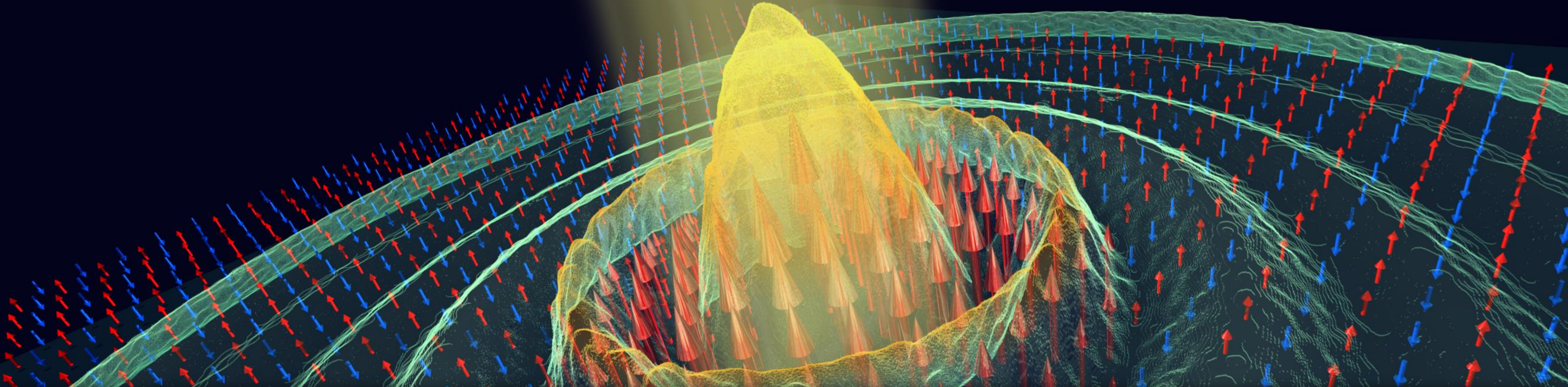
Engineering magnetic states with light through nonlinear lattice excitation



Ankit Disa

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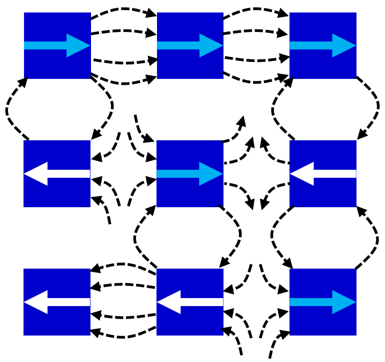
School of Applied & Engineering Physics, Cornell University, Ithaca, NY, USA

SPICE-Ultrafast Antiferromagnetic Writing Workshop, May 10, 2022

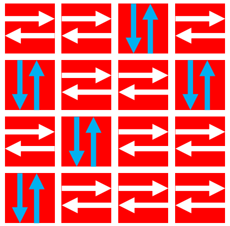
Antiferromagnetic writing

How can we manipulate order in an antiferromagnet?

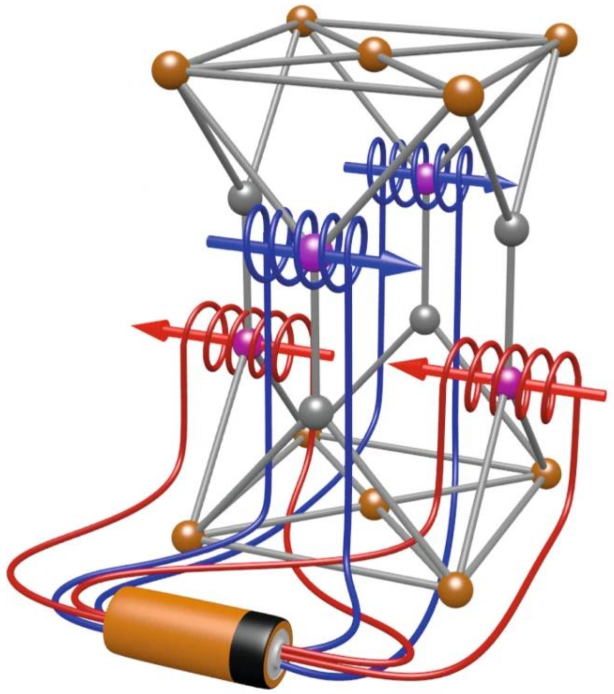
Ferromagnetic



Antiferromagnetic

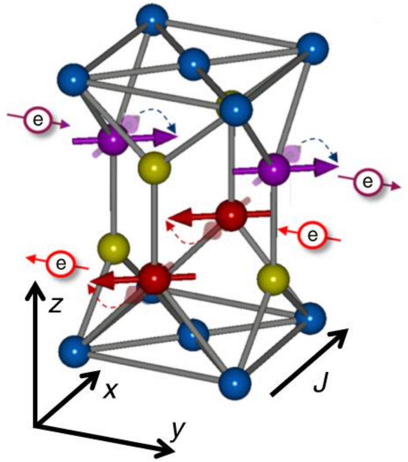


- Denser
- More robust
- Faster



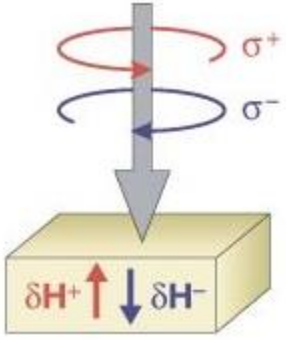
A structural approach to magnetic control

Electrical control



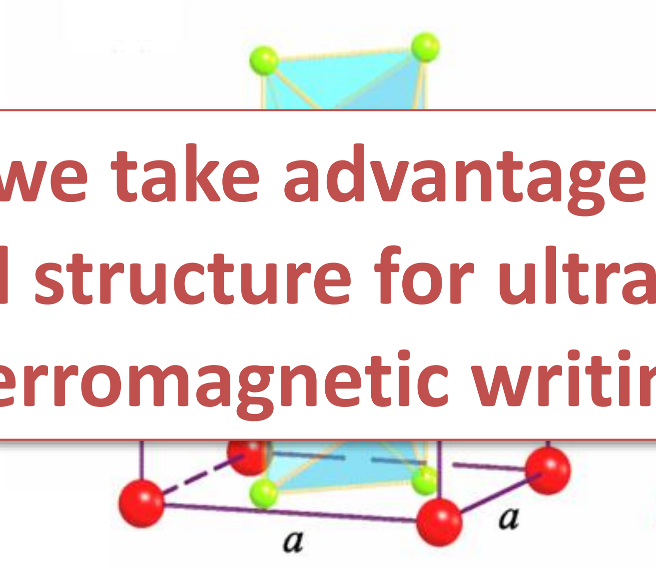
J. Gordinho, *et al.* Nat. Comms. **9**, 4686 (2018).

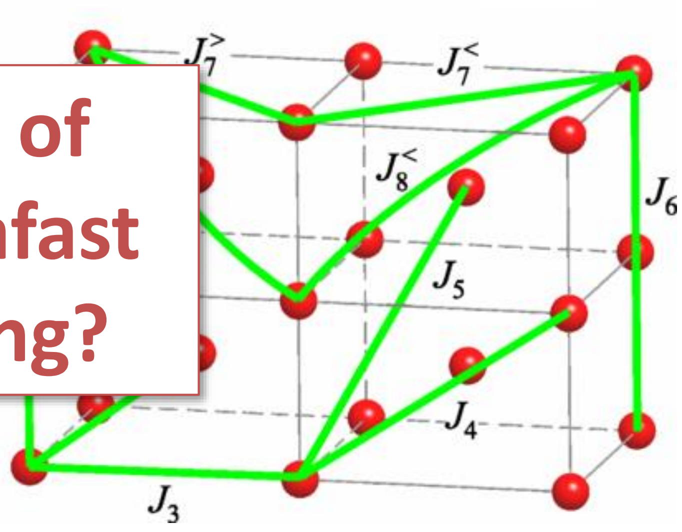
Optical control



A.V. Kimel, *et al.* Nature **435**, 655 (2007).

- **Crystal structure directly determines local magnetic states and their interactions**





Can we take advantage of crystal structure for ultrafast antiferromagnetic writing?

Symmetry, crystal field
→ Anisotropy

Bond lengths, angles
→ Exchange

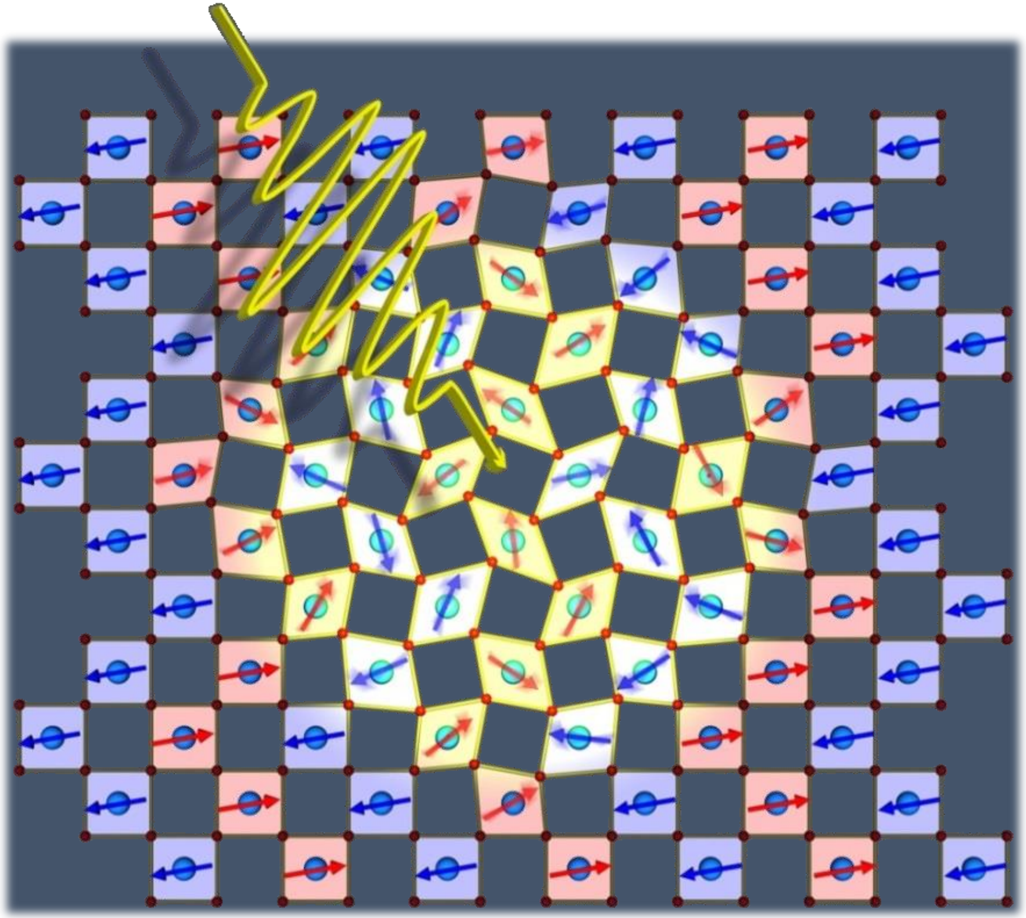
Engineering the crystal structure with light

Drive large amplitude structural distortions with laser pulses

→ Resonantly excite optical phonons (~2-200 meV, ~0.5-50 THz)

~MV/cm electric fields → 5-10% atomic displacements

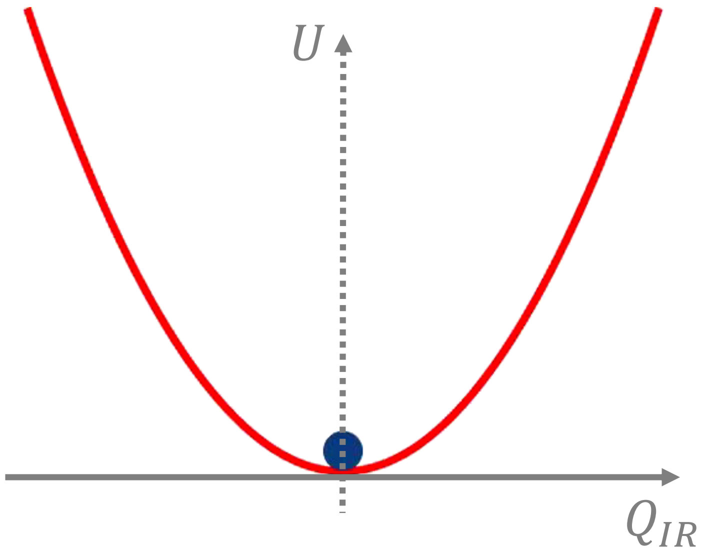
Leads to highly nonlinear response of lattice → targeted structural distortions

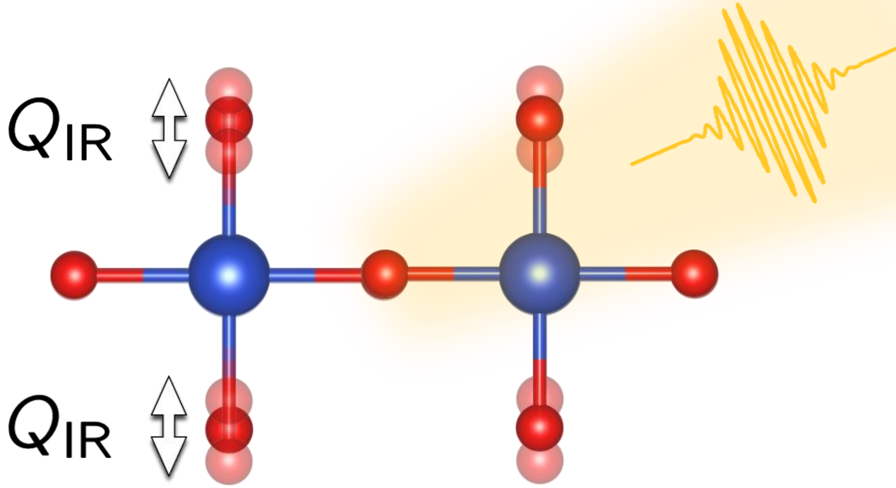


Linear excitation of lattice modes

- **Light couples to infrared-active modes:**

$$U_{lattice} = \frac{1}{2}\omega_{IR}^2 Q_{IR}^2 - z^* Q_{IR} E_{laser}$$



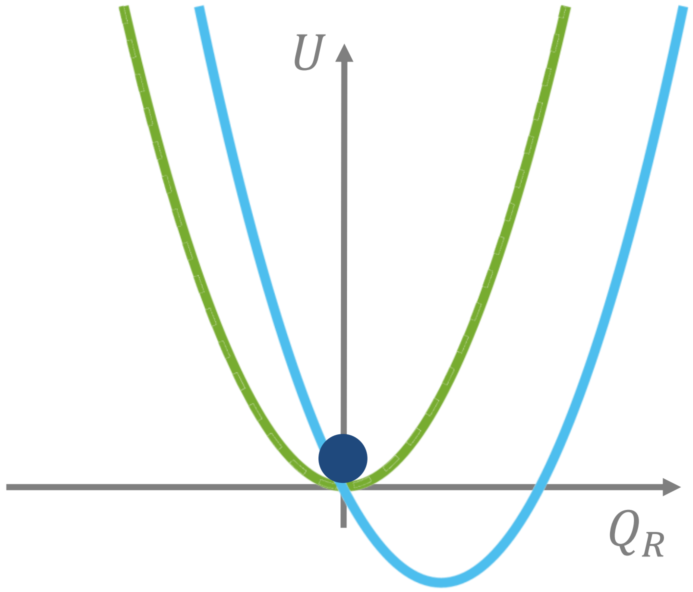


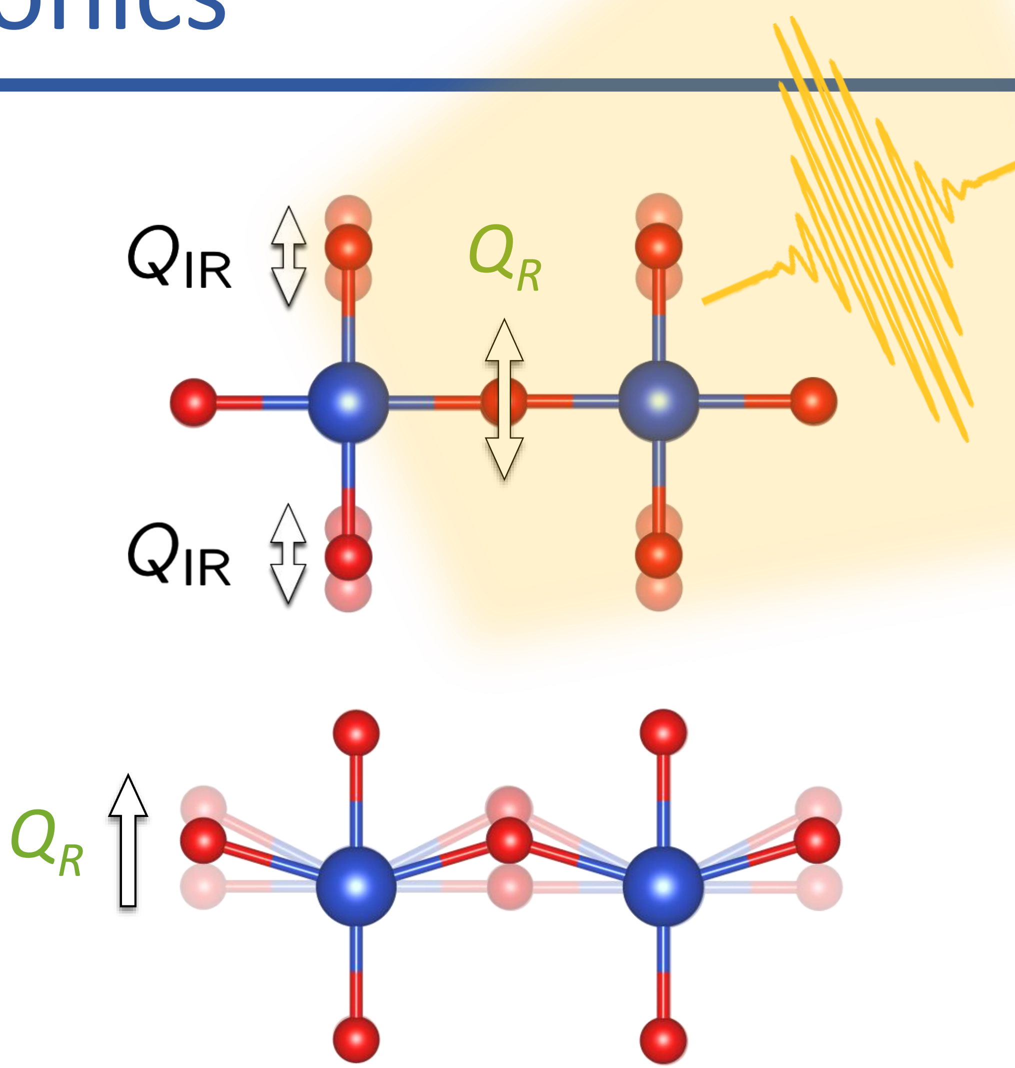
No average change to the lattice

"Nonlinear phononics"

- **Large IR motions can couple to other modes:**

$$U_{lattice} = \frac{1}{2}\omega_{IR}^2 Q_{IR}^2 + \frac{1}{2}\omega_R^2 Q_R^2 - g Q_{IR}^2 Q_R + \cdots$$

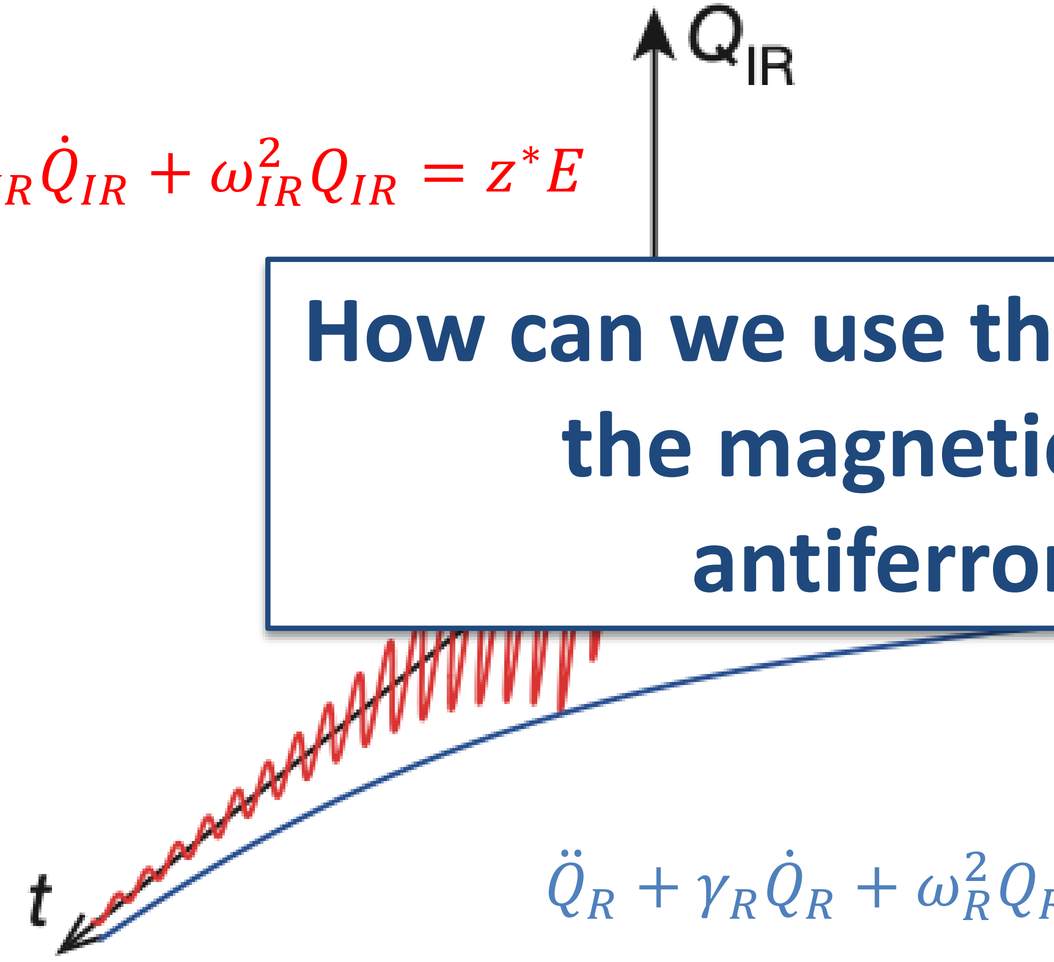




Net lattice displacement of coupled mode

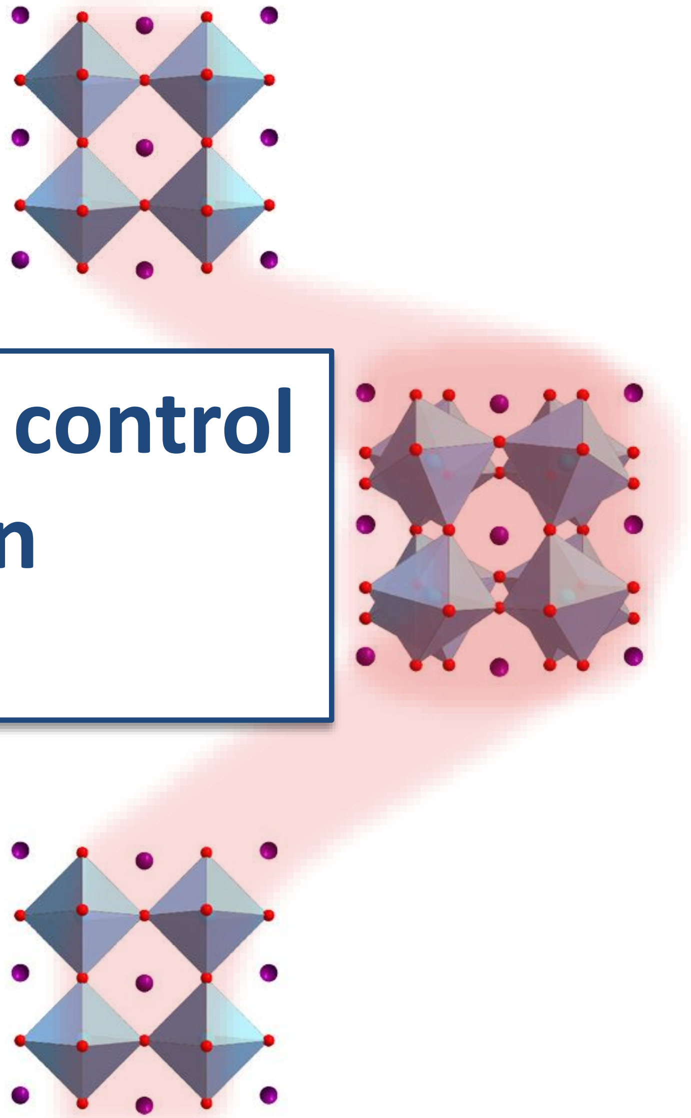
Engineering new crystal structures with light

$$\ddot{Q}_{IR} + \gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = z^* E$$



How can we use this effect to control the magnetic state in an antiferromagnet?

$$\ddot{Q}_R + \gamma_R\dot{Q}_R + \omega_R^2 Q_R = gQ_{IR}^2$$

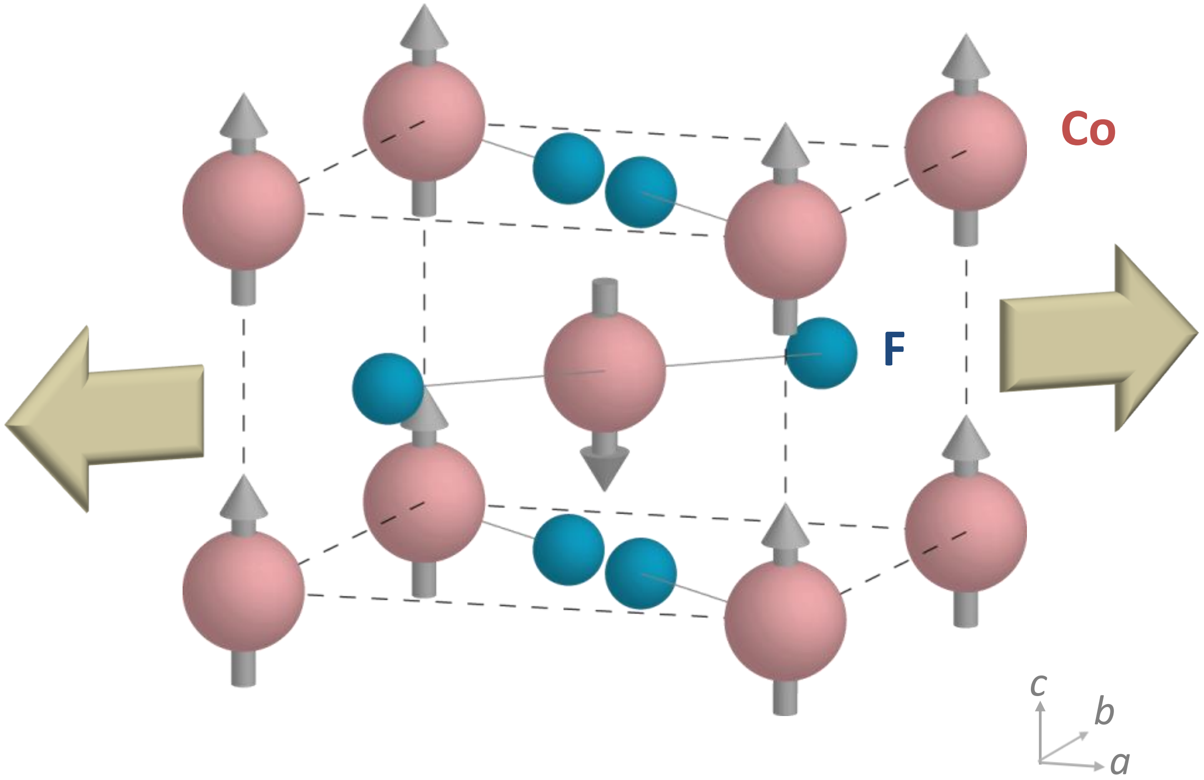


Strain control: piezomagnetism

CoF_2

(T_N = 38 K)

σ_{xy}

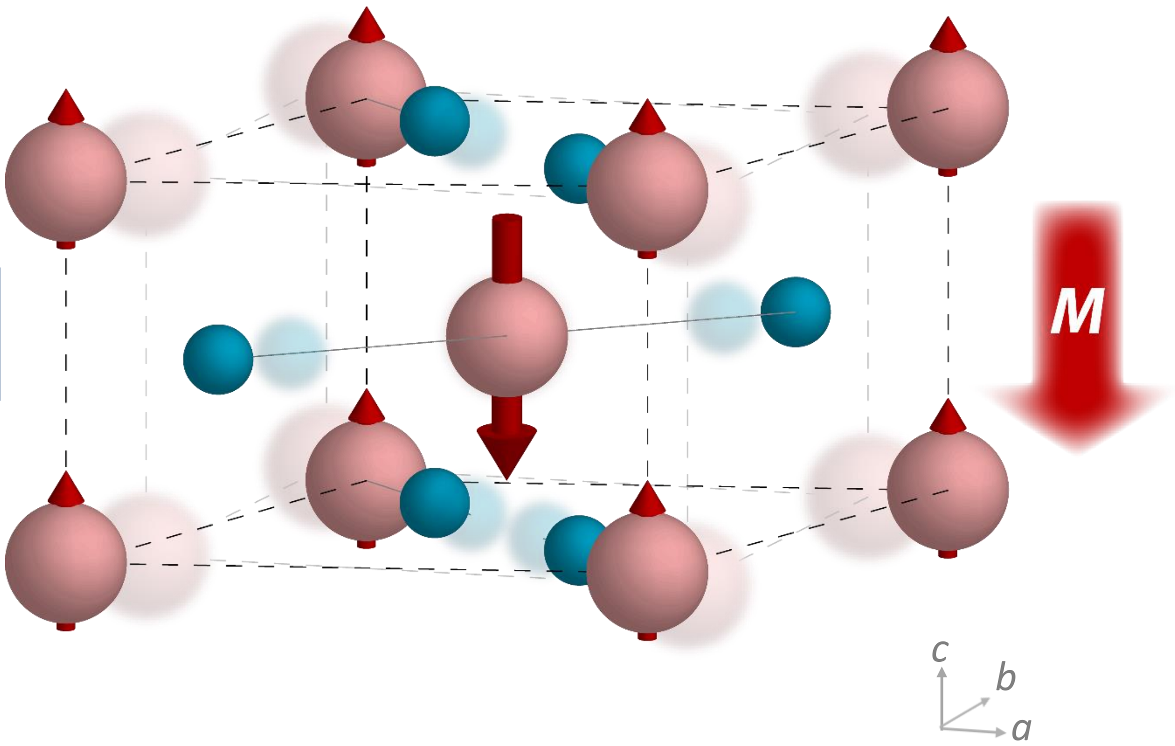


Strain control: piezomagnetism

CoF_2

(T_N = 38 K)

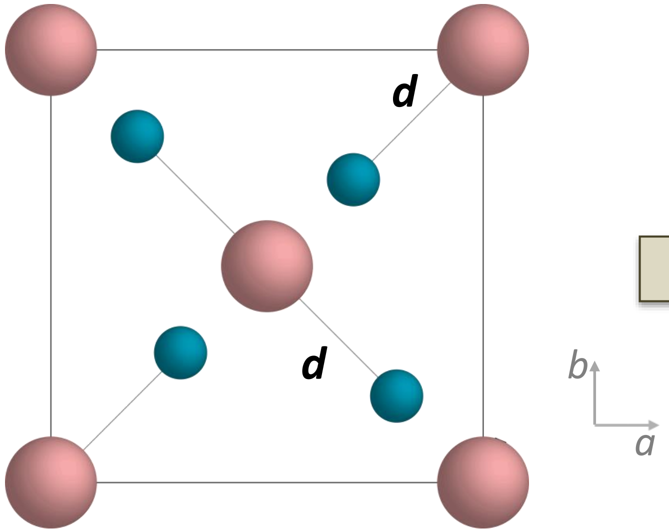
$$M_z \propto \sigma_{xy}$$

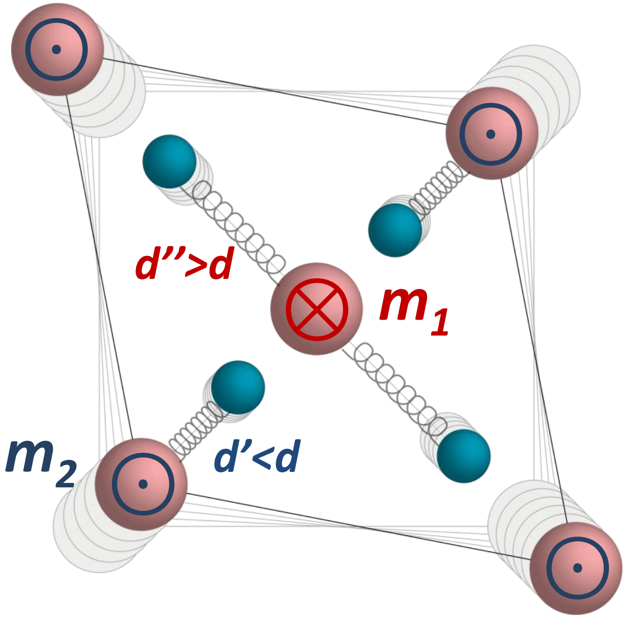


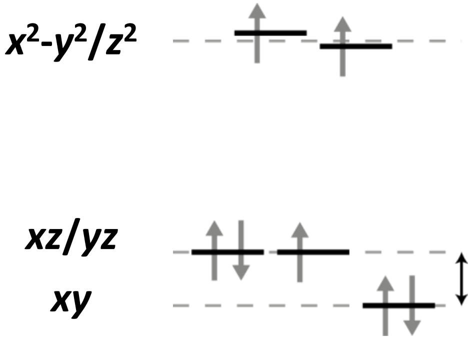
Origin of piezomagnetism in CoF_2

Co orbital moment:

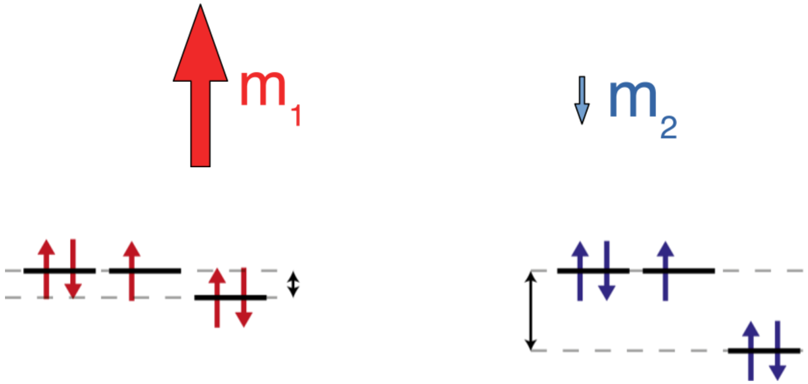
$$\langle m_L \rangle \sim d$$







$\Delta_{t_{2g}} \sim \frac{1}{m_L}$

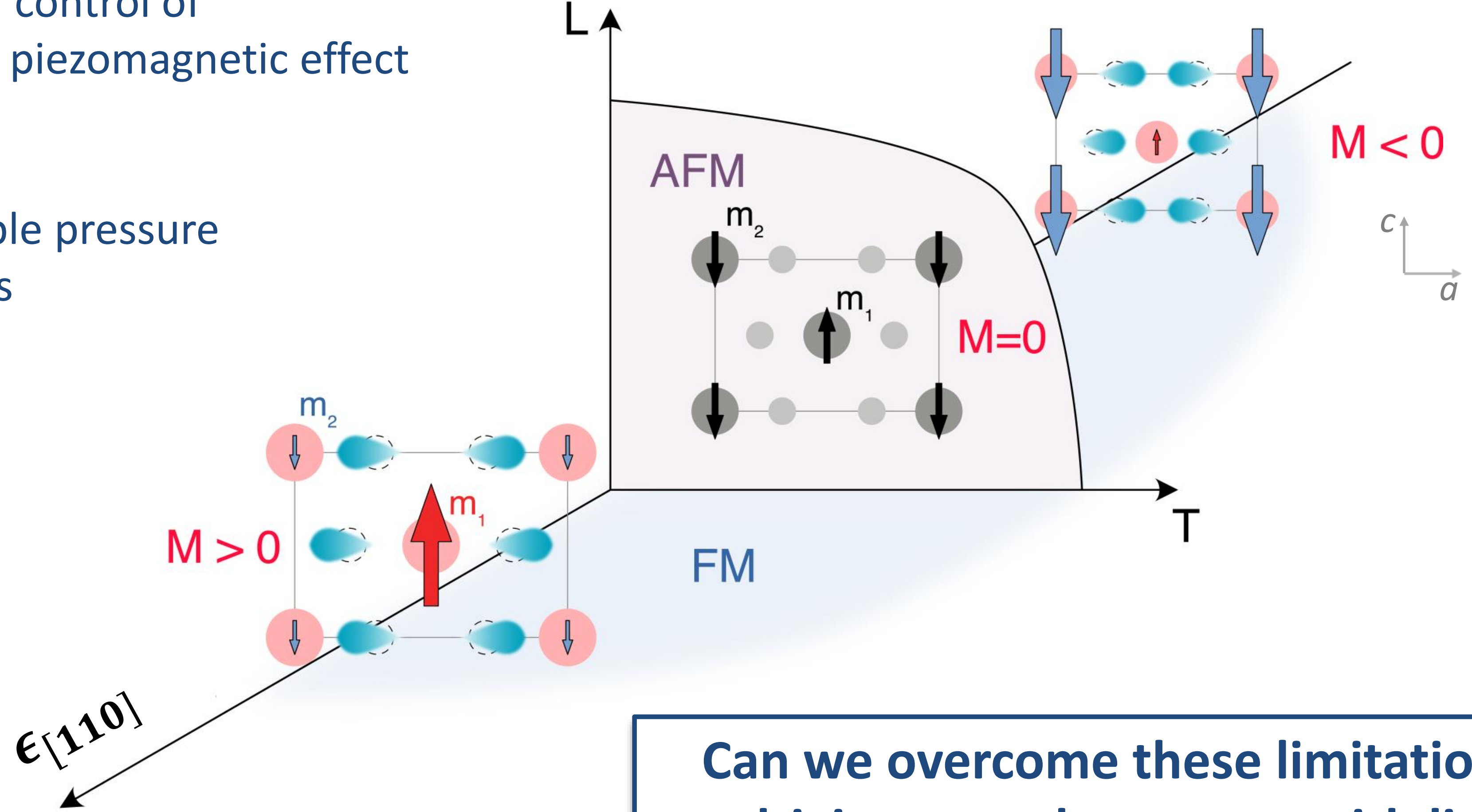


T. Moriya, J. Phys. Chem. Solids, **11**, 73 (1959) A. Borovik-Romanov, JETP, **11**, 4 (1960)

Uniaxial strain control of magnetization

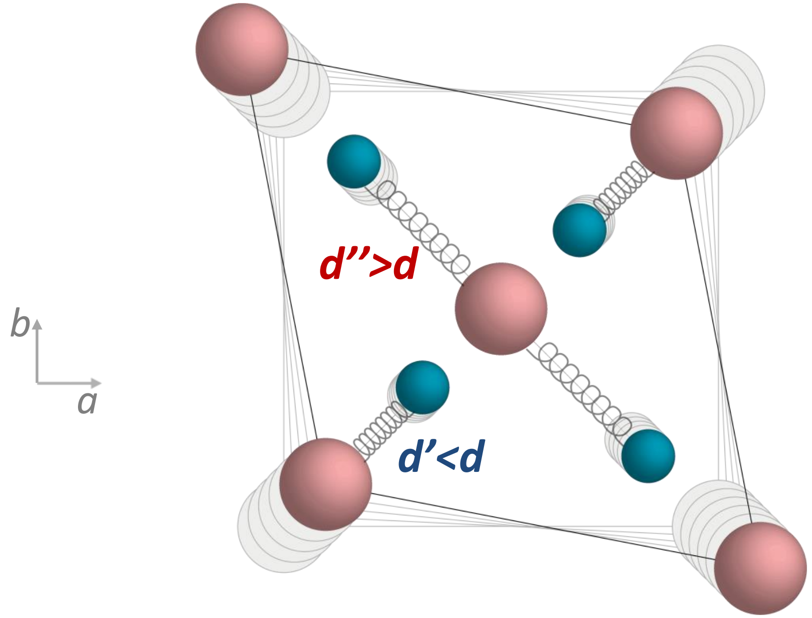
- Bi-directional strain control of magnetization with piezomagnetic effect

Disadvantages:
- Limited by achievable pressure
- Acoustic time scales



Can we overcome these limitations by driving crystal structure with light?

Symmetry of strain



Stress along [110]

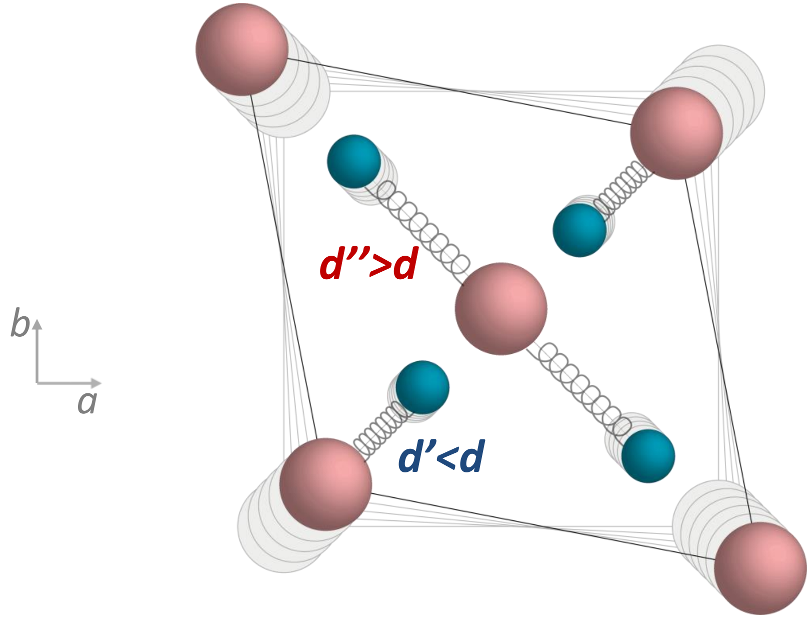
- **Piezomagnetic effect**

$$M_n = \Lambda_{nij}\sigma_{ij}$$

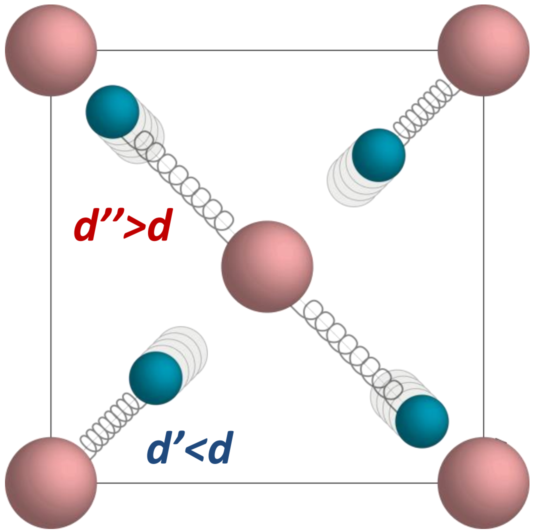
$$\Lambda_{nij} = \begin{bmatrix} 0 & 0 & 0 & \Lambda_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & \Lambda_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & \Lambda_{36} \end{bmatrix}$$

σ_{xy} → B_{2g} symmetry

Symmetry of strain



Stress along [110]



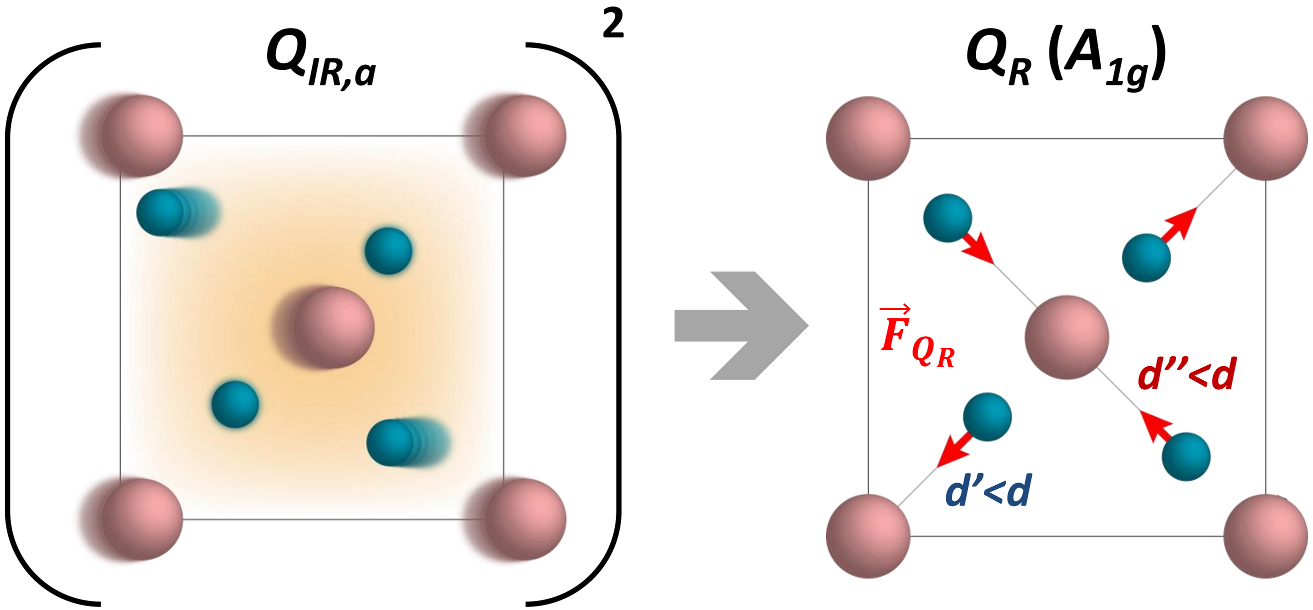
B_{2g} Raman phonon

$M_z \propto Q_R$

- B_{2g} Raman mode provides same lattice distortions as uniaxial strain
- **Must break underlying symmetry of the lattice**

Nonlinear phonon coupling in CoF_2

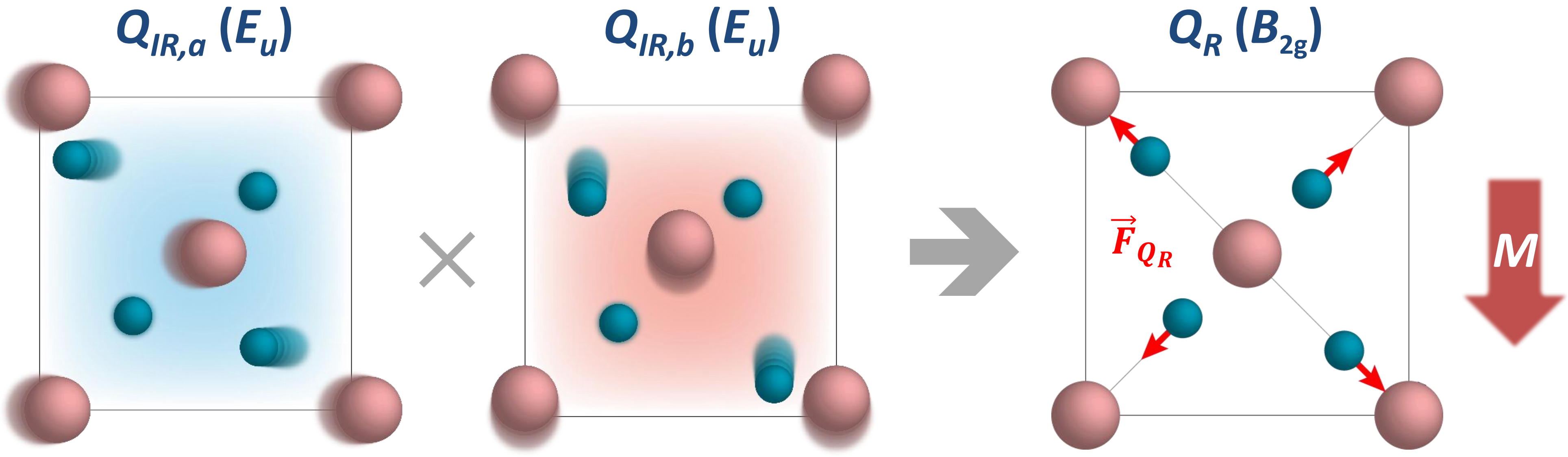
Two-phonon nonlinear phonon interaction: $U_{NL} \propto Q_{IR}^2 Q_R$



- For single IR phonon excitation, Q_R preserves lattice symmetry
→ Does not generate magnetization

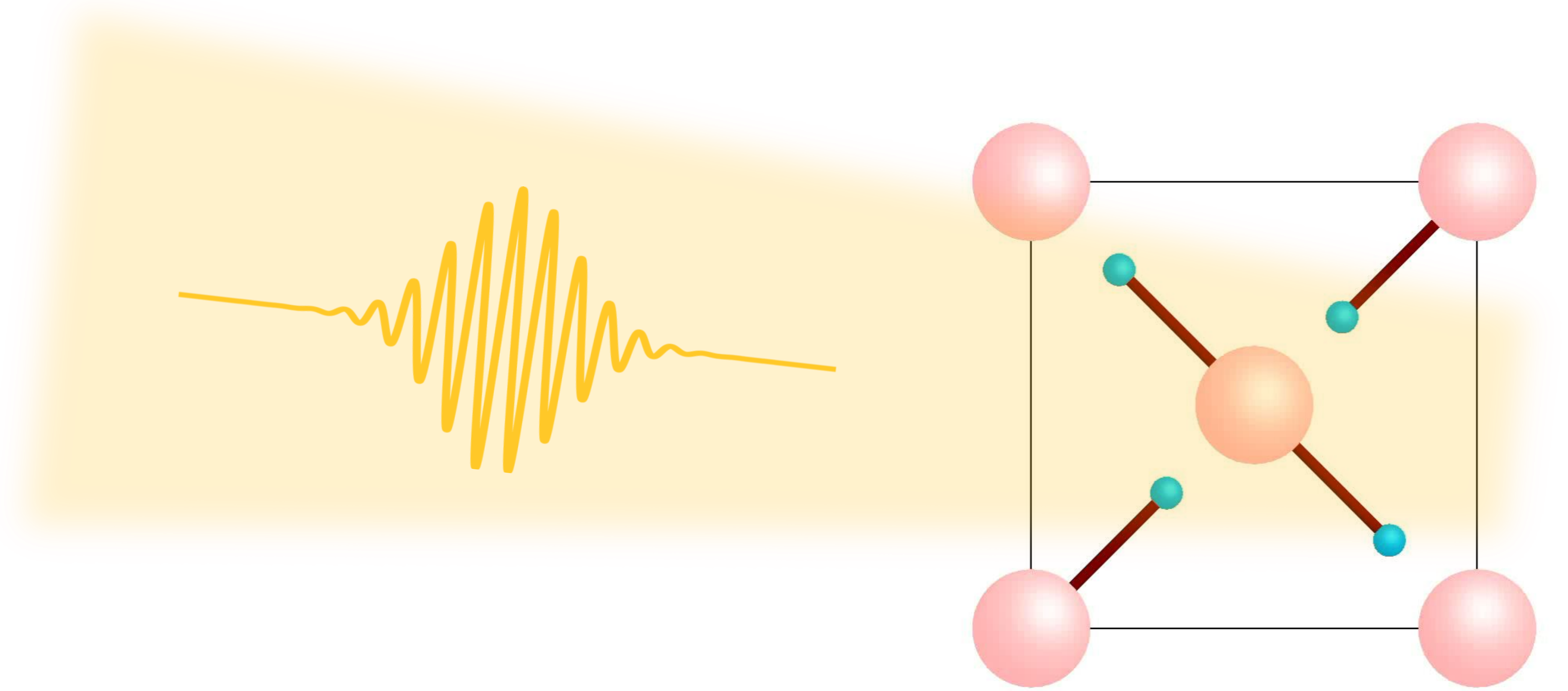
Symmetry breaking from phonons

Three-phonon nonlinear interaction: $\boldsymbol{U_{lattice}} \propto \boldsymbol{Q_{IR,1}Q_{IR,2}Q_R}$



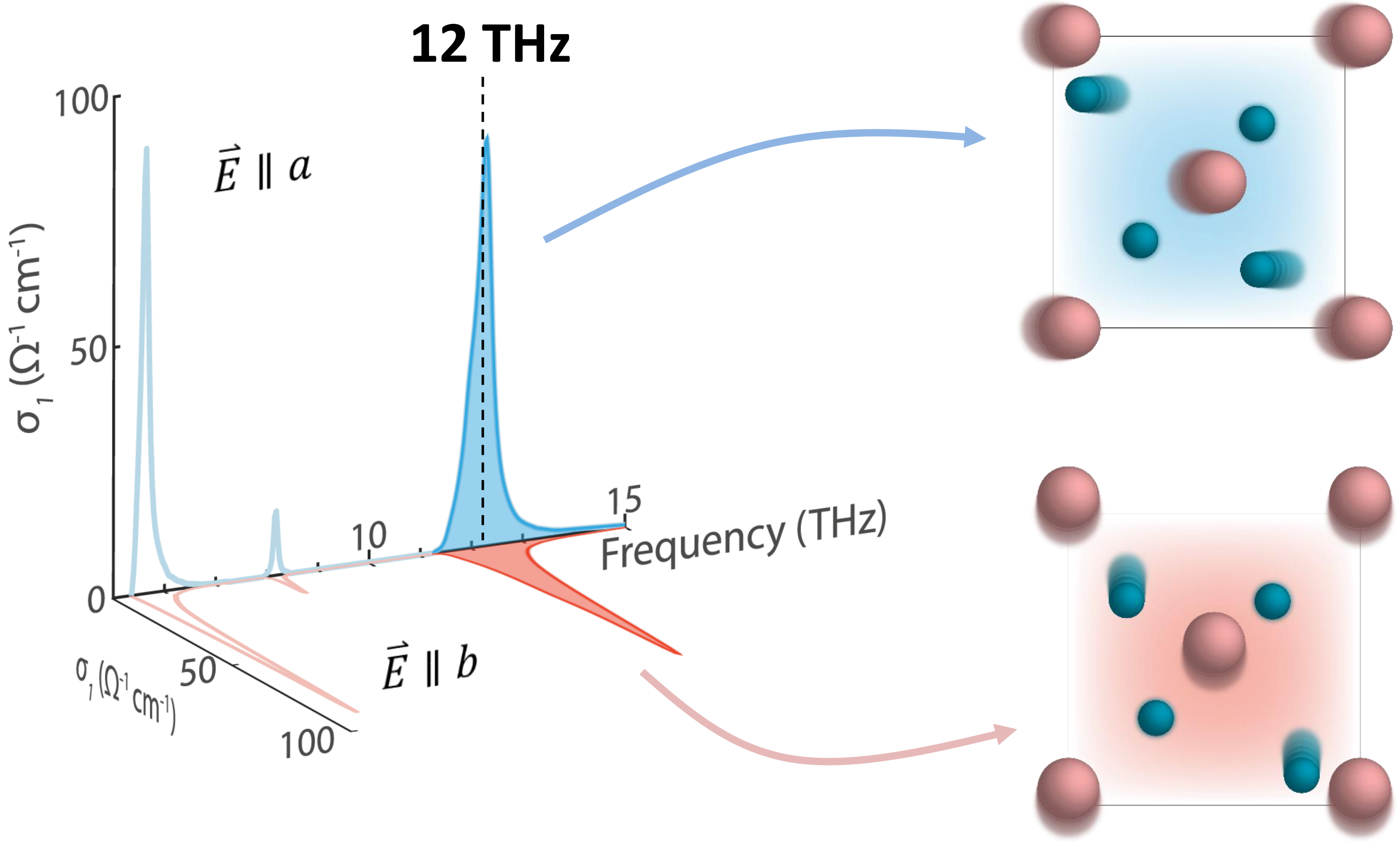
- Simultaneously excite degenerate IR phonons along *a* and *b* to generate magnetization

Symmetry breaking from phonons



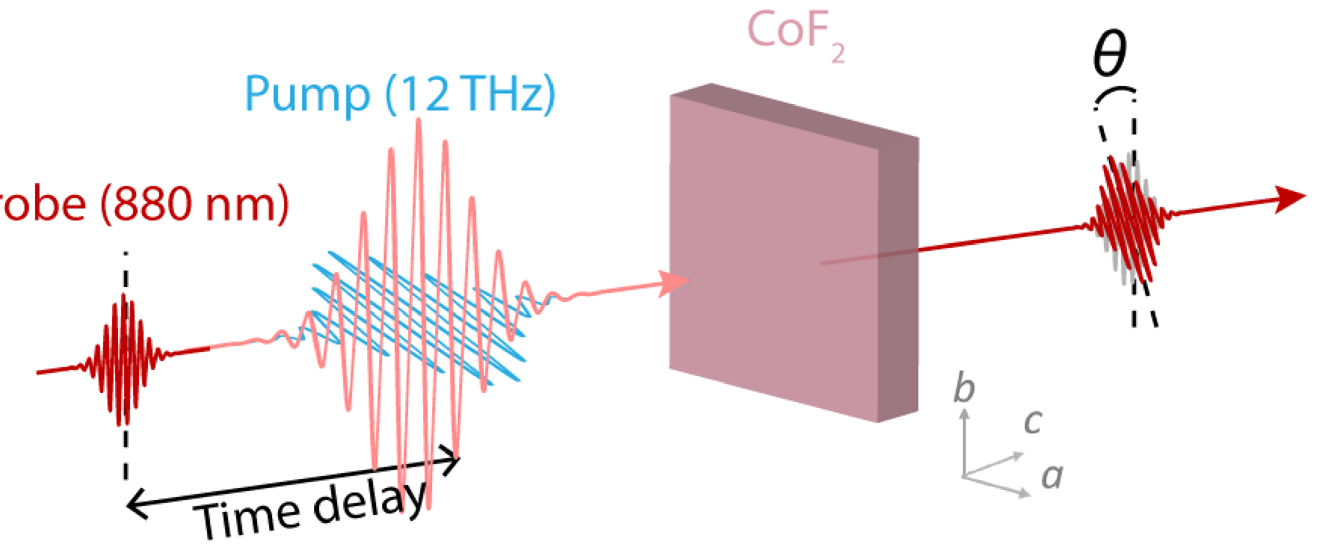
A.S. Disa, *et al.* Nature Phys. **16**, 937–941 (2020)

Resonantly driving phonons



Experimental setup

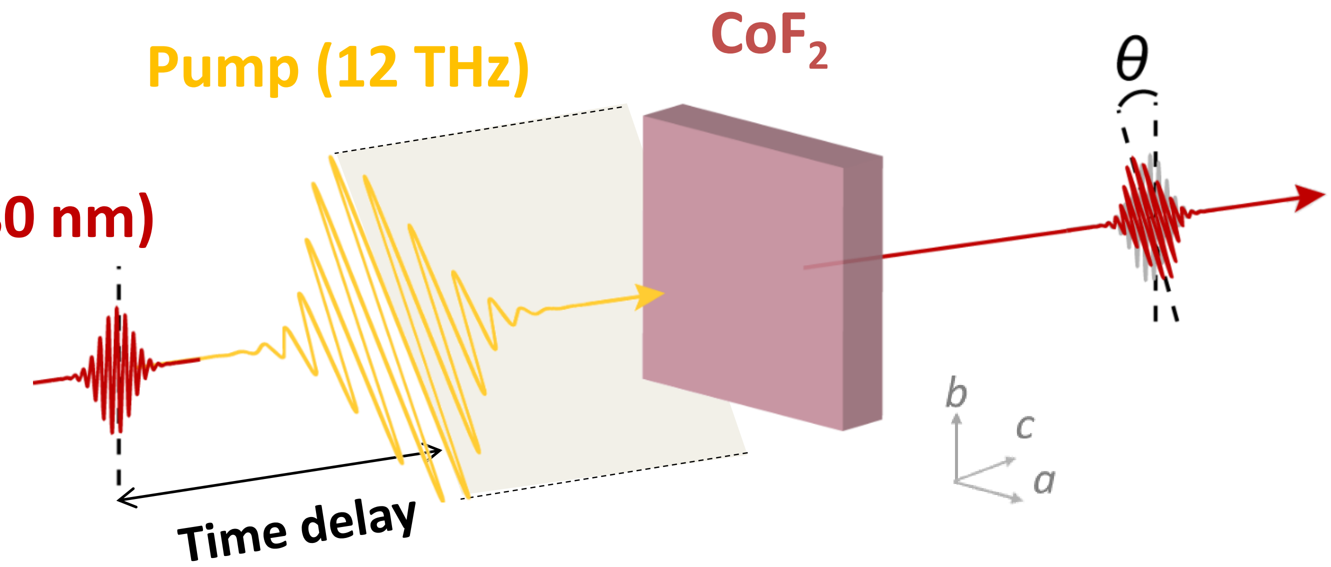
- Simultaneously drive *a* and *b* phonons by pumping along [110]



- Measure time-resolved Faraday effect: $\theta(t) \propto M_z(t)$

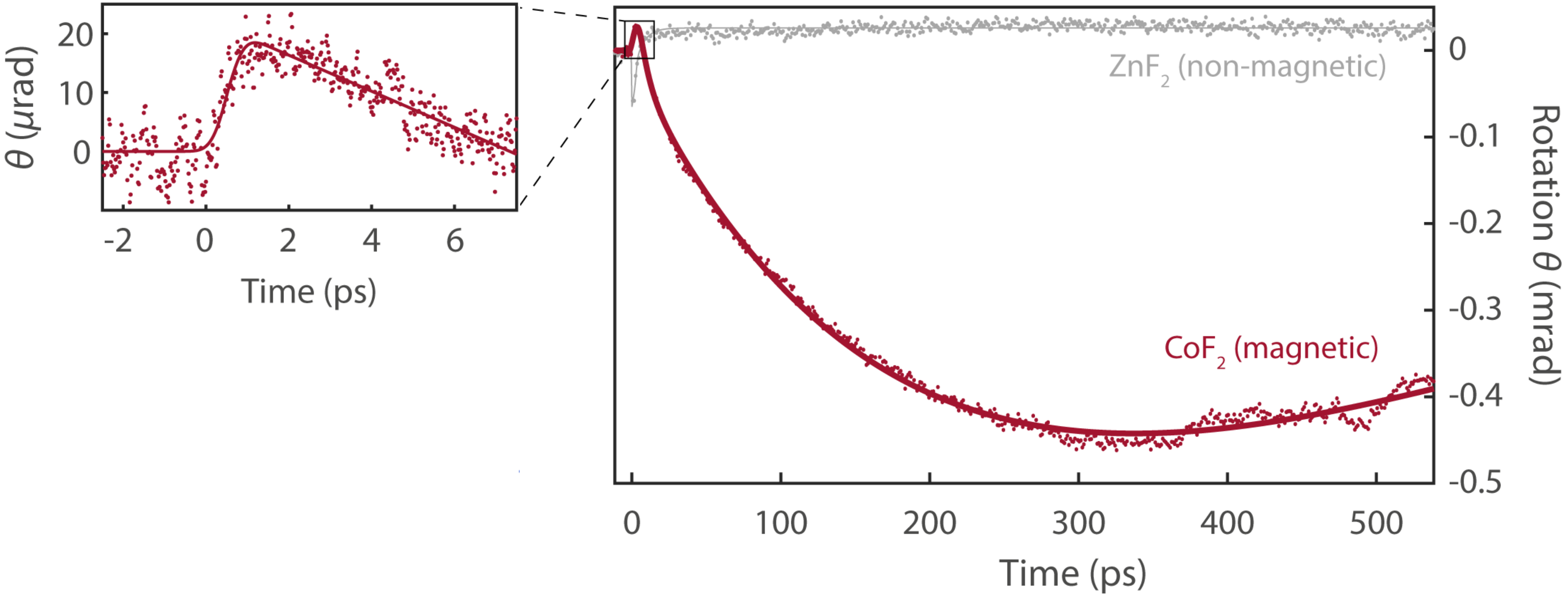
Experimental setup

- Simultaneously drive *a* and *b* phonons by pumping along [110]



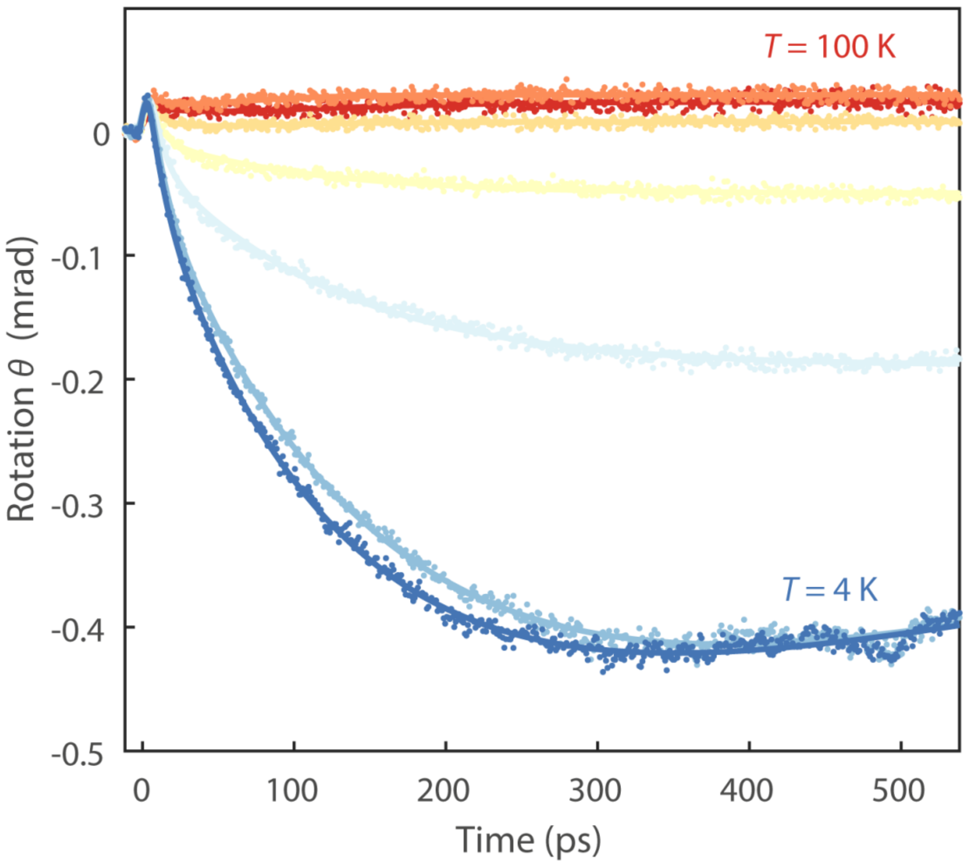
- Measure time-resolved Faraday effect: $\theta(t) \propto M_z(t)$

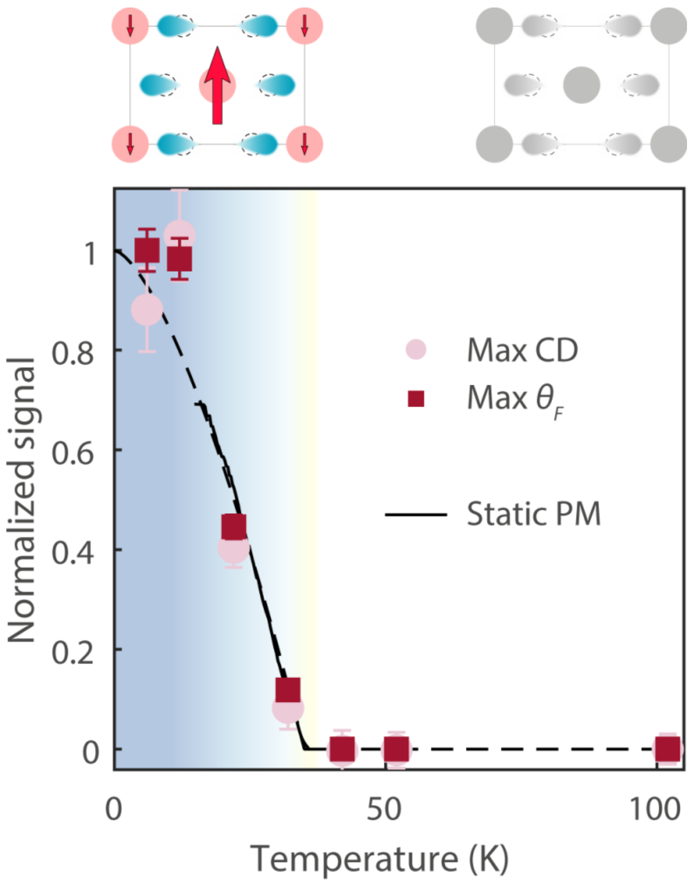
Pump-induced Faraday rotation



- Long-term Faraday signal: **signature of pump-induced magnetization**
  - Same behavior seen in circular dichroism signal

Temperature dependence

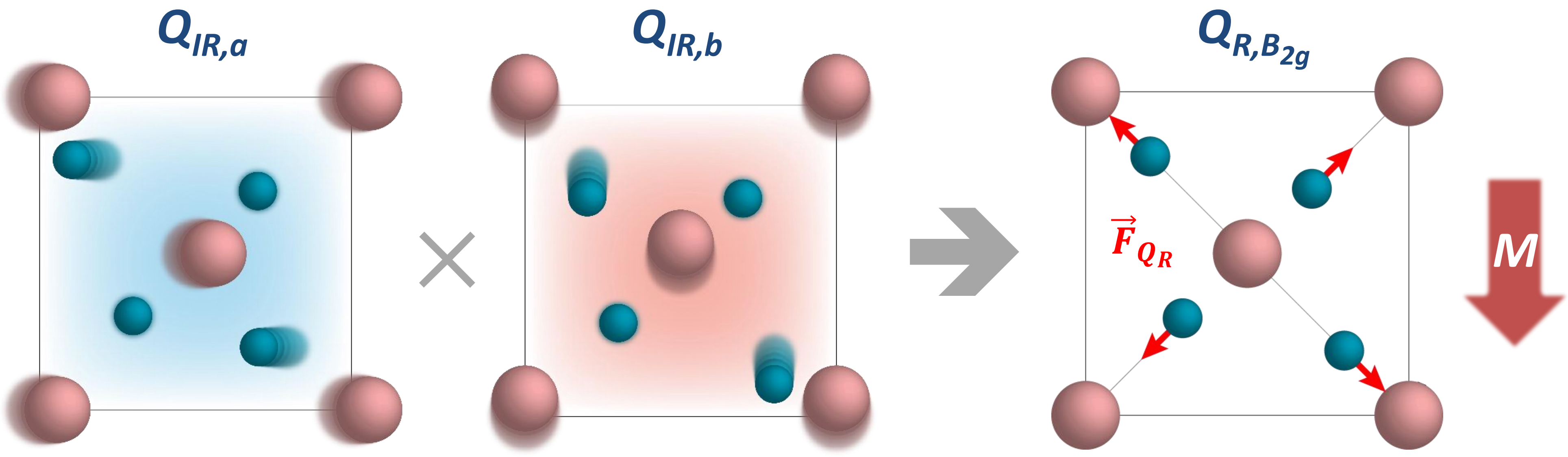




- Pump-induced effect follows static piezomagnetic response

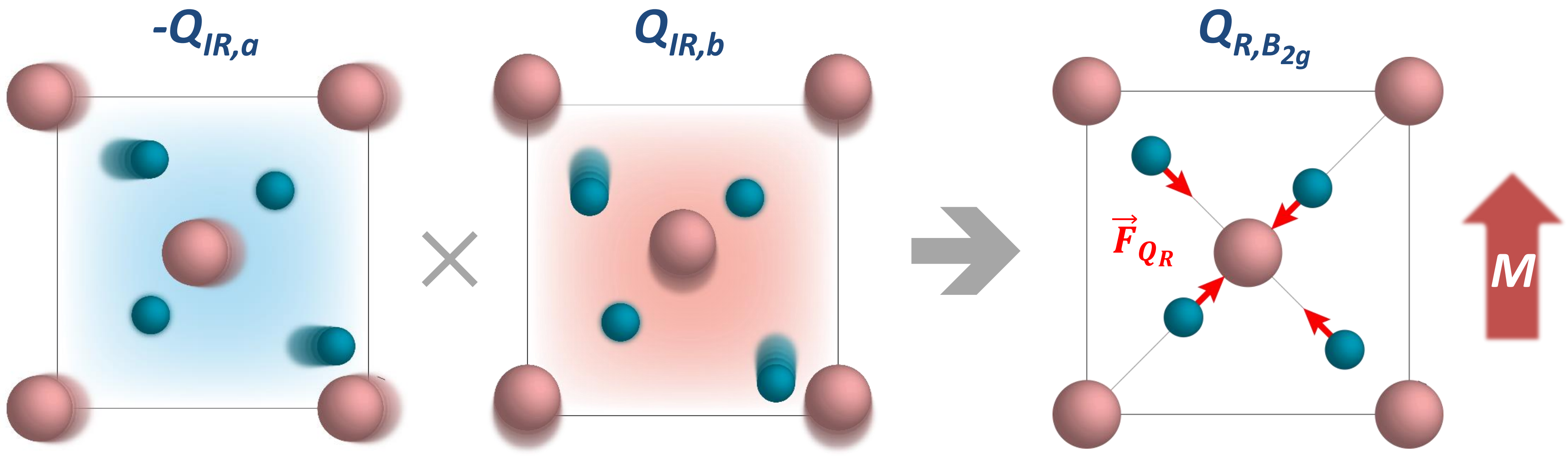
Switchable magnetization

Three-phonon nonlinear interaction: $U_{lattice} \propto Q_{IR,1} Q_{IR,2} Q_R$



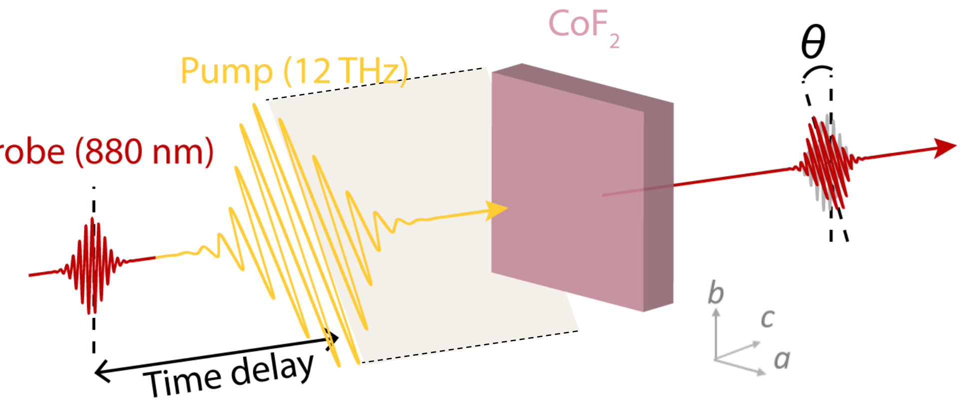
Switchable magnetization

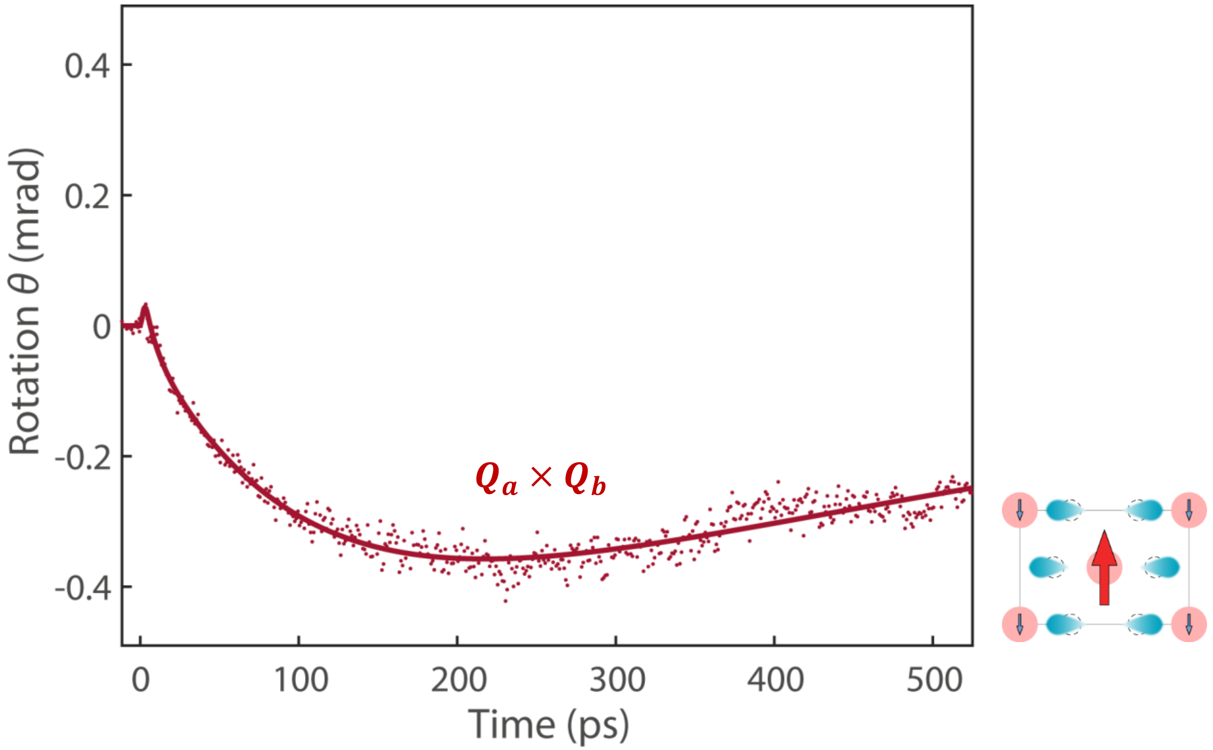
Three-phonon nonlinear interaction: $U_{lattice} \propto Q_{IR,1} Q_{IR,2} Q_R$



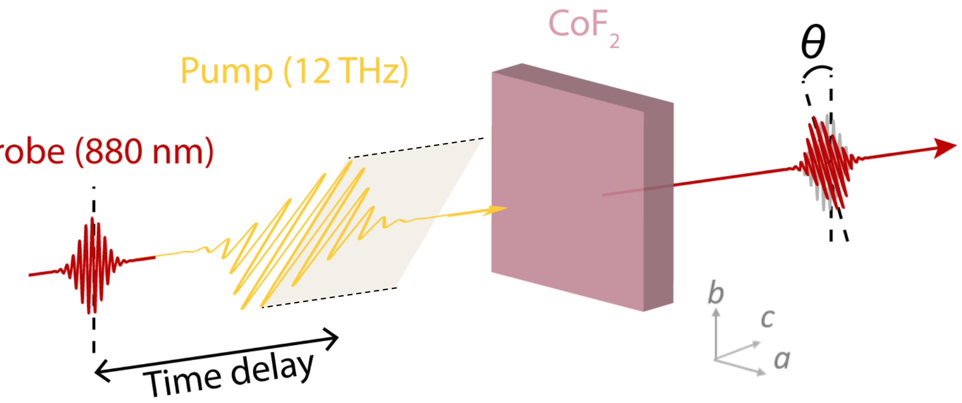
- Can change direction of magnetization relative phase of phonon excitation (polarization of pump)

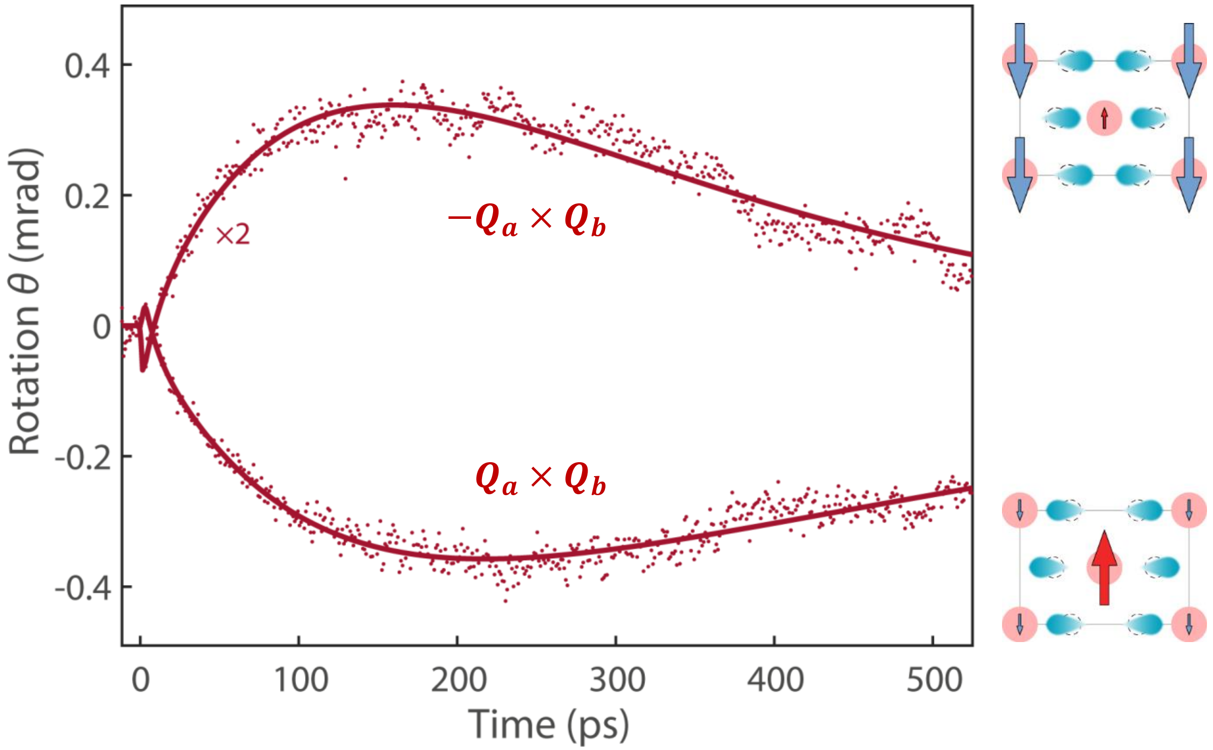
Controlling magnetization direction





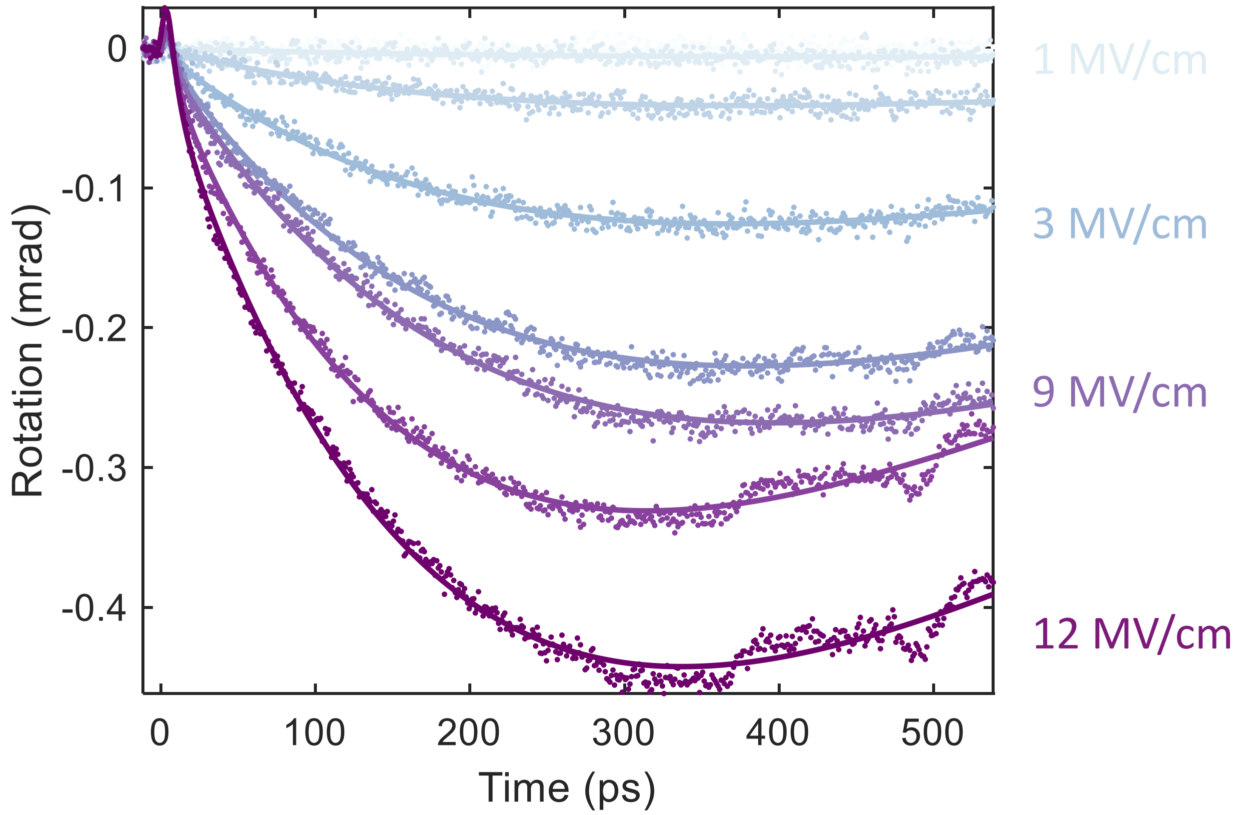
Controlling magnetization direction



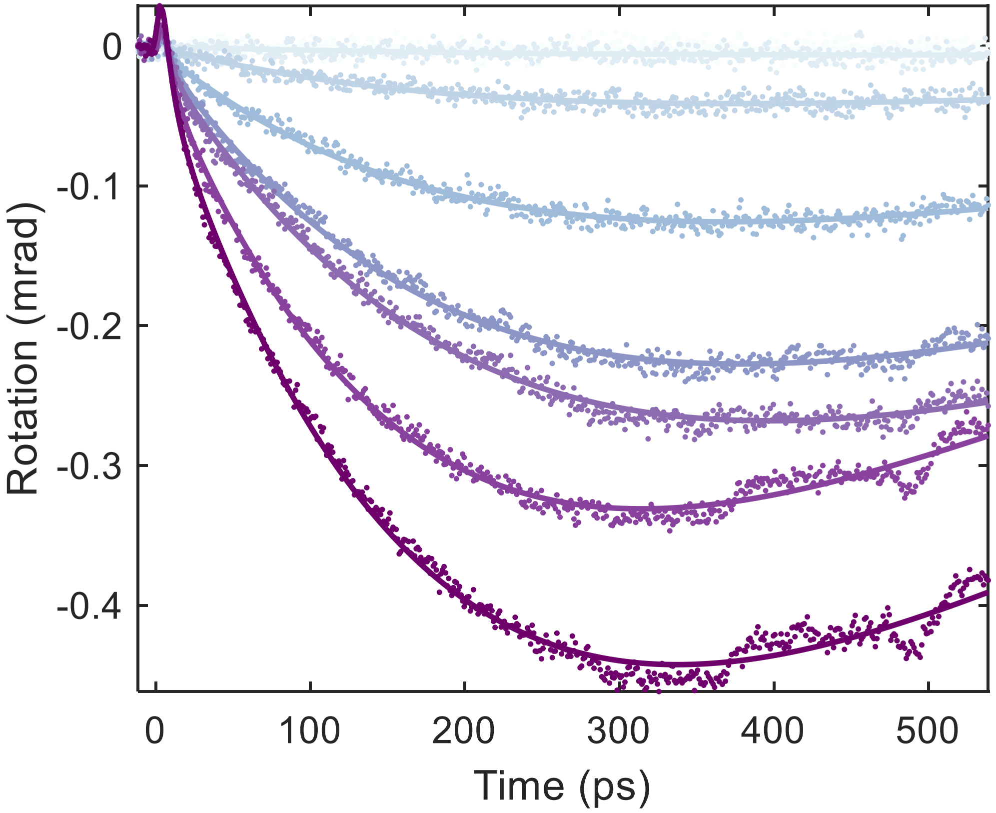


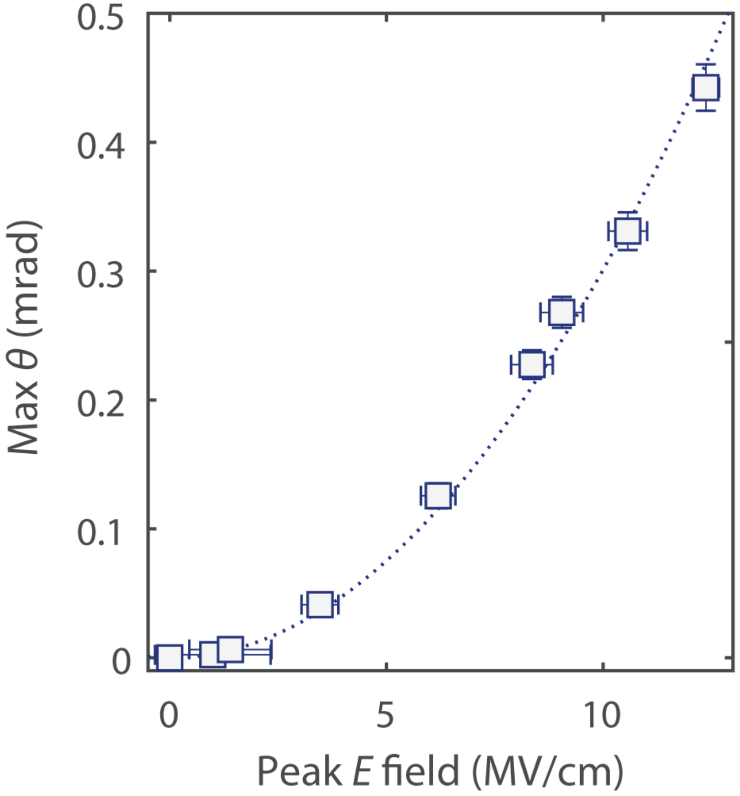
- Optical control over direction and magnitude of induced magnetization

Dependence on pump strength



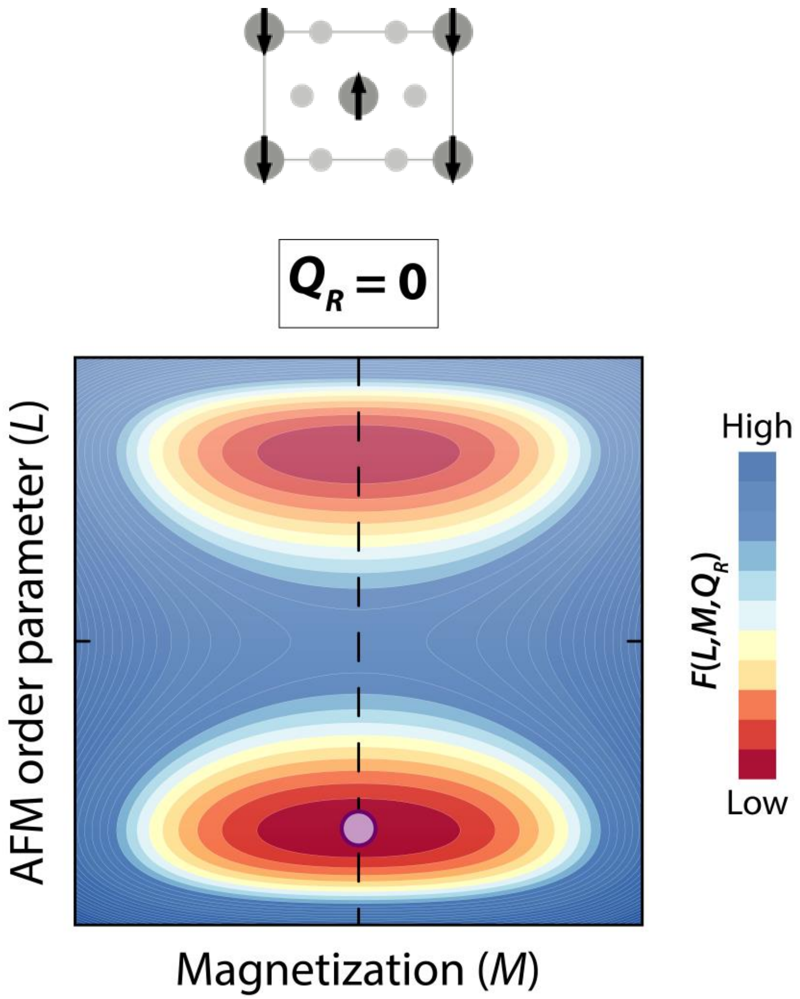
Dependence on pump strength





- Induced magnetization $\propto E^2 \rightarrow Q_R \propto Q_{IR,1} Q_{IR,2}$ ✓

Phenomenological model of dynamics



$$F(L, \delta m, Q_R) \approx -\frac{|a_L|}{2}L^2 + \frac{|b_L|}{4}L^4 + \frac{|\widetilde{a_M}|}{2}\delta m^2 + \frac{|\widetilde{a_R}|}{4}Q_R^2 + \lambda Q_R L \delta m,$$

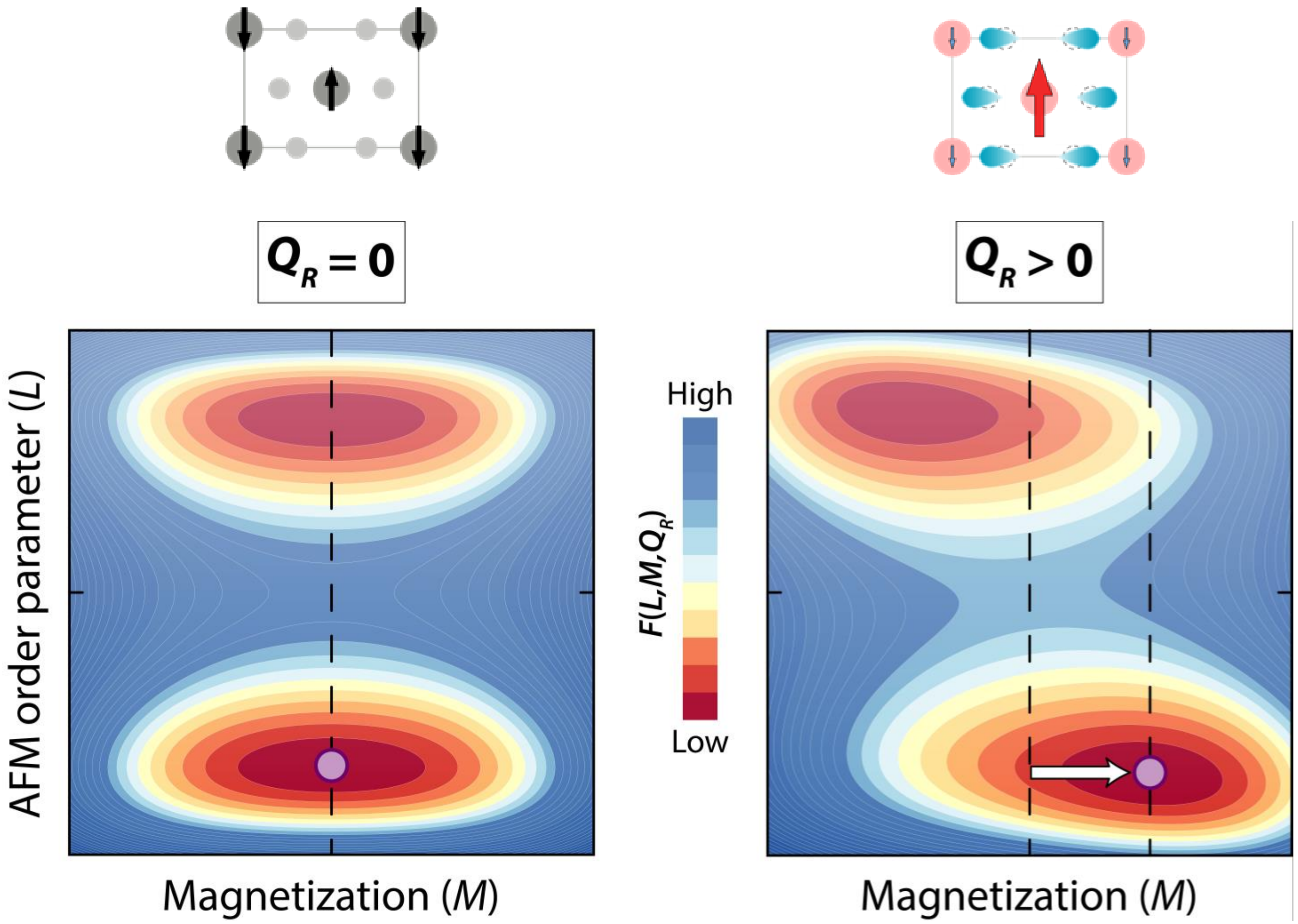
$$L = \left(m_1^0 + \frac{\delta m}{2}\right) - \left(m_2^0 + \frac{\delta m}{2}\right) = L_0$$

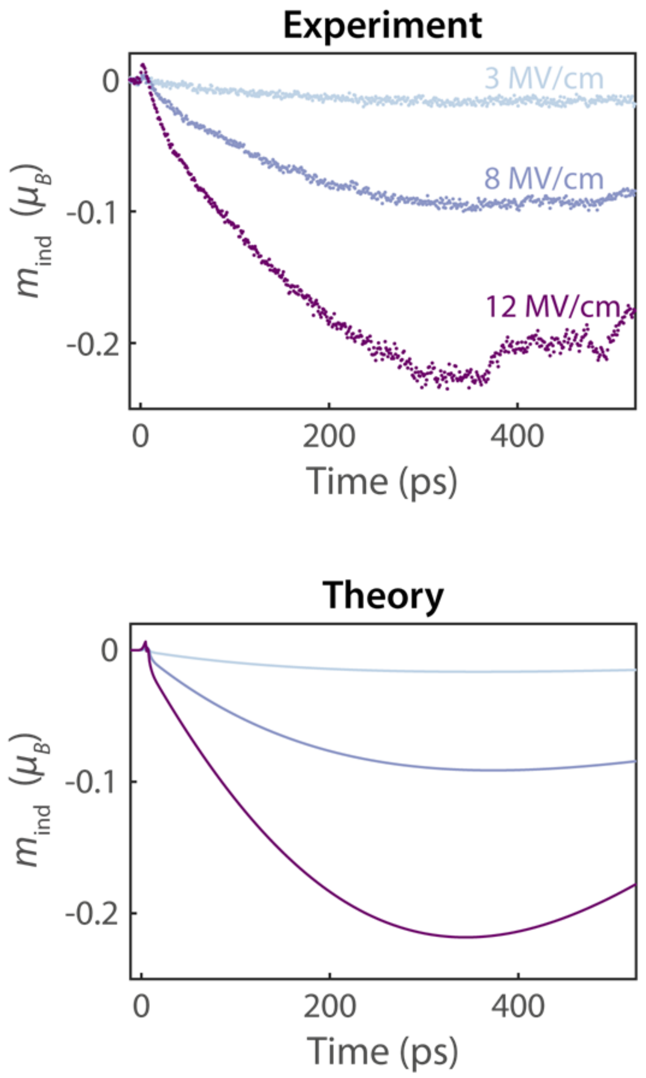
Fixed

$$M = \left(m_1^0 + \frac{\delta m}{2}\right) + \left(m_2^0 + \frac{\delta m}{2}\right) = \delta m,$$

New order parameter

Phenomenological model of dynamics



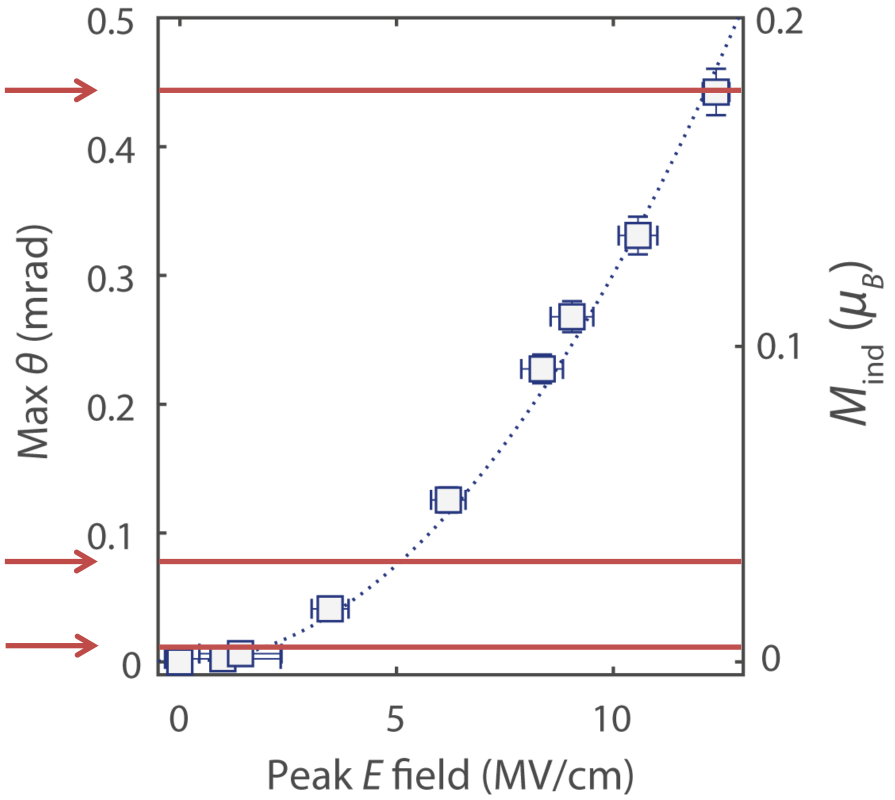


Strength of induced magnetization

Equivalent to ~50 GPa uniaxial stress →

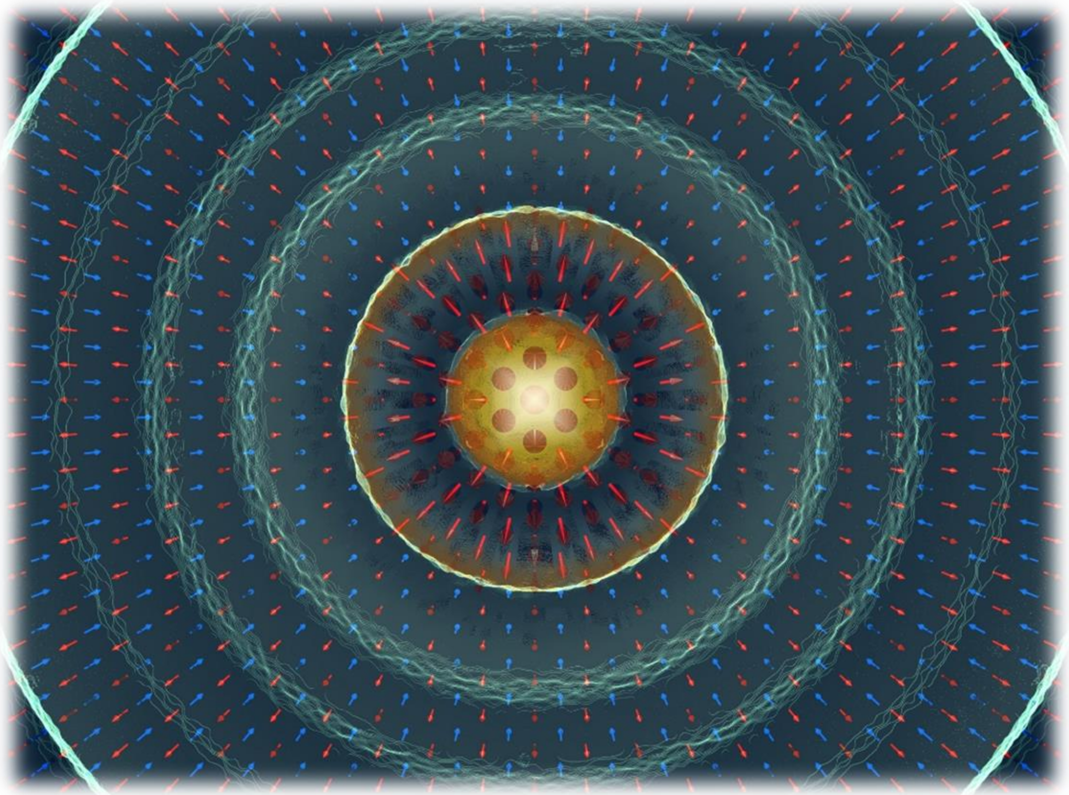
Tensile strength (strain at which crystal breaks) →

Largest strain-induced magnetization achieved →



Induced magnetic properties by nonlinear phonon excitation ~100× statically achievable

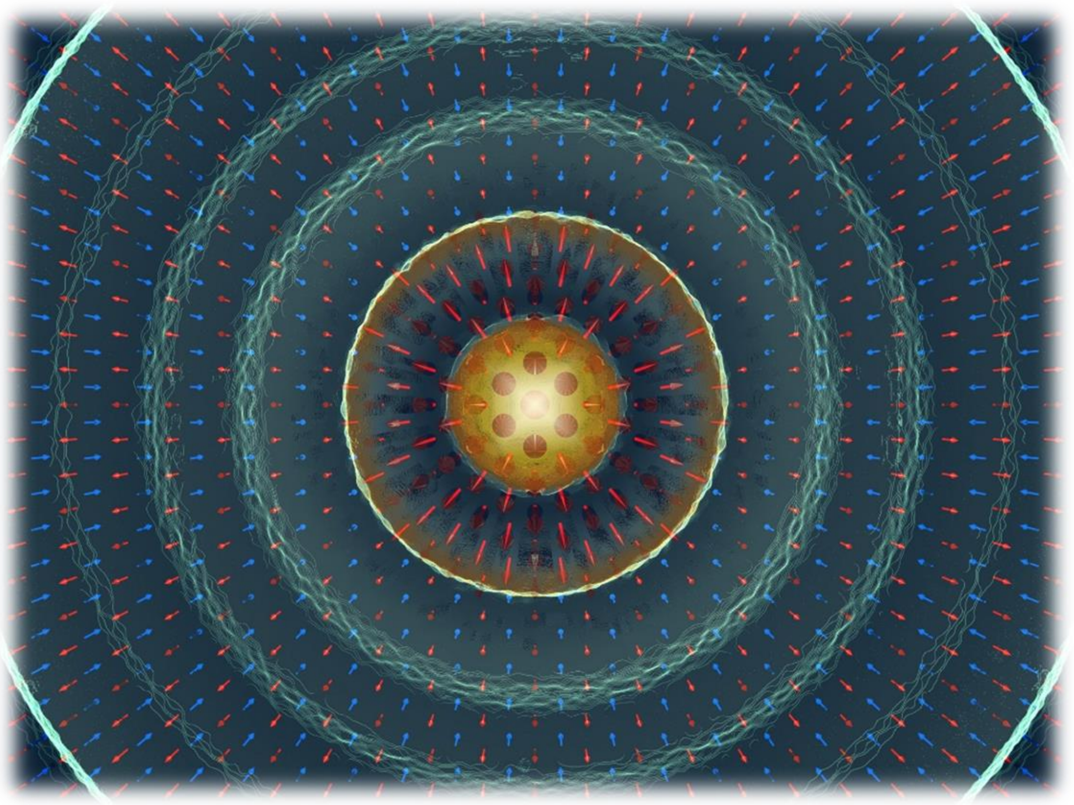
Recap: Optical lattice control of magnetism



Open questions:

- Why are the dynamics so slow? How can we speed up the effect?
- What’s happening to the angular momentum?
- How can we more accurately describe the longitudinal “switching”?

Enhancing non-equilibrium magnetism

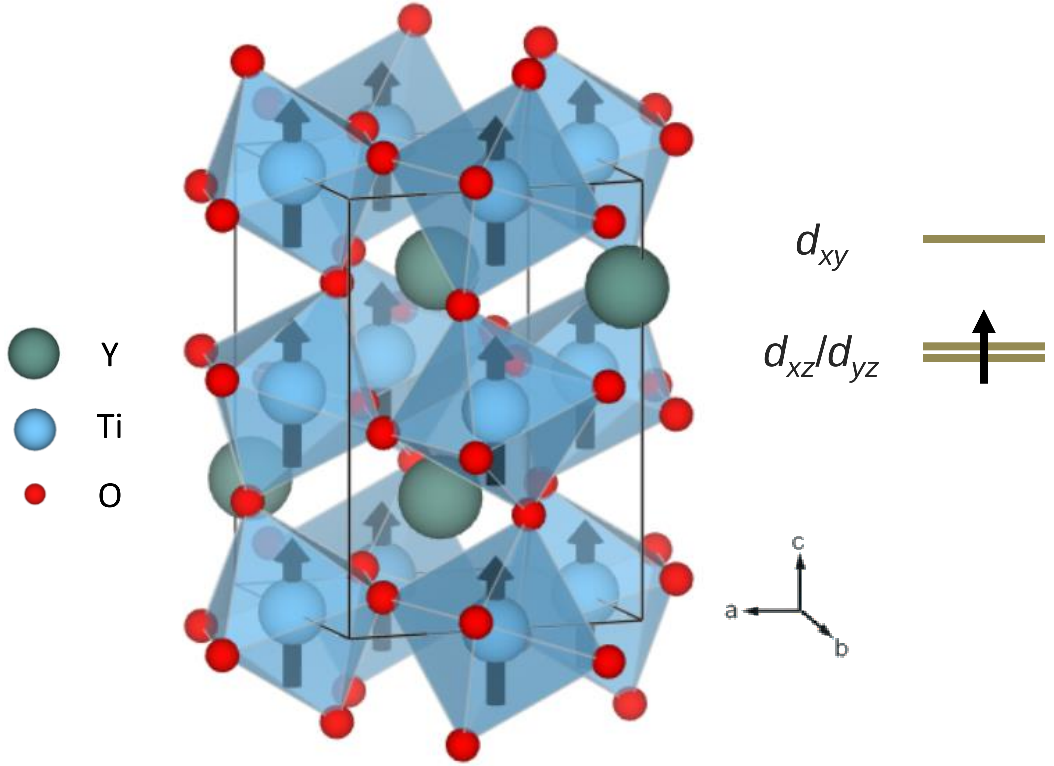


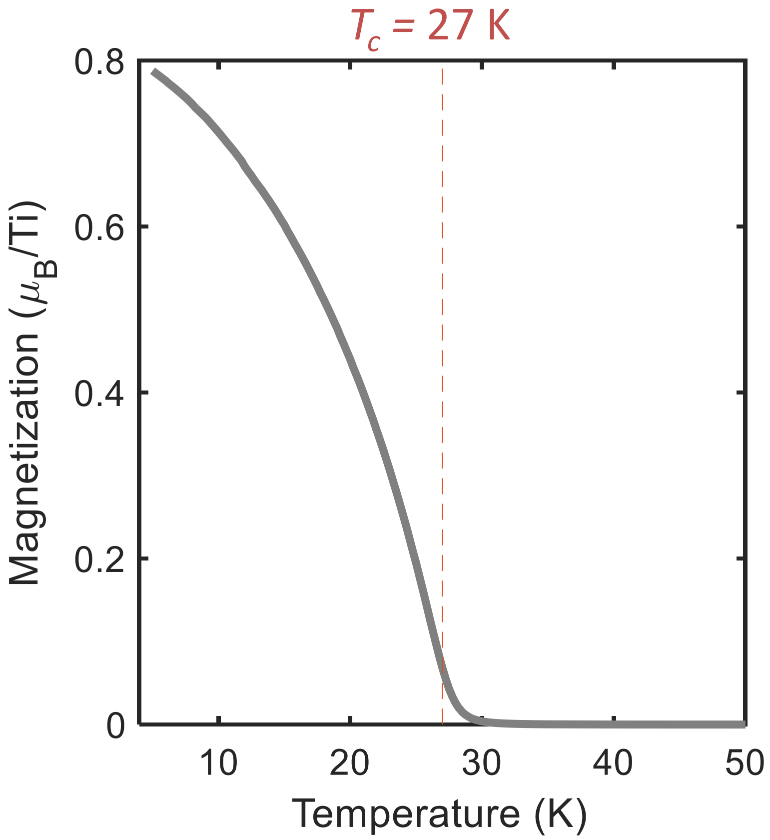
Can we extend non-equilibrium behavior to higher temperatures?

- Demonstrated control of magnetic state through crystal lattice ***below equilibrium T_c***

$YTiO_3$ – a fluctuating ferromagnet

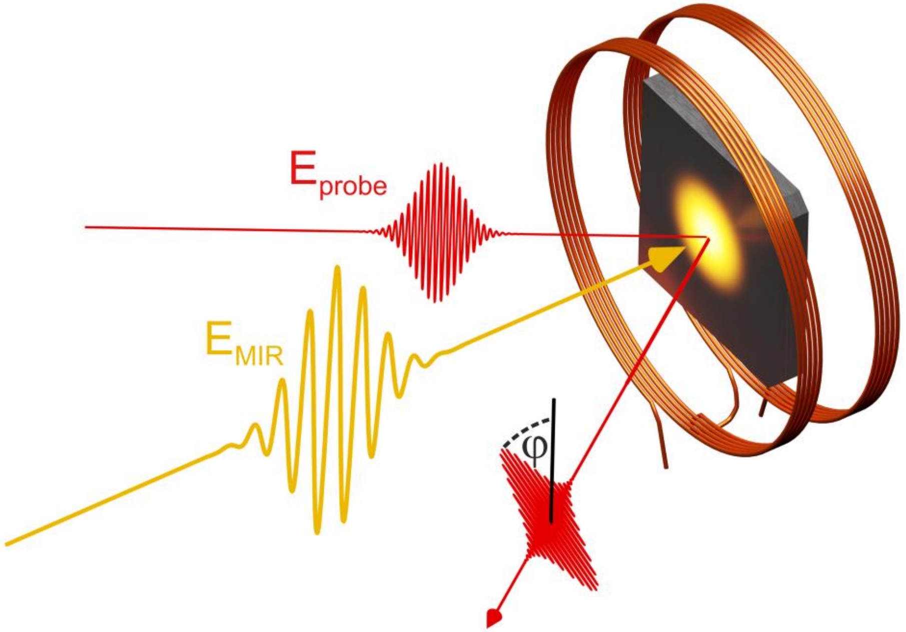
Ferromagnetic Mott insulator

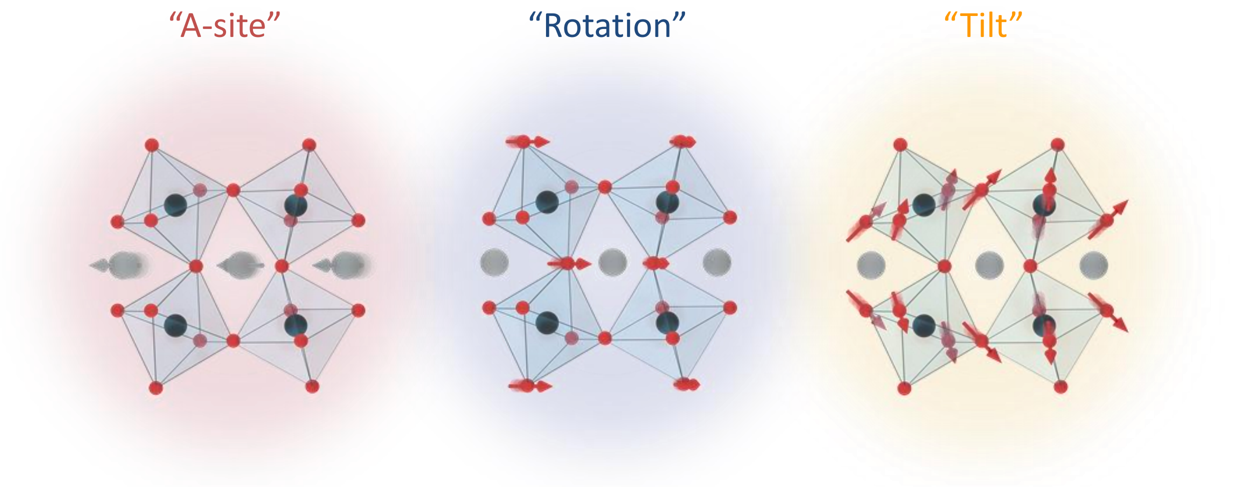


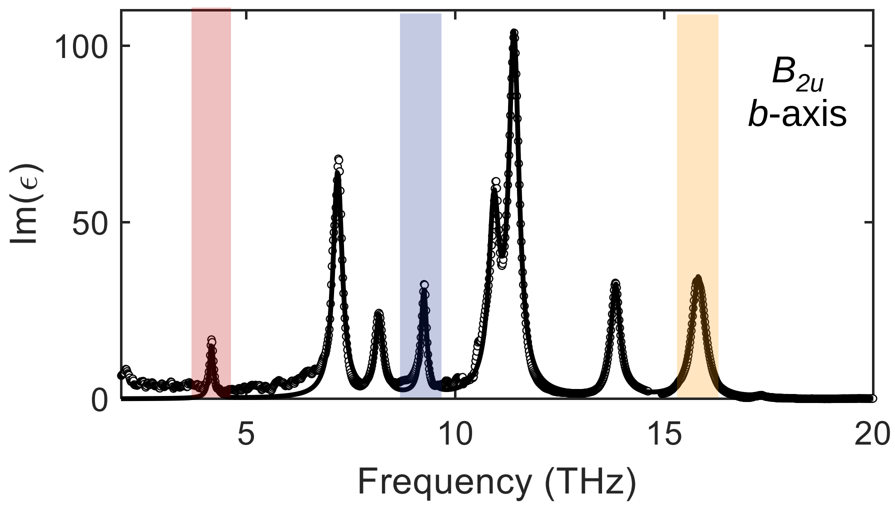


- **Magnetism highly coupled to crystal lattice and orbital configuration**

Engineering magnetism through the lattice





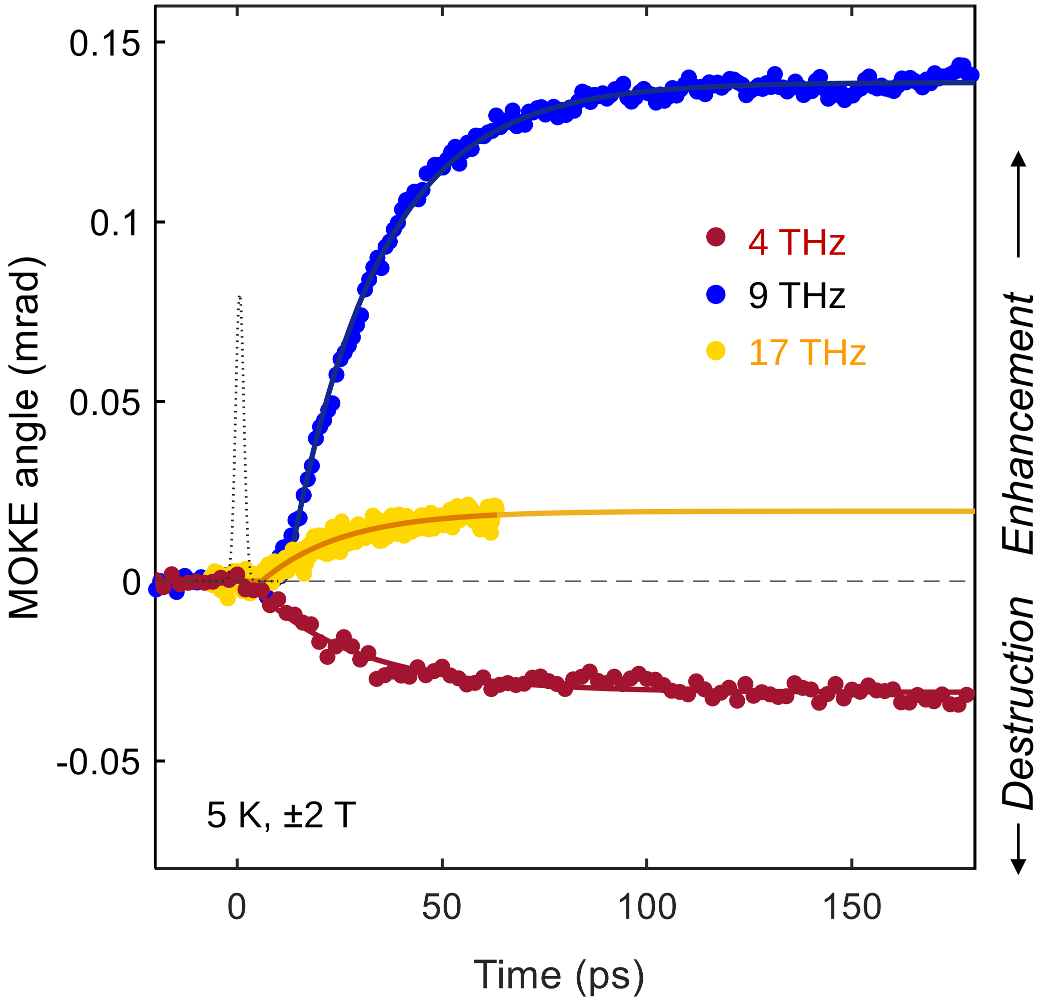


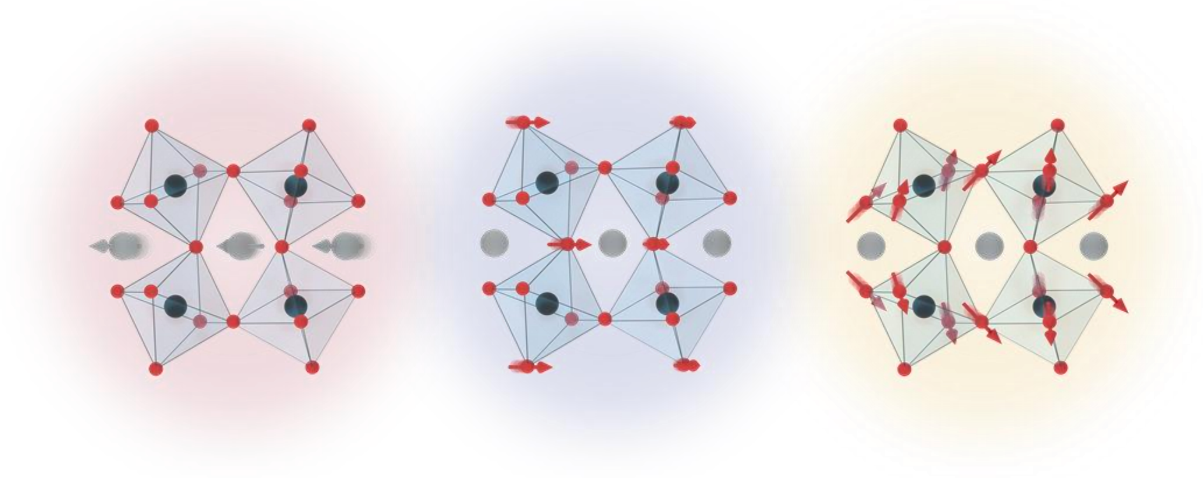
- **Time-resolved MOKE experiment**

$$\Delta\varphi(+H) - \Delta\varphi(-H) \propto \Delta M$$

Disa, *et al.*, arXiv:2111.13622

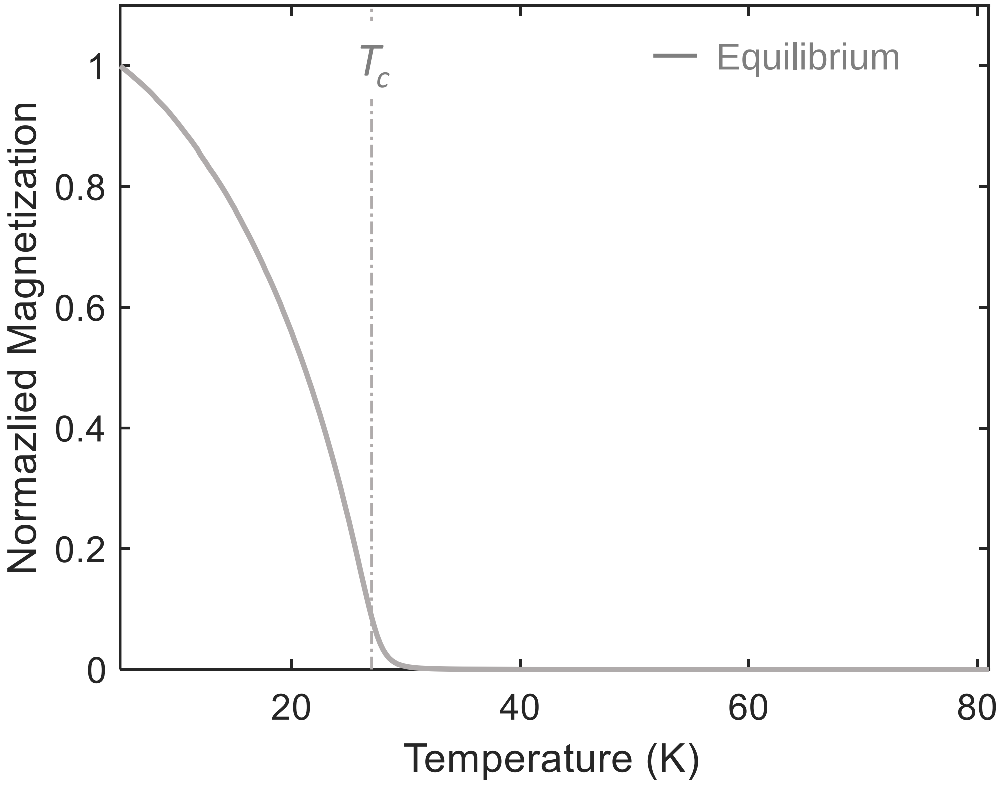
Effect of pumping different phonons



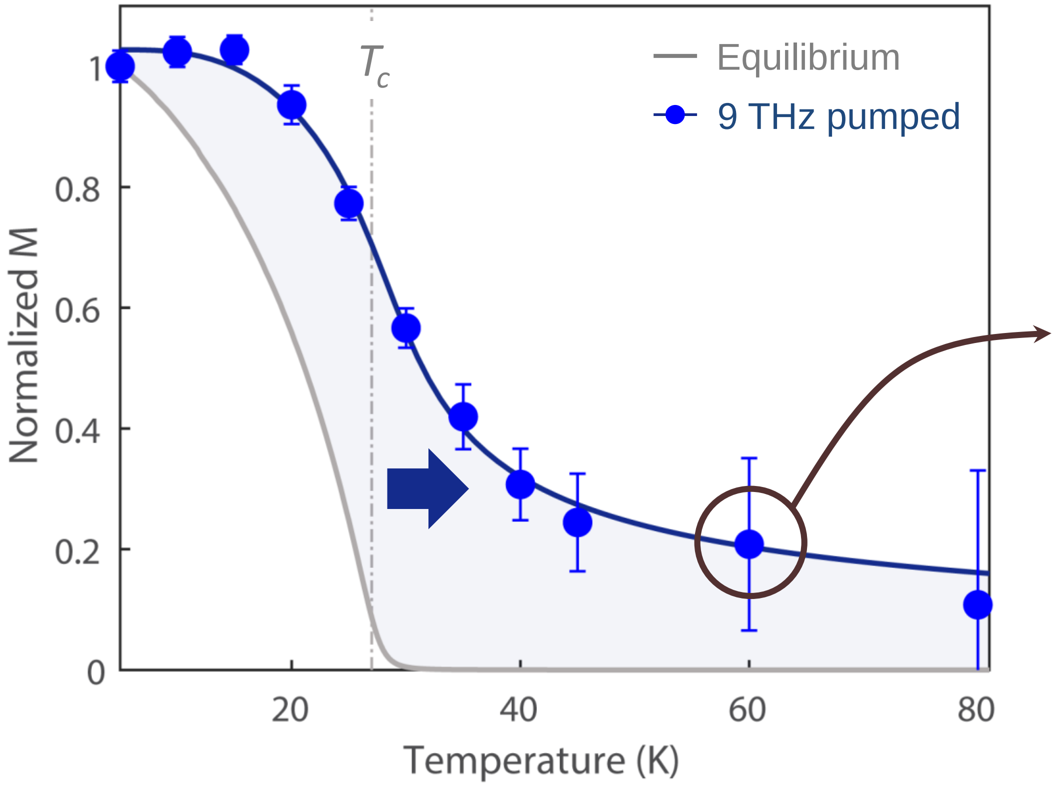


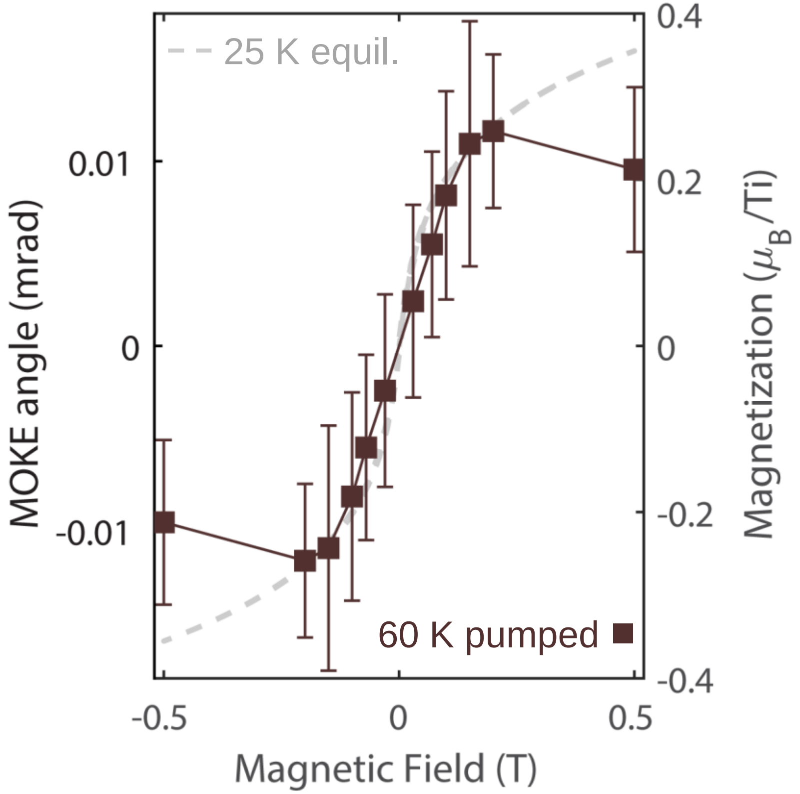
- **Phonon-selective manipulation of ferromagnetism** (below T_c)

Enhancement of magnetism **above** T_c



Enhancement of magnetism **above** T_c

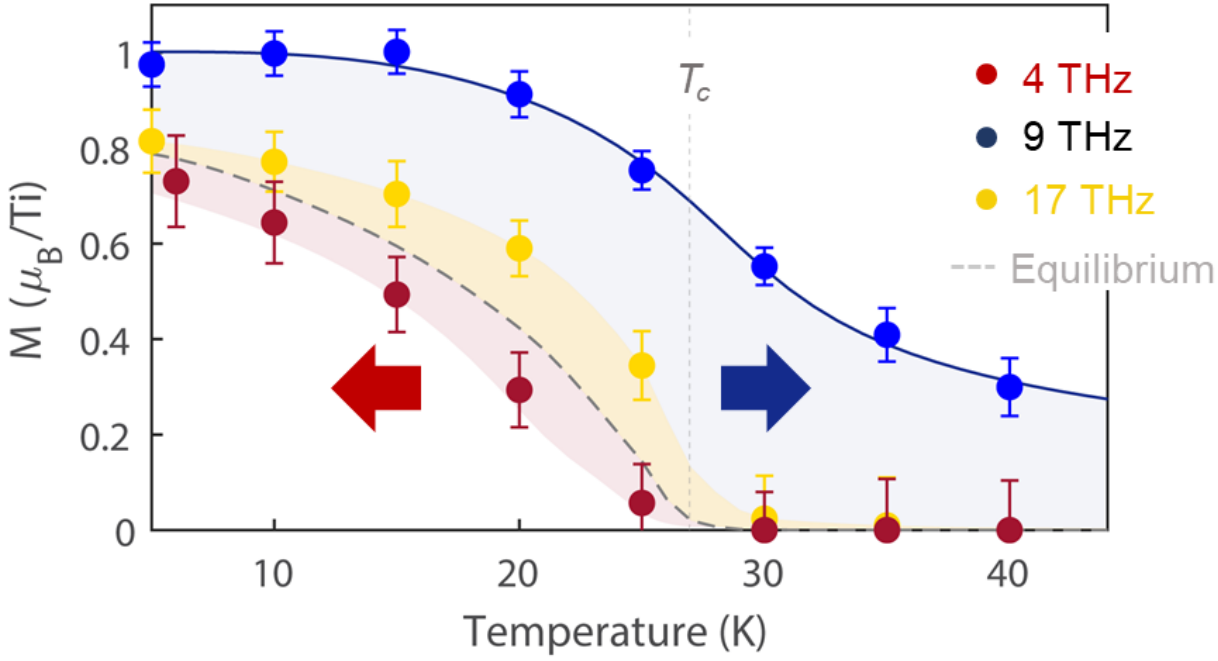


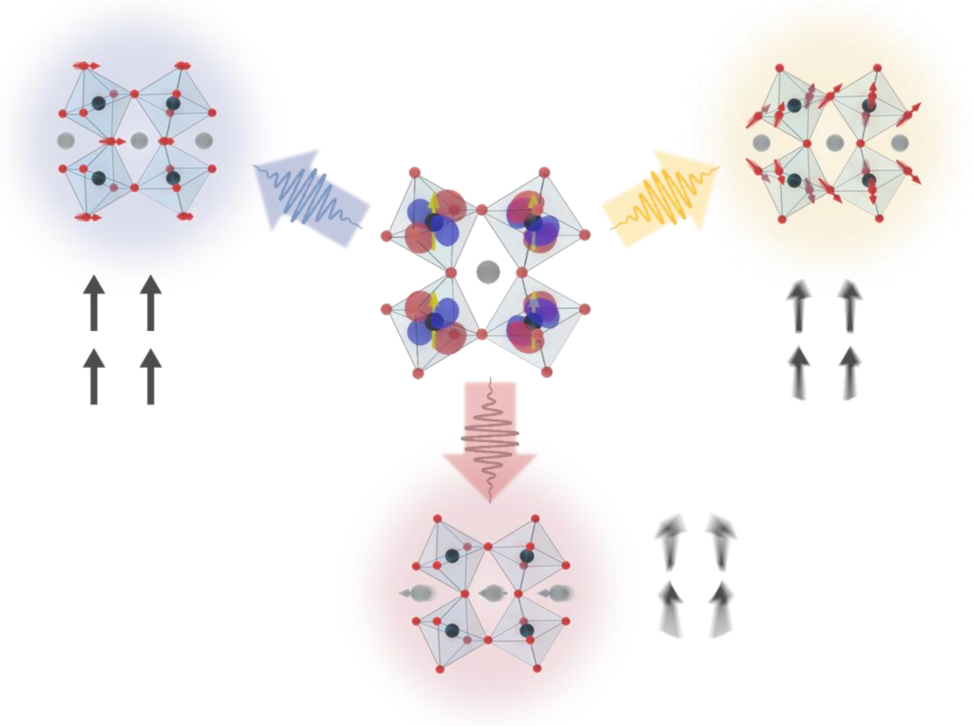


- Pump-induced magnetization up to **more than 3 × T_c**
- Non-equilibrium ferromagnetic state follows short-range spin correlations

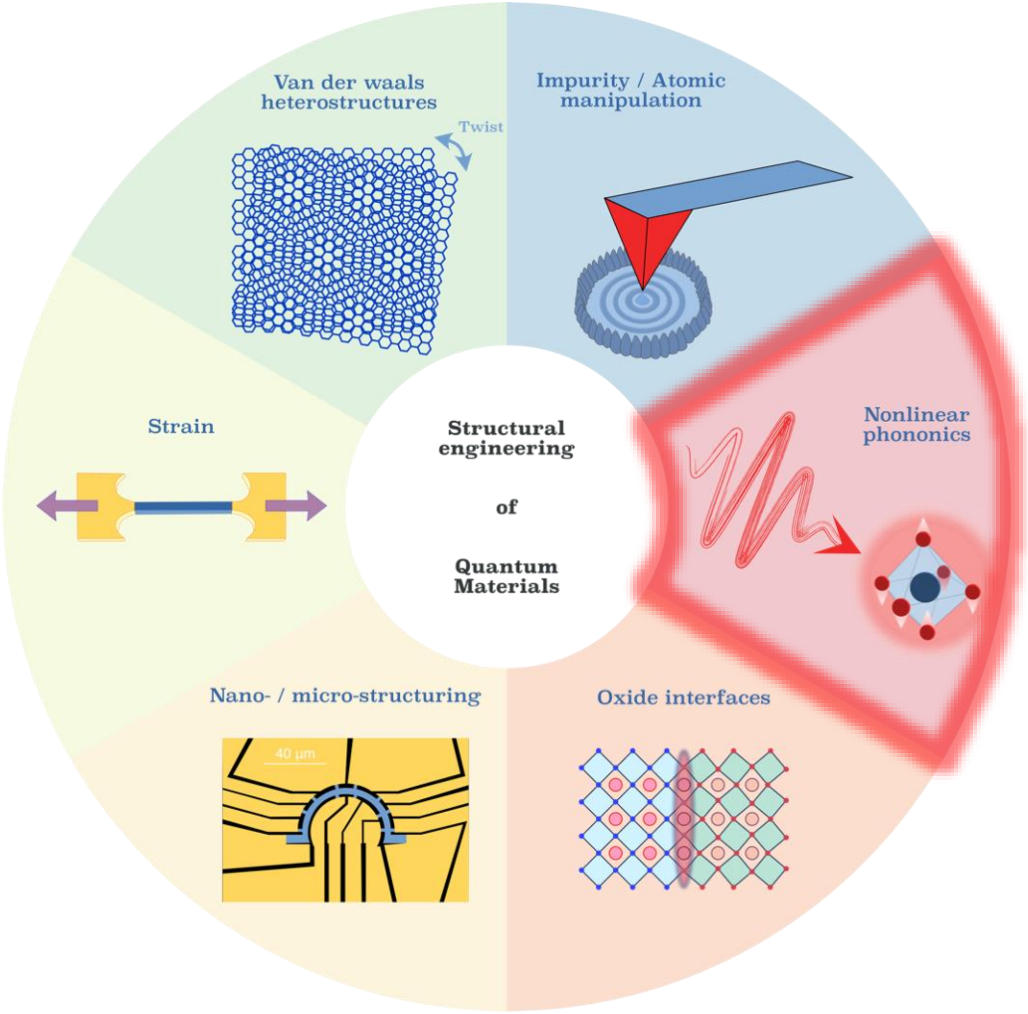
Picture of non-equilibrium ferromagnetism

- **Phonon driving enhances or weakens ferromagnetism through orbital state**

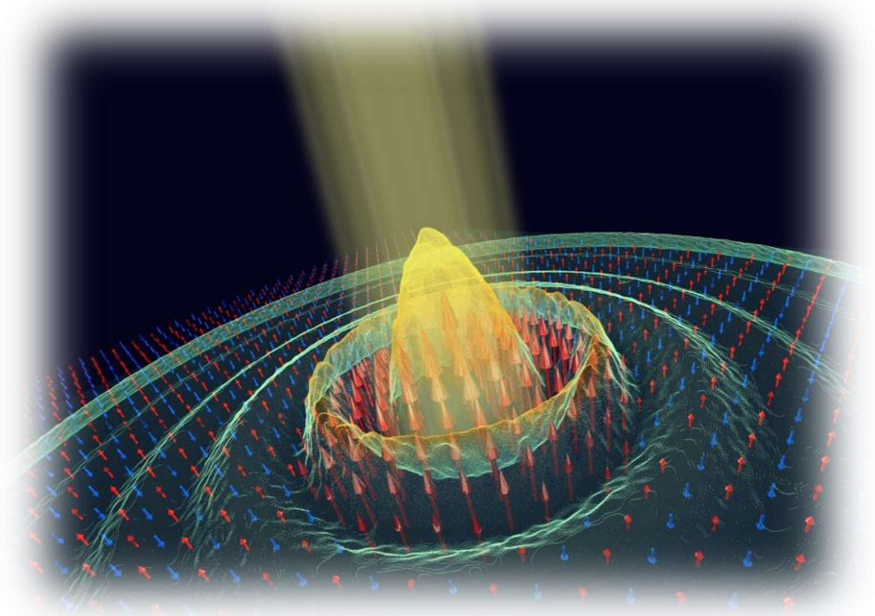




Take-home message



Driving the crystal lattice with light provides a powerful means to control magnetic order and induce non-equilibrium functionalities



A.S. Disa, *et al.* Nature Phys. **17**, 1087 (2021)

A.S. Disa, *et al.* Nature Phys. **16**, 937–941 (2020)

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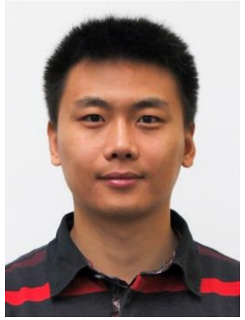
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- Moving to Cornell Applied & Engineering Physics in July 2022

- Students or postdocs with experience/interest in:
  - Ultrafast lasers
  - THz spectroscopy
  - Oxide heterostructures

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