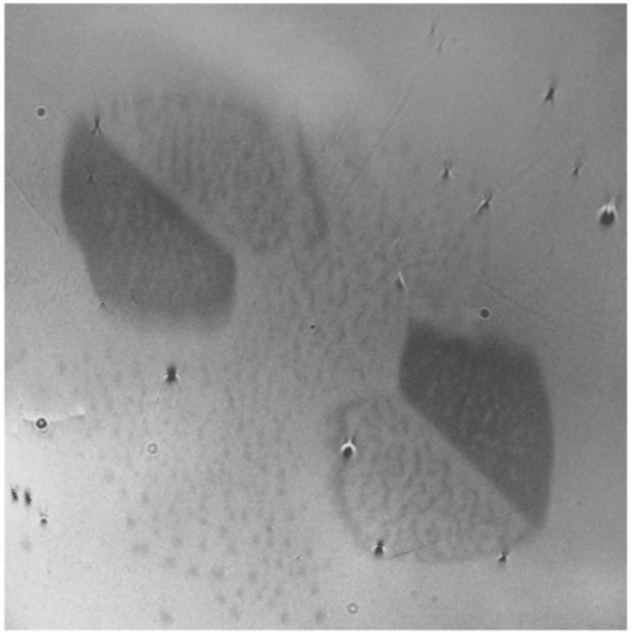
$$E_{me} = b_1\left(\varepsilon_{xx}m_x^2+\varepsilon_{yy}m_y^2\right)+2b_2\varepsilon_{xy}m_x m_y$$

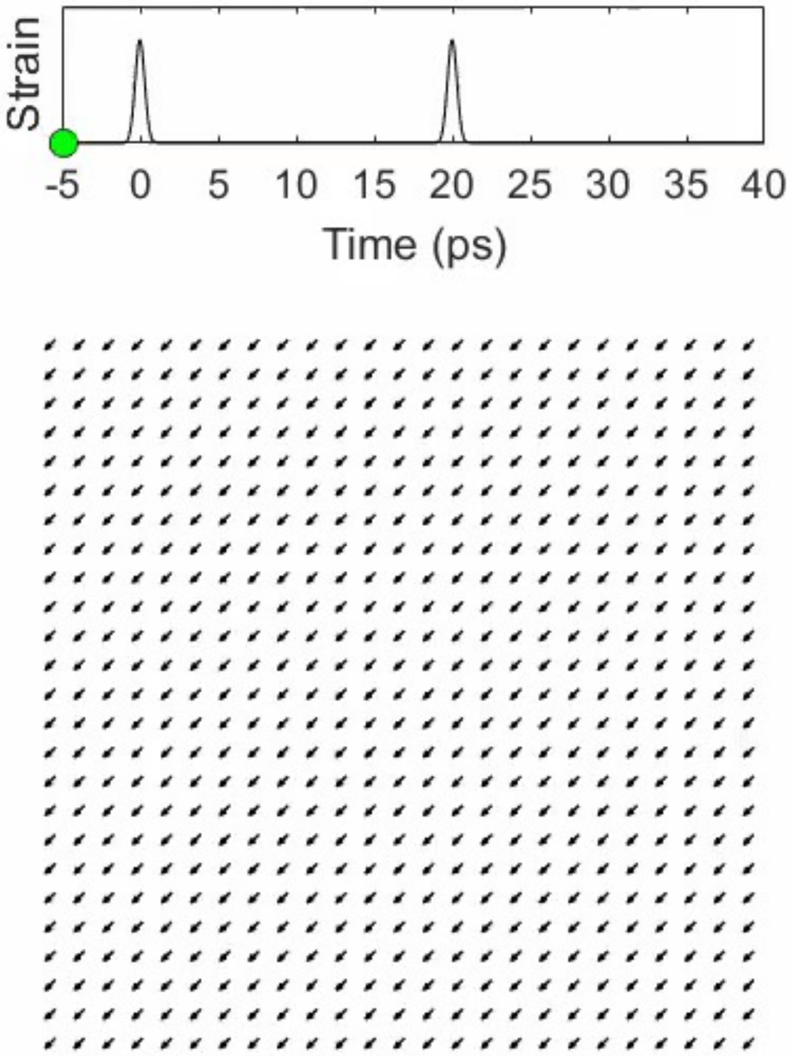




L.D. Landau and E.M. Lifshitz, *Course of Theoretical Physics, vol. 7: Theory of Elasticity (3rd ed.)* (Elsevier, Amsterdam, 1986).

Micromagnetic simulations of the reversal

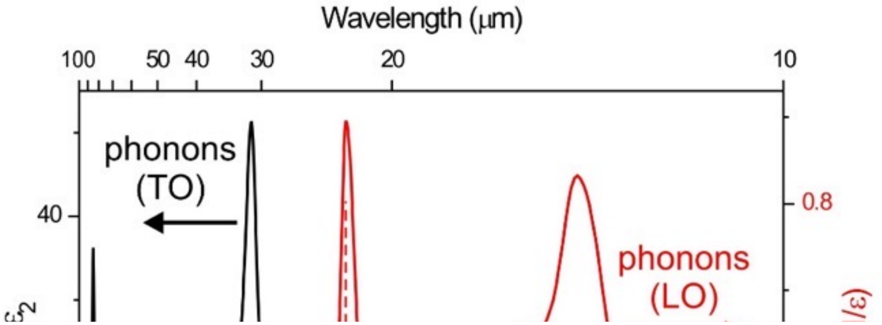


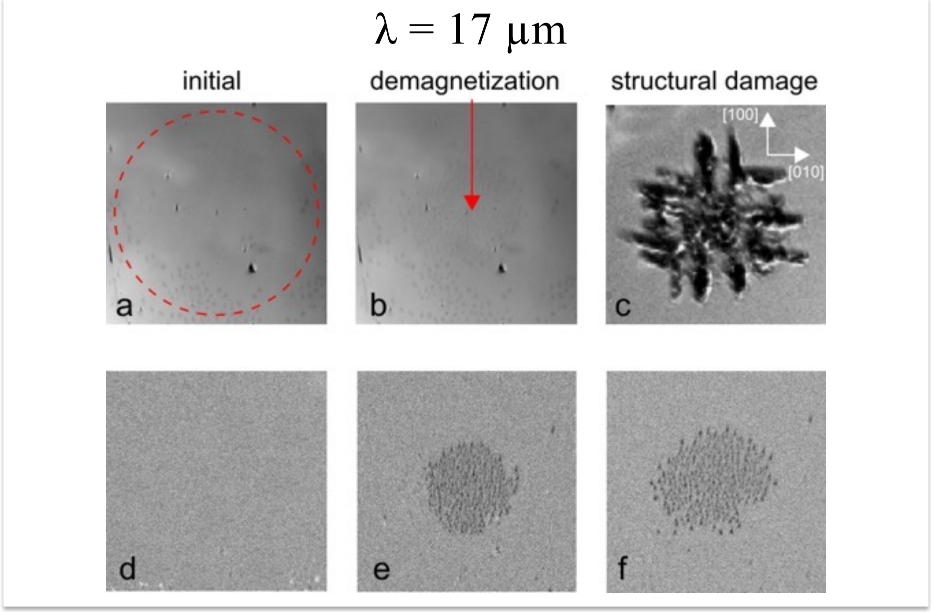


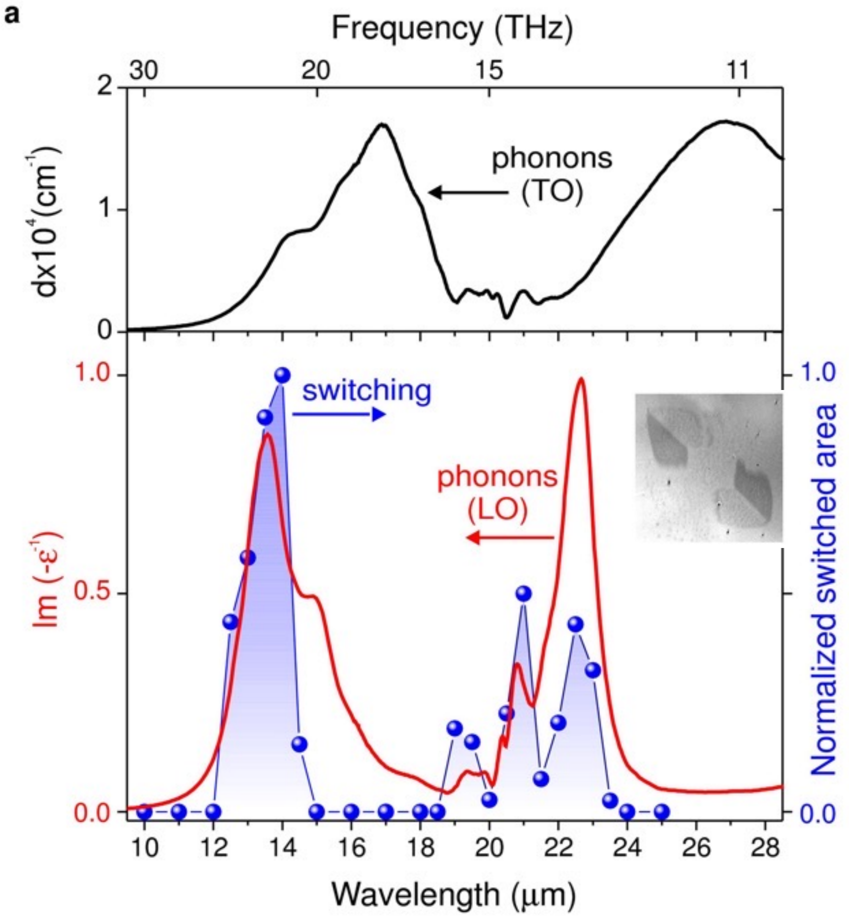
We saw the same switching pattern after multiple pulses… How come?

Spectral dependence of switching

Phonon spectra measured with infrared spectroscopic ellipsometry (MPI Stuttgart)



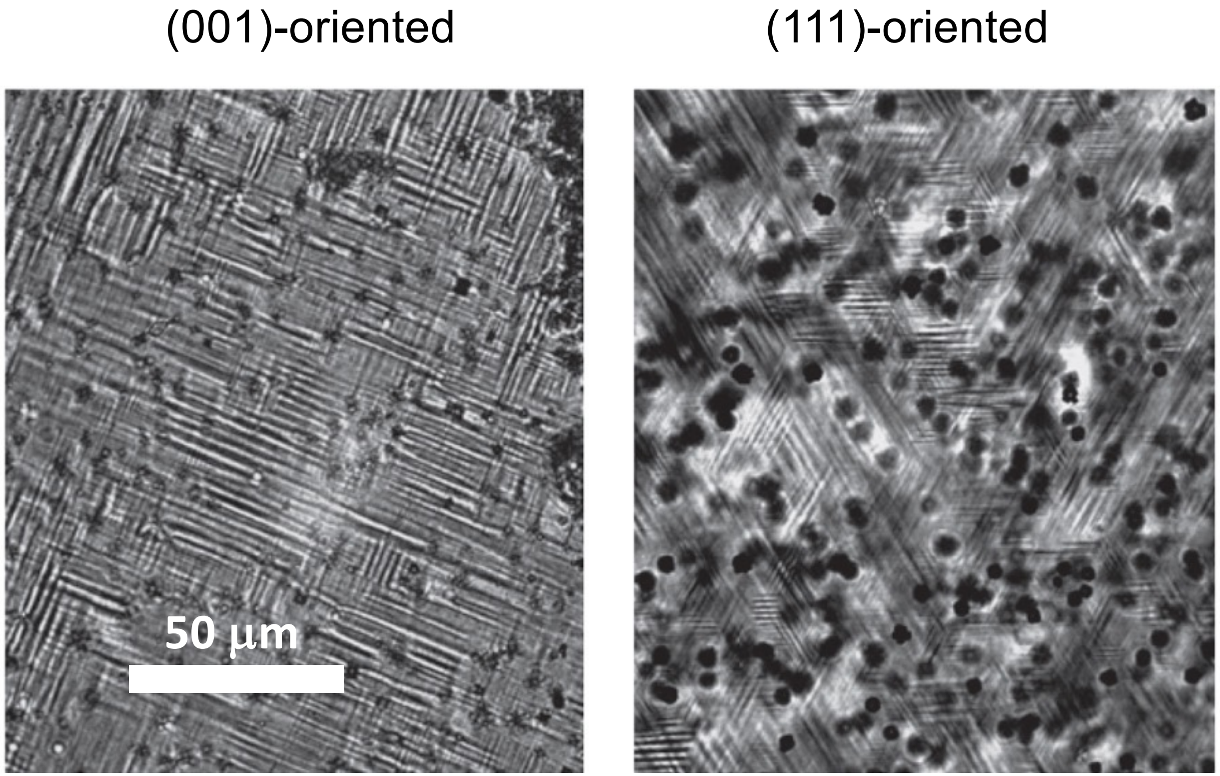




Good correlation between the spectral dependence of **longitudinal** phonons and the switching

How universal is such switching?

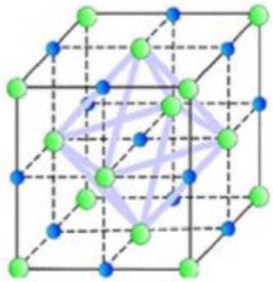
Antiferromagnetic NiO



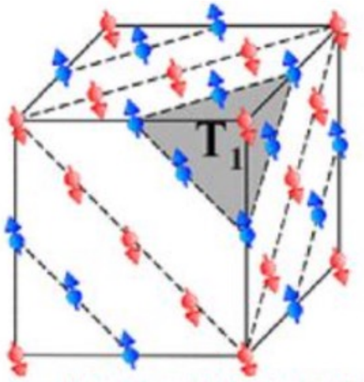
bulk single crystals, 0.5 mm thick

T-domains are easily visible through linear magnetic birefringence

Phonons in NiO

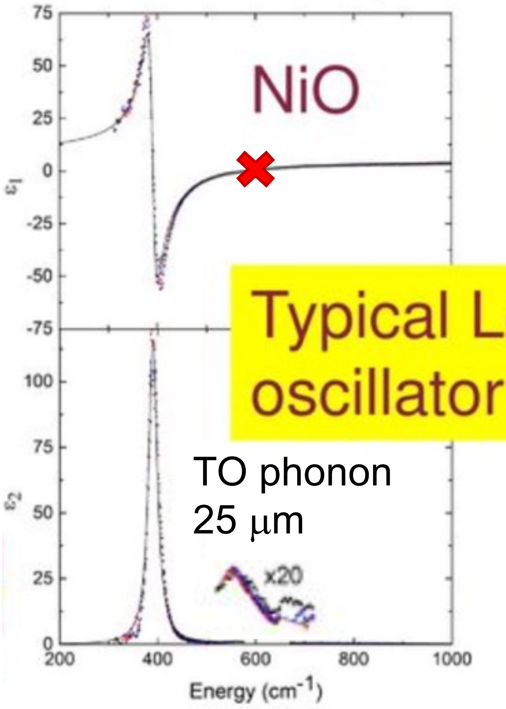


NiO or NaCl:
Rocksalt lattice



Néel temperature $T_N = 523$ K

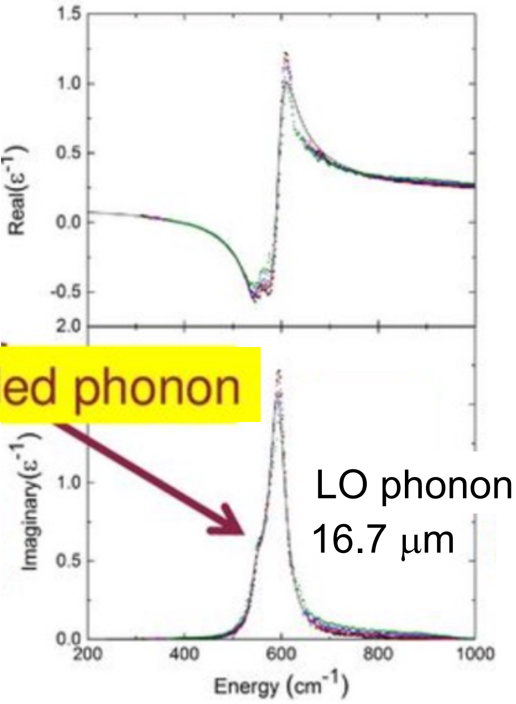
small rhombohedral distortion;
some zone-folding effects



Typical Lorentz oscillator

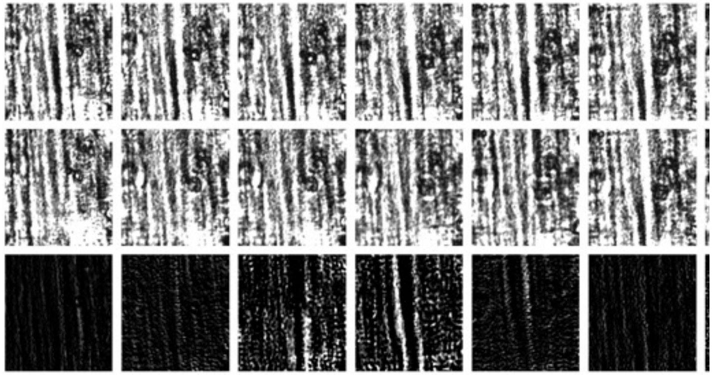
TO phonon
25 µm

Zone-folded phonon



LO phonon
16.7 µm

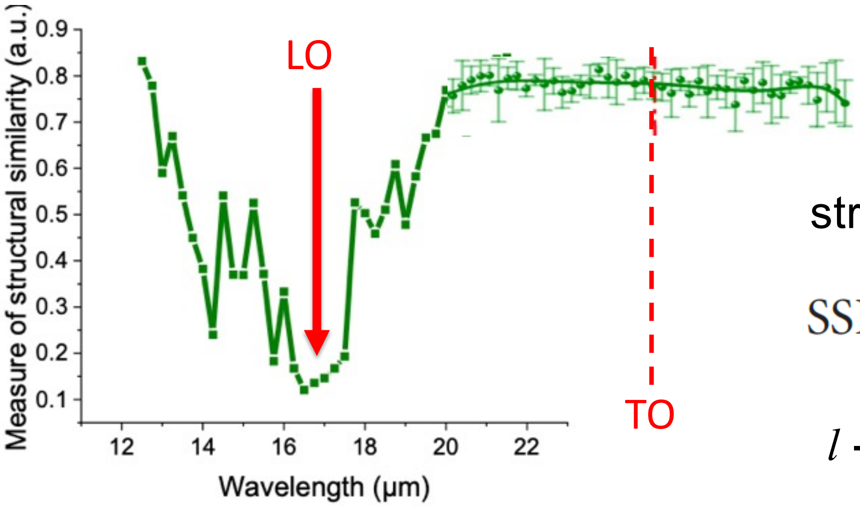
Modification of domain structure in antiferromagnetic NiO



before

after

difference



structural similarity index measure (SSIM)

$$\mathrm{SSIM}(\mathbf{x},\mathbf{y}) = [l(\mathbf{x},\mathbf{y})]^{\alpha} \cdot [c(\mathbf{x},\mathbf{y})]^{\beta} \cdot [s(\mathbf{x},\mathbf{y})]^{\gamma}$$

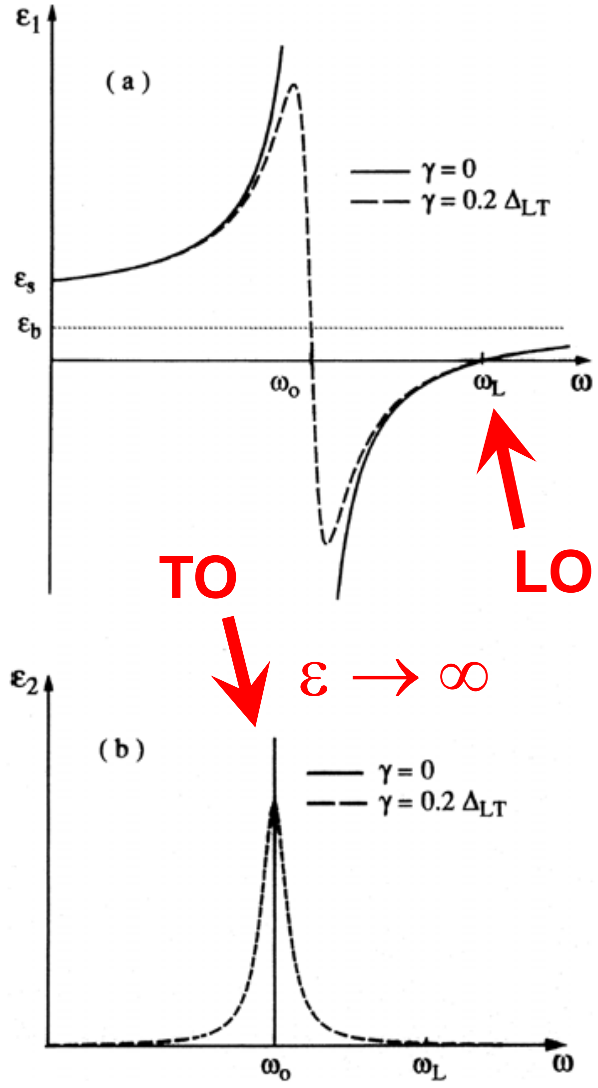
l - luminance, c – contrast, s - structure

P. Stremoukhov et al, New J. Physics **24**, 023009 (2022).

Mechanism?

- it looks **very universal**: (almost) everything can be switched by excitation at the LO phonon frequency
- however, **LO phonon is not IR active**
- so how come it works better than the TO one??

Transverse and longitudinal optical phonons



TO

$\varepsilon \to \infty$

LO

$\varepsilon = 0$

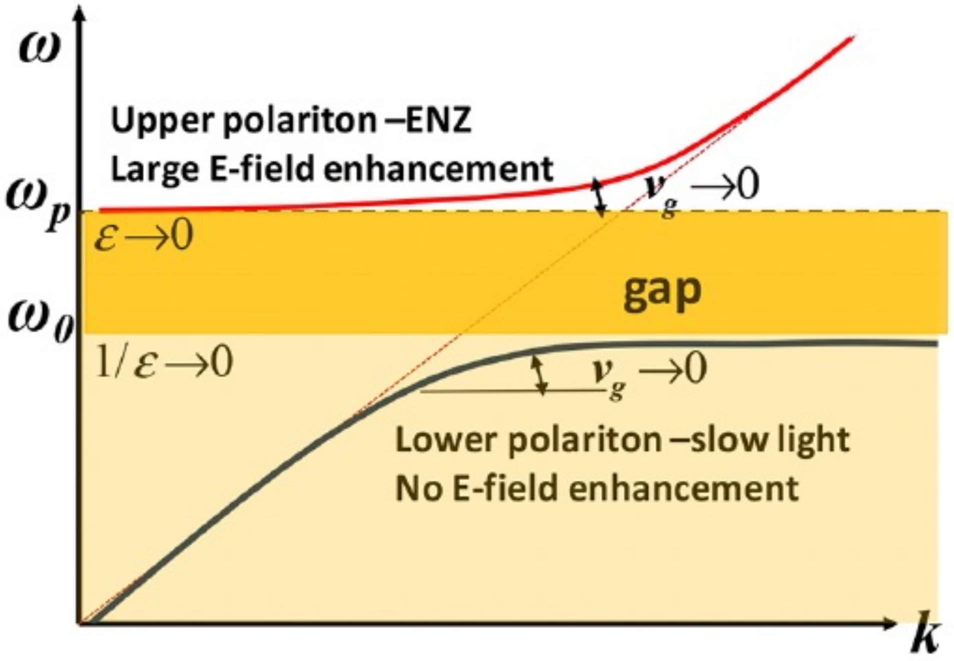
Lyddane-Sachs-Teller relation

$$\frac{\varepsilon(0)}{\varepsilon(\infty)} = \frac{\omega_{\mathrm{LO}}^2}{\omega_{\mathrm{TO}}^2}.$$

microscopically, it is the same phonon,

but not macroscopically!

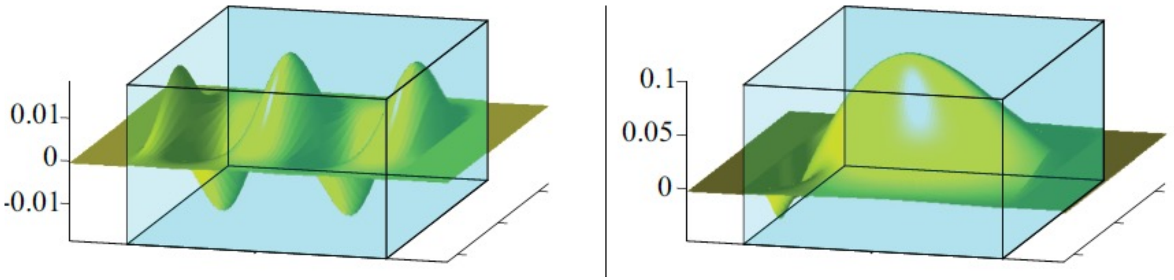
Enhancement of light-matter interaction at ε-near-zero condition



- ➢ infinite wavelength ('static' optics) => no zero's
- ➢ enhancement of optical field
- ➢ zero group velocity => long interaction time
- ➢ strong nonlinearity of pulse propagation
- ➢ transverse wave is OK

Kinsey & Khurgin,
Opt. Mater. Express **9**, 2793 (2019).

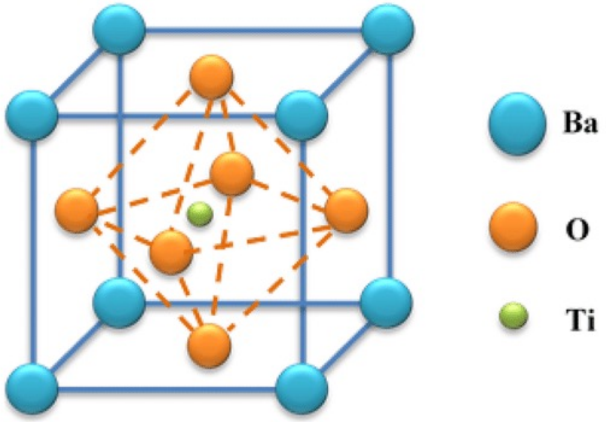
$$D \neq \varepsilon E$$



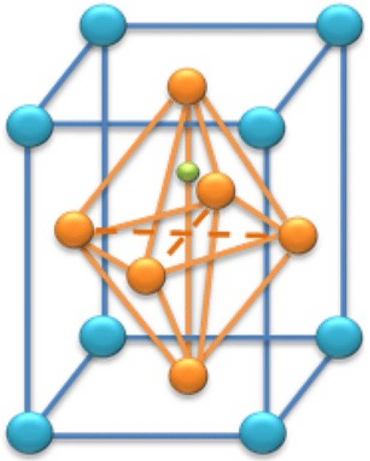
Ciattoni et al, Laser Phot. Reviews **10**, 517 (2016).

Ferroelectric $BaTiO_3$

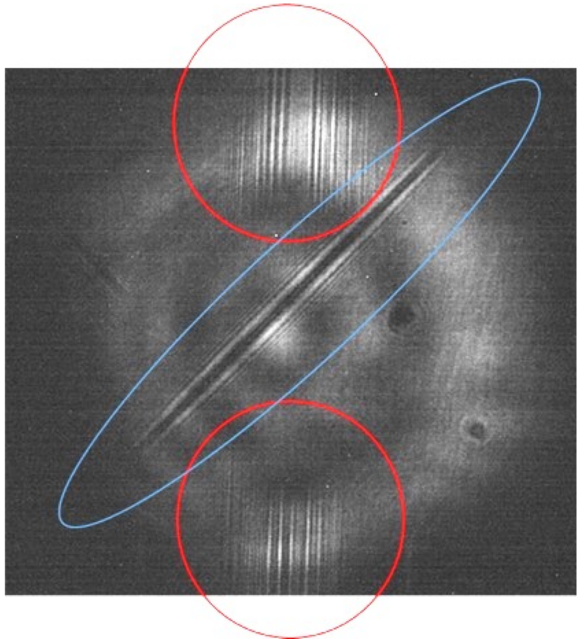
M. Kwaaitaal et al, to be published



paraelectric phase



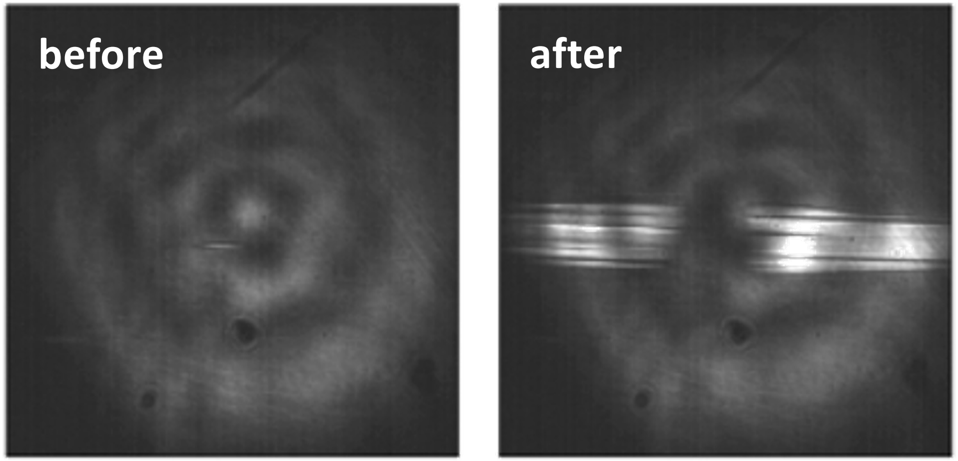
ferroelectric phase



180° domains
90° domains

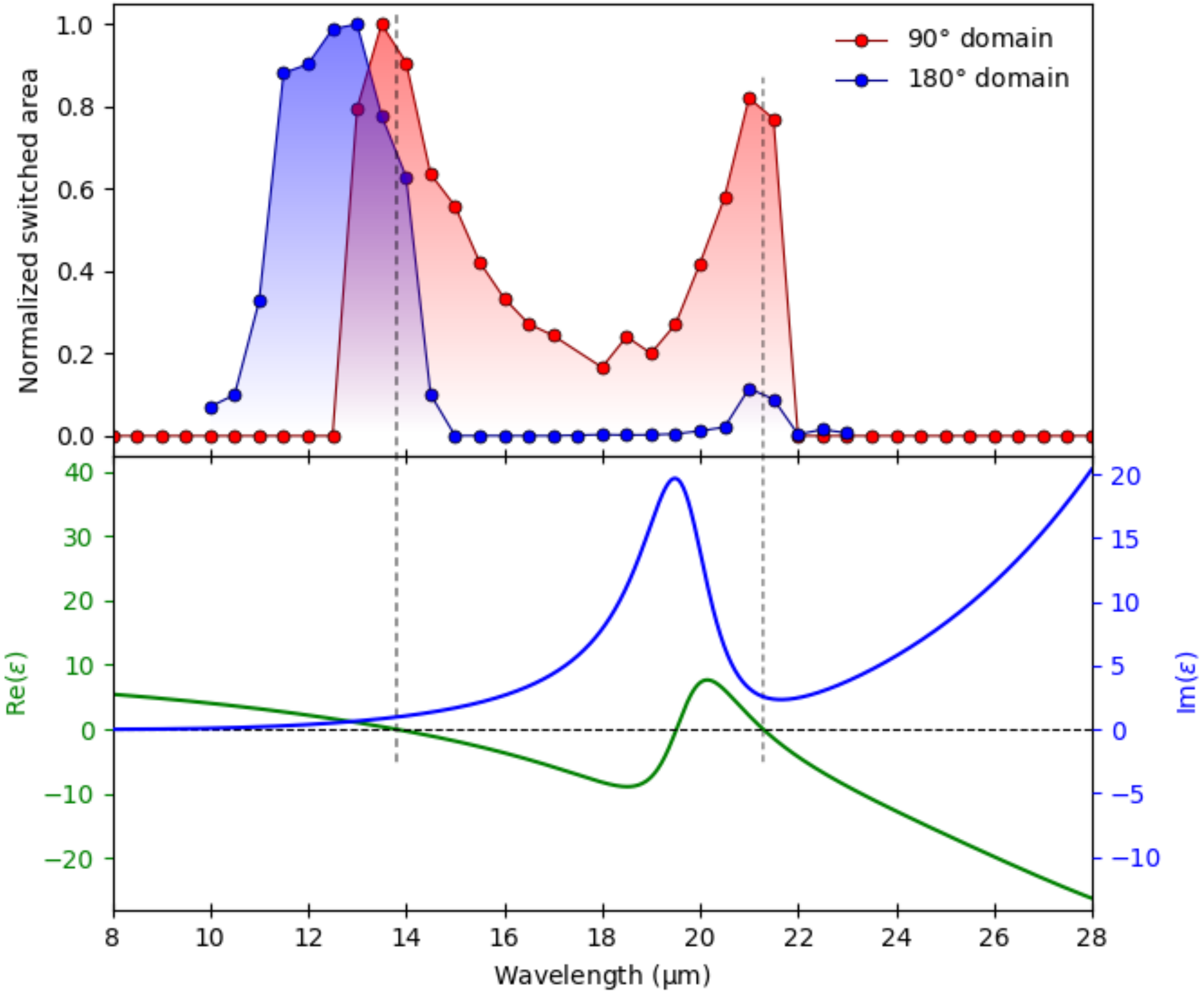
1 shot at 13 μm wavelength

polarization in-plane



180° domains

Again, epsilon-near-zero effect is obvious!



Kwaaitaal et al,
arXiv:2305.11714 (2023)

Nonlinear phononics, IR-resonant Raman scattering, or what?

PHYSICAL REVIEW X **11,** 021067 (2021)

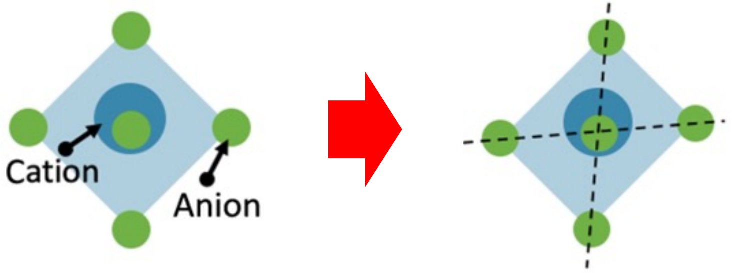
Ultrafast Control of Material Optical Properties via the Infrared Resonant Raman Effect

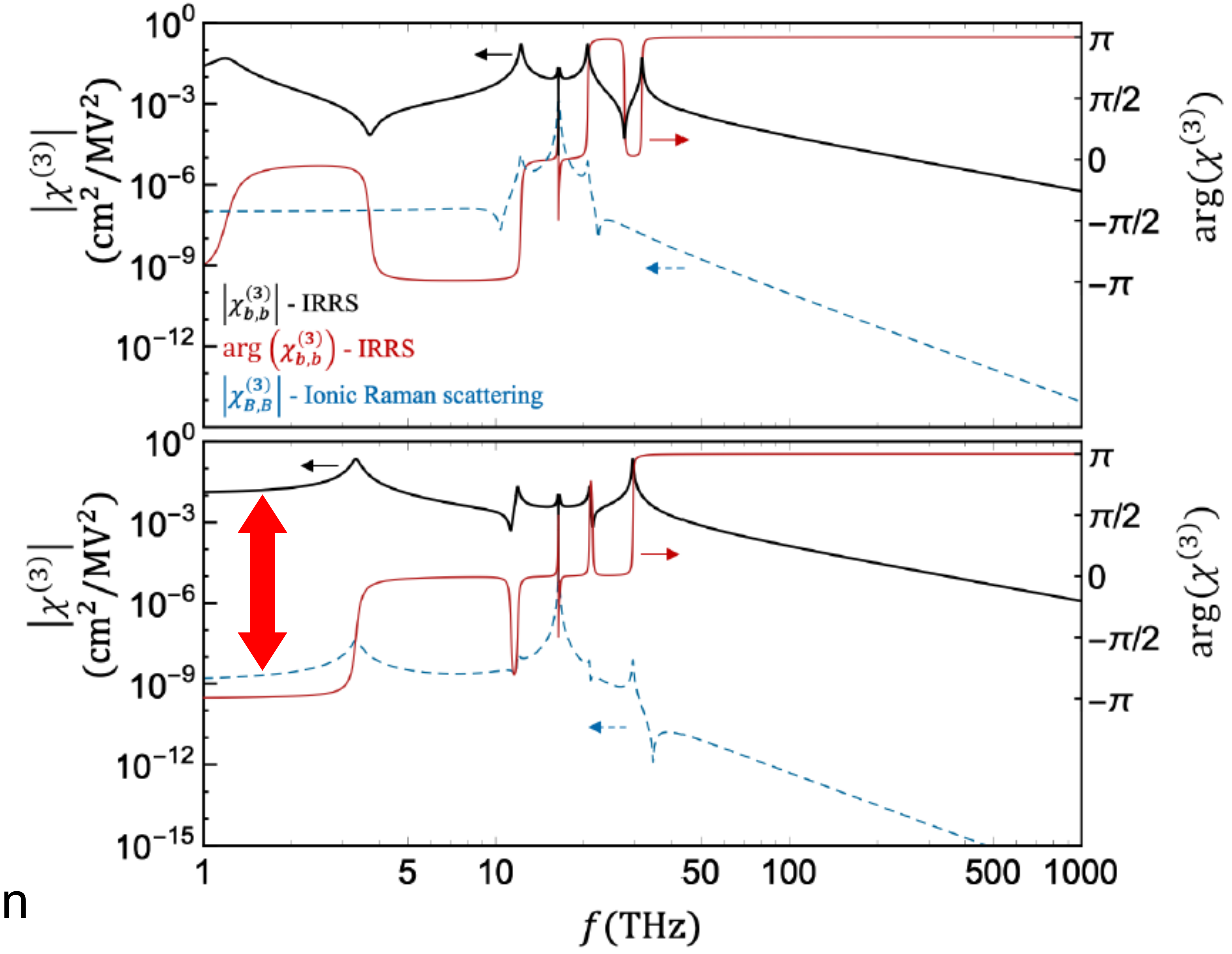
Guru Khalsa* and Nicole A. Benedek
cience and Engineering, Cornell University, Ithaca,

Jeffrey Moses
Engineering Physics, Cornell University, Ithaca,

large-amplitude ionic polarization

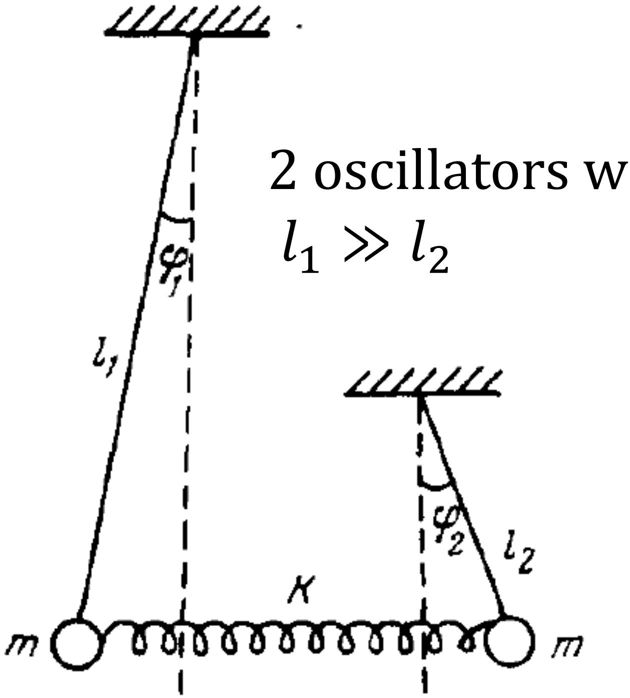
rectified pulse of microscopic deformations





not a phonon as such, but polarization

Intermezzo: Nonlinear coupled oscillators



2 oscillators with
$l_1 \gg l_2$

Magnetoelastic interaction is nonlinear in many cases, but in some cases it is more nonlinear than in others

Ozhogin and Preobrazhenskiĭ, Sov. Phys. Usp. **31**, 713 (1988).

amplitudes relate by $\varphi_2 = \xi \frac{\omega_1 l_1}{\omega_2 l_2} \varphi_1$, $\xi = \frac{K}{m\omega_1\omega_2}$

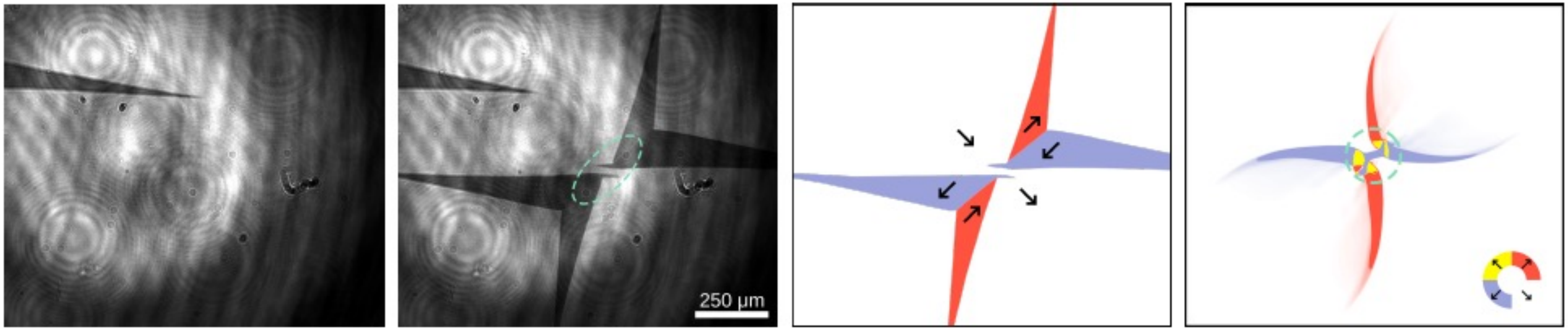
because $\varphi_2 \gg \varphi_1$ we can have $\varphi_2 \sim 1$

with $\varphi_1 \ll 1$

- if one component is nonlinear, the complete system is also nonlinear
- at resonance, energy is distributed equally between the two systems
- elastic system ~few eV/atom versus magnetic system < 1 meV/atom
- in system with weak magnetic anisotropy, very-large-amplitude dynamics is easy to reach

Different magnetization & anisotropy – similar pattern

$(LuYBi)_3(FeGa)_5O_{12}$ magnetic garnet, (001) substrate, 6 μm thick,
very weak magnetic anisotropy, in-plane magnetization



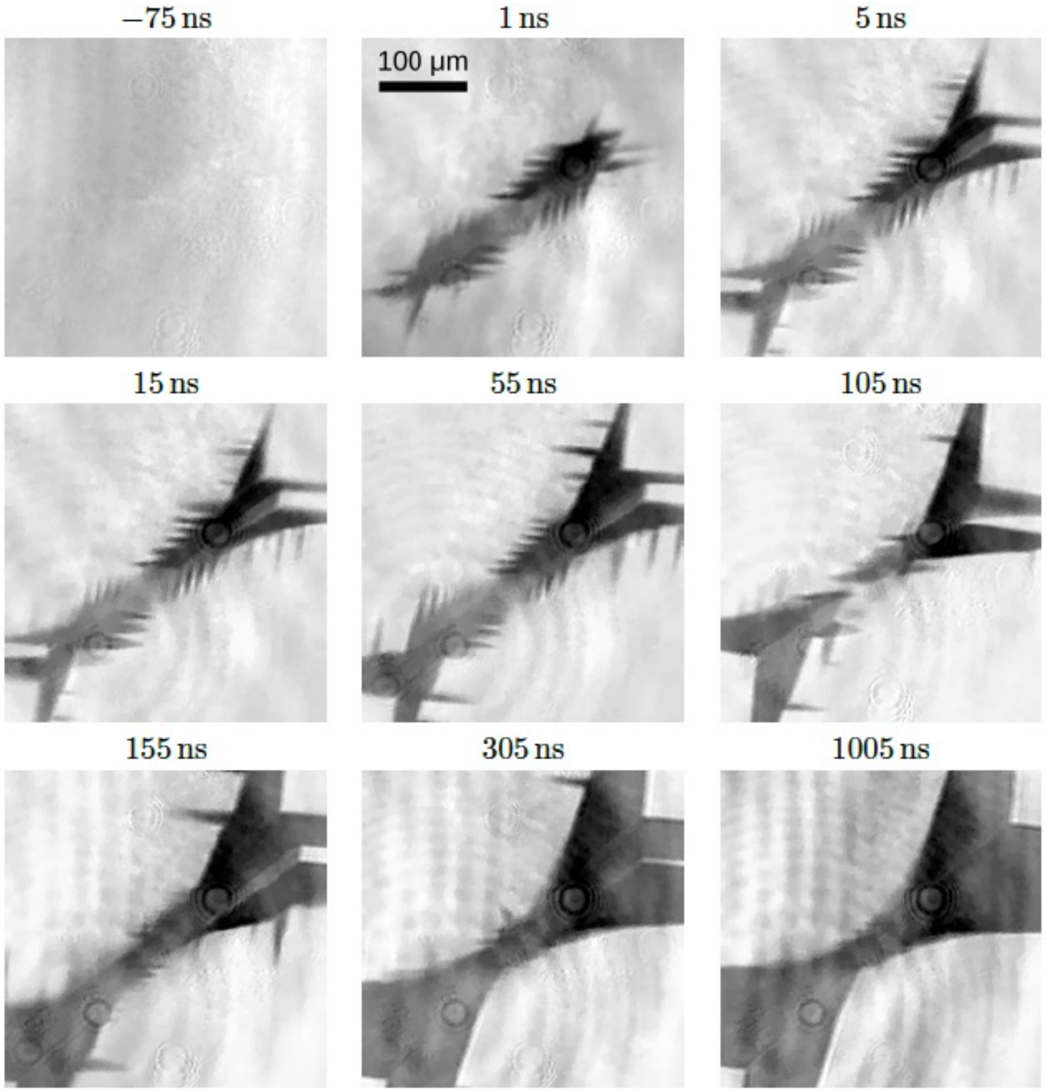
before

after

domain pattern

simulations

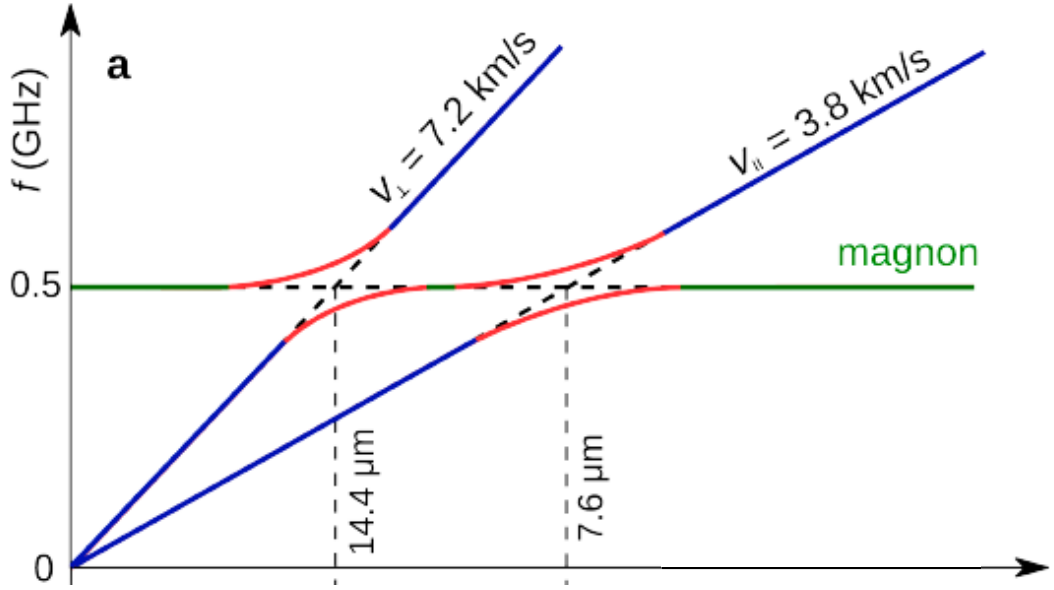
The dynamics of domain development



- single-shot time-resolved imaging
- time resolution: 5 ns (Nd:YAG laser)
- FWHM of IR pump: ~300 µm
- domain pattern with period <20 µm
- propagation speed ~1 km/s

M. Gidding et al, Nature Commun. **14**, 2208 (2023).

These periodic ripples? what is the origin of their size?

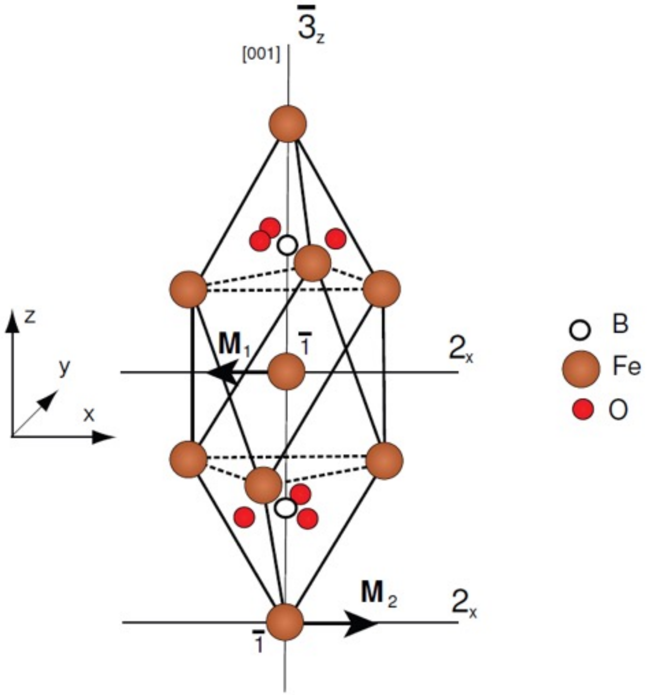


The fact that we actually observe domains with reversed magnetization supports the idea of condensation in the sense that the waves become phase-locked and **coherently contribute to the excitation** amplitude.

Their large velocity also supports the magneto-elastic origin

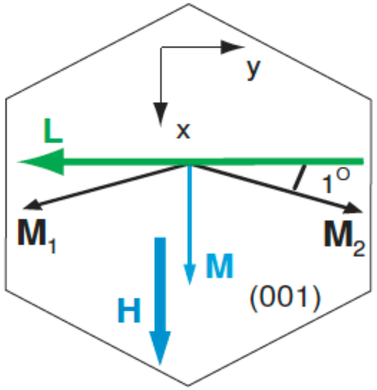
M. Gidding et al, Nature Commun. **14**, 2208 (2023).

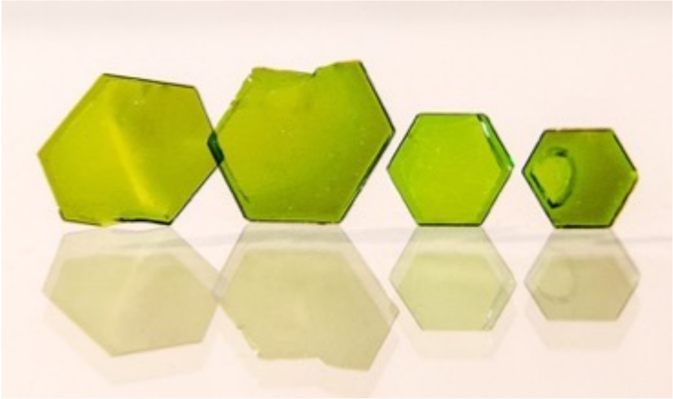
Canted antiferromagnet $FeBO_3$



Néel temperature $T_N = 348$ K

easy-plane anisotropy

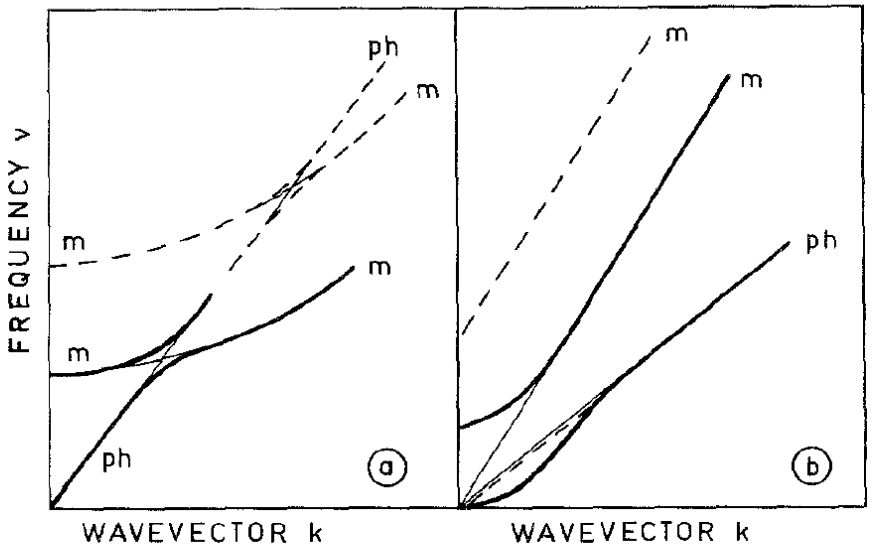




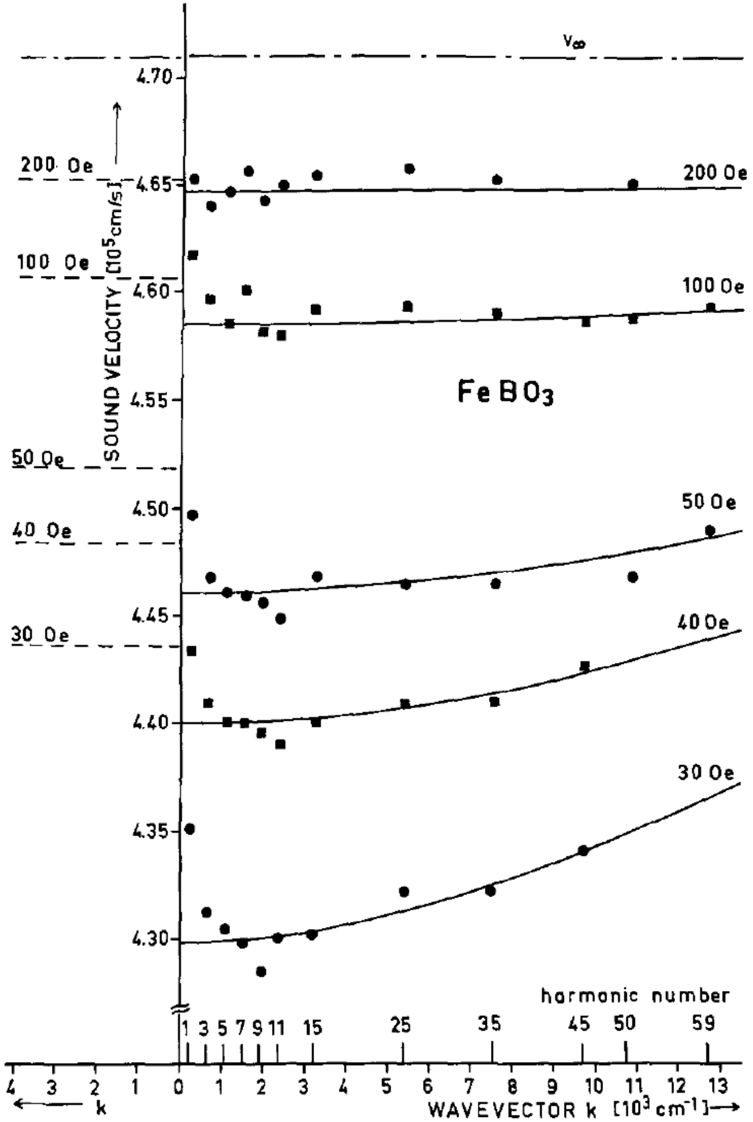
$$H_{A\ \text{in-plane}} = 0.26\ \text{Oe}$$

$$H_{A\ \text{out-of-plane}} = 1.7\ \text{kOe}$$

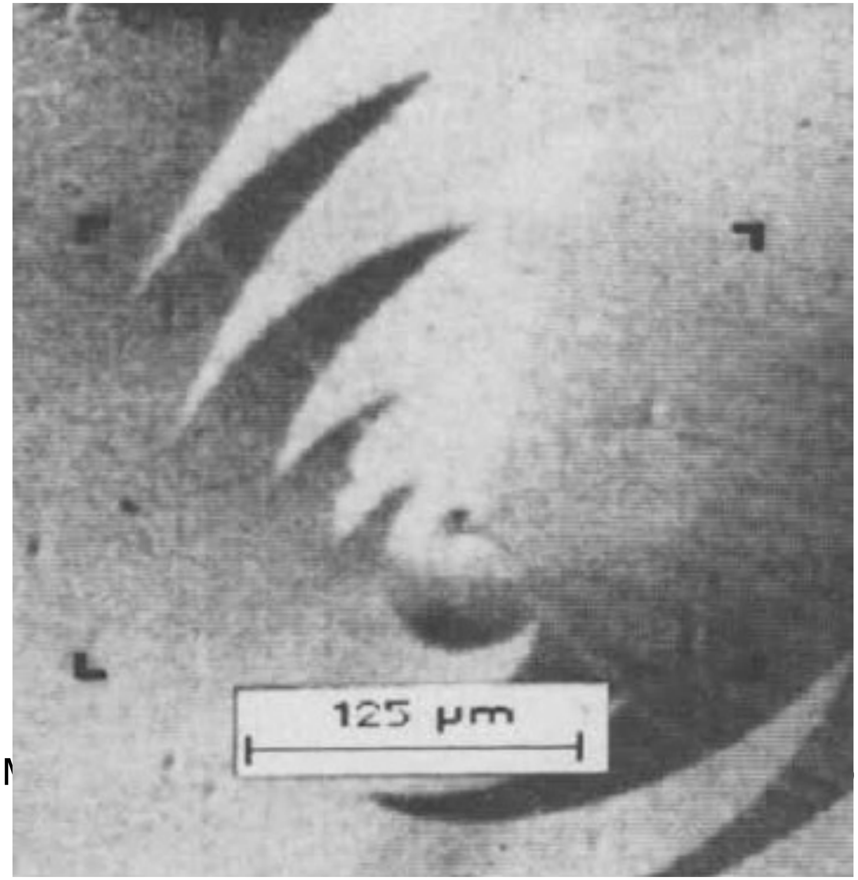
Magnetically tunable sound in $FeBO_3$



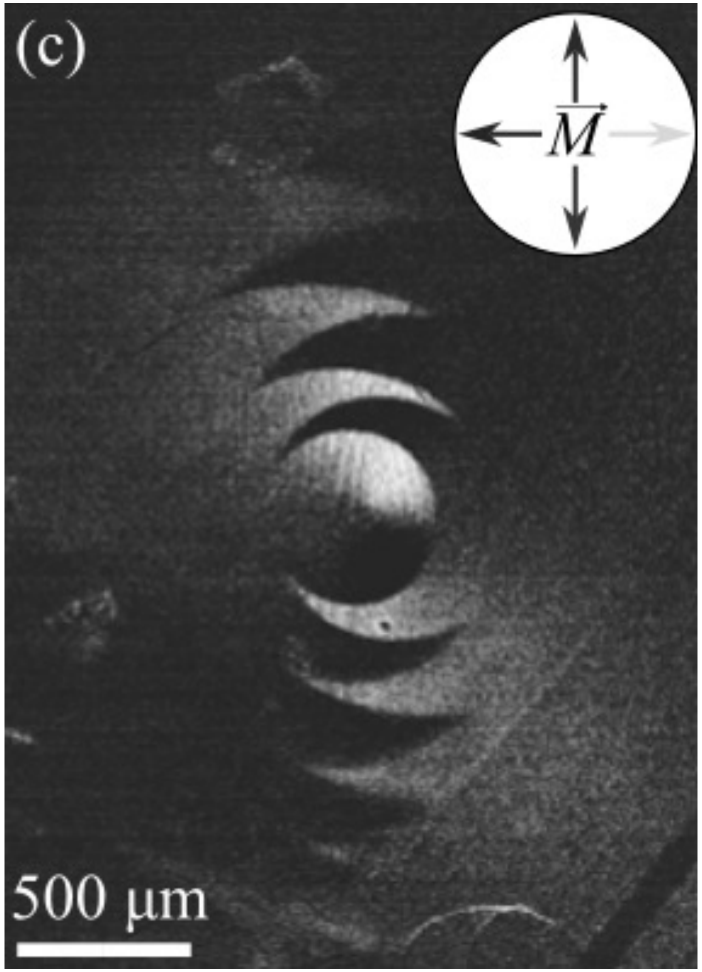
Wettling & Jantz, Appl. Phys. **23**, 195 (1980)



Domain structures in $FeBO_3$ – static and induced



W. Rave, R. Schäfer, and A. Hubert,
JMMM **65**, 7 (1987).



T. Janssen et al, Phys. Rev. B **108**, L140405 (2023)

Summary

- Circular-polarized TO phonons can lead to magnetic switching via **ultrafast Barnett effect**
- Strong **excitation at the frequency of LO phonons** leads to high-amplitude deformations and transient magnetic anisotropy
- Peculiar shape of **switched pattern is a fingerprint** confirming the mechanism
- Large amplitudes of elastic excitation lead to nonlinear behaviour, with **solitons and pattern formation**