How to engineer non-equilibrium crystal and magnetic structures with light

Ankit Disa

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

Max Planck Institute for the Structure and Dynamics of Matter (Hamburg, Germany)

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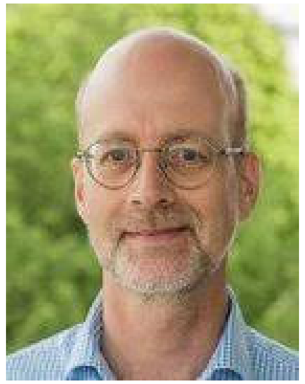
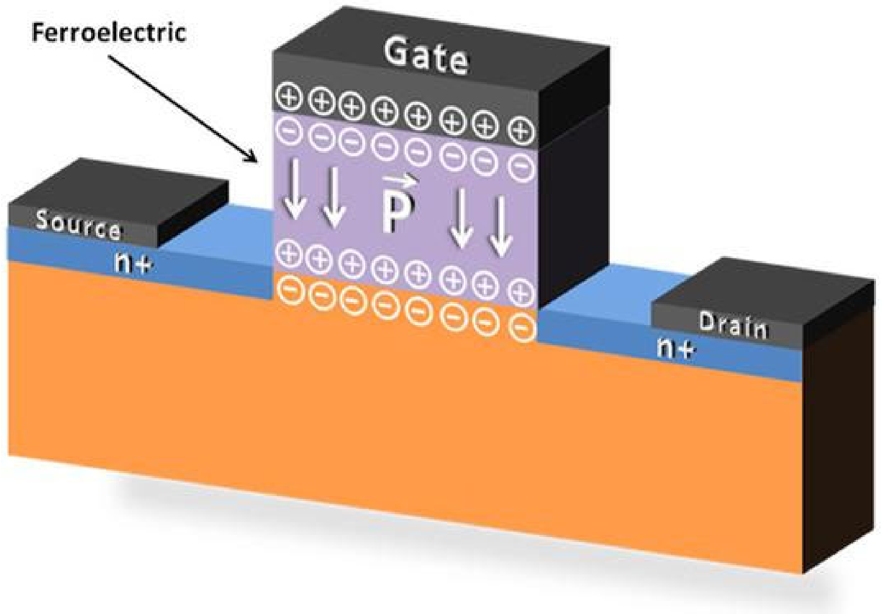
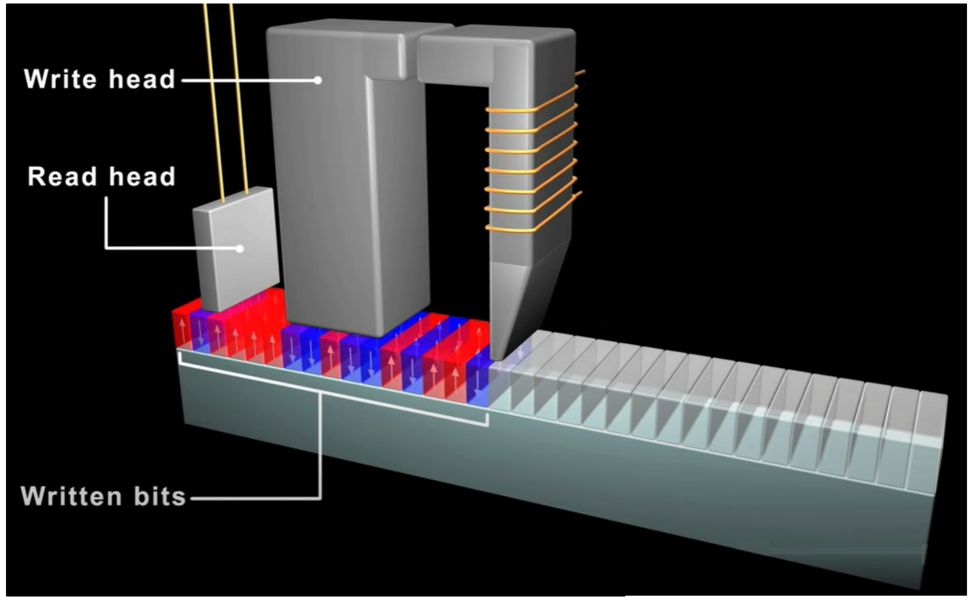

Bernhard Keimer

Controlling functional behavior





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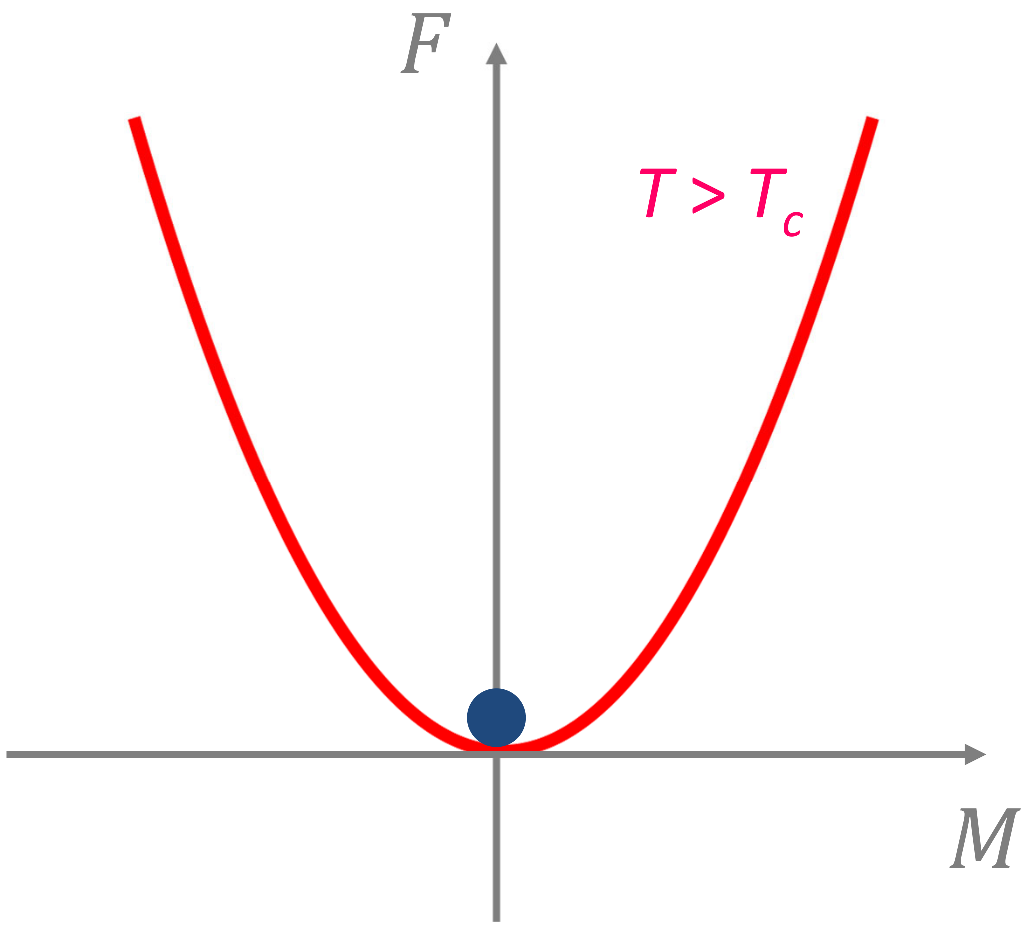
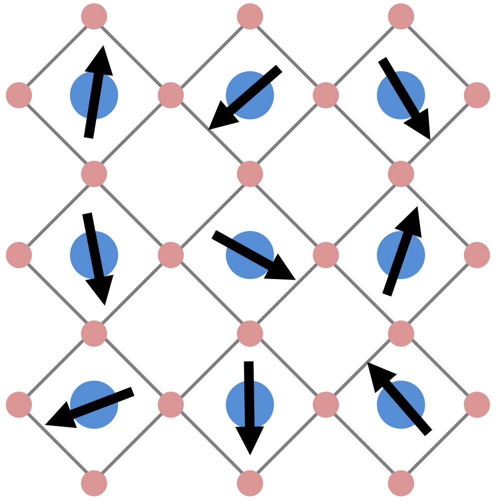
Simple approach to functional control



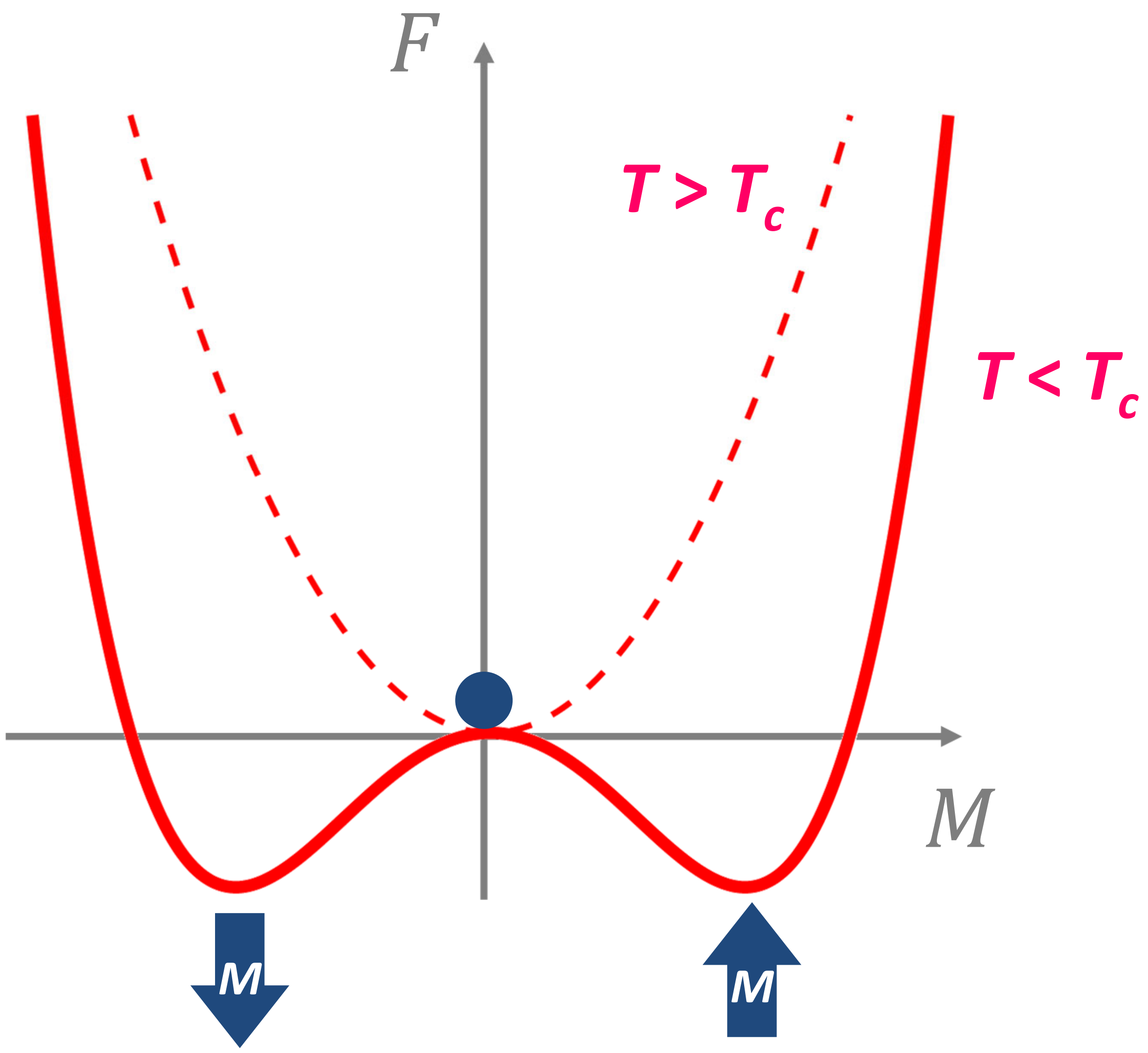
- Thermodynamic free energy:

$$F(T, M) \approx F_0 + \alpha(T - T_c)M^2 + bM^4 + \cdots$$

Paramagnet



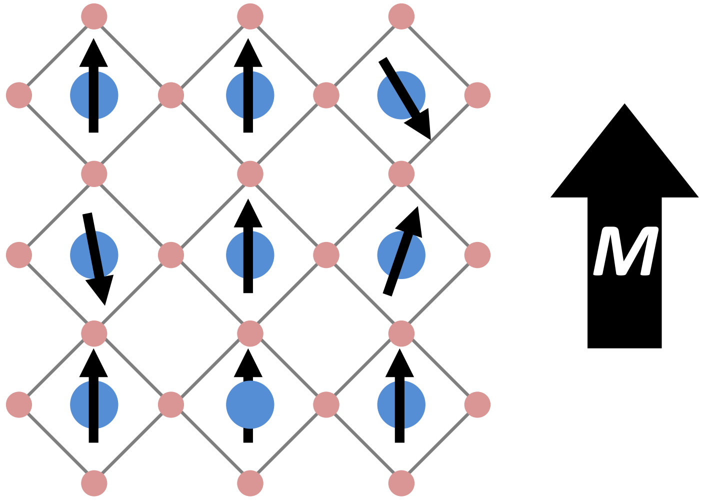
Simple approach to functional control



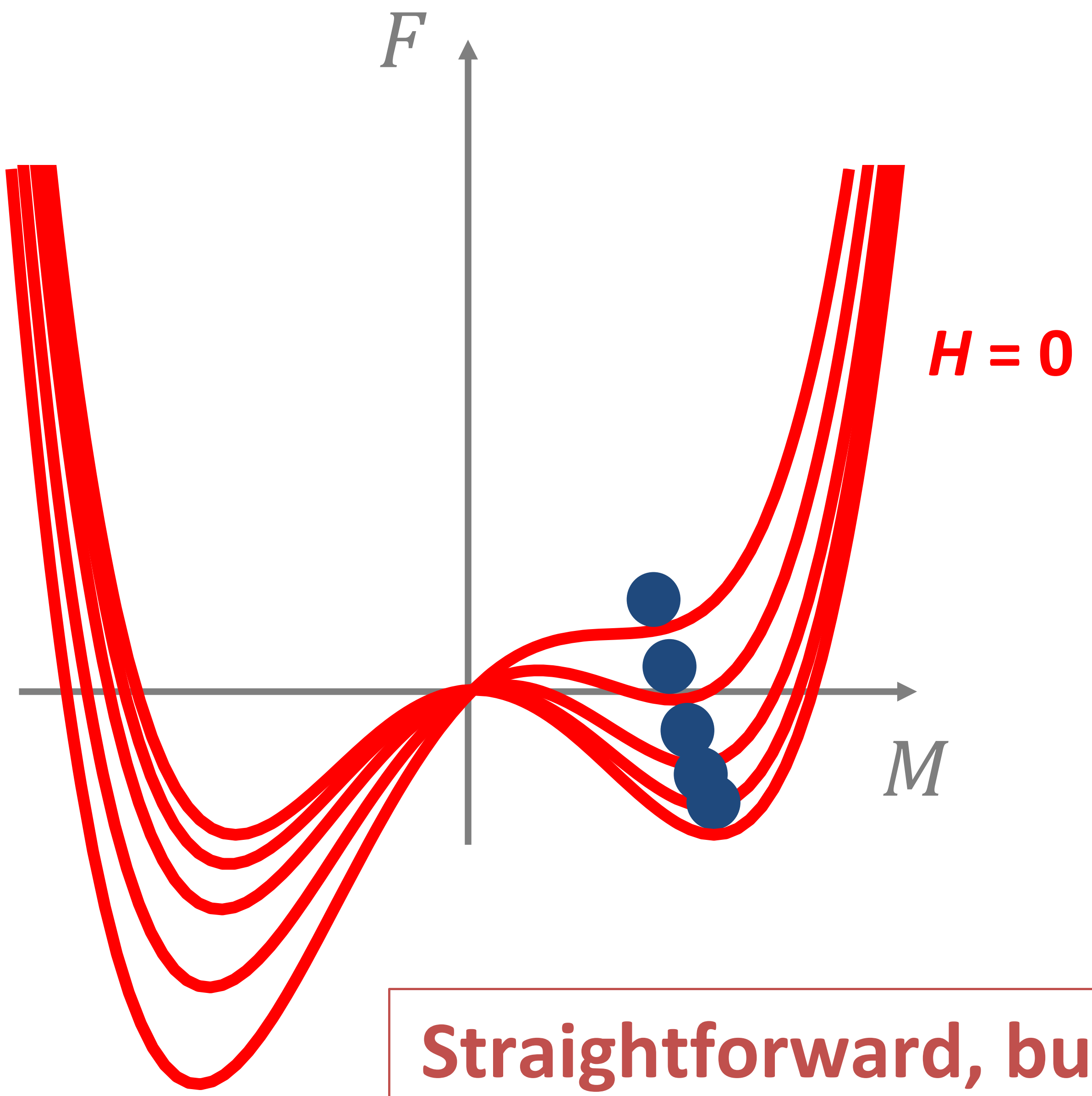
- Thermodynamic free energy:

$$F(T, M) \approx F_0 + \alpha(T - T_c)M^2 + bM^4 + \cdots$$

Ferromagnet



Simple approach to functional control

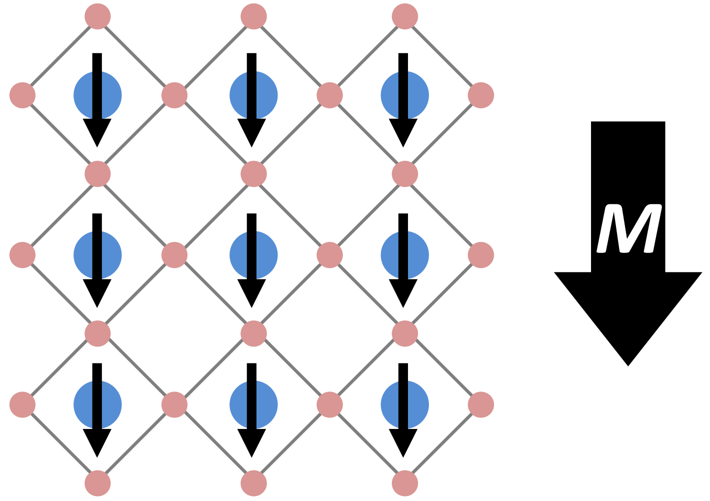


- Thermodynamic free energy:

$$F(T,M,H) \approx F_0 + \alpha(T - T_c)M^2 + bM^4 + \cdots$$
$$-\mu_0 H \cdot M$$

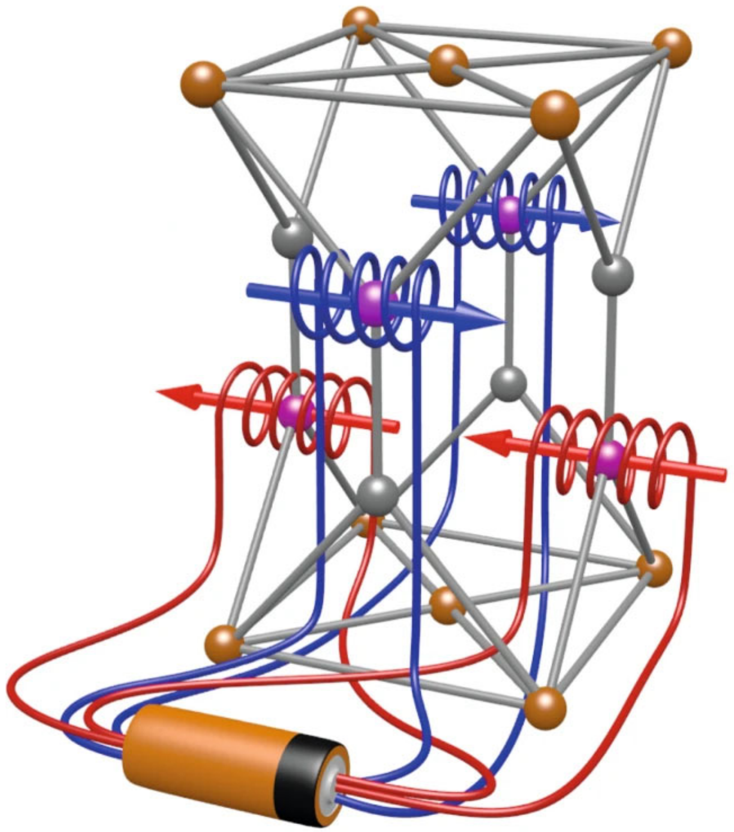
Coupling to external field

Ferromagnet



Straightforward, but limited to certain systems

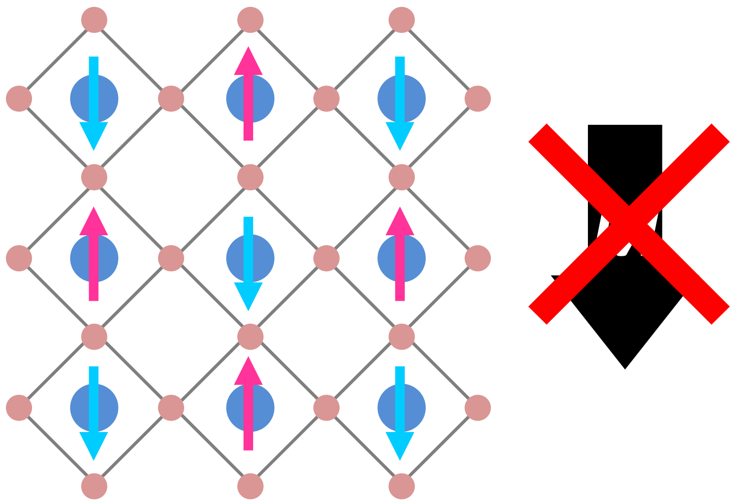
More complicated case: Antiferromagnets



- Thermodynamic free energy:

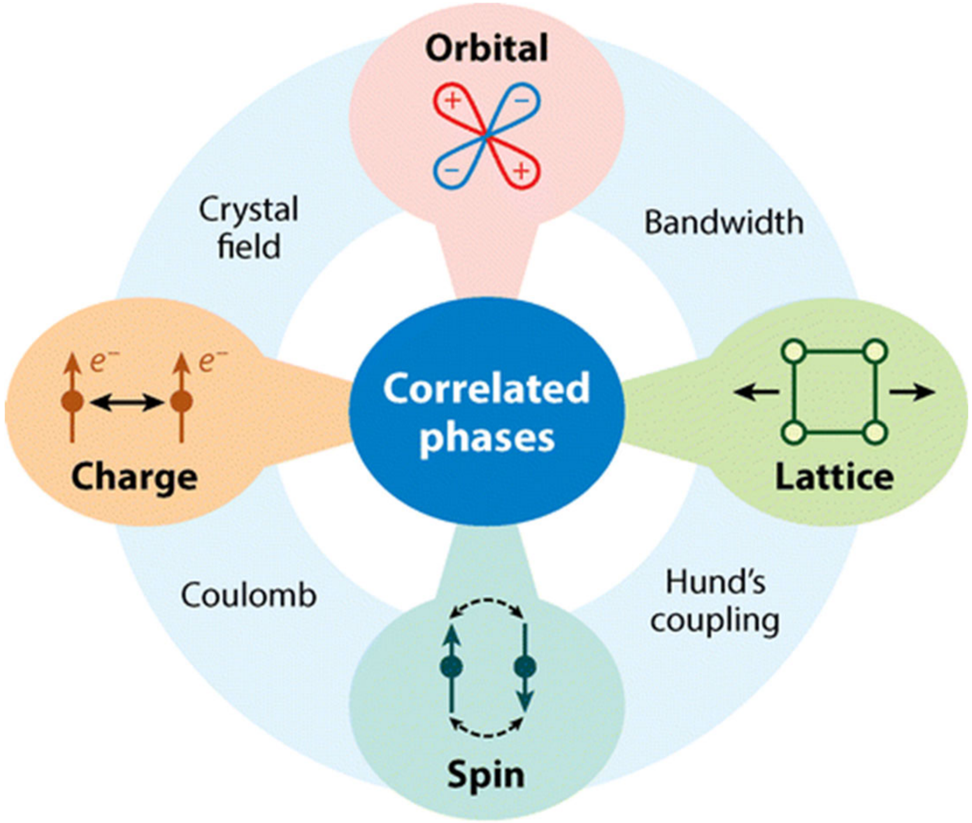
$$F(T,M,H) \approx F_0 + a(T-T_c)M^2 + bM^4 + \cdots$$
$$-\mu_0 H\cdot M$$

Antiferromagnet



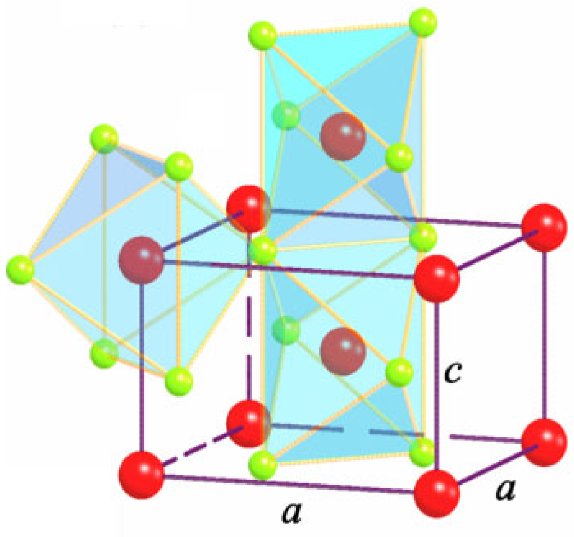
Alternative approach for controlling macroscopic phases of materials?

A structural approach to functional control

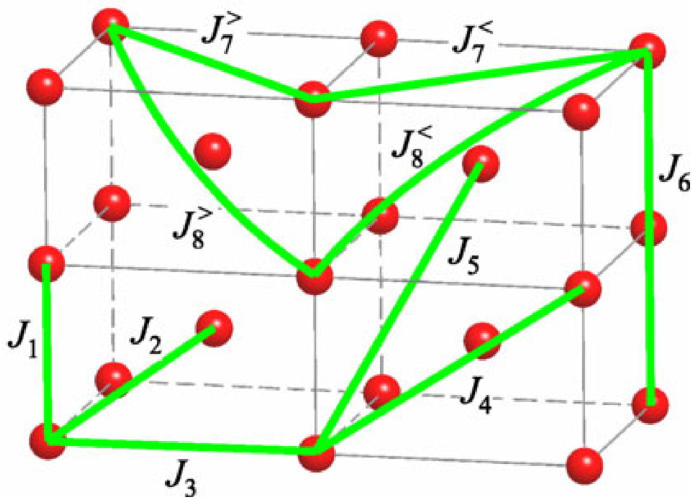


Provides control over wide array of electronic/magnetic phases

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



Symmetry, crystal field
→ Anisotropy

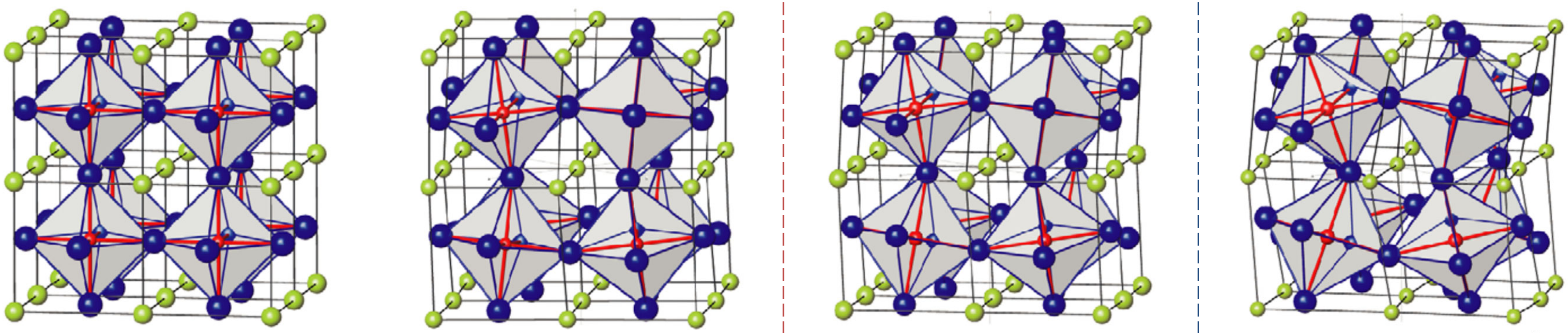


Bond lengths, angles
→ Exchange

Example of extreme structural sensitivity

Transition metal oxides (*d^1* electronic configuration):

$SrVO_3$ | $CaVO_3$ | $LaTiO_3$ | $YTiO_3$

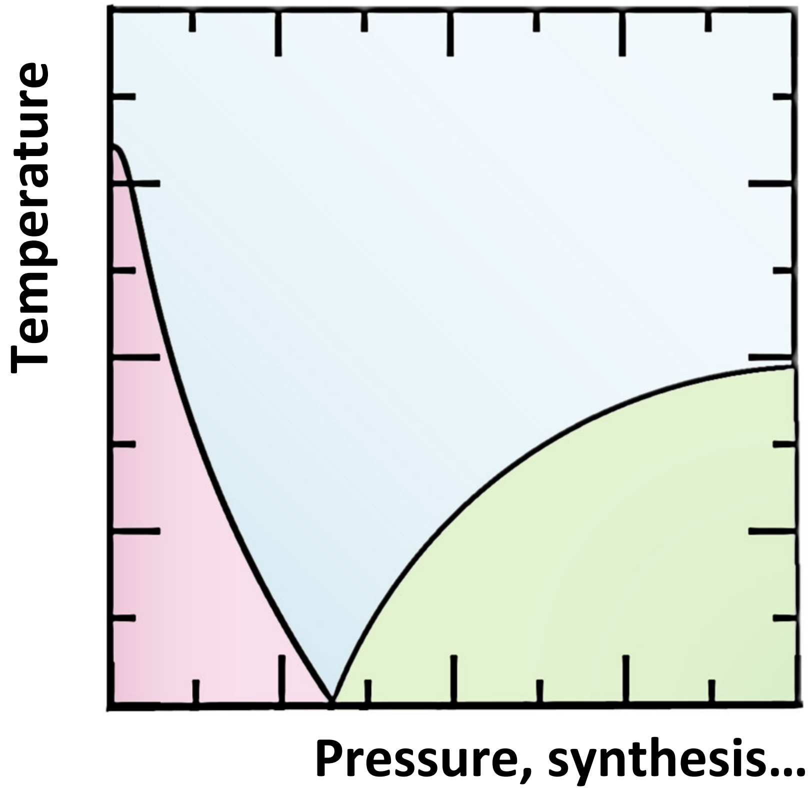


Increasing structural distortion

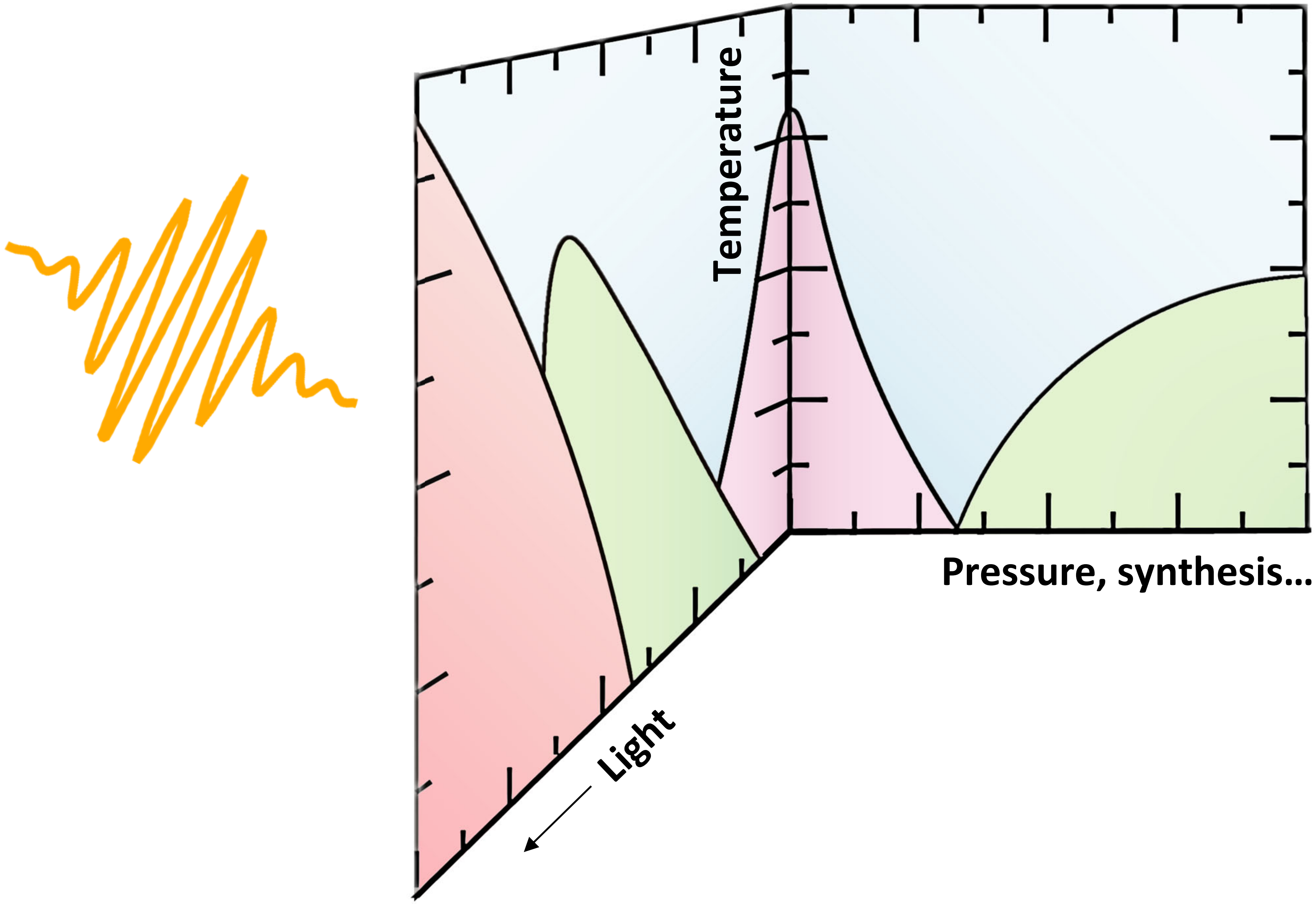
Metallic | Insulating

Paramagnetic | Antiferromagnetic | Ferromagnetic

Controlling electronic/magnetic phases



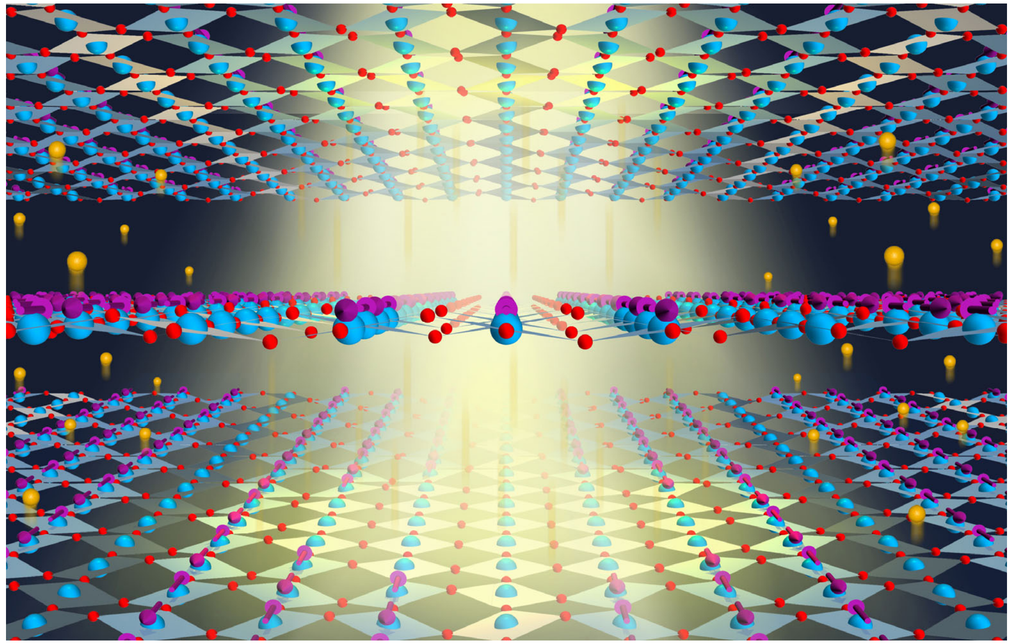
Controlling electronic/magnetic phases



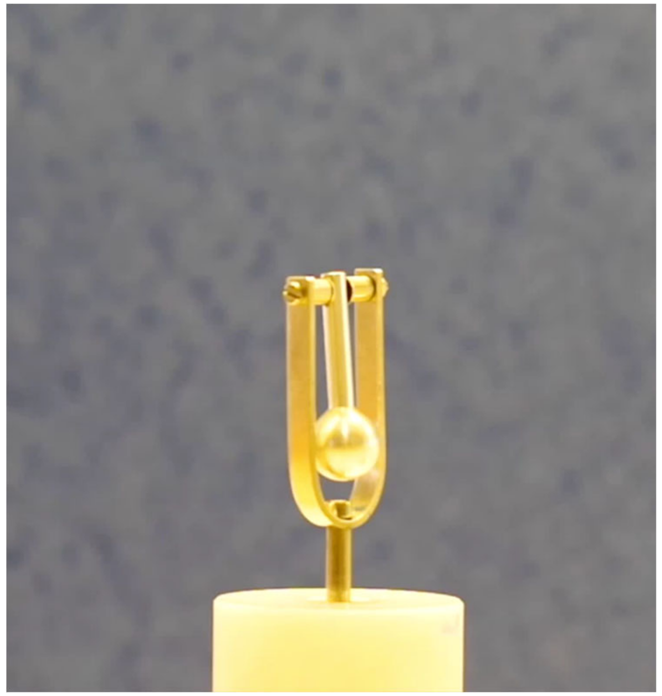
Can we:

- **Control emergent phenomena on ultrafast time scales?**
- **Go beyond the equilibrium phase diagram?**
- **Induce new phases and functionalities?**

Optical control of functional behavior



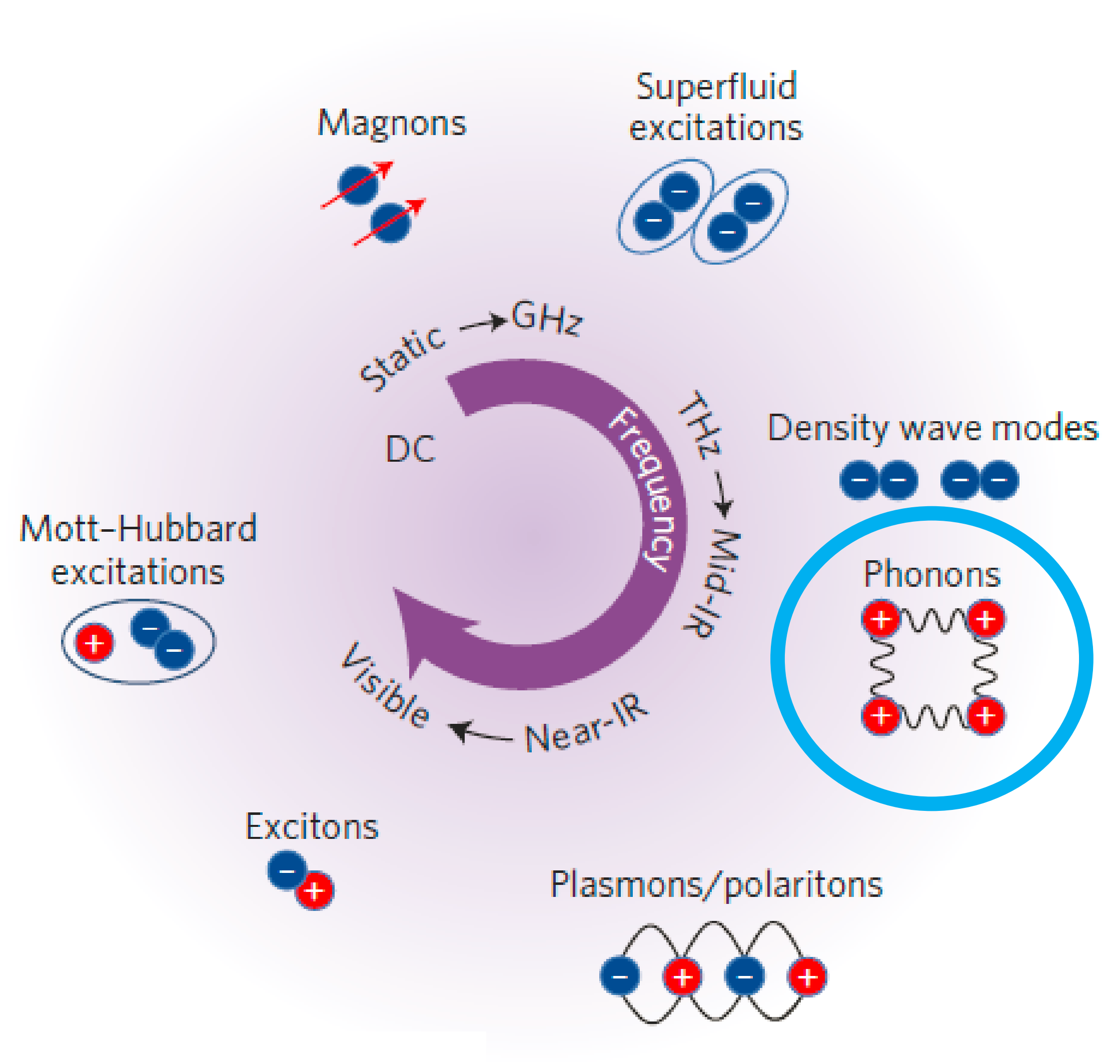
Ex) Kapitza's pendulum



- **Use light to dynamically control macroscopic properties and phases**
- **Driven systems often form states that cannot be realized in equilibrium**

Mode selective control

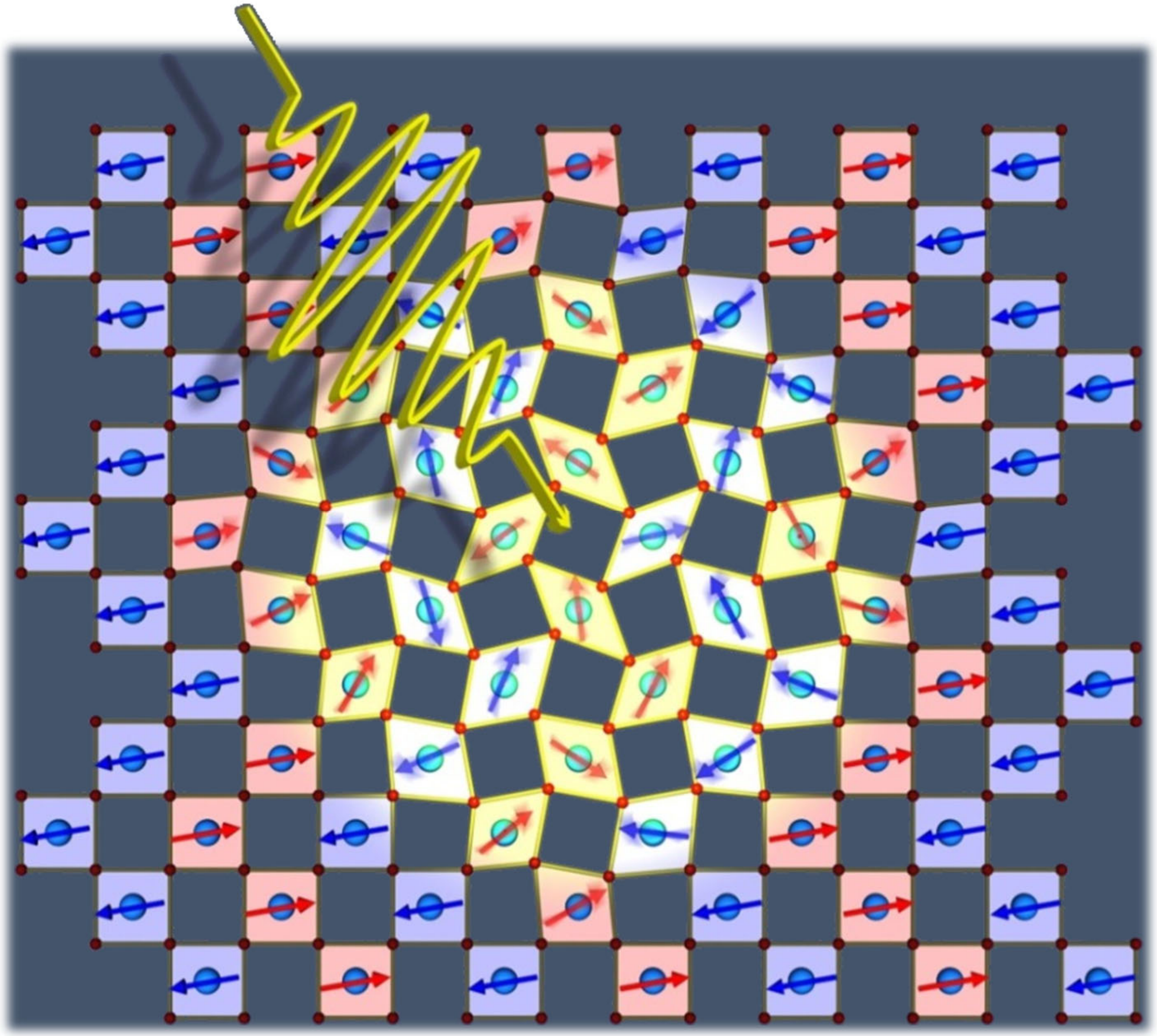
Selectively excite specific degrees of freedom by driving collective modes in solids at their natural energy scales



Engineering structure with light

Drive large amplitude structural distortions with laser pulses

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



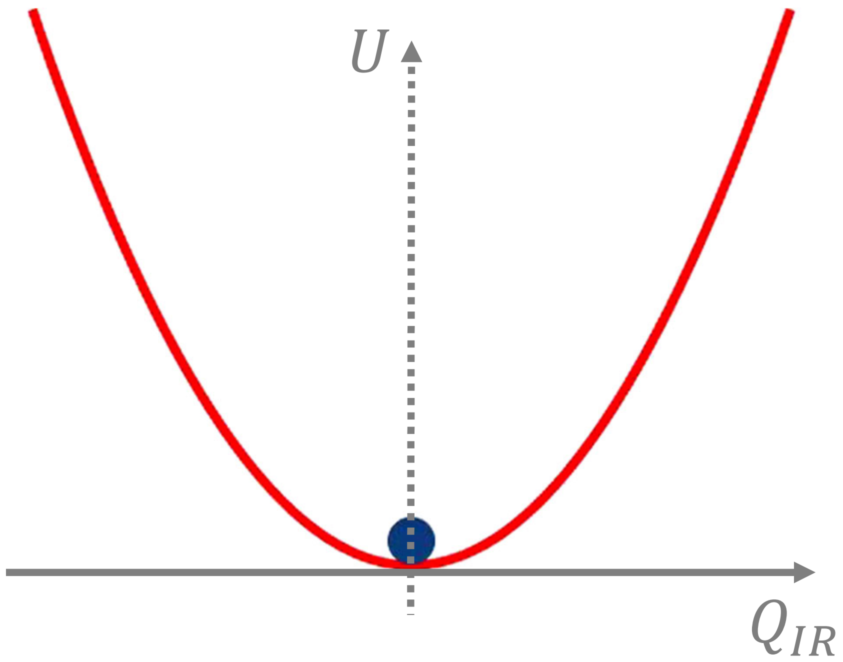
~MV/cm electric fields → 5-10% atomic displacements

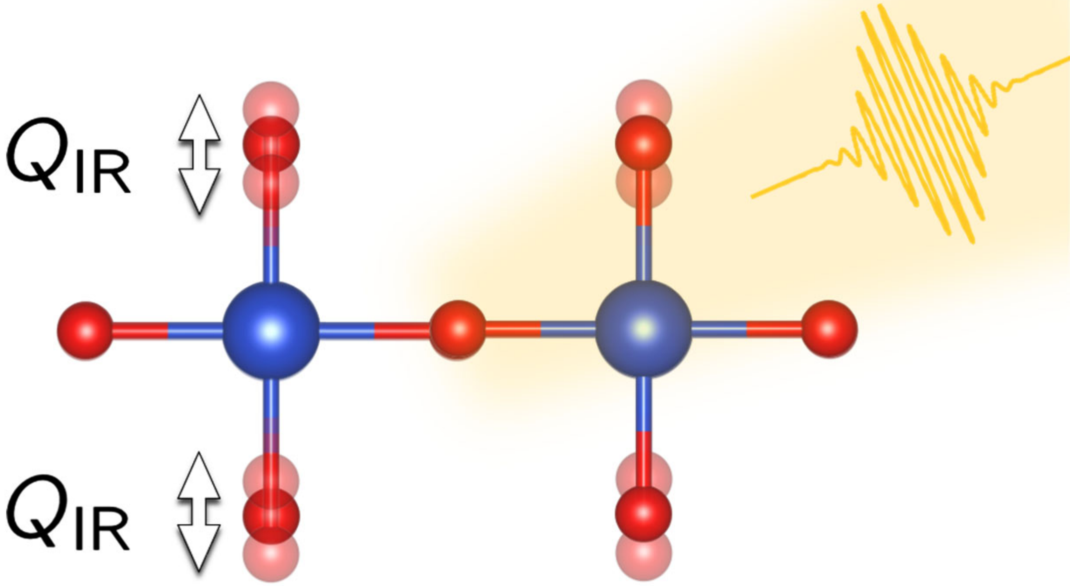
Leads to highly nonlinear response of crystal lattice to access new functionalities

Linear excitation of lattice modes

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

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



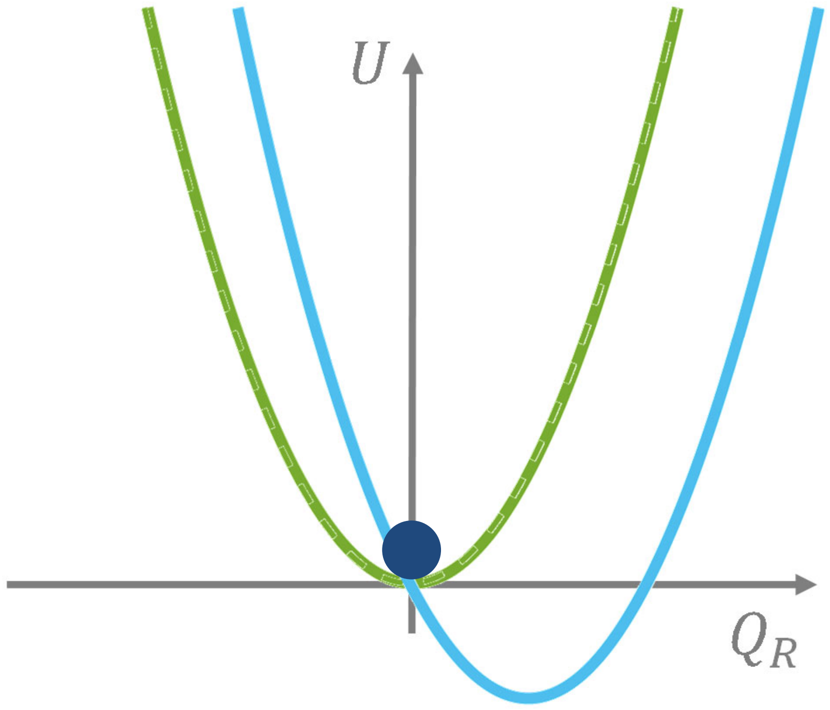


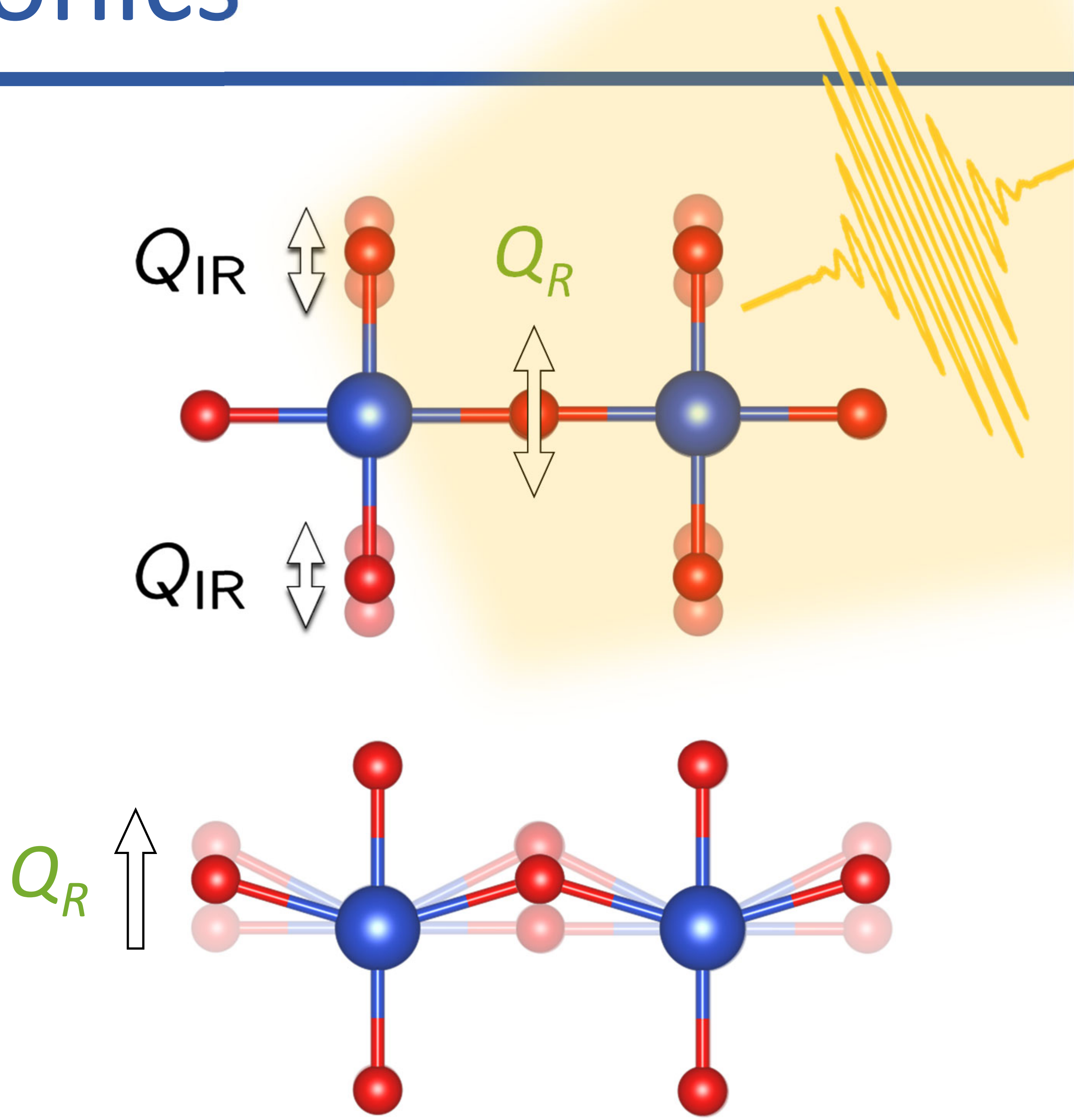
No average change to the lattice

"Nonlinear phononics"

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

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

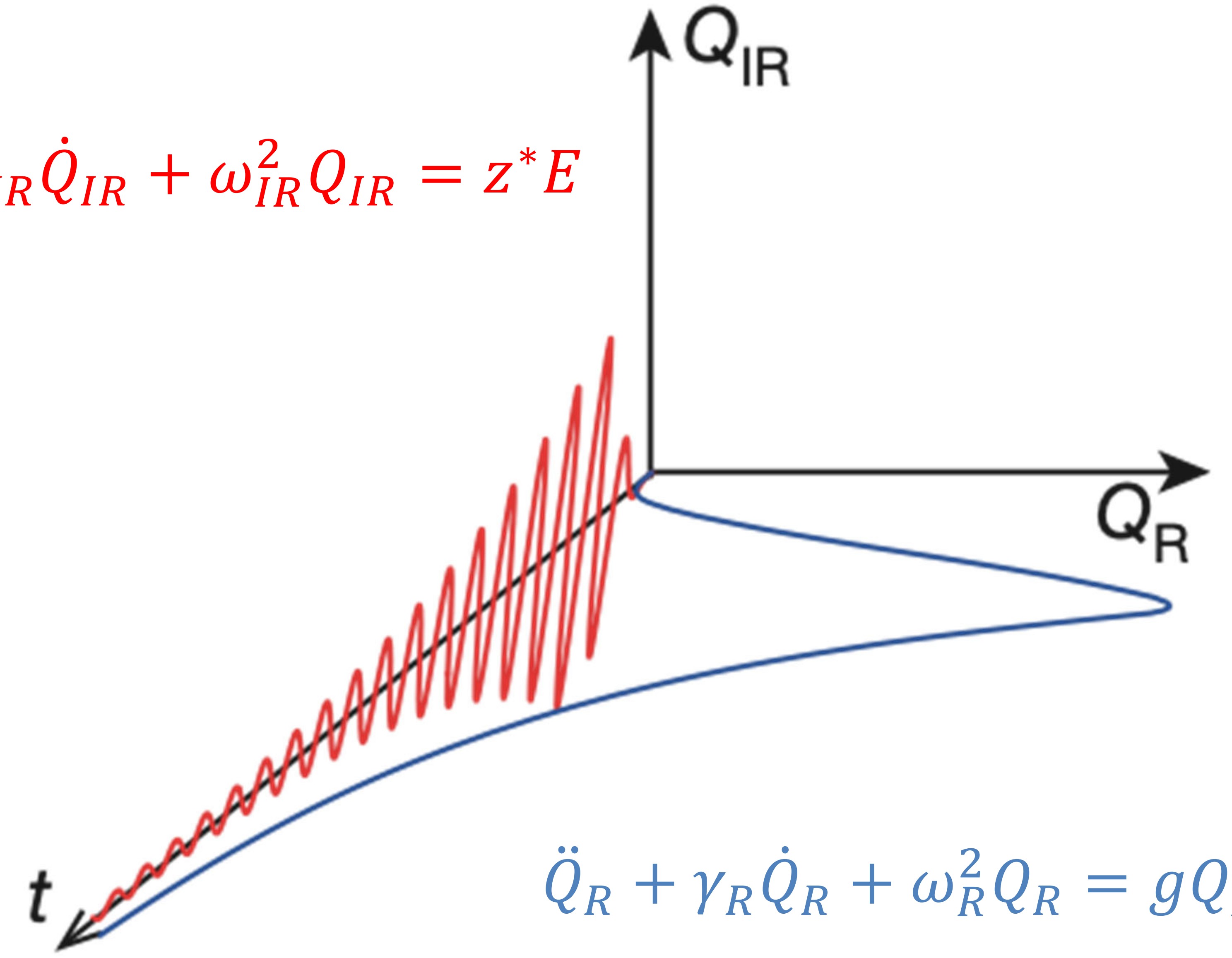




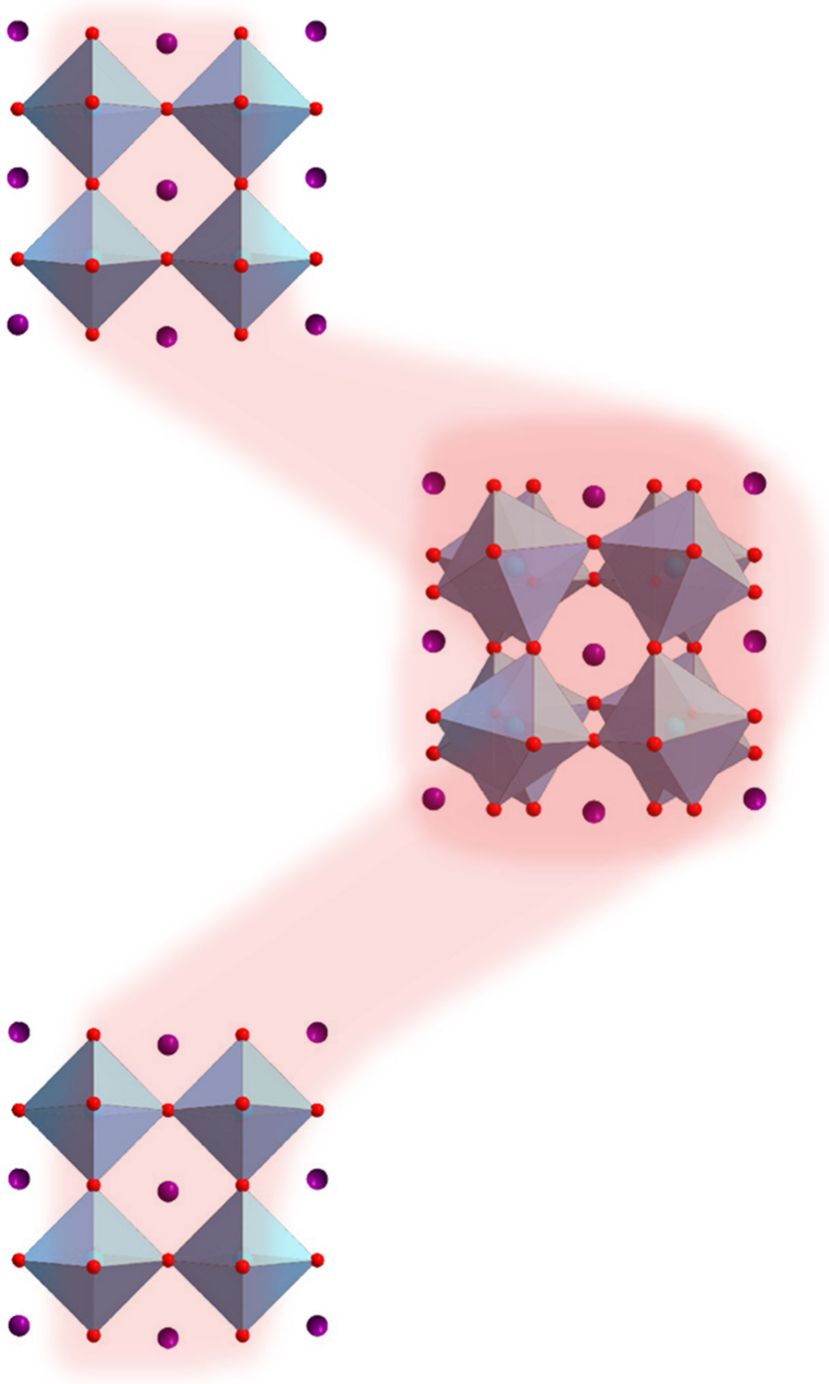
Net lattice displacement of coupled mode

Engineering new crystal structures with light

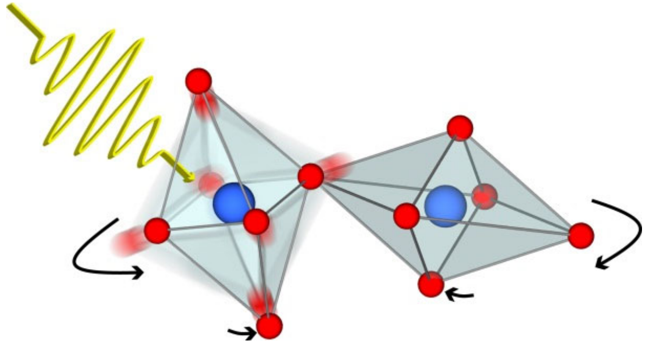
$$\ddot{Q}_{IR} + \gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = z^* E$$



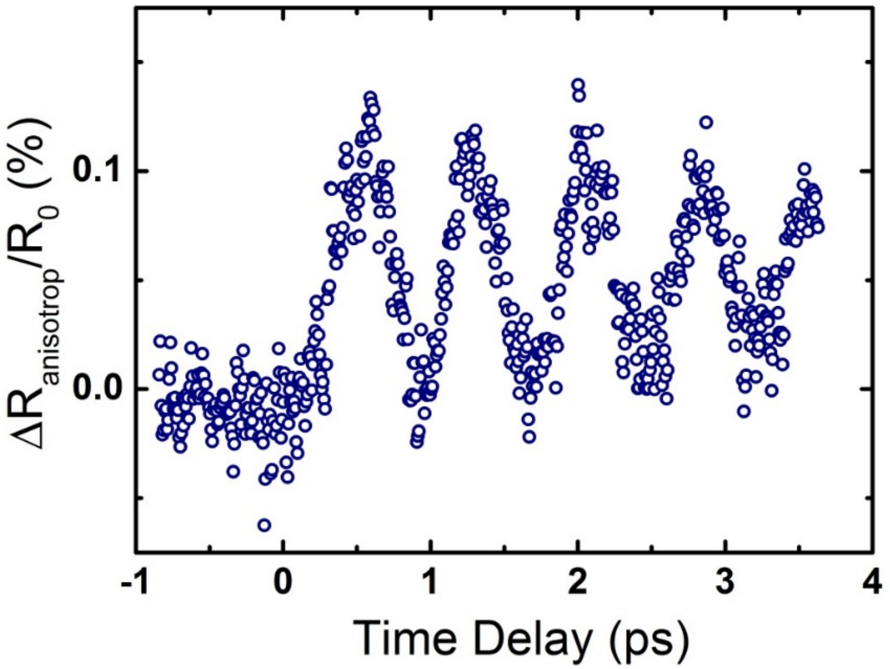
$$\ddot{Q}_R + \gamma_R \dot{Q}_R + \omega_R^2 Q_R = g Q_{IR}^2$$

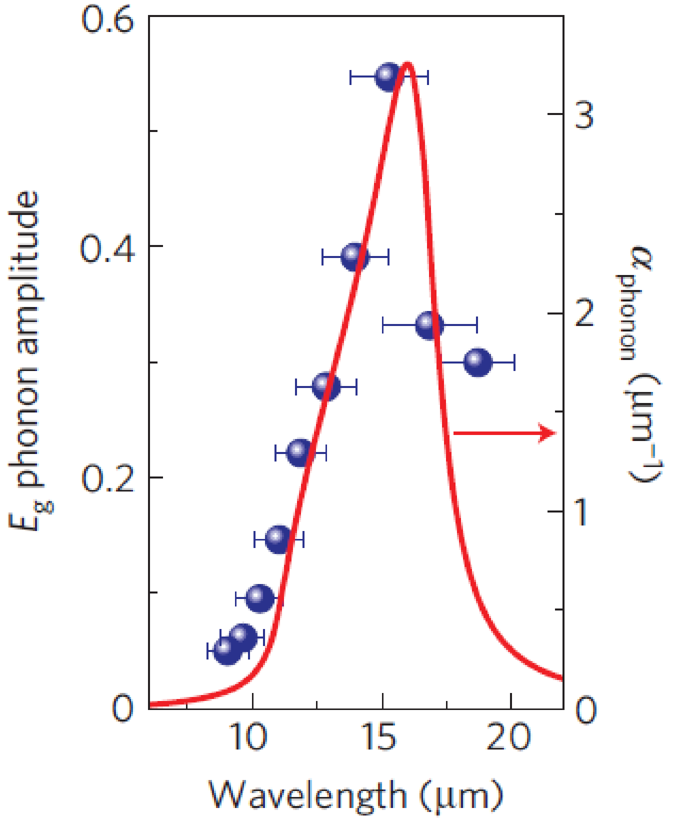


Experimental verification



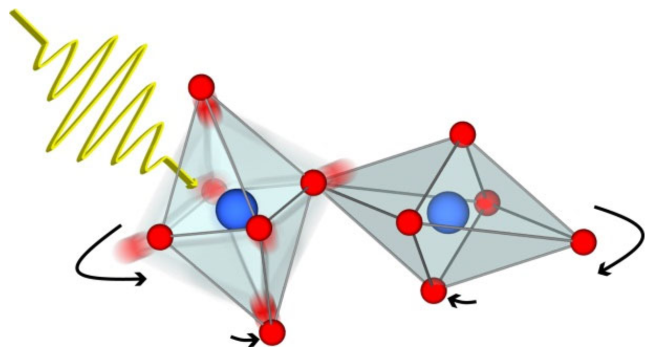
$La_{0.7}Sr_{0.3}MnO_3$



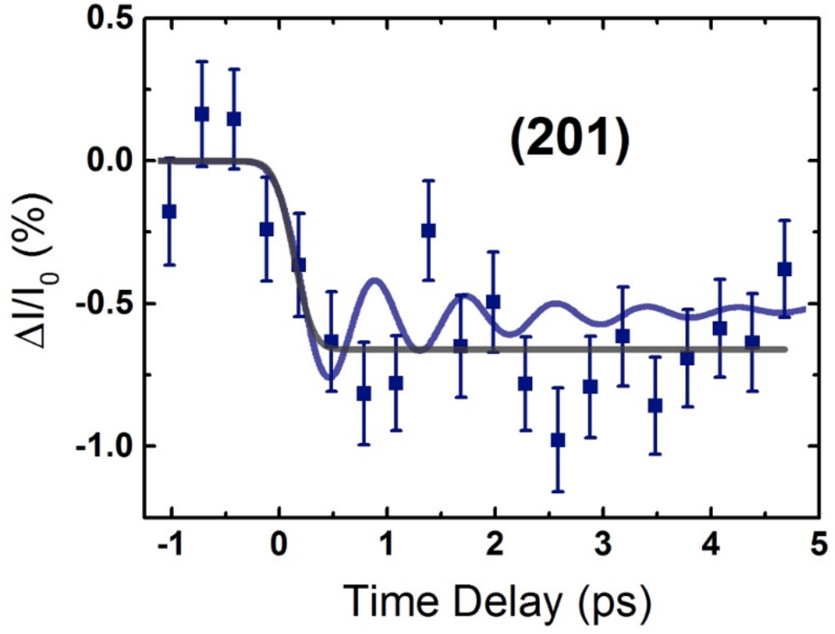


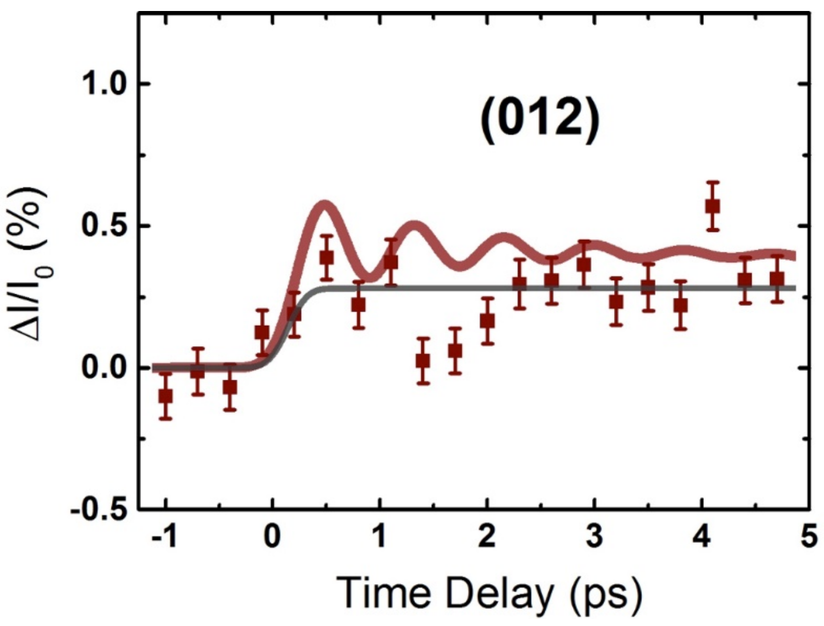
- Resonant coupling between IR mode and Raman mode demonstrated by time-resolved optical measurements

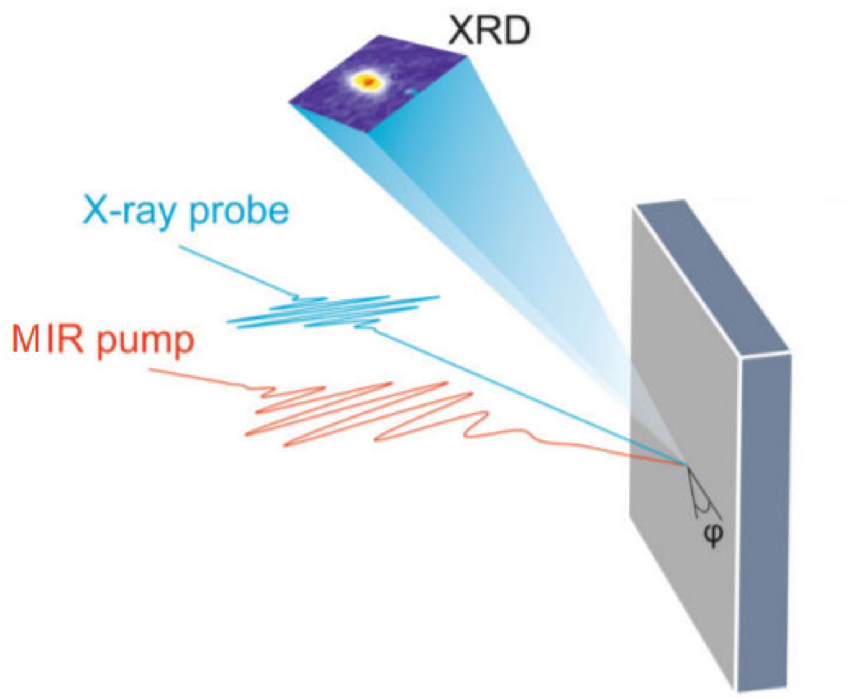
Experimental verification



$La_{0.7}Sr_{0.3}MnO_3$



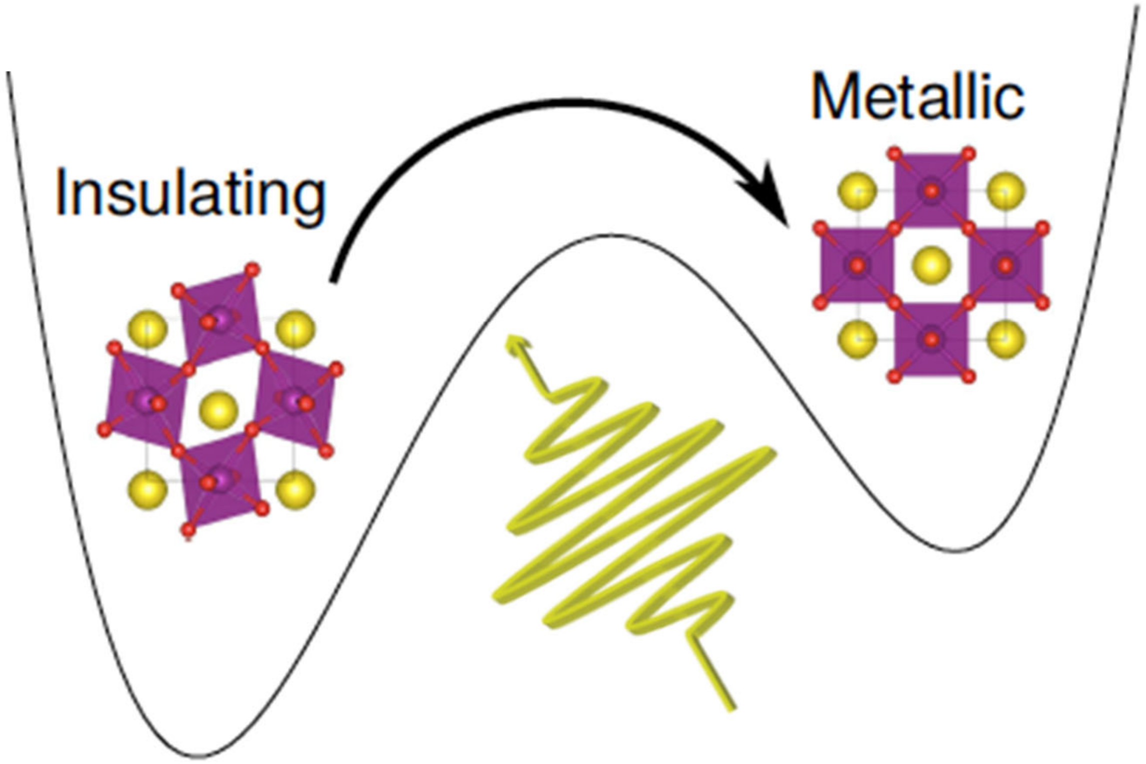




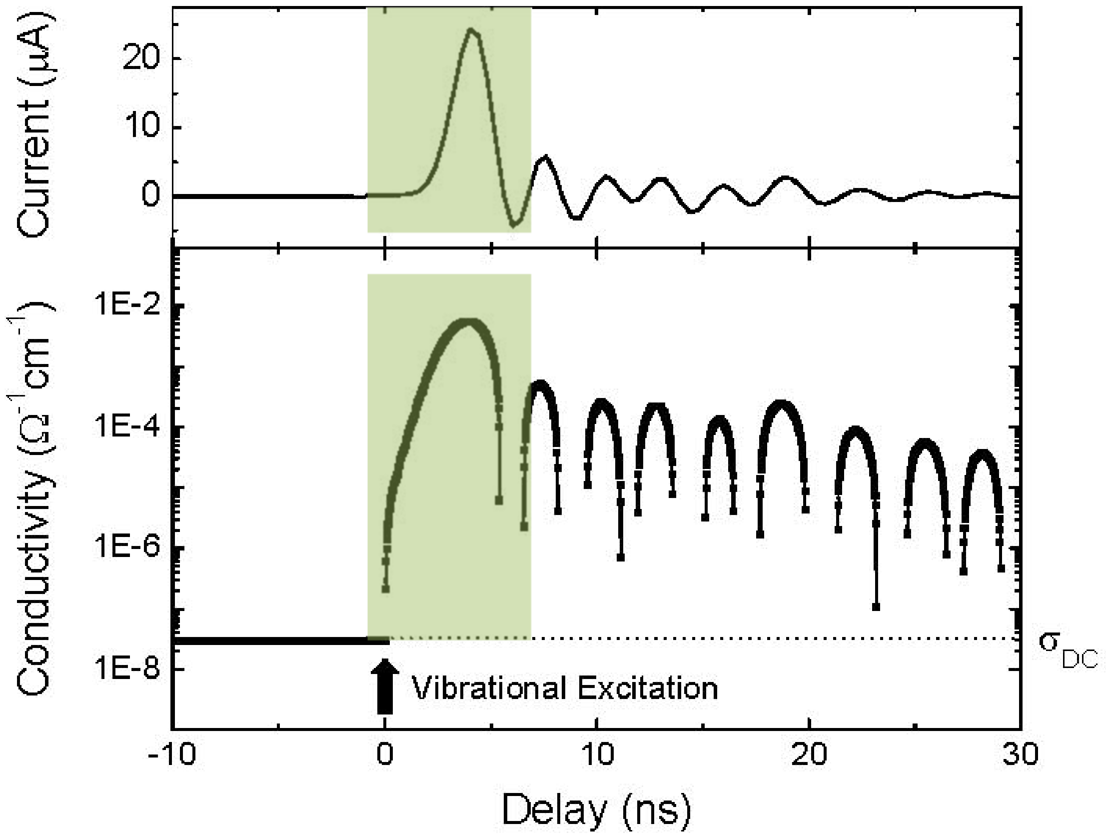
- Displacive excitation of Raman mode confirmed by time-resolved structural measurements

Ex) Vibrationally driven insulator-metal transition

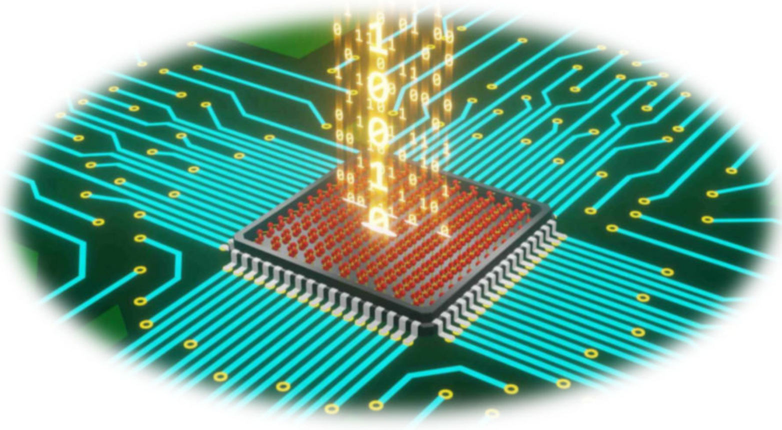
Correlated insulator: **$Pr_{0.7}Ca_{0.3}MnO_3$**

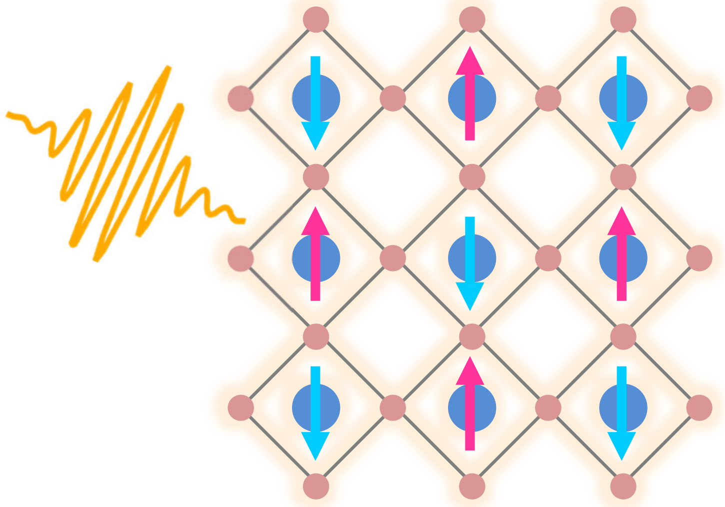


- Direct observation of induced metallic phase from time-resolved conductivity

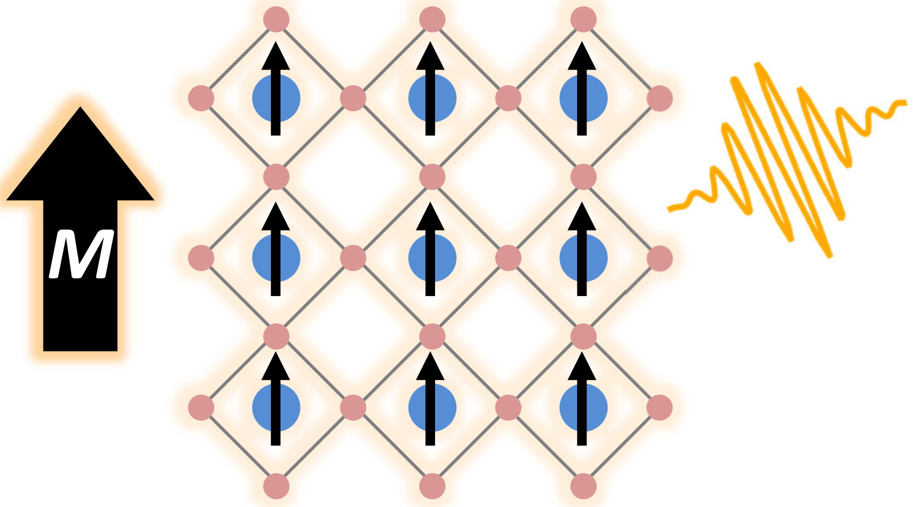


How can we optically engineer crystal structures to induce, enhance, and control non-equilibrium magnetic states?





1. Controlling order in an antiferromagnet

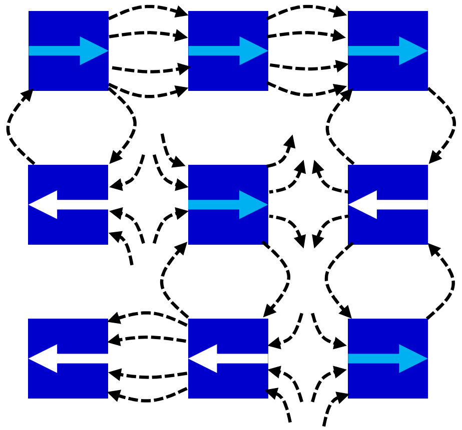


2. Enhancing ferromagnetism at high temperatures

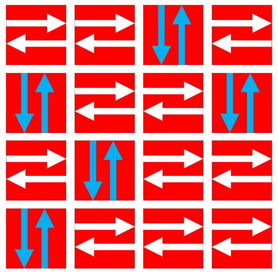
Antiferromagnetic writing

Can we manipulate order in antiferromagnet with light?

Ferromagnetic



Antiferromagnetic



- **Denser**
- **Faster**
- **More robust**

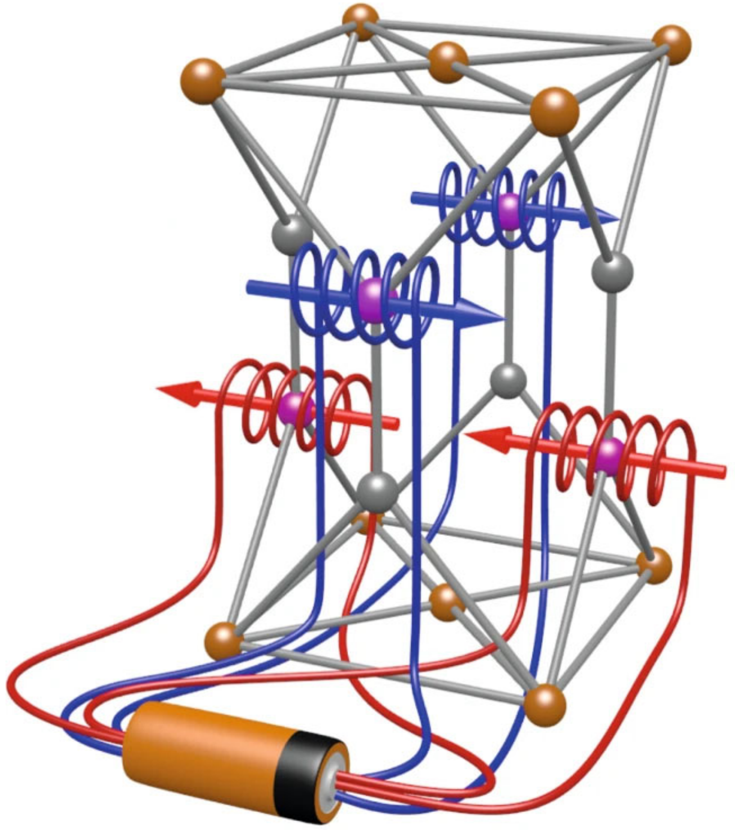


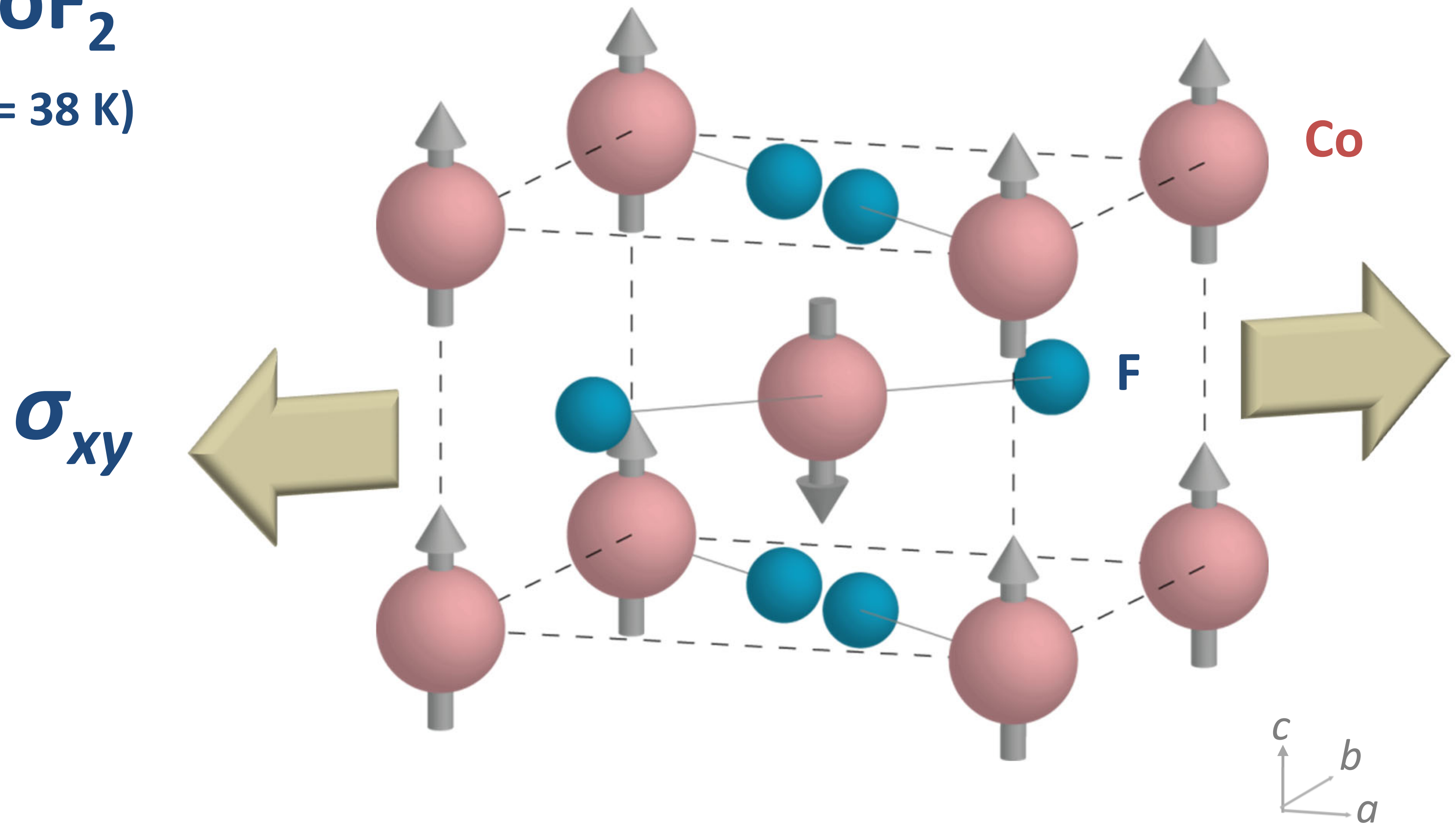
Image: tlo-kyoto.co.jp

T. Jungwirth, *et al.* Nat. Phys. **14**, 200 (2018)

A prototypical antiferromagnet

CoF_2

(T_N = 38 K)

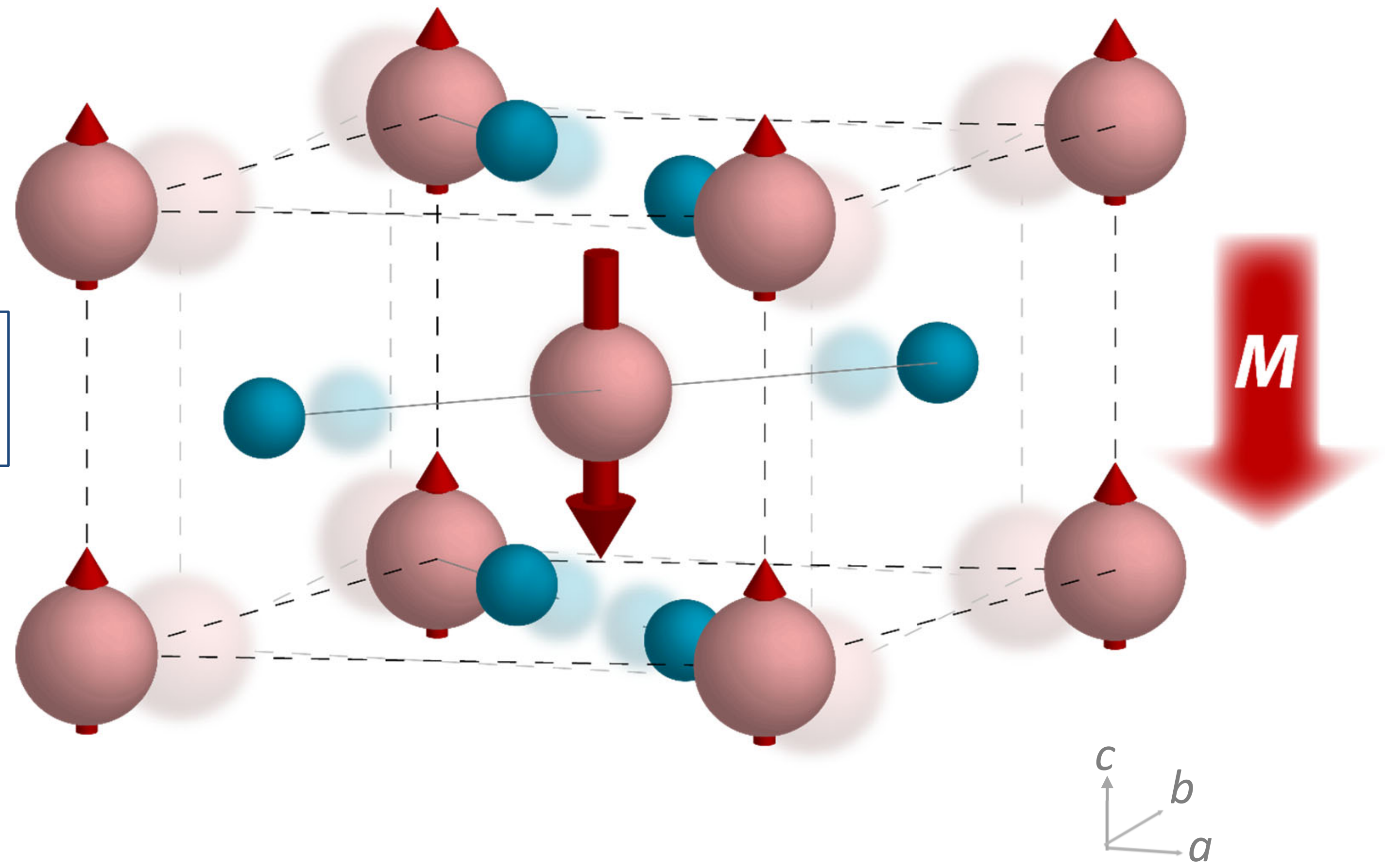


Strain control: piezomagnetism

CoF_2

(T_N = 38 K)

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

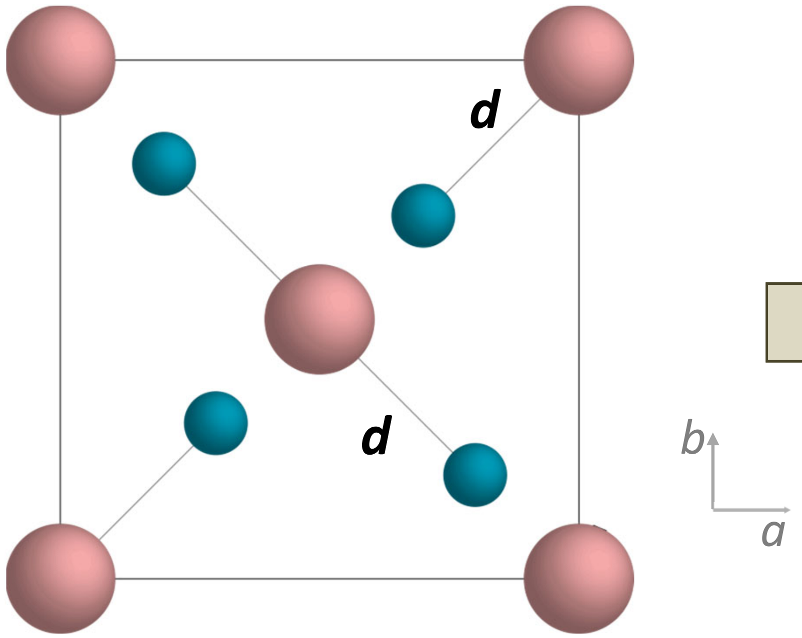


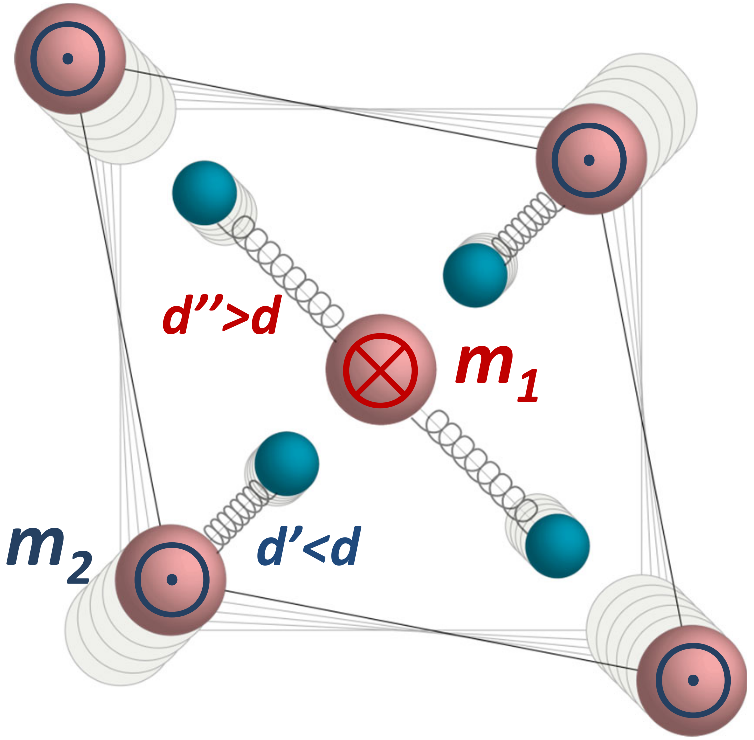
Origin of piezomagnetism in CoF_2

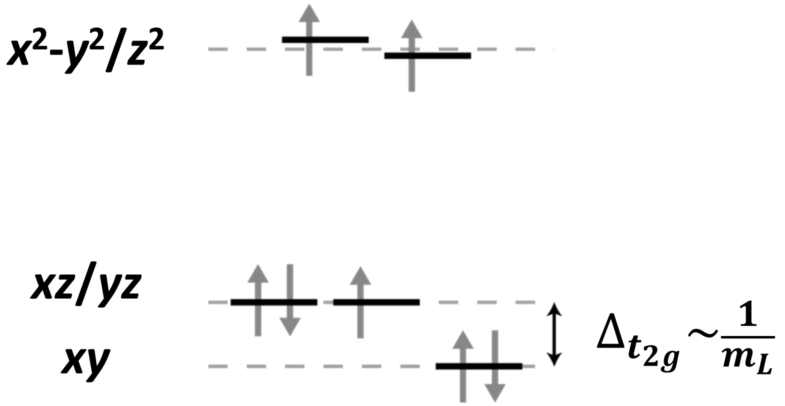
Top view

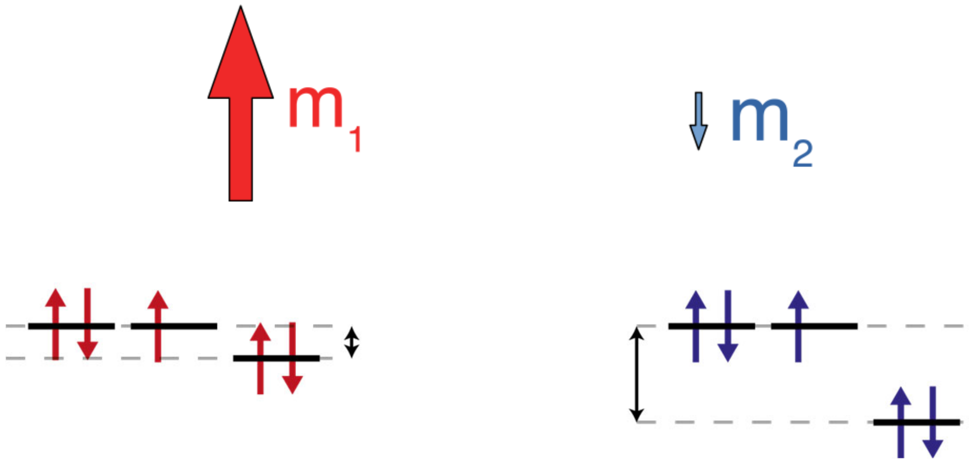
Co orbital moment:

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





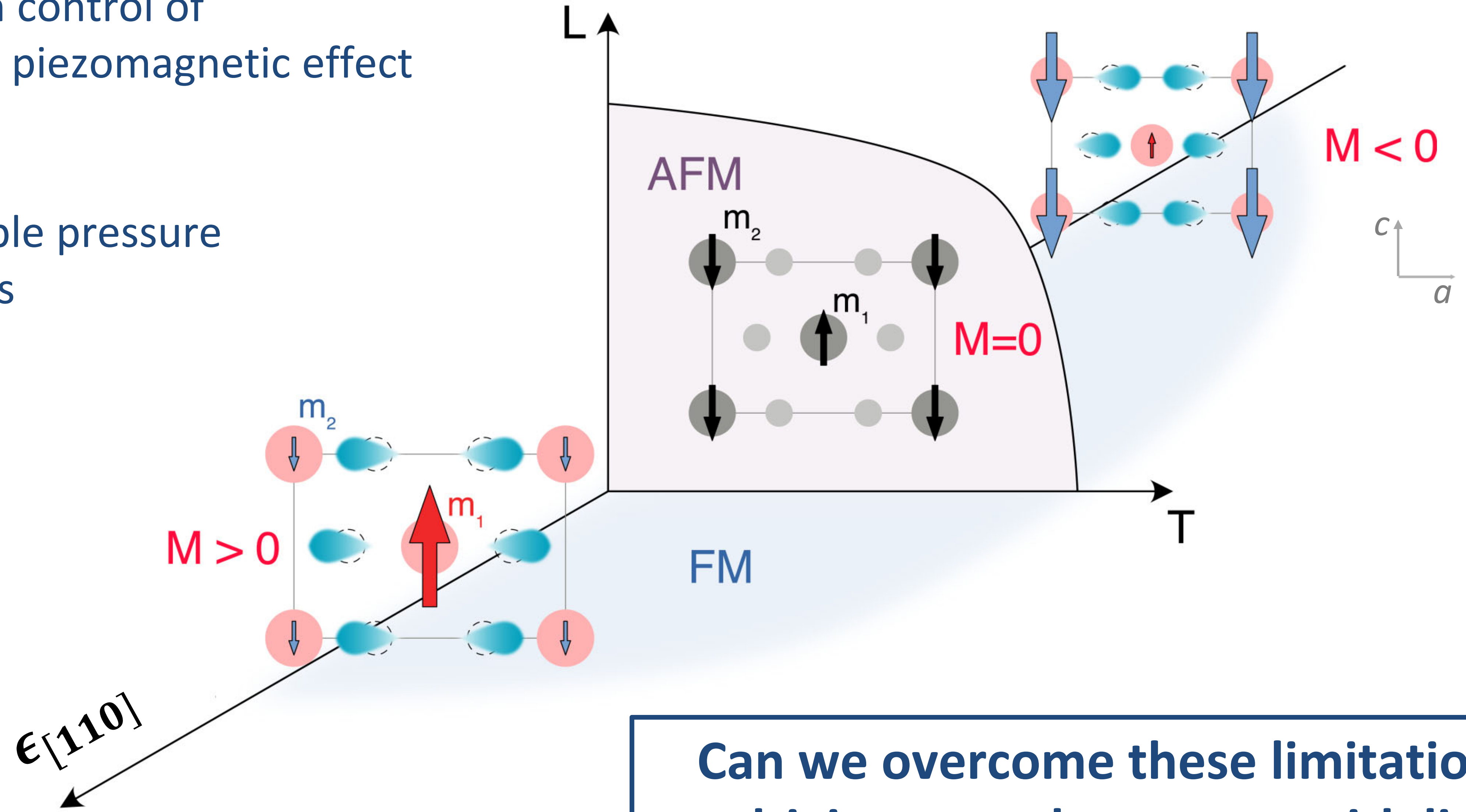




Uniaxial strain control of magnetization

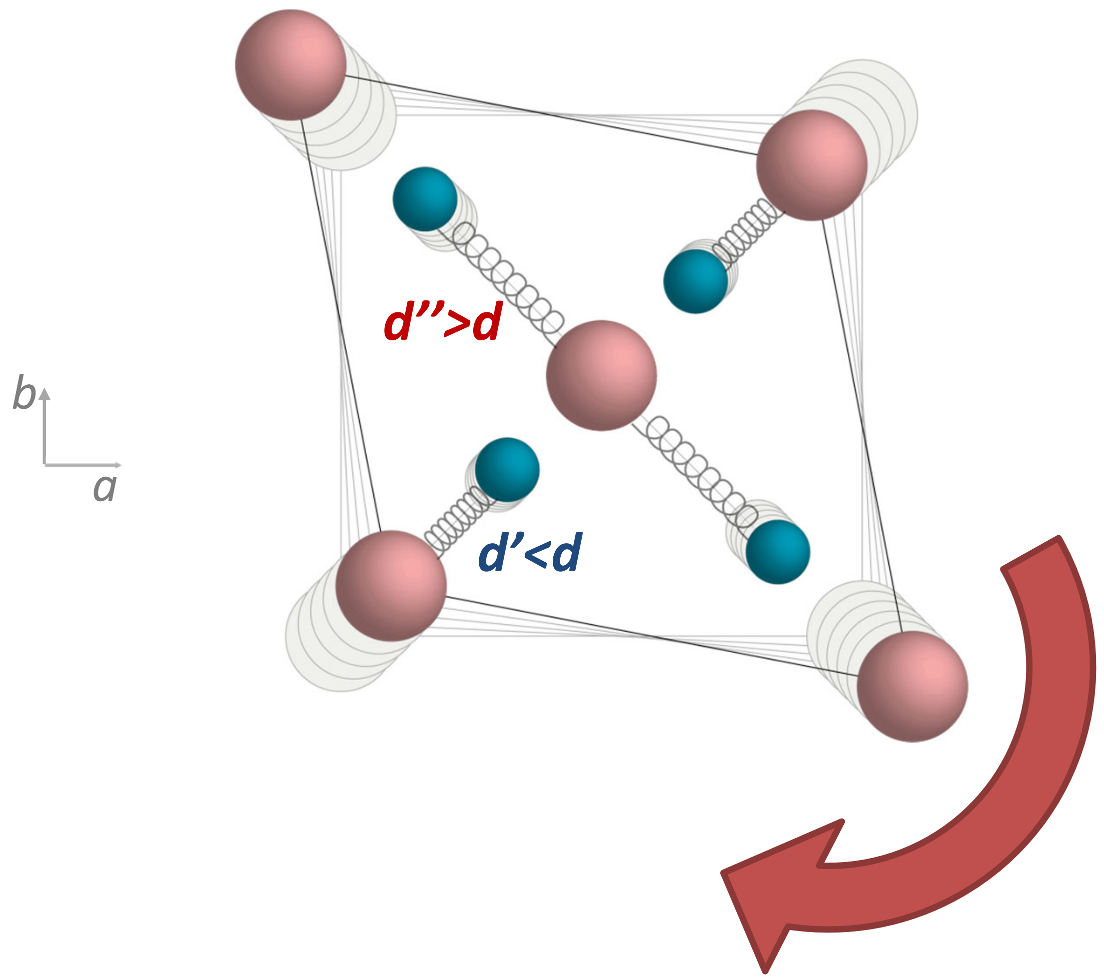
- Bi-directional strain control of magnetization with piezomagnetic effect

<u>Disadvantages:</u>
- Limited by achievable pressure
- Acoustic time scales



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

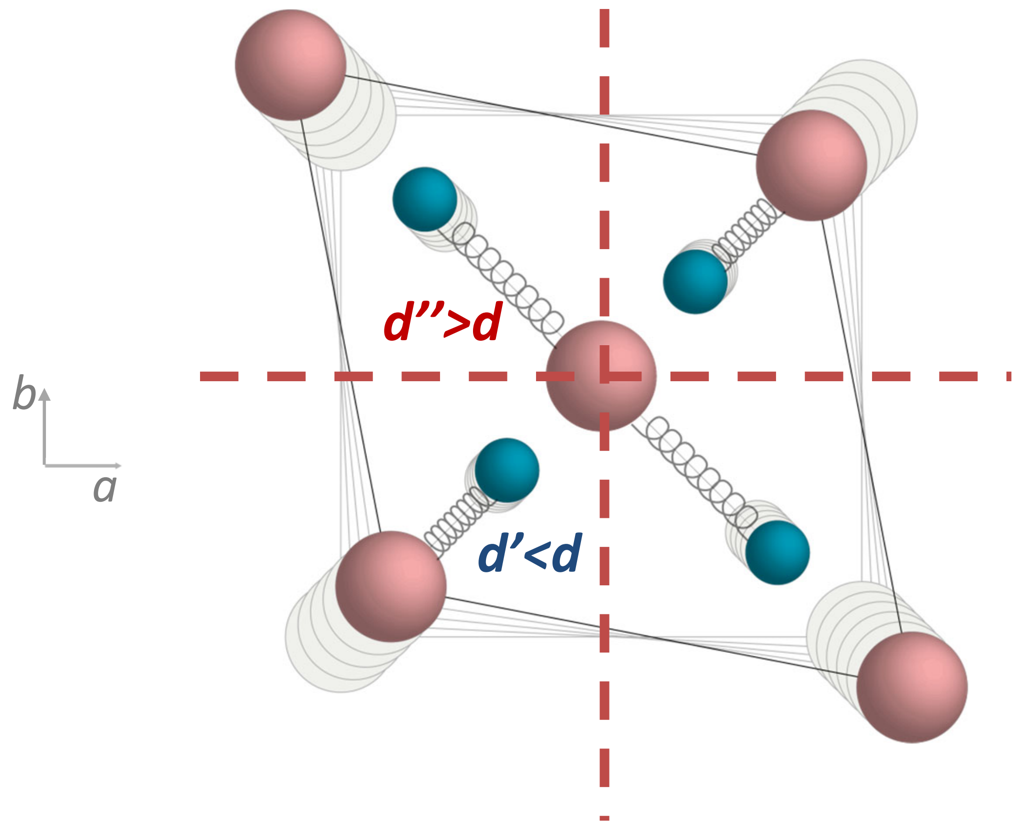
Emulating uniaxial strain



Symmetry of strain:

- Breaks C_4 rotation symmetry (antisymmetric)

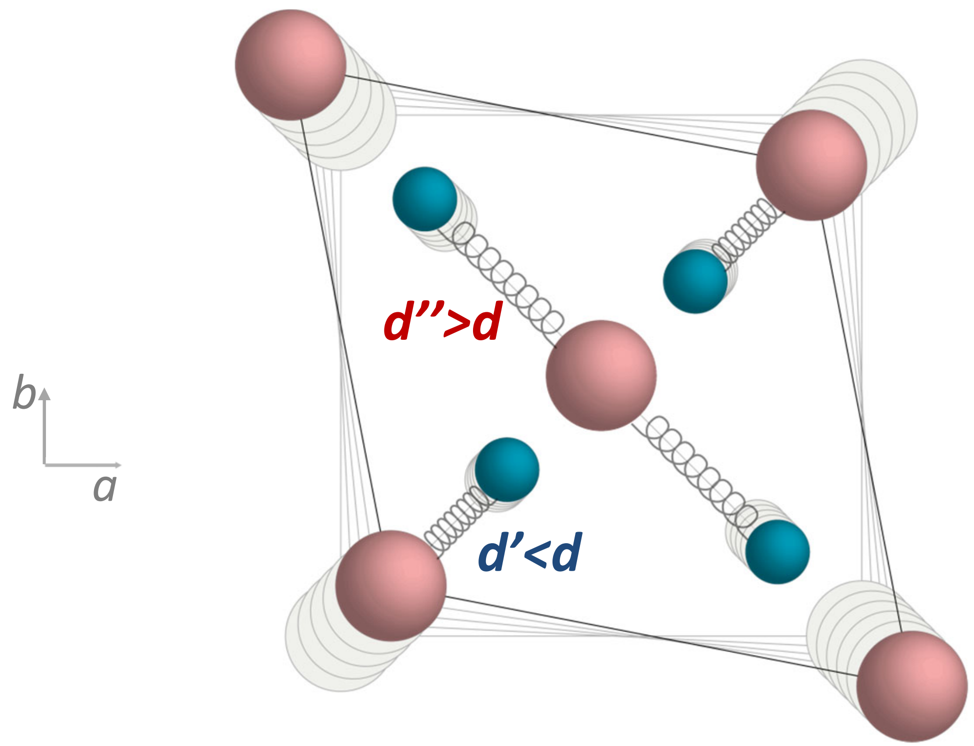
Emulating uniaxial strain



Symmetry of strain:

- Breaks C_4 rotation symmetry (antisymmetric)
- Breaks xy mirror planes (antisymmetric)

Emulating uniaxial strain

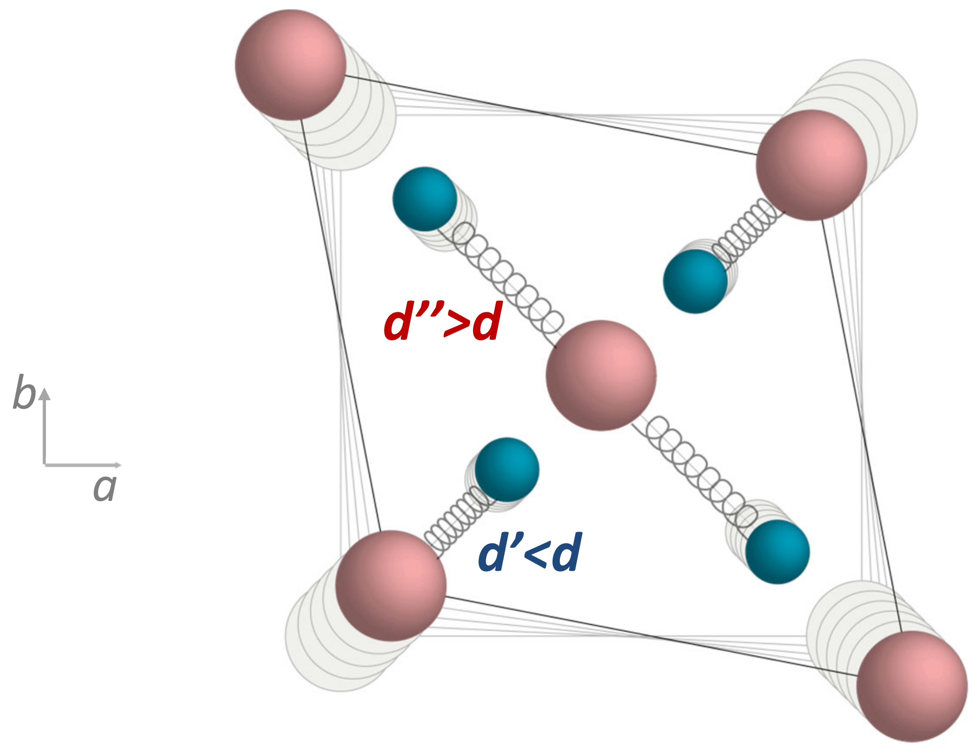


Symmetry of strain:

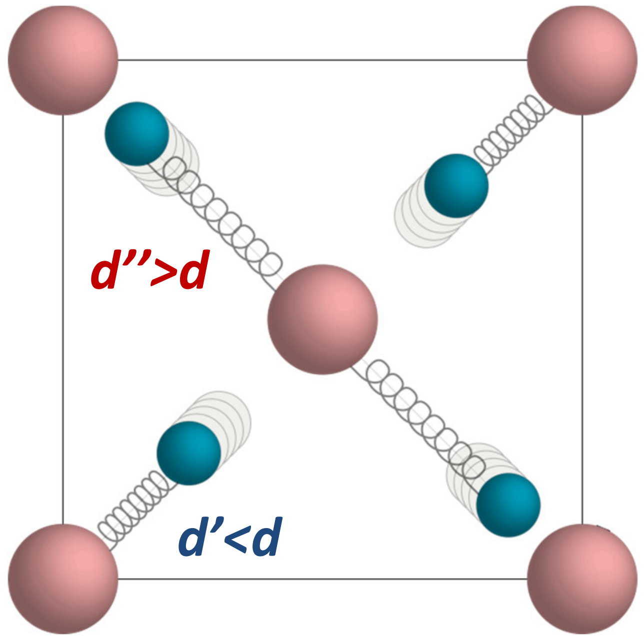
- Breaks C_4 rotation symmetry (antisymmetric)
- Breaks xy mirror planes (antisymmetric)
- Preserves inversion

➔ B_{2g} **symmetry**

Emulating uniaxial strain



Stress along [110]



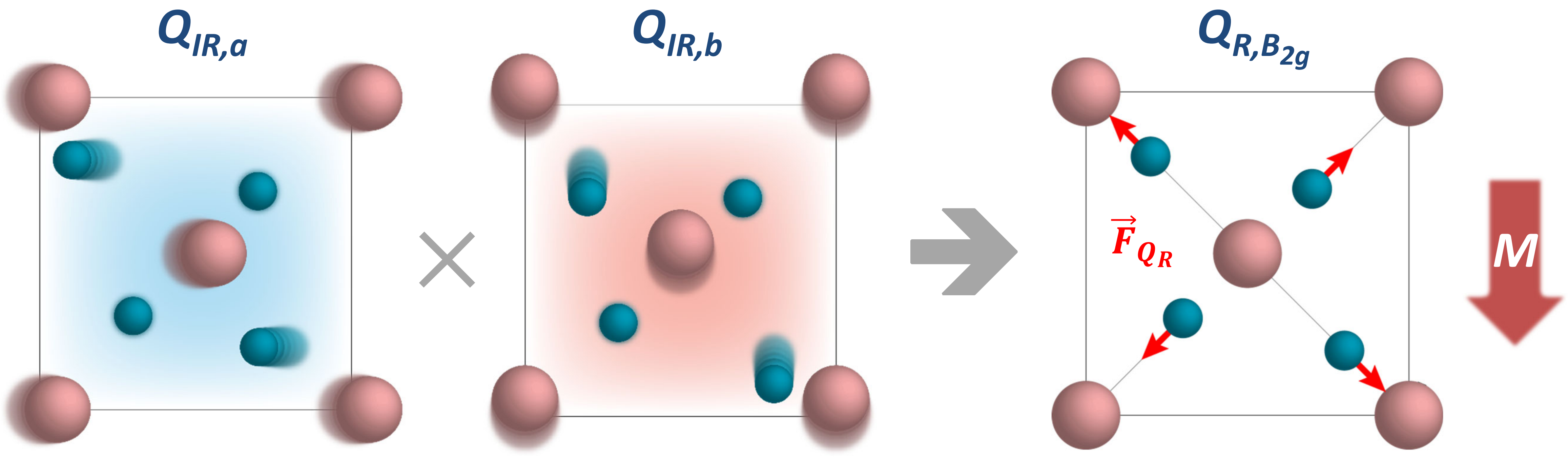
B_{2g} Raman phonon

$$M_z \propto Q_R$$

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

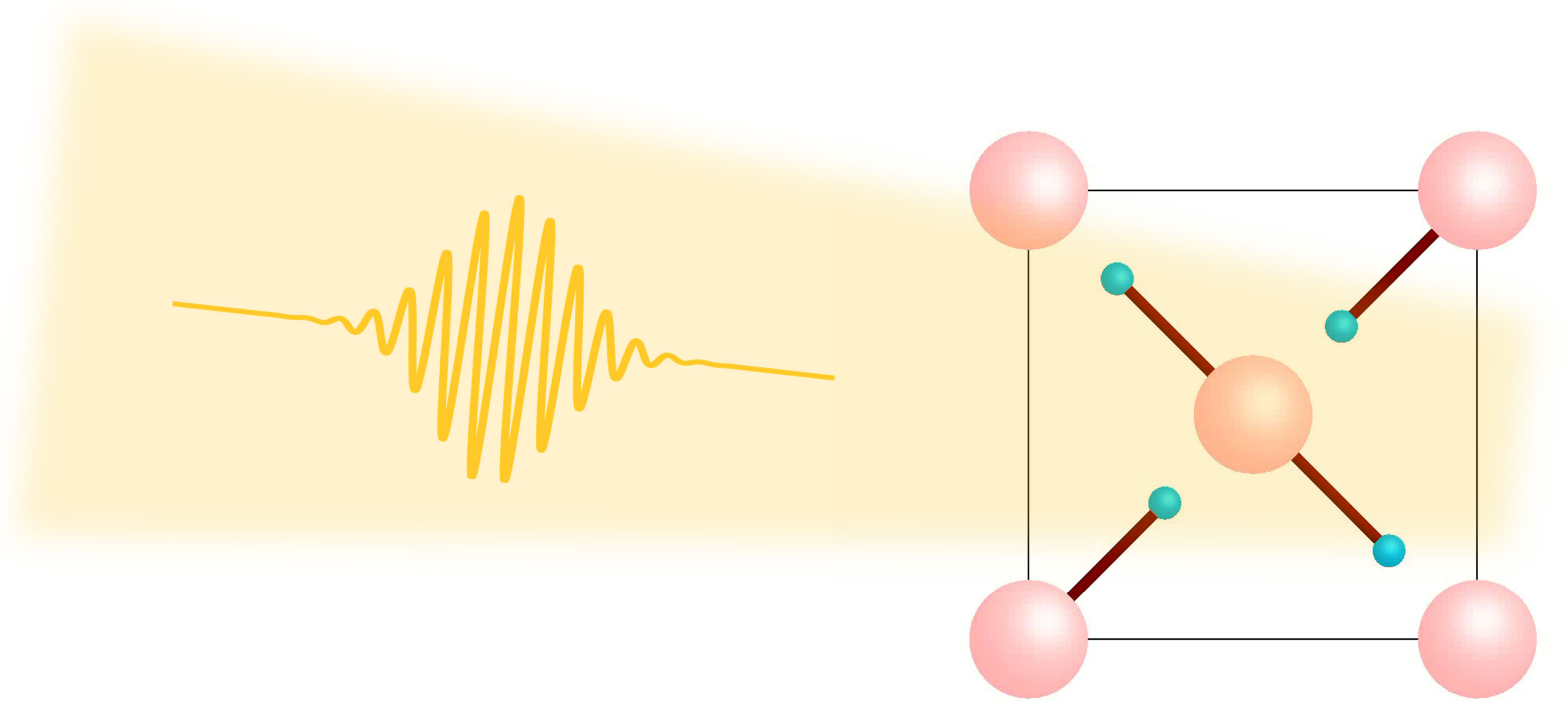
Symmetry breaking from phonons

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

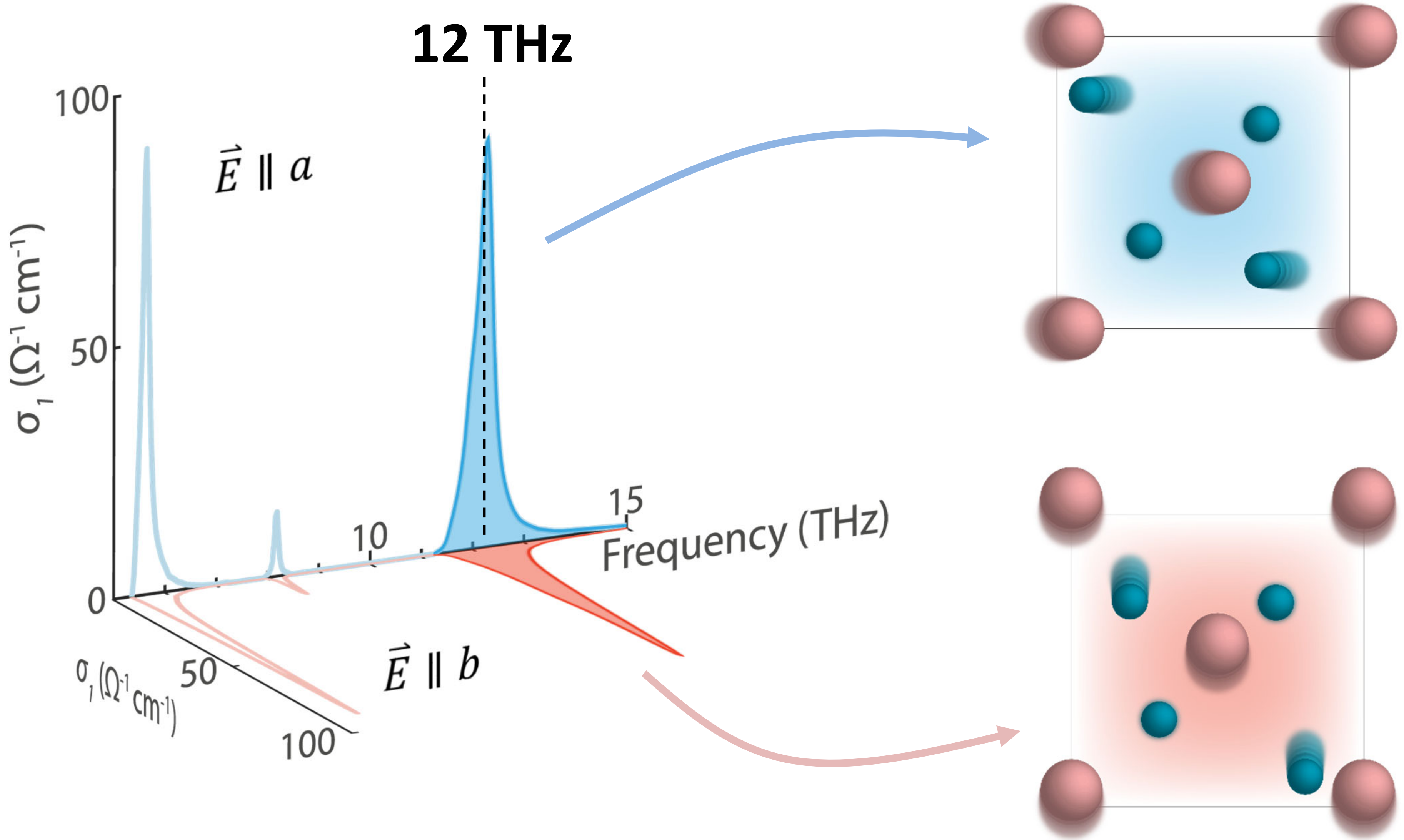


- Simultaneously excite degenerate IR phonons along *a* and *b* to generate magnetization

Symmetry breaking from phonons

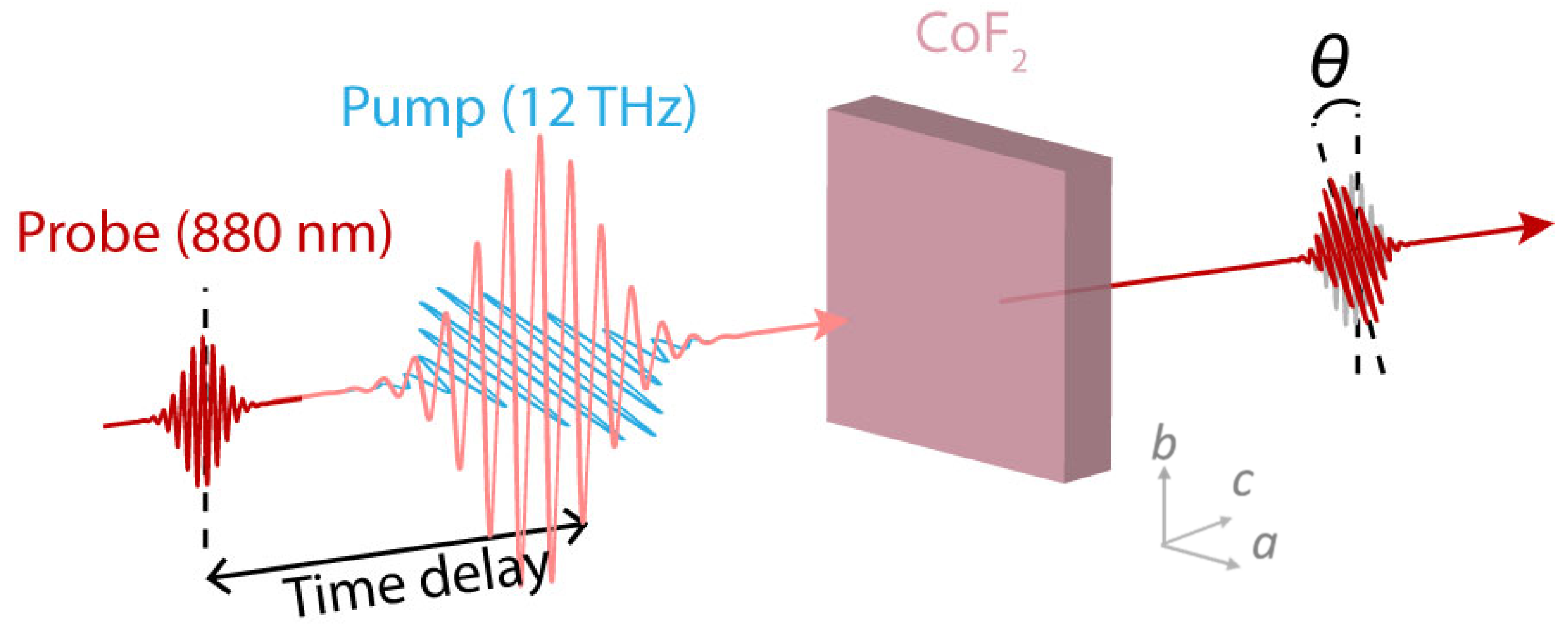


Resonantly driving phonons



Experimental setup

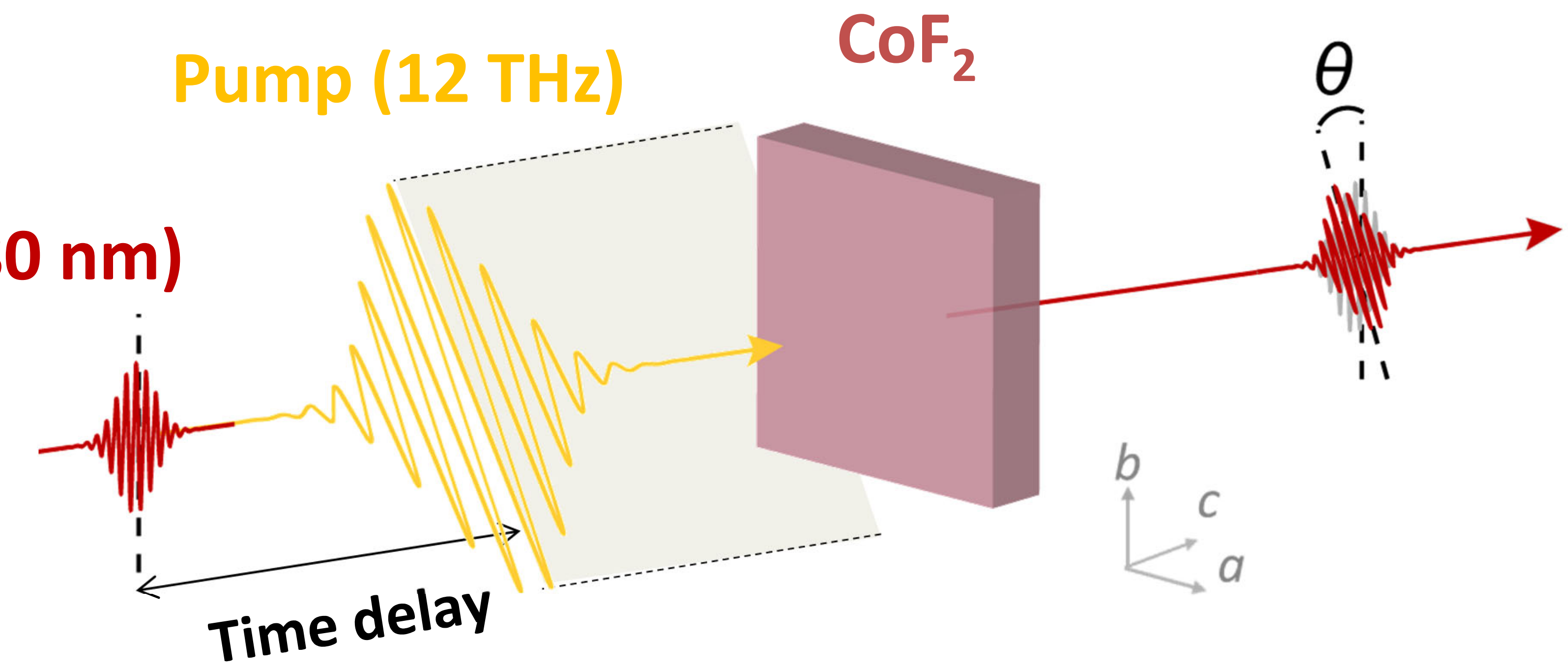
- Simultaneously drive *a* and *b* phonons by pumping along [110]



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

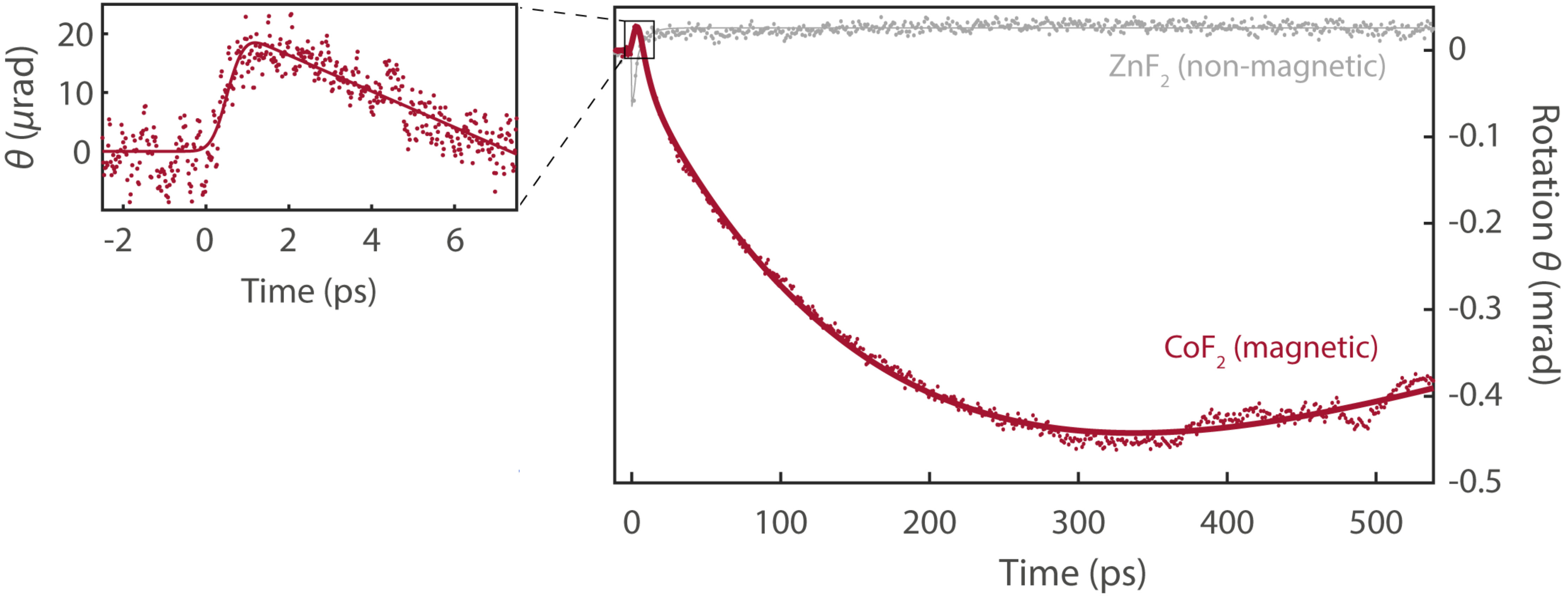
Experimental setup

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



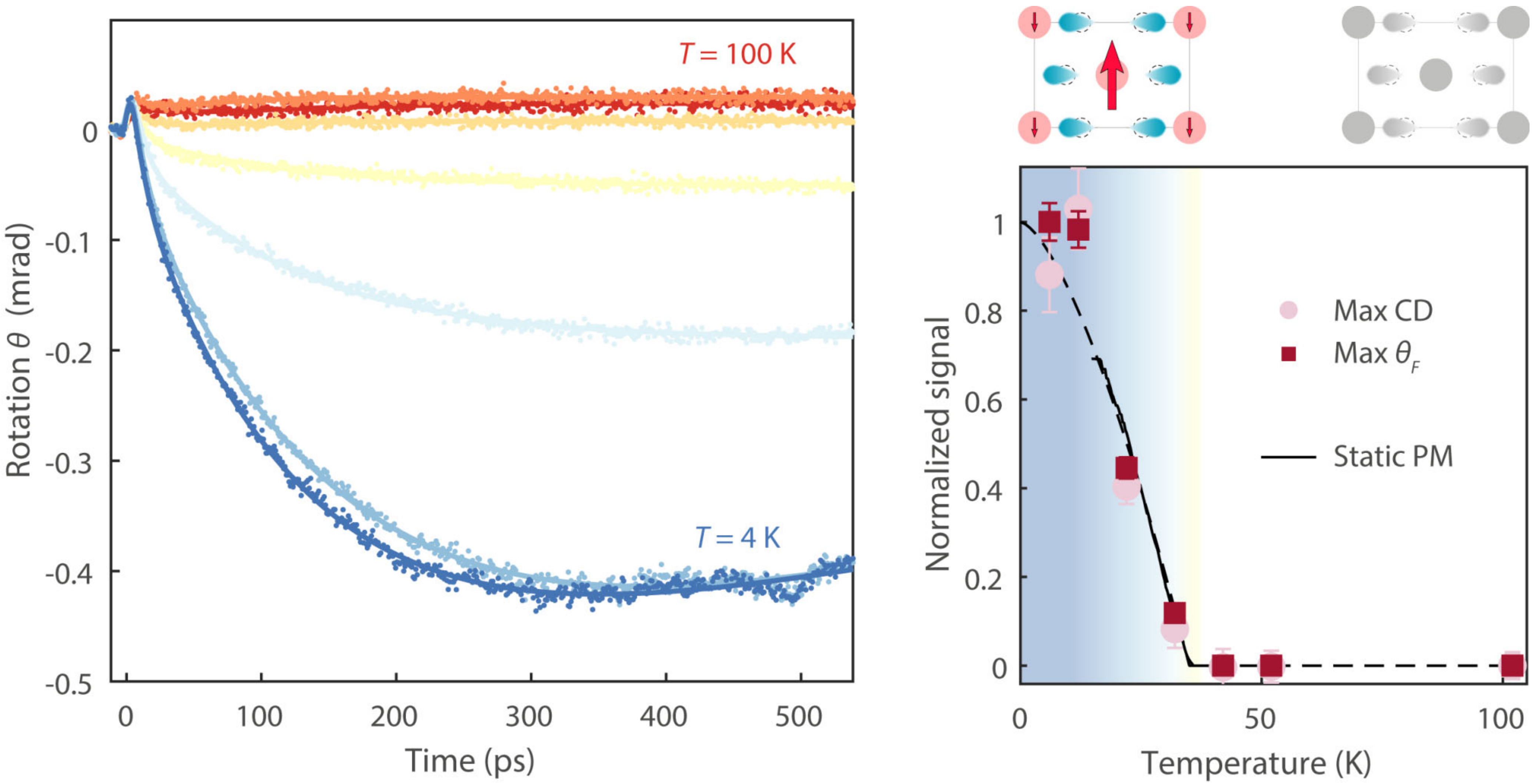
- Measure time-resolved Faraday effect: $\theta(t) \propto M_z(t)$

Pump-induced Faraday rotation



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

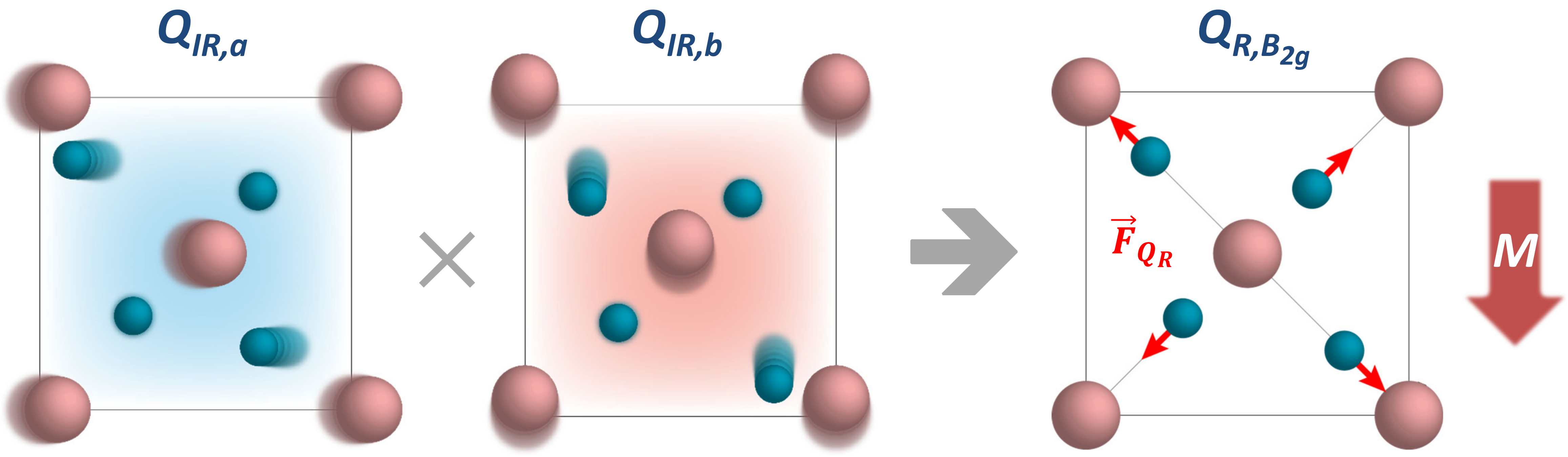
Temperature dependence



- Pump-induced effect follows static piezomagnetic response

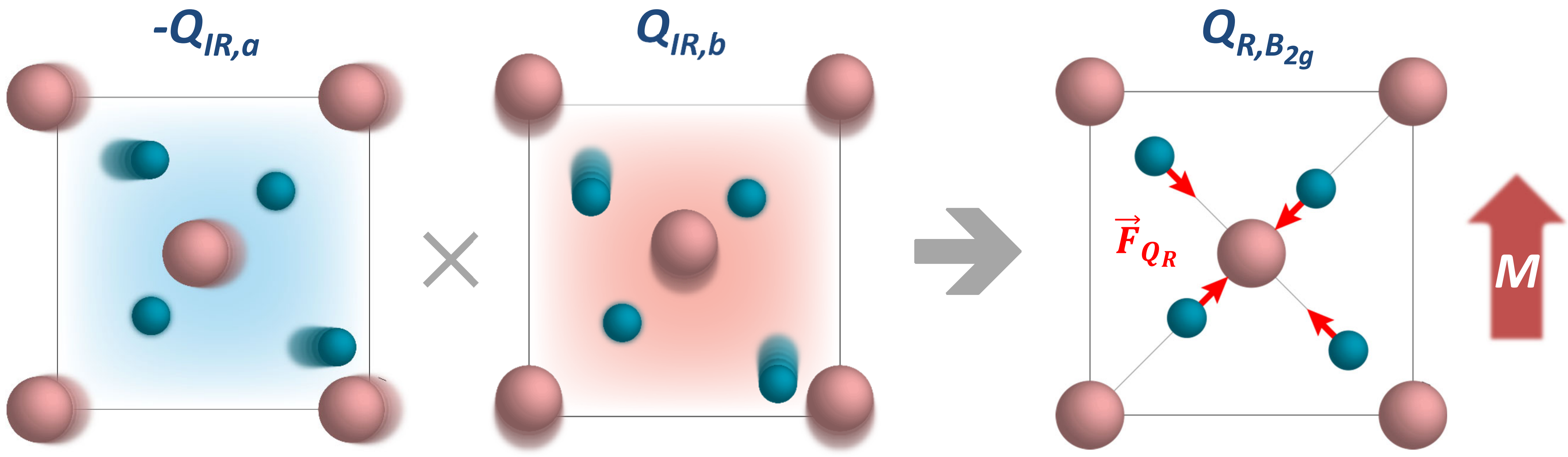
Switchable magnetization

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



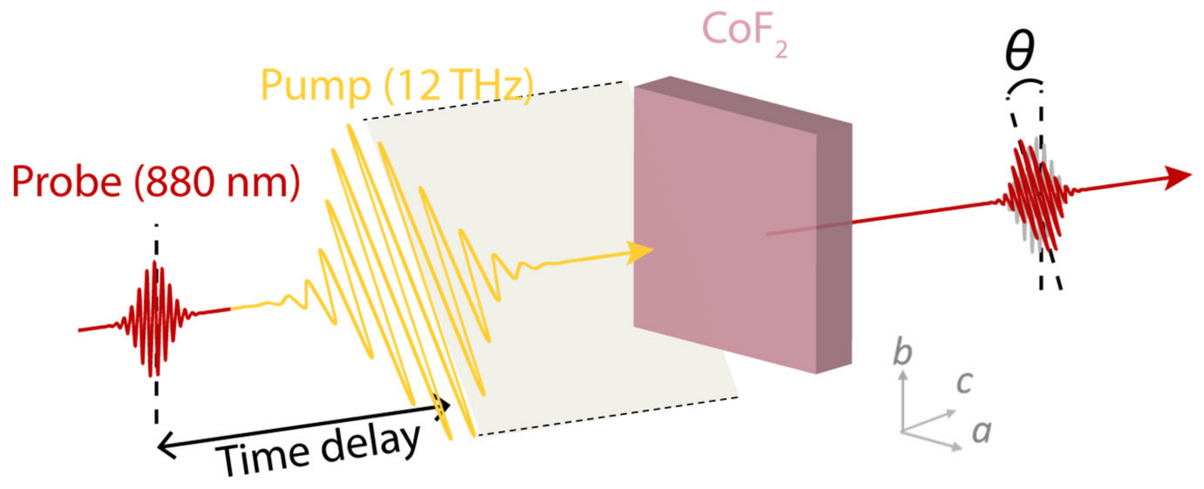
Switchable magnetization

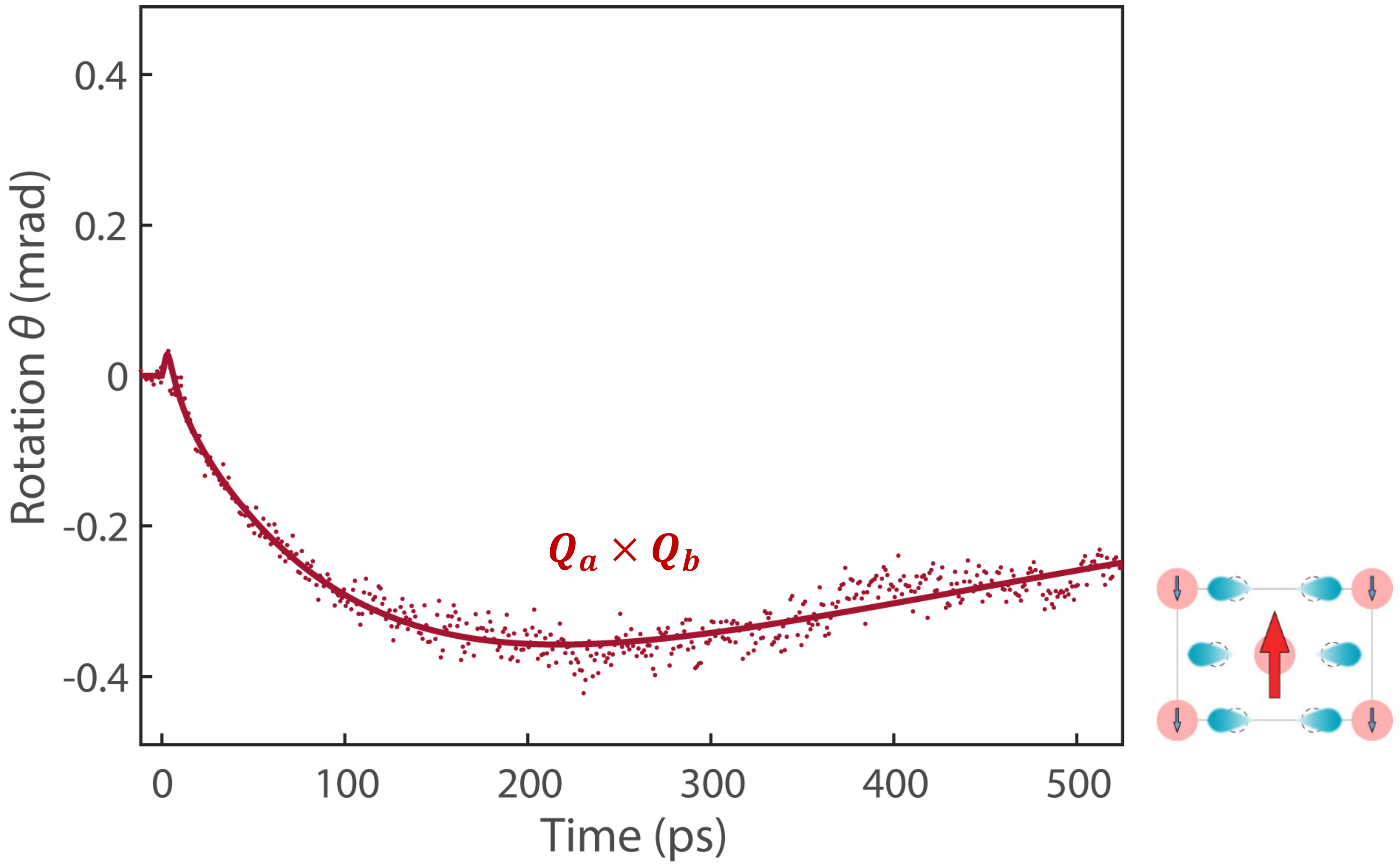
Three-phonon nonlinear interaction: $U_{lattice} \propto Q_{IR,1} Q_{IR,2} Q_R$



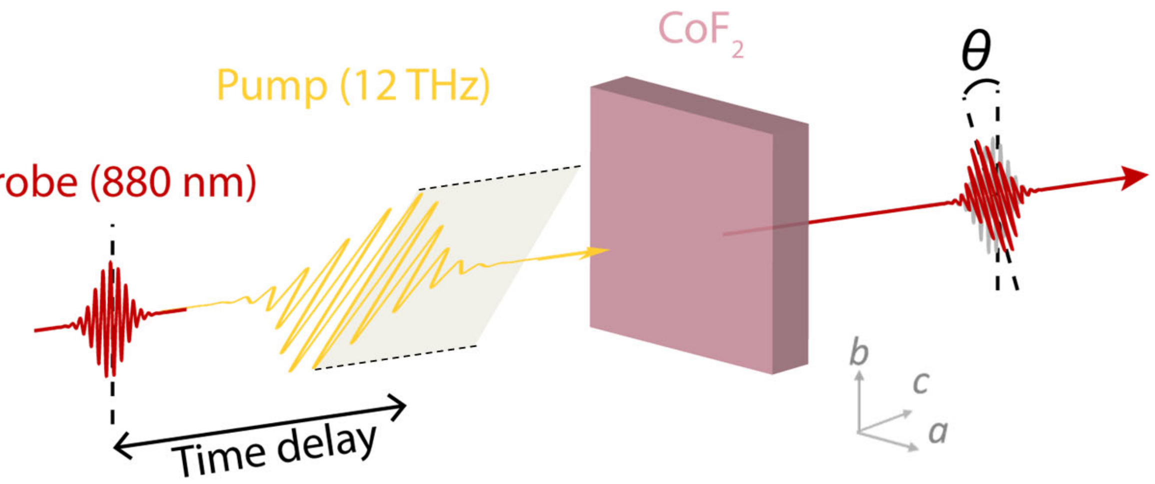
- Can change direction of magnetization relative phase of phonon excitation (polarization of pump)

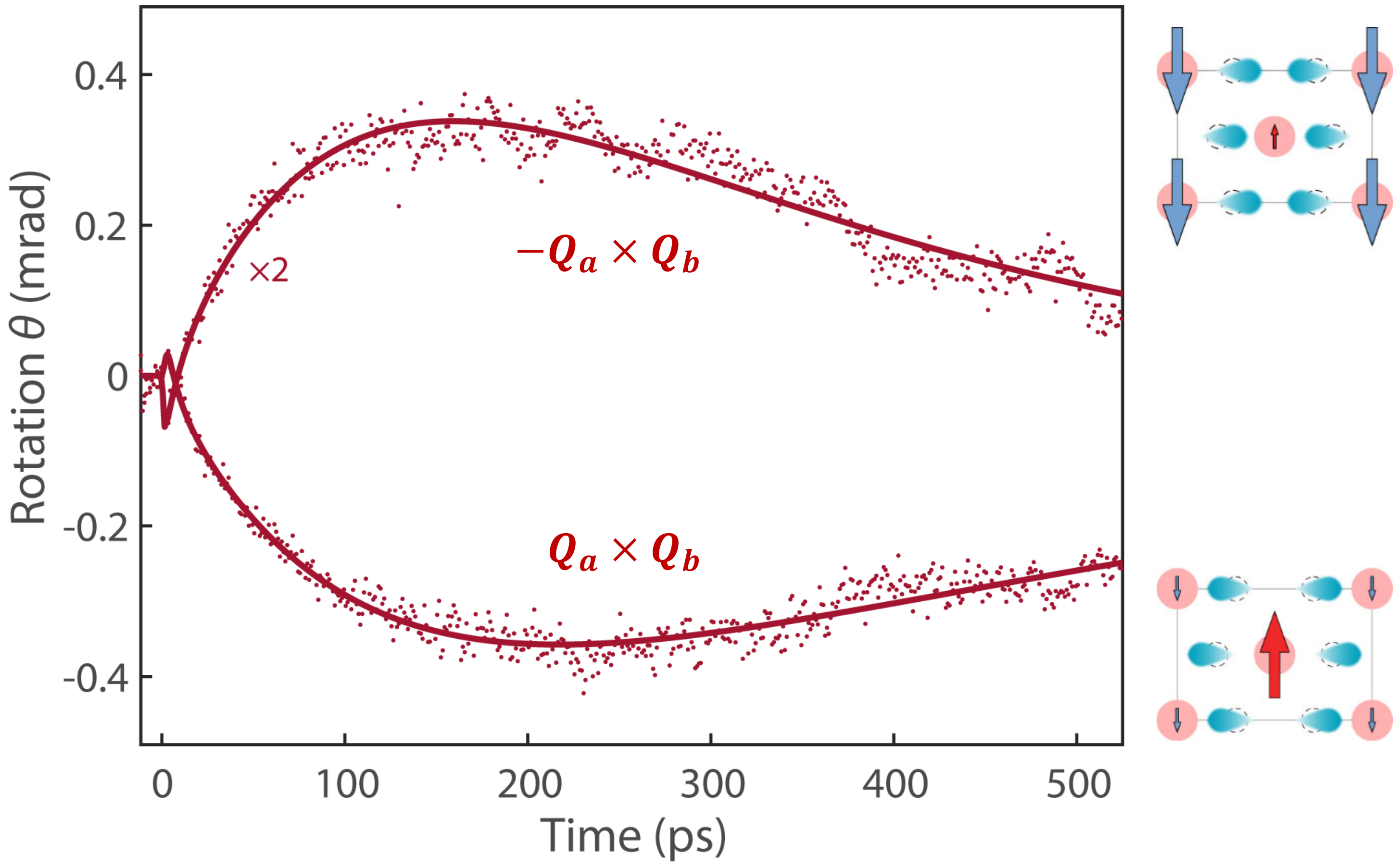
Controlling magnetization direction





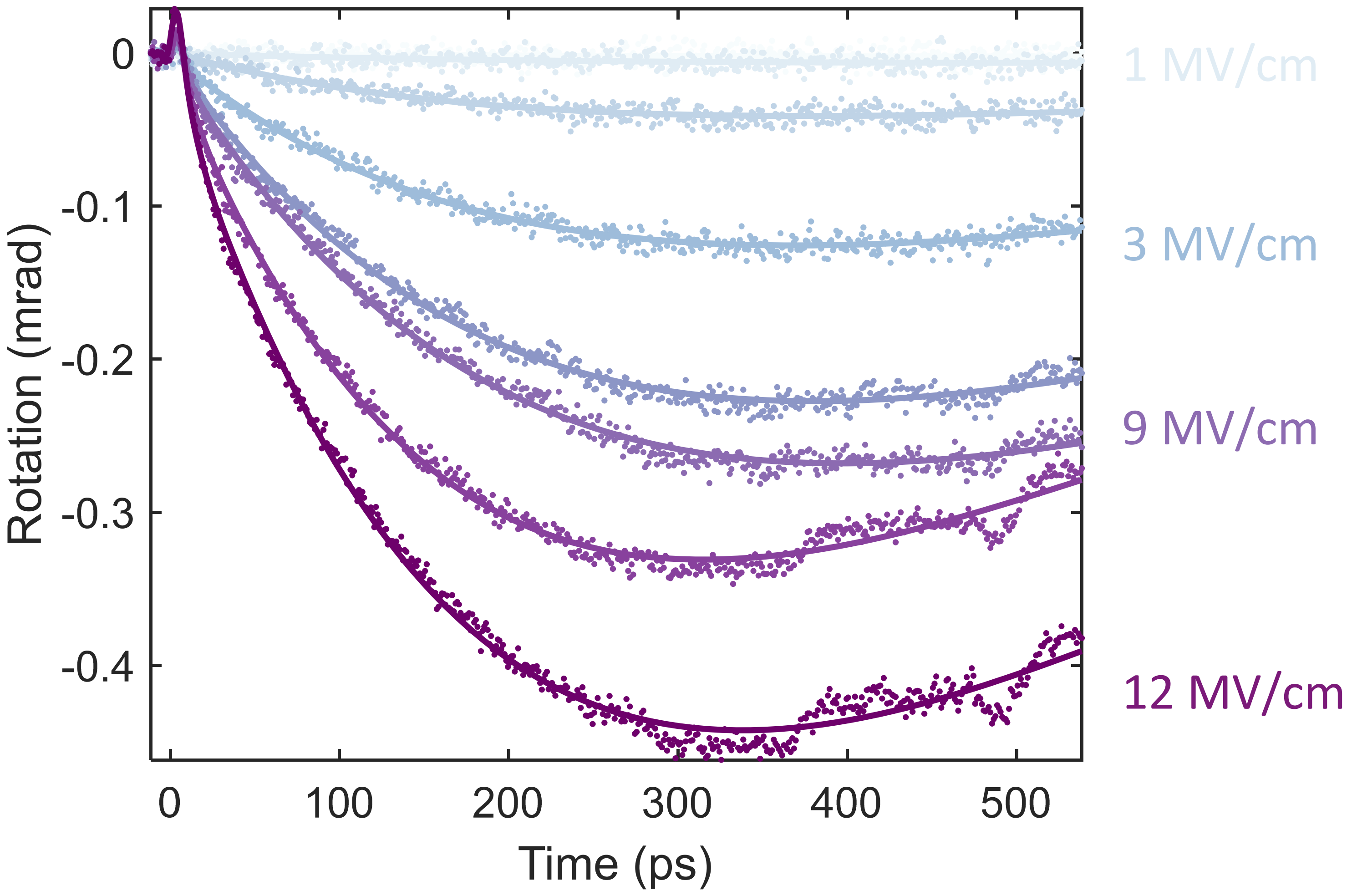
Controlling magnetization direction



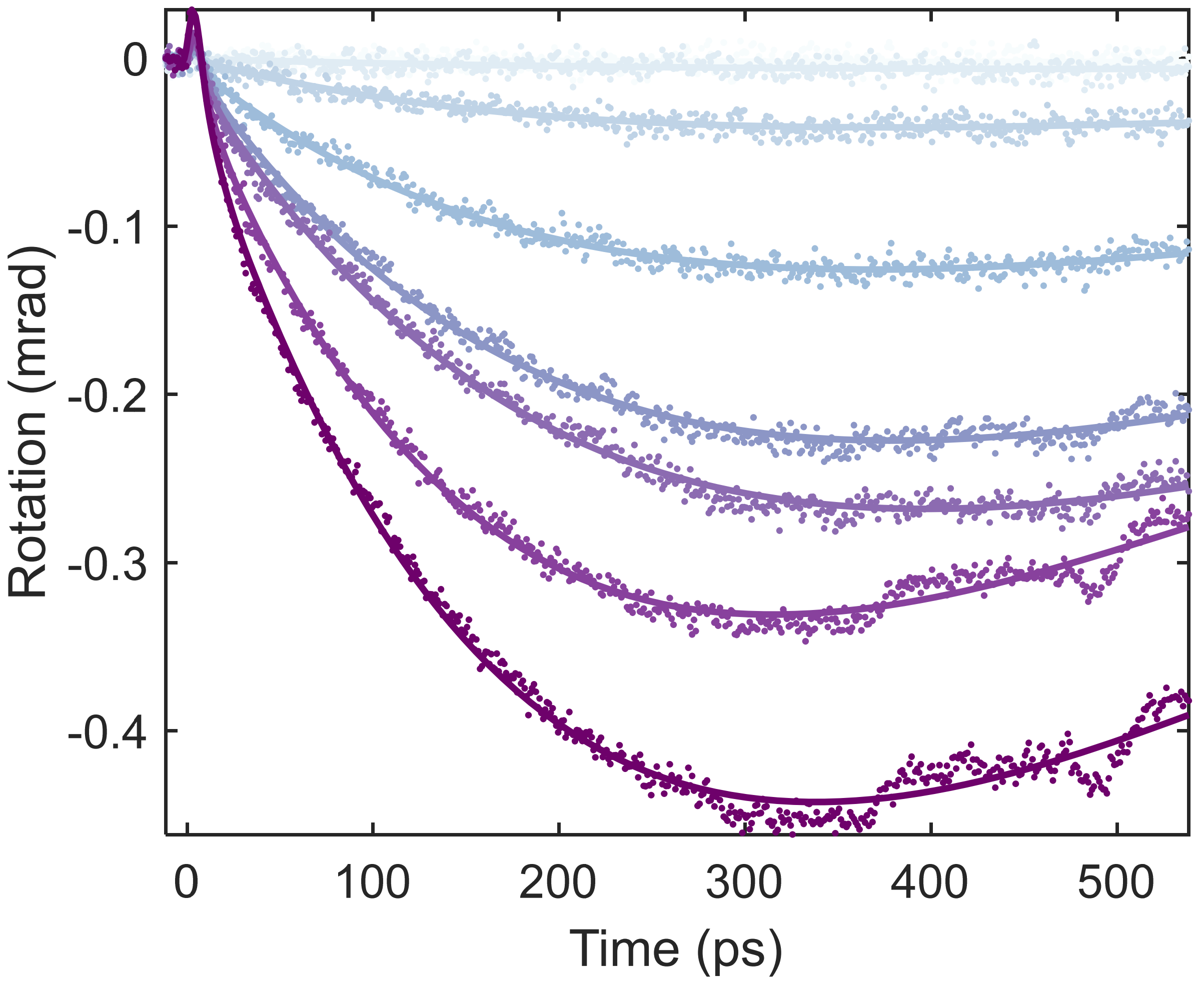


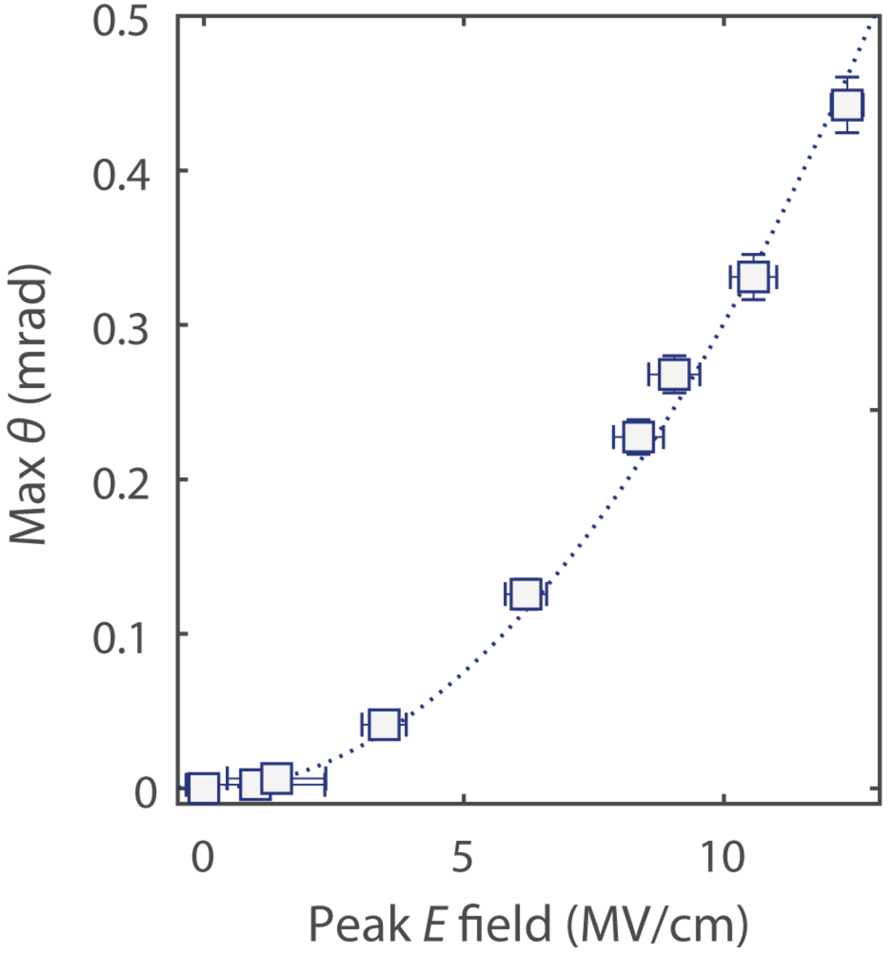
- Optical control over direction and magnitude of induced magnetization

Dependence on pump strength



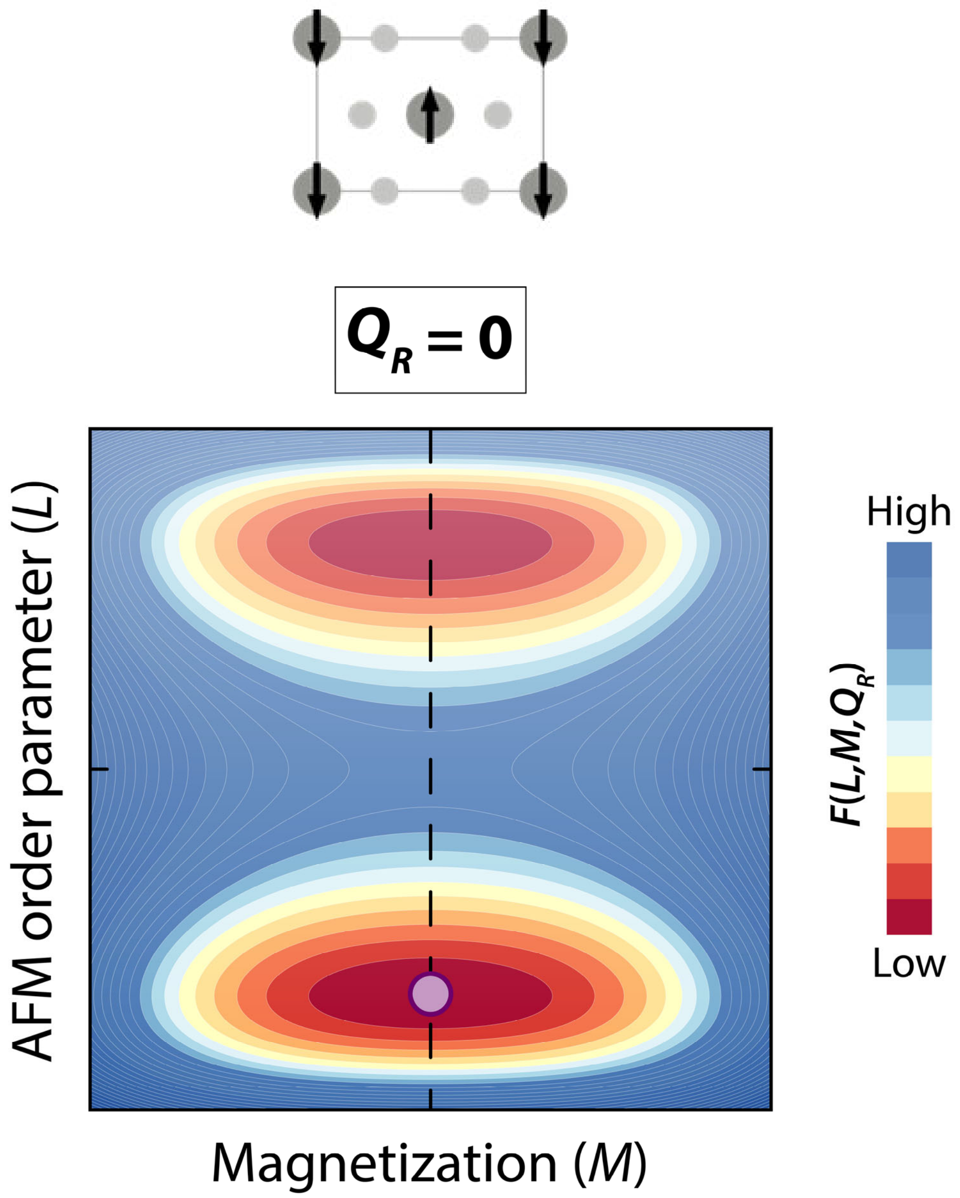
Dependence on pump strength





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

Phenomenological model of dynamics



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

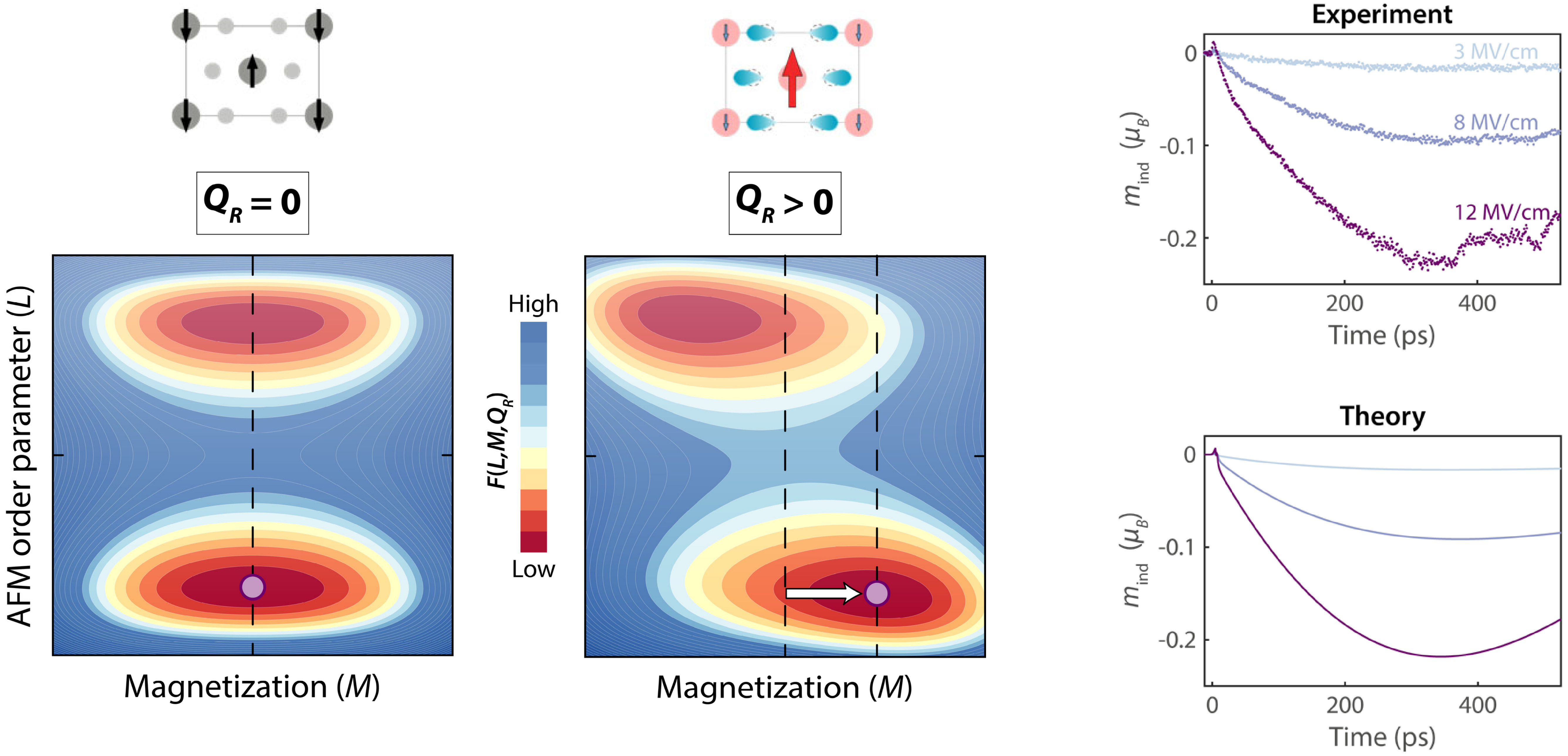
$$L = \left(m_1^0 + \frac{\delta m}{2}\right) - \left(m_2^0 + \frac{\delta m}{2}\right) = L_0$$

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

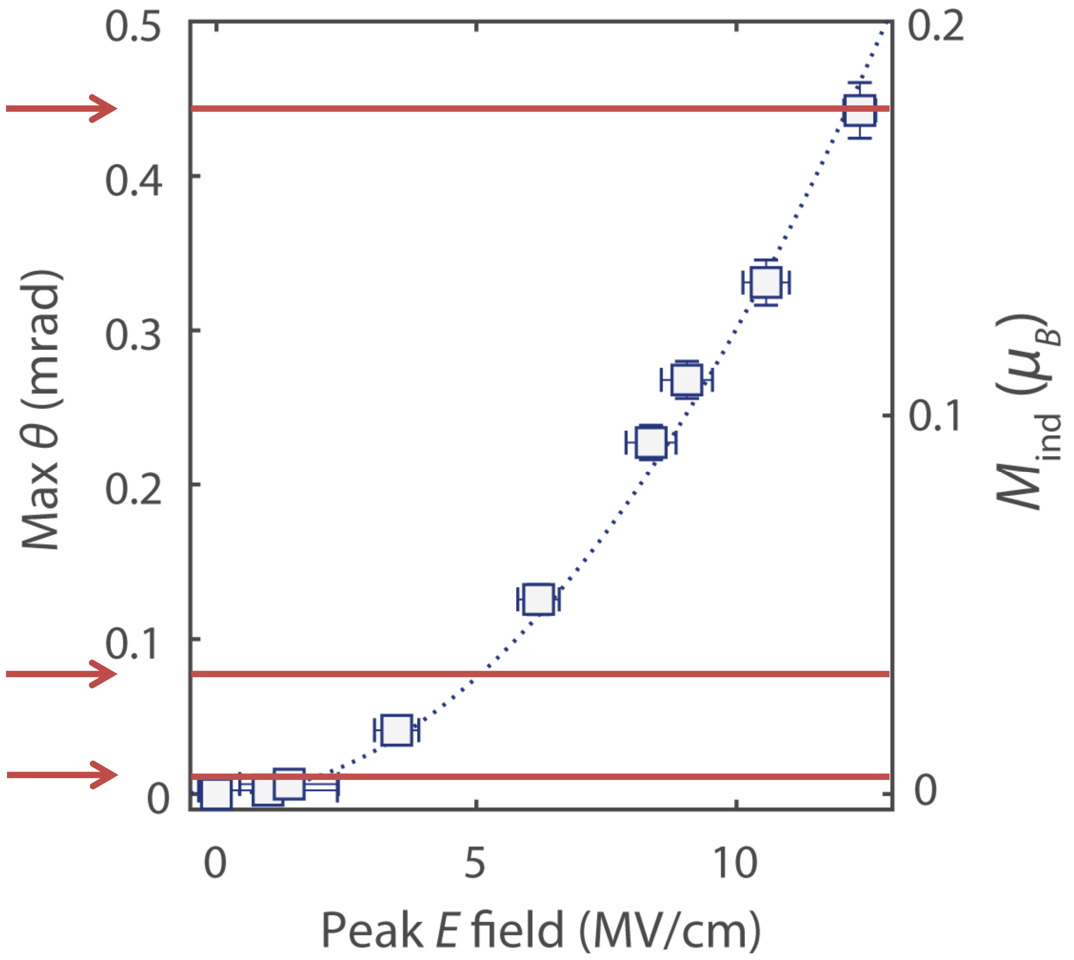


Strength of induced magnetization

Equivalent to ~50 GPa uniaxial stress

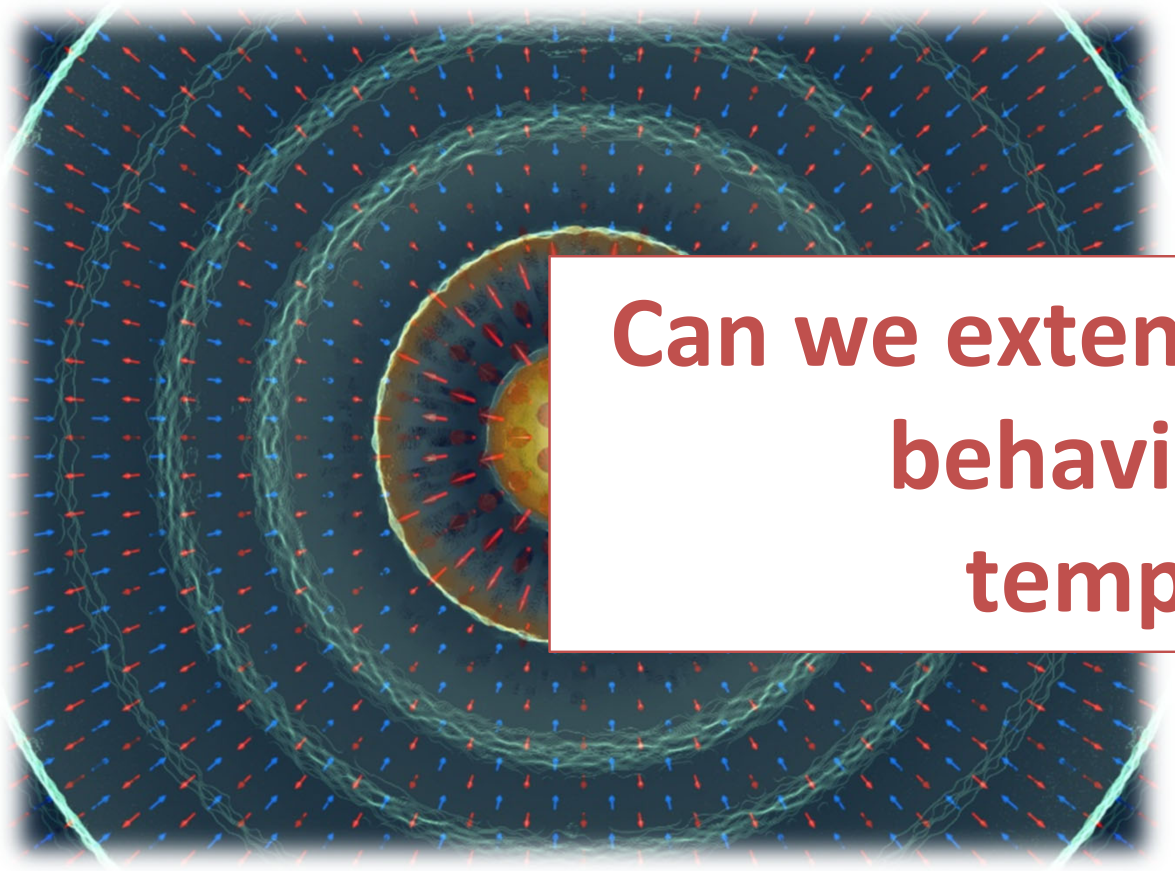
Tensile strength (strain at which crystal breaks)

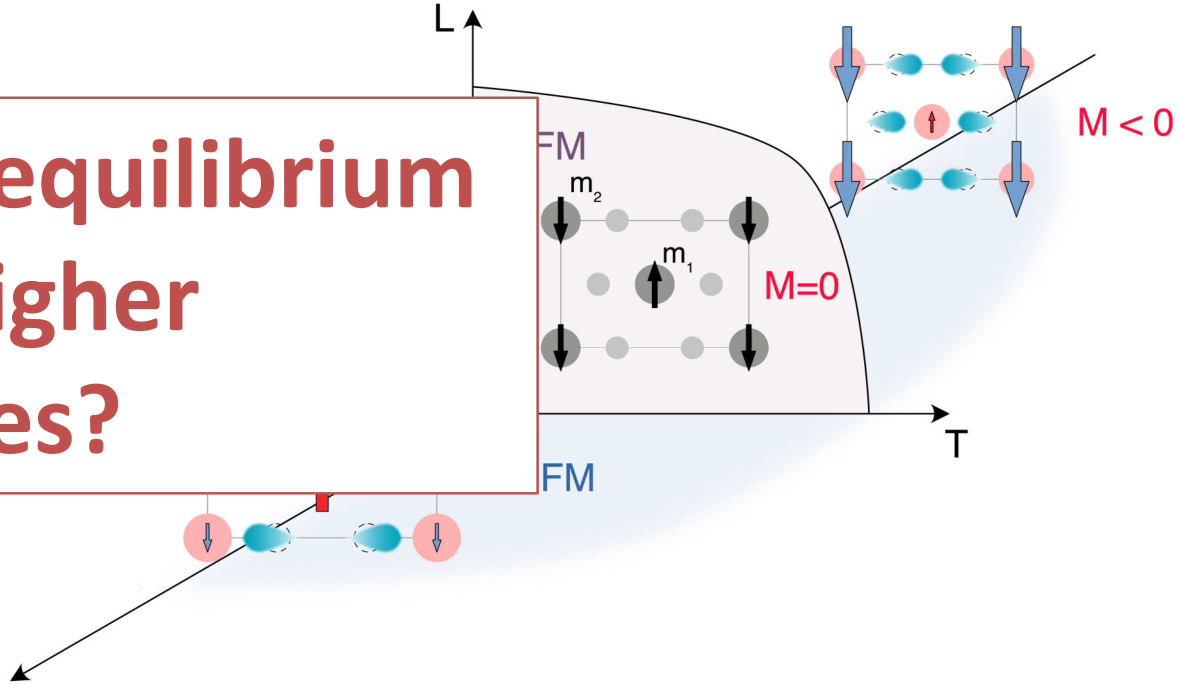
Largest strain-induced magnetization achieved



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

Non-equilibrium control of magnetism



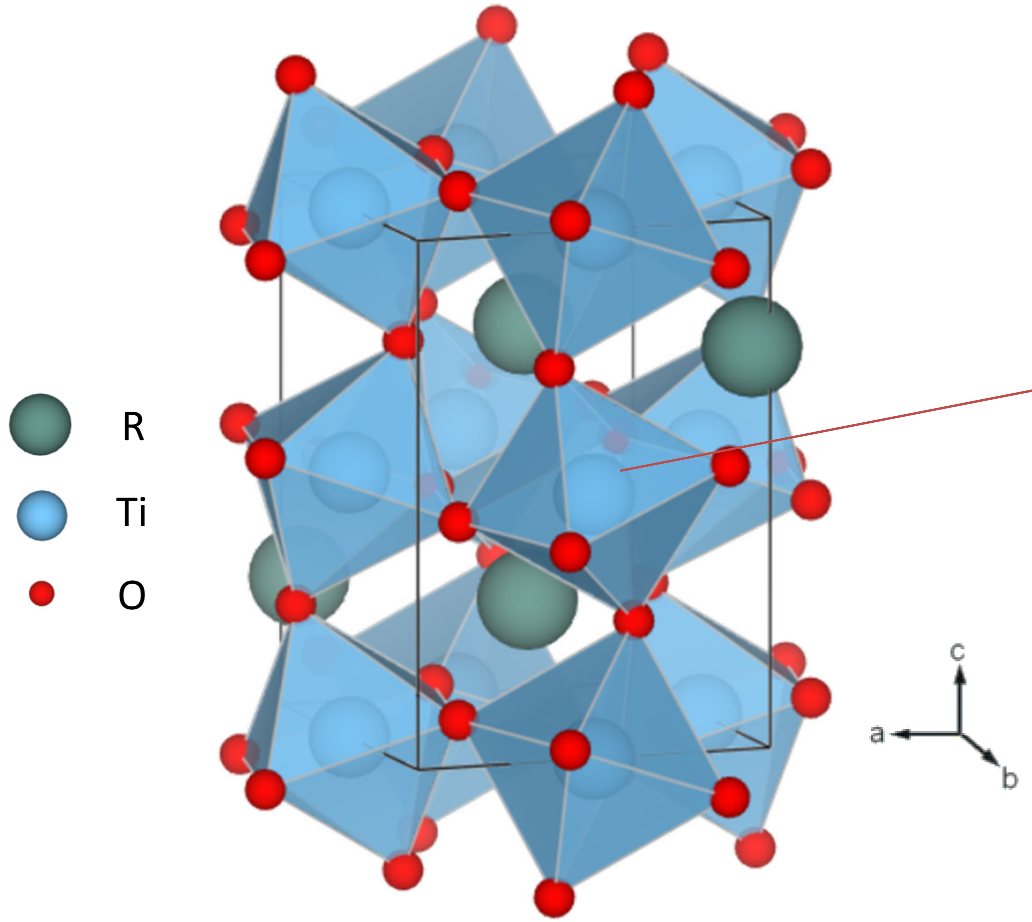


Can we extend non-equilibrium behavior to higher temperatures?

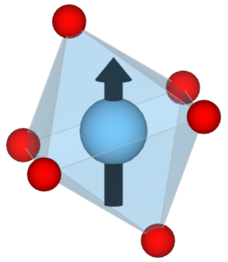
- Demonstrated control of magnetic state through crystal lattice ***below equilibrium*** T_c

Rare-earth titanates – correlated magnets

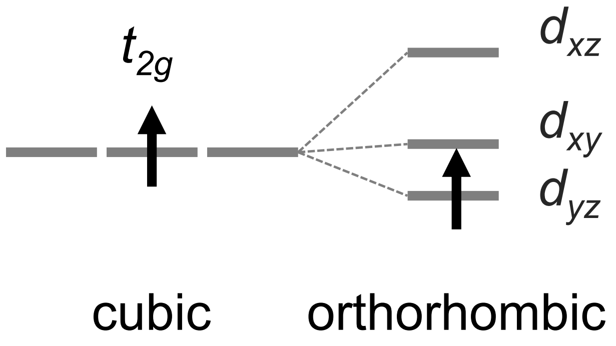
***R*TiO$_3$** (*R* = Y, La, …)

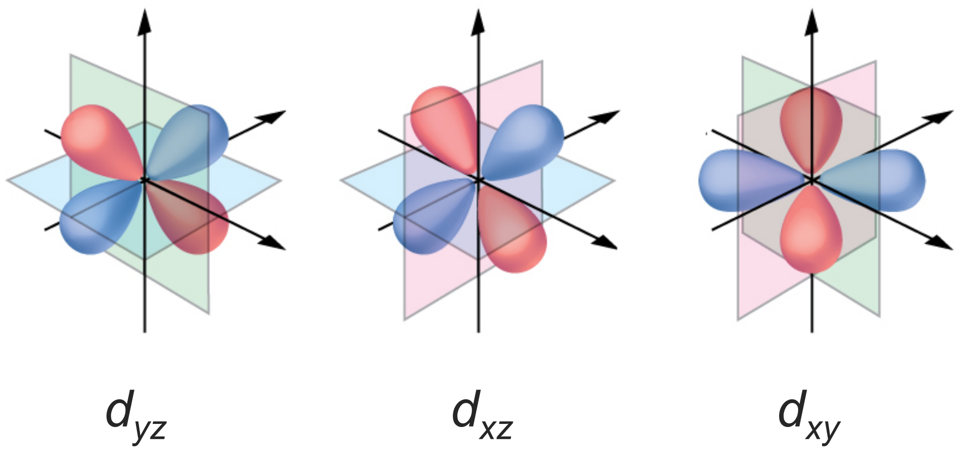


Ti 3*d*^{1} : spin = ½



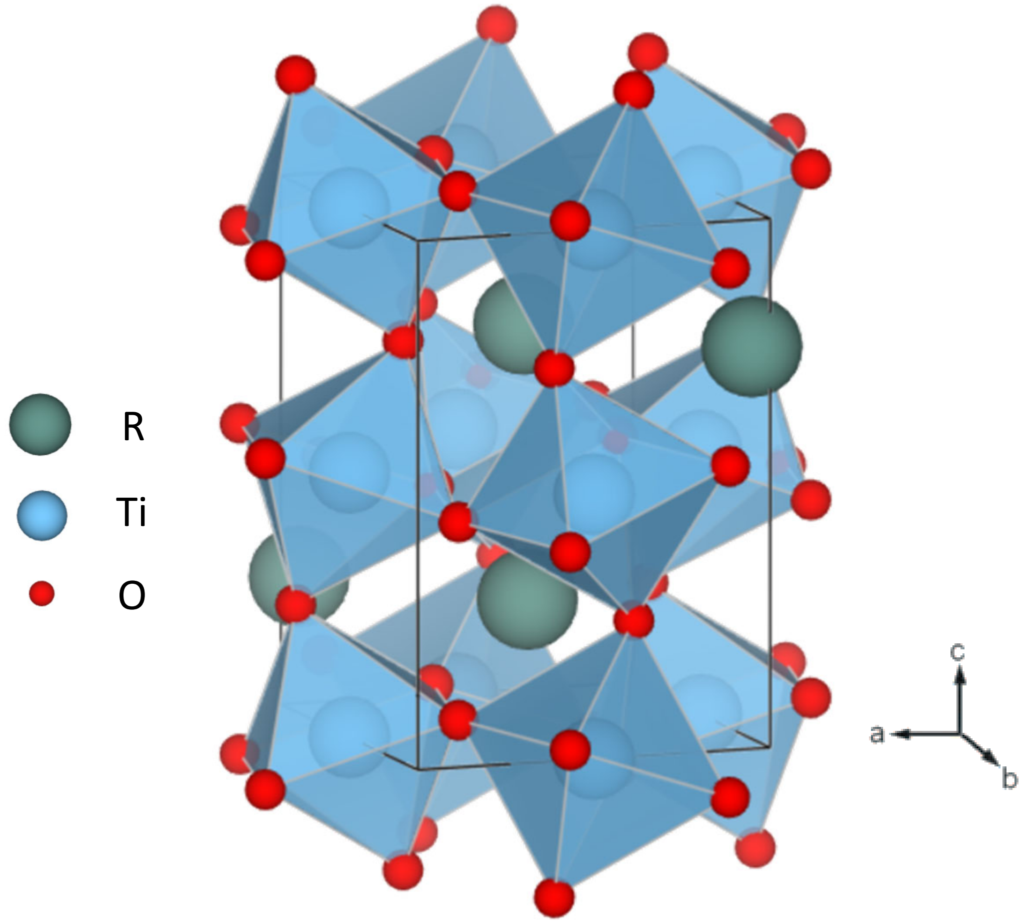
High degree of orbital degeneracy



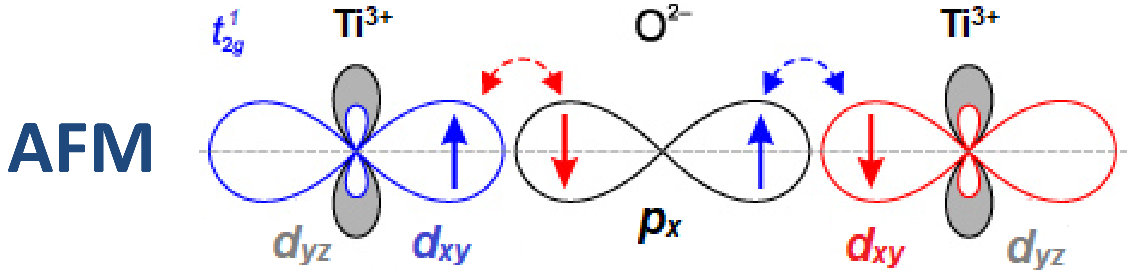


Spin-orbital-lattice interactions

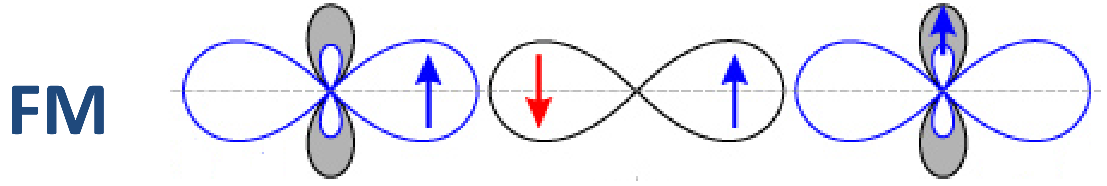
***R*TiO_3** (*R* = Y, La, …)



Same orbitals



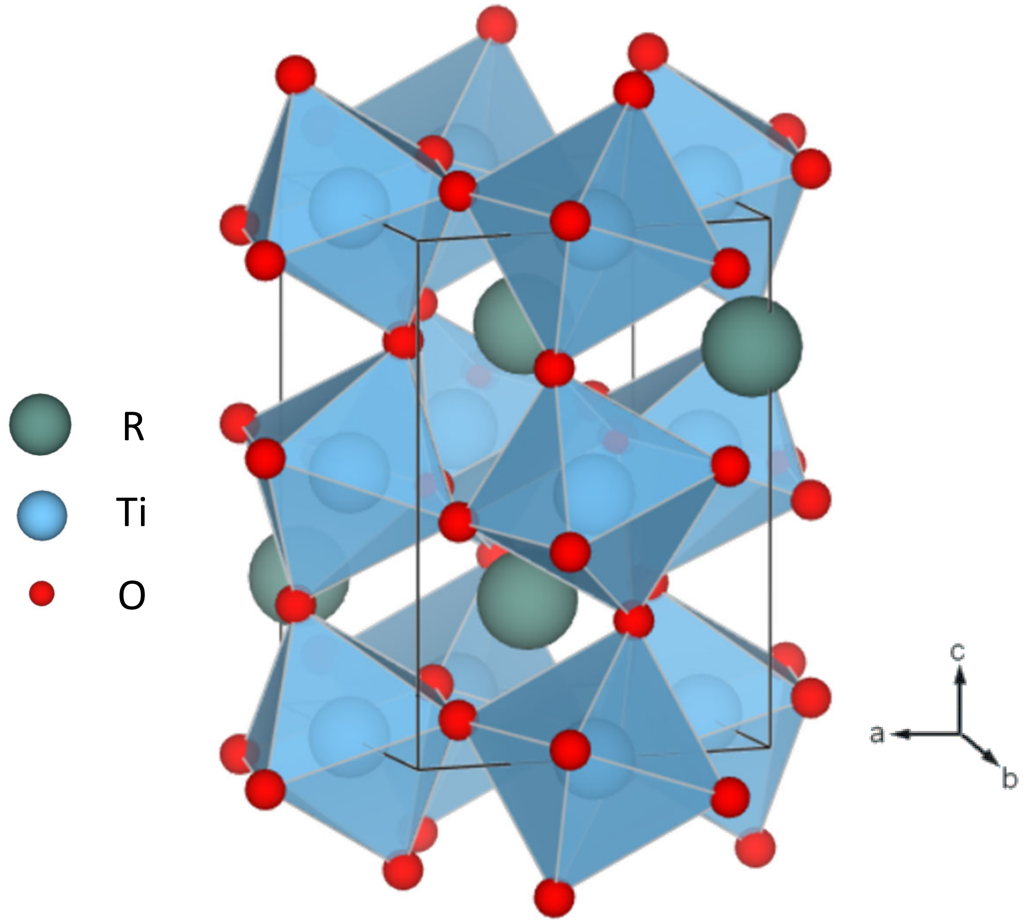
Different orbitals

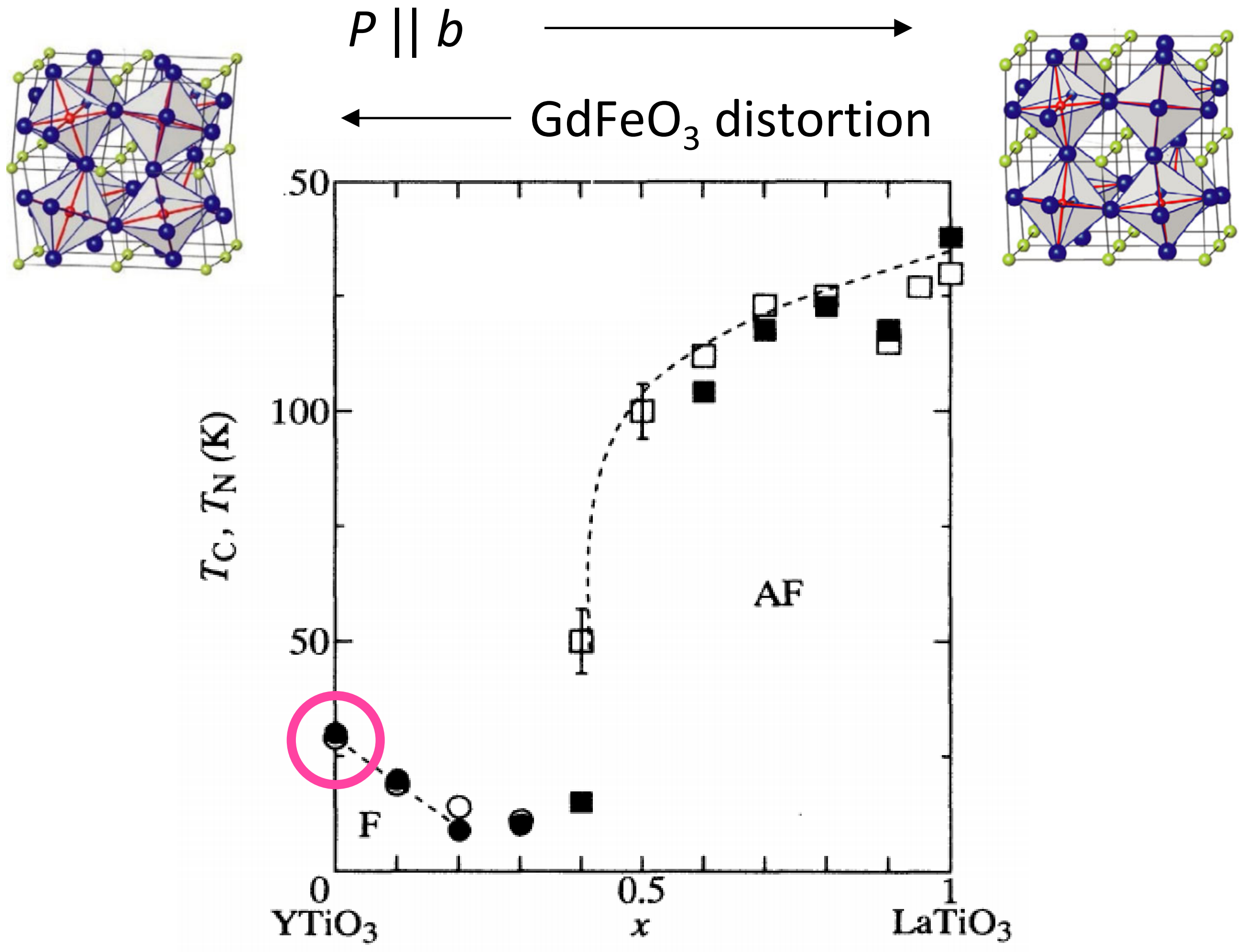


Magnetism highly coupled to crystal lattice and orbital configuration

Magnetic phase diagram

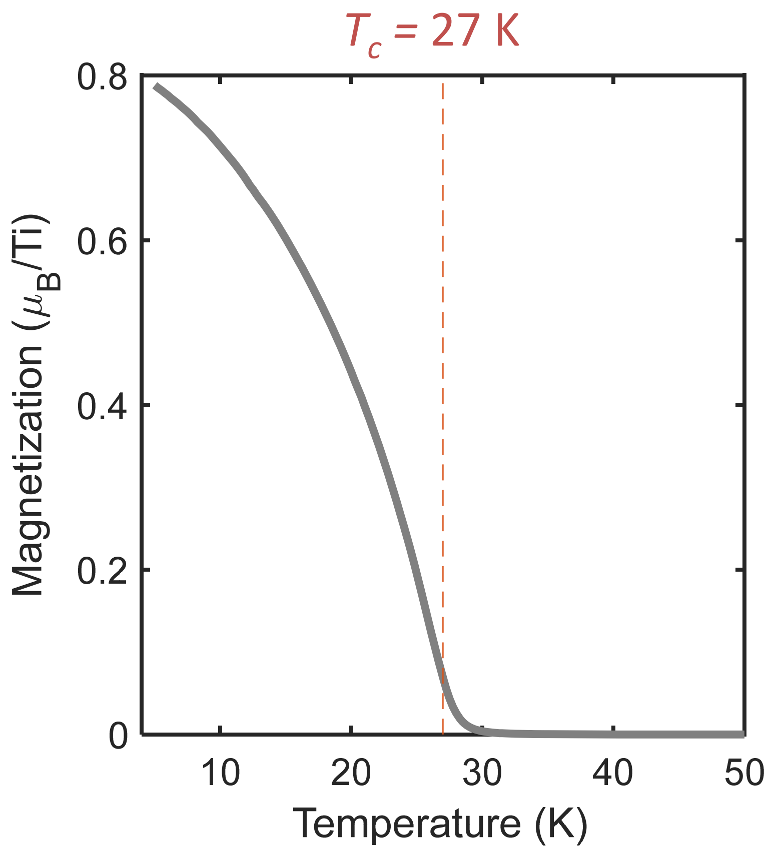
***R*TiO₃** (*R* = Y, La, …)

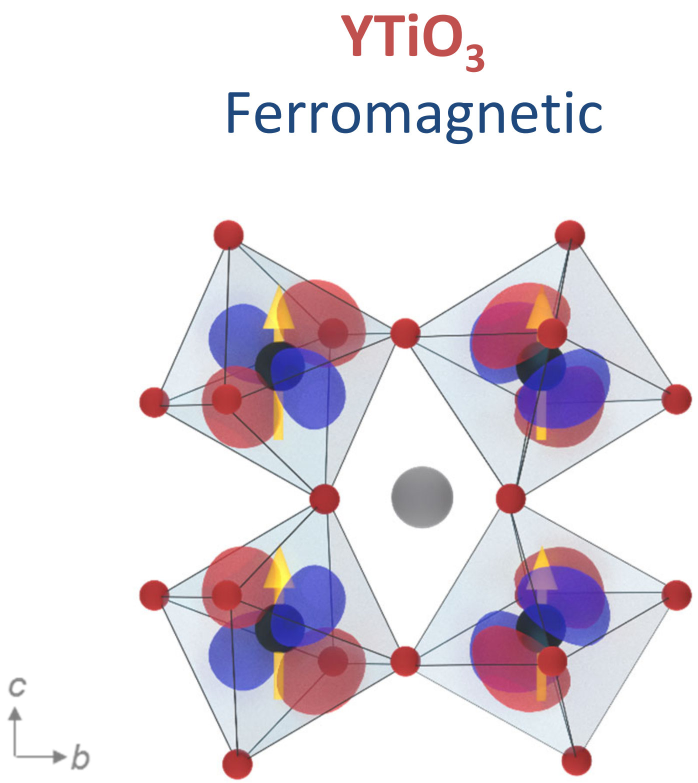


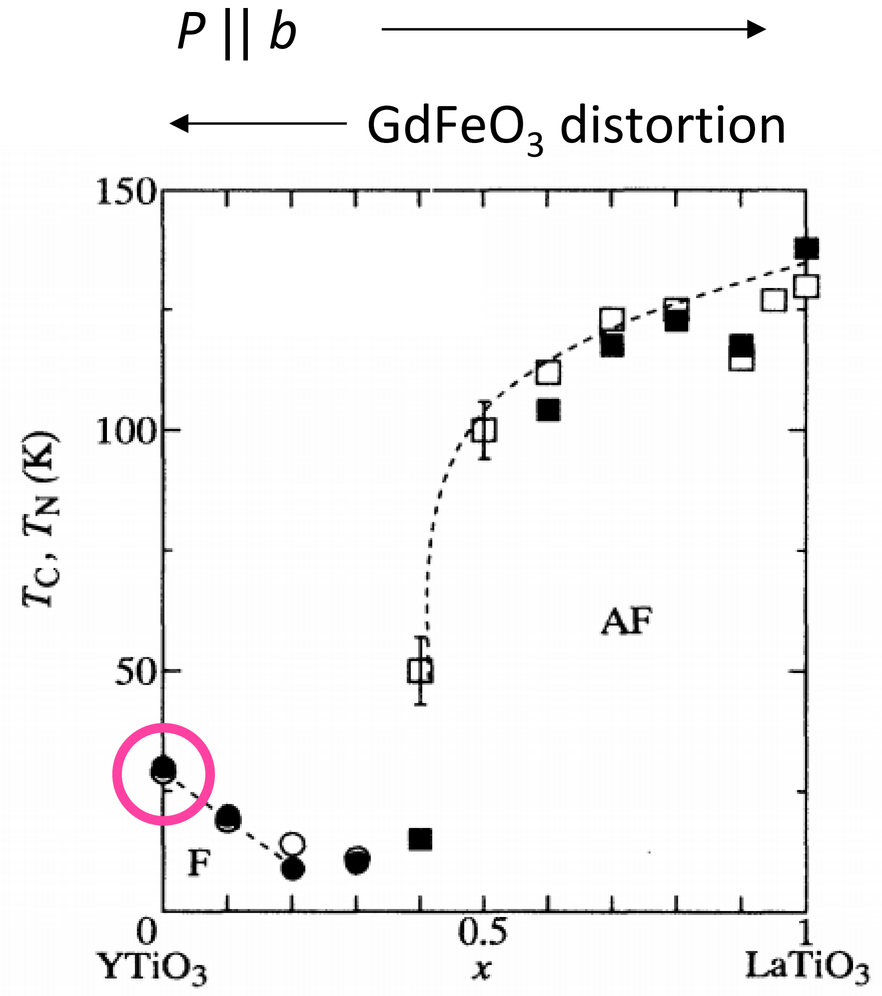


- **Magnetic order tunable by structural distortions**

$YTiO_3$ – a fluctuating ferromagnet

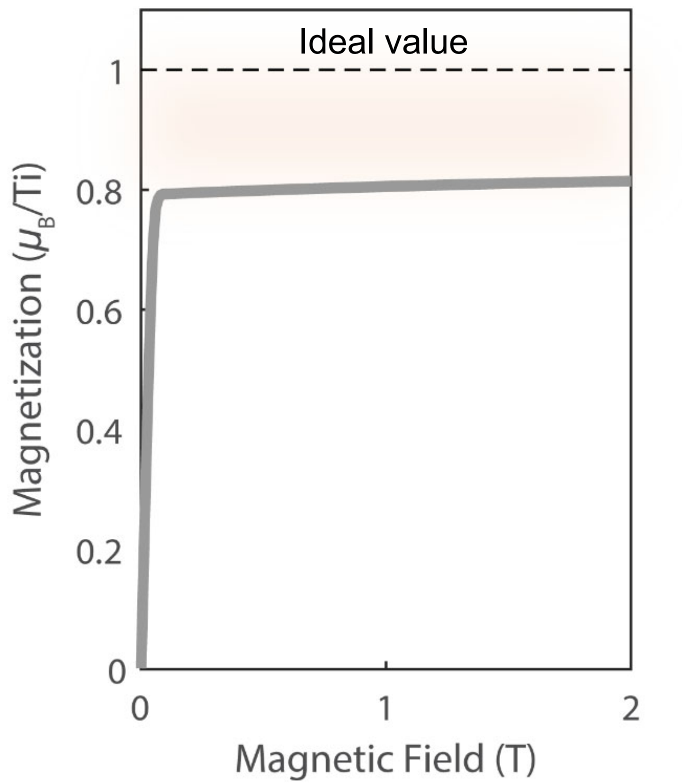




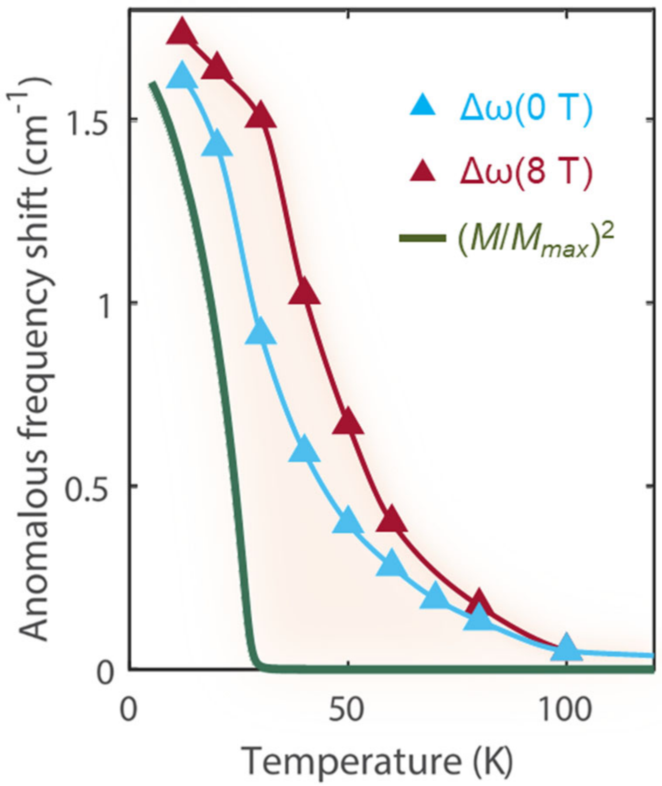


Evidence for strong fluctuations in $YTiO_3$

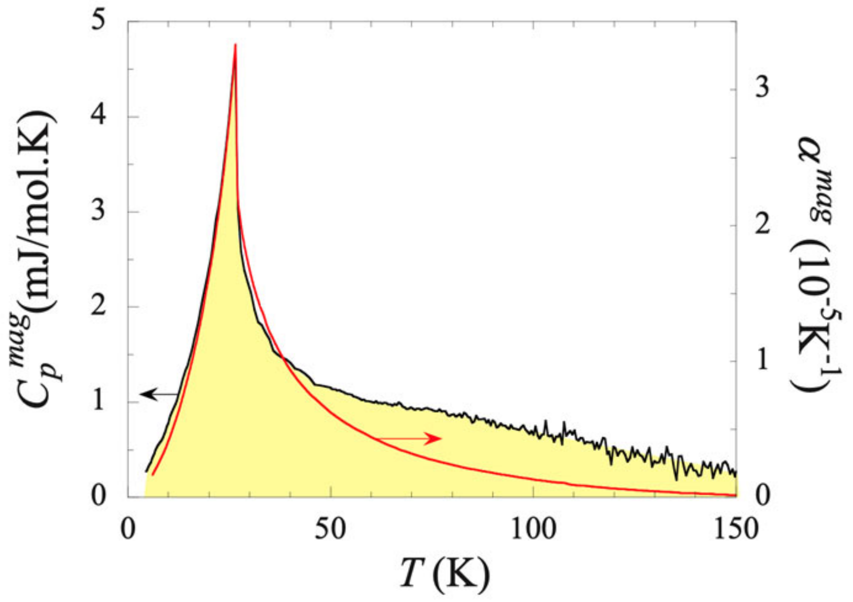
Low-temperature magnetization



Spin-lattice coupling

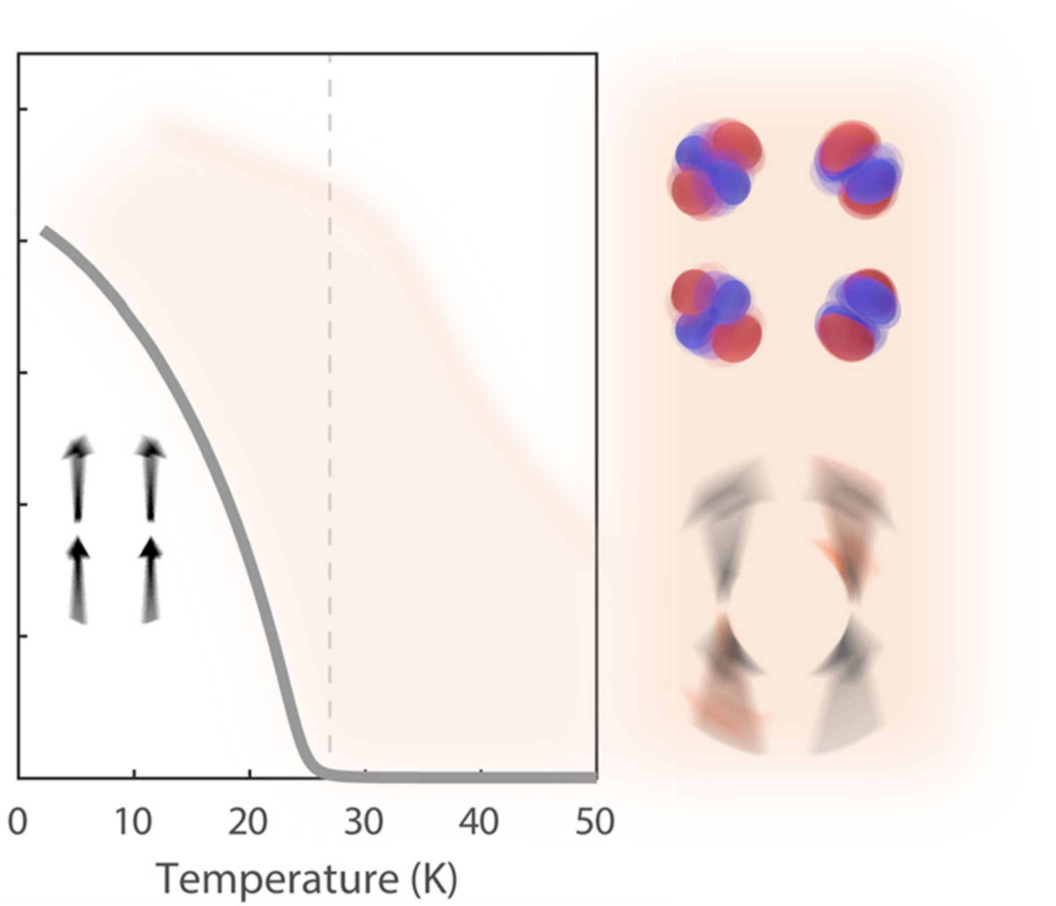


Thermodynamic measurements



- **Suppressed moment below T_c**
- **Magnetic correlations up to ~5 × T_c**

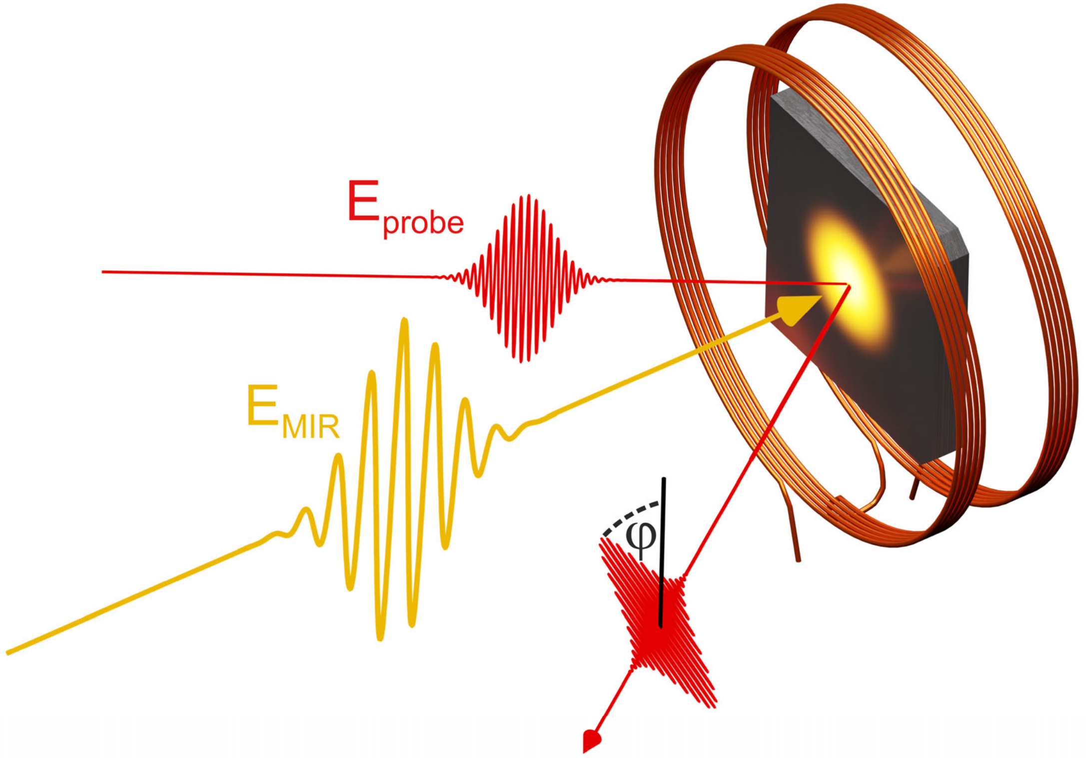
Enhancing magnetism through the lattice

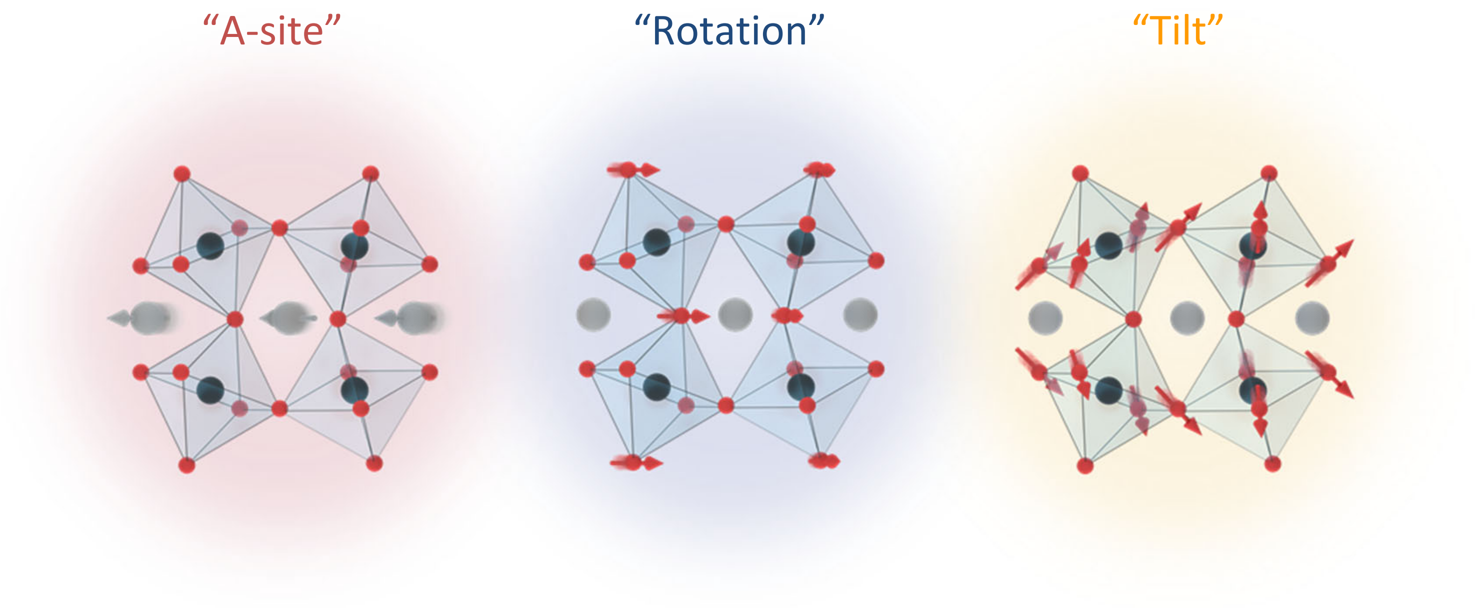


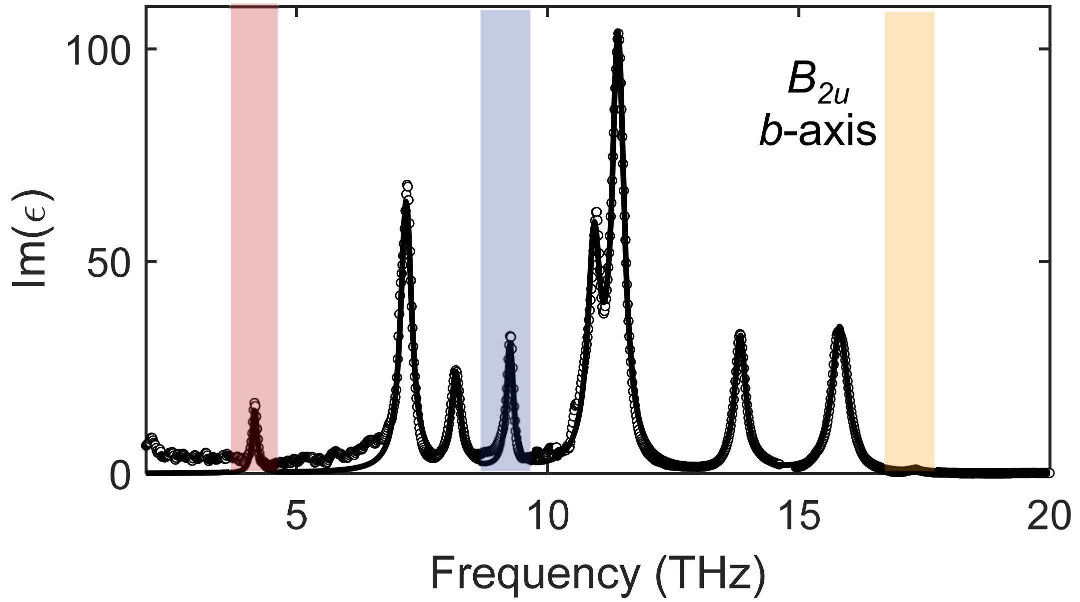
Can we optically control magnetic order through the lattice?

Can we stabilize ferromagnetism above equilibrium T_c?

Experimental design

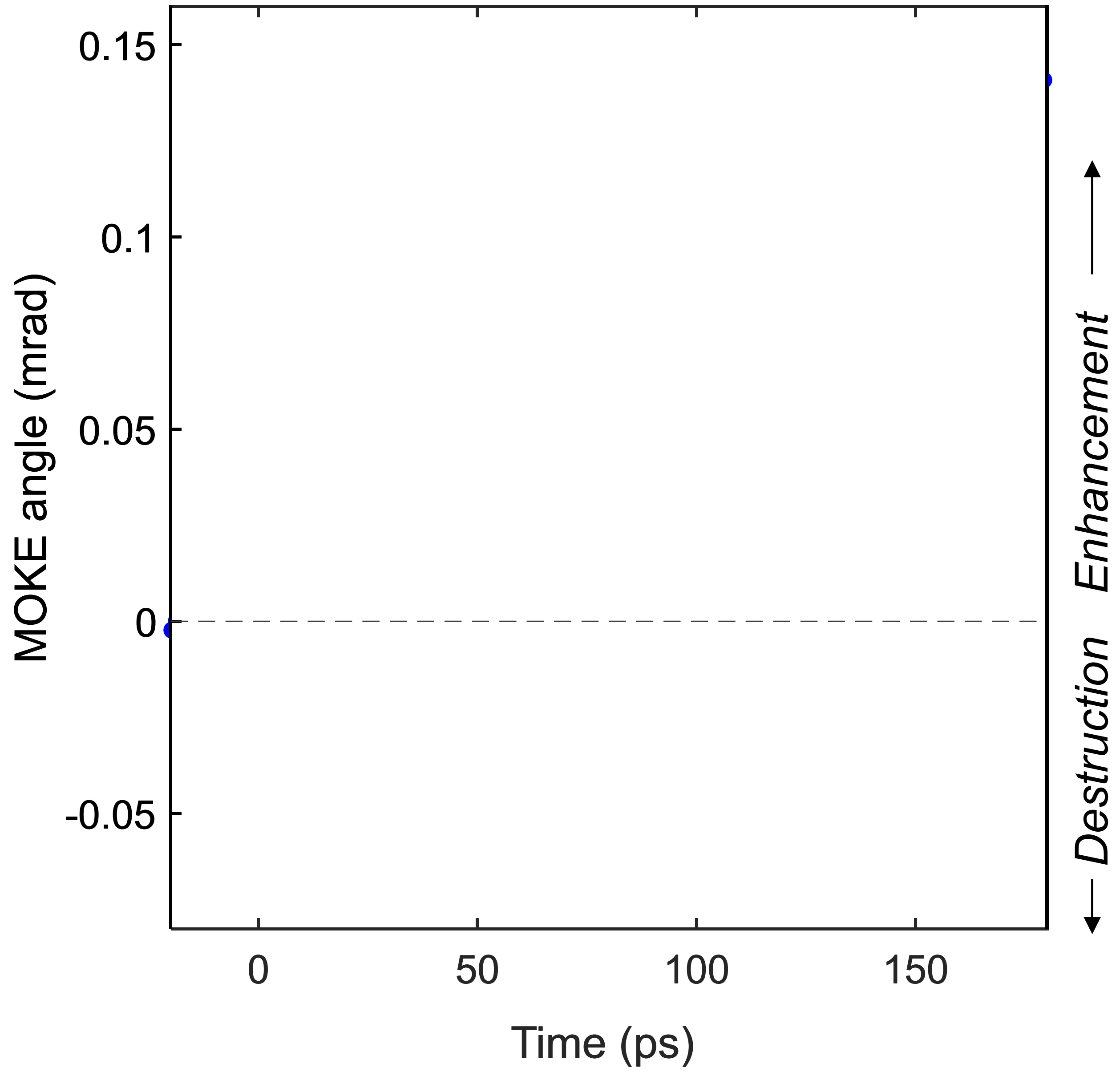


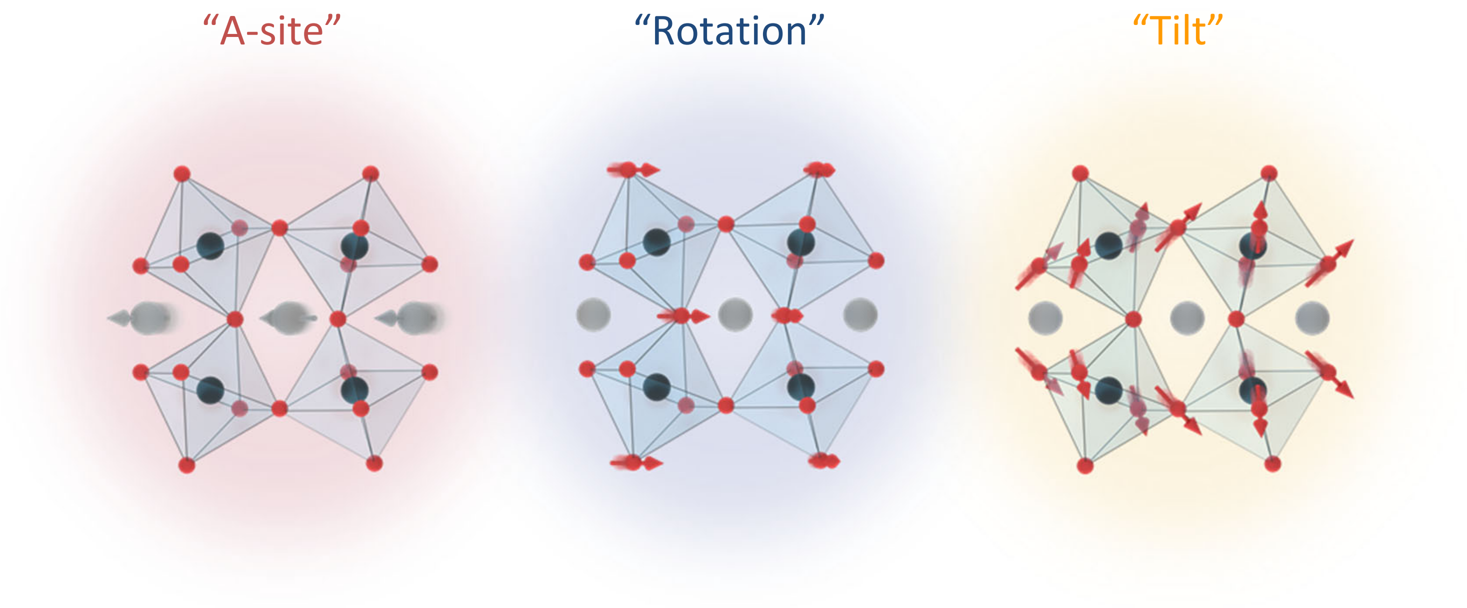


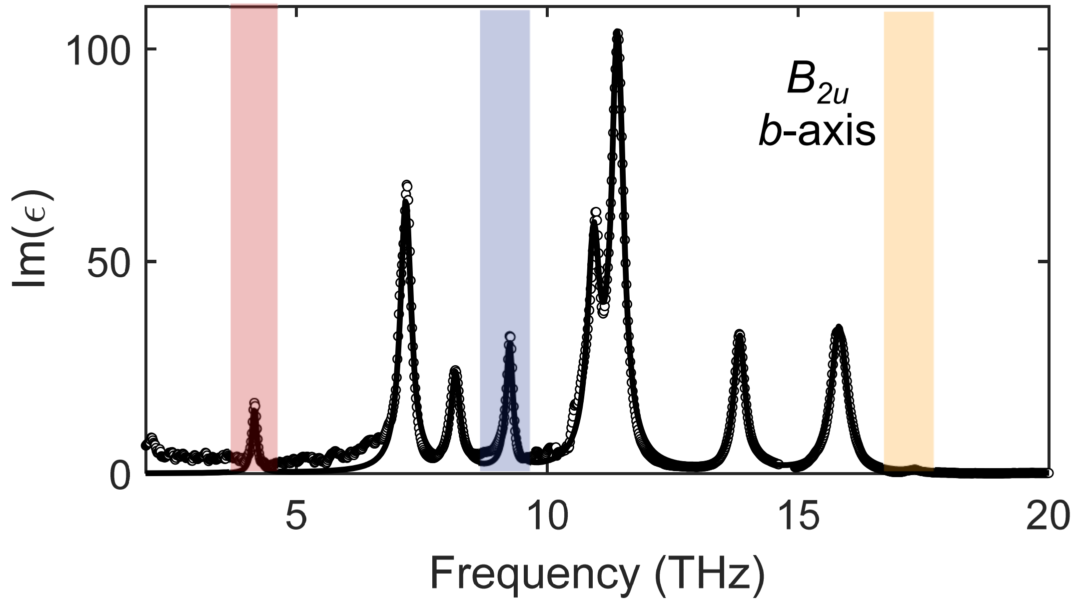


- **Time-resolved MOKE experiment**
  $\Delta\varphi(+H) - \Delta\varphi(-H) \propto \Delta M$

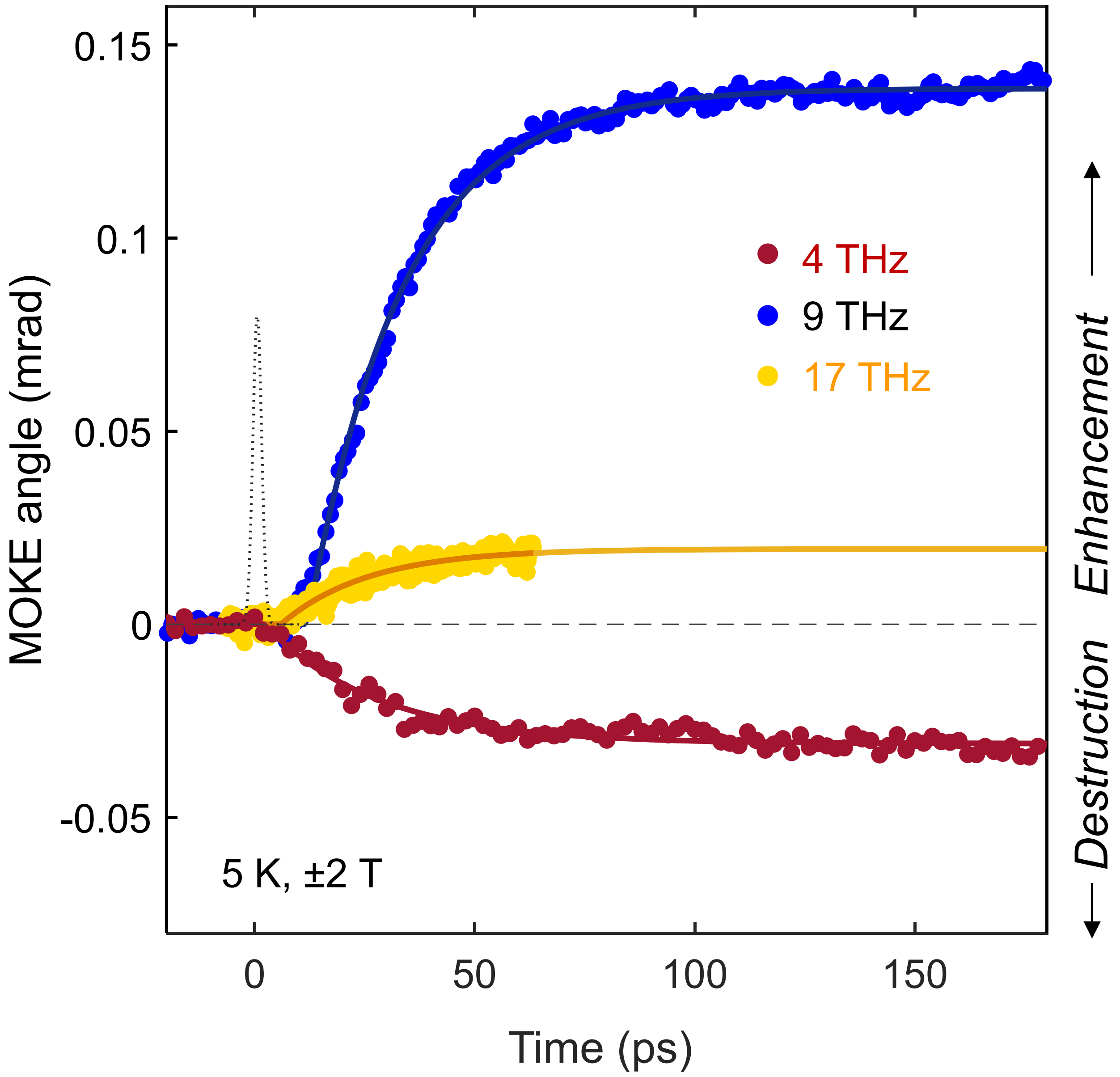
Effect of pumping different phonons

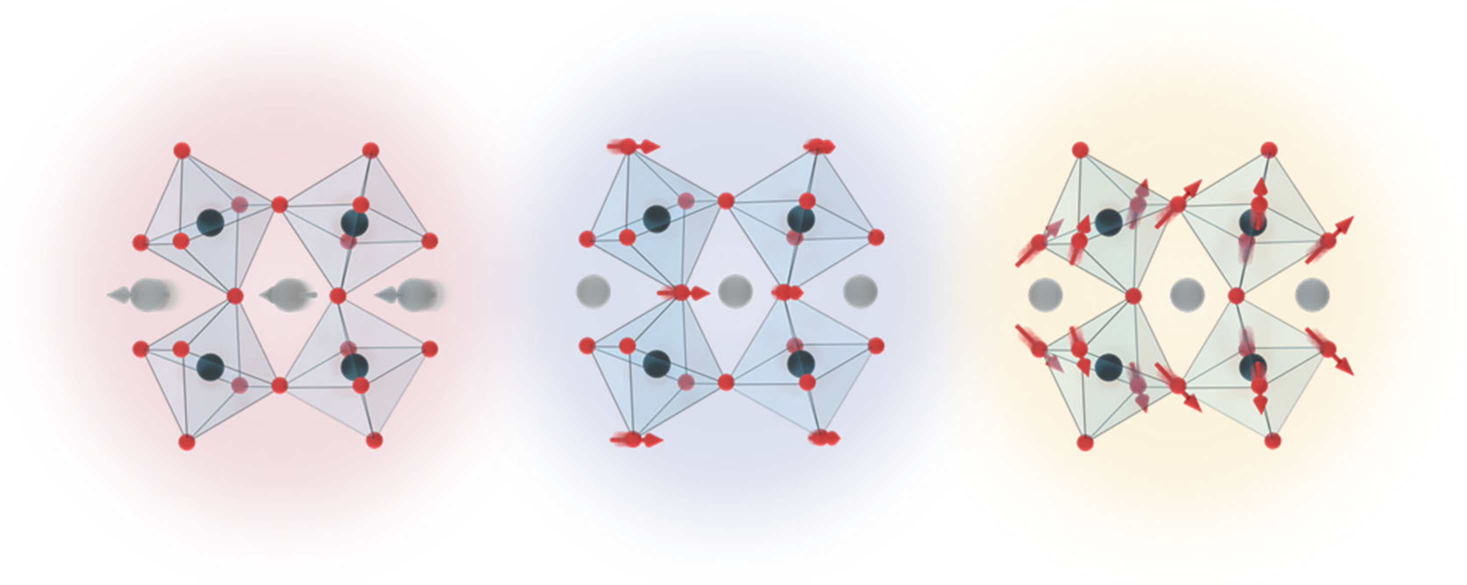






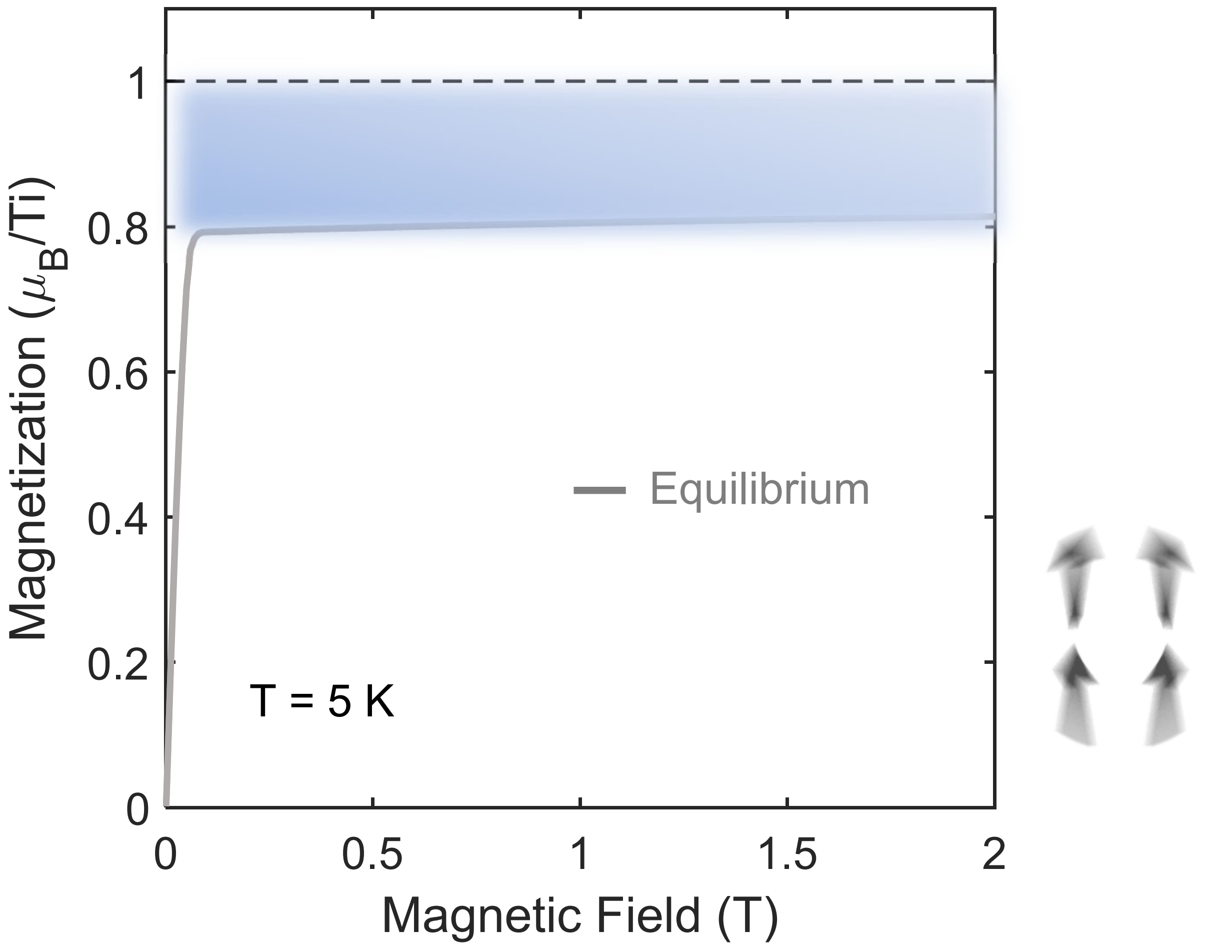
Effect of pumping different phonons





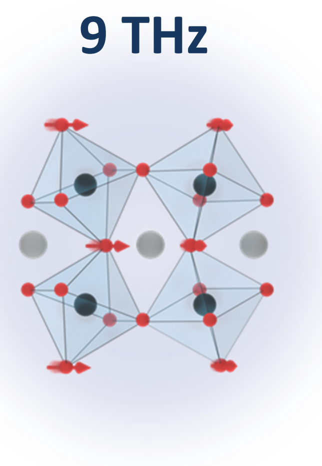
- **Phonon-selective manipulation of ferromagnetism**

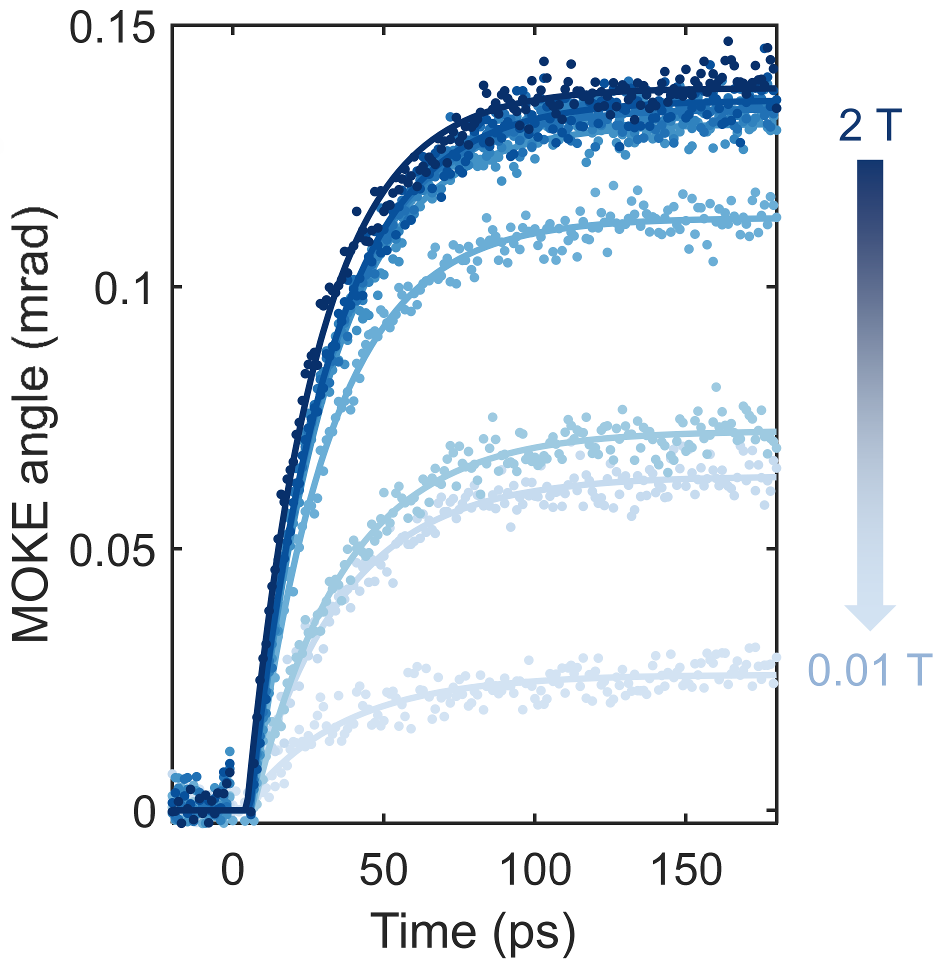
Magnetic field dependence **below** T_c

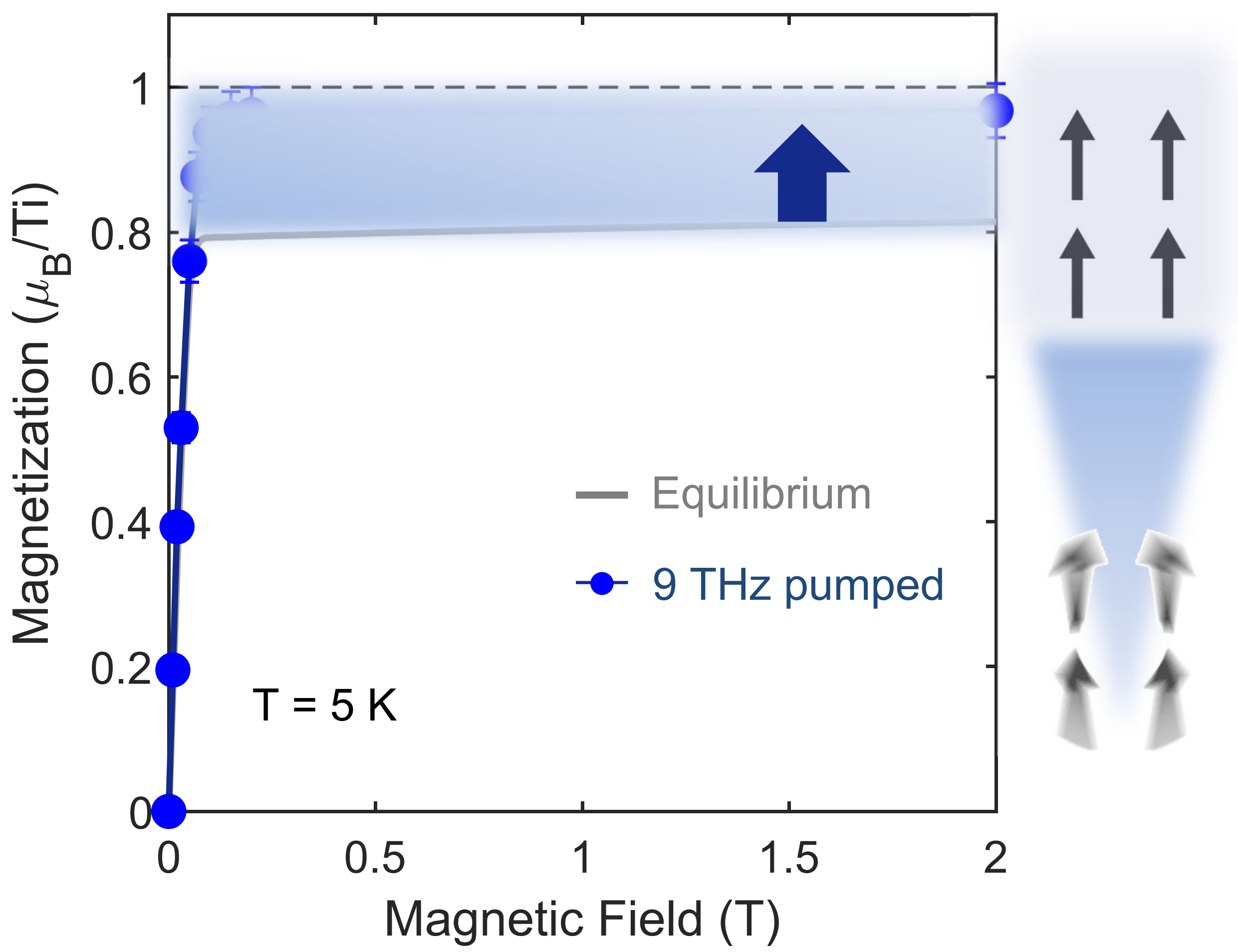


- Moment saturates well below spin- ½ limit even for $T << T_c$

Magnetic field dependence **below** T_c



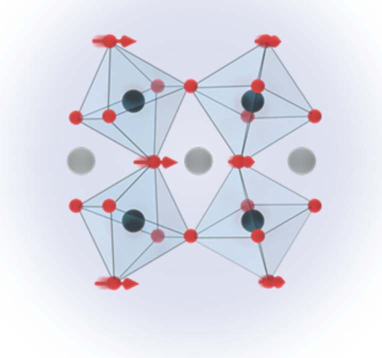




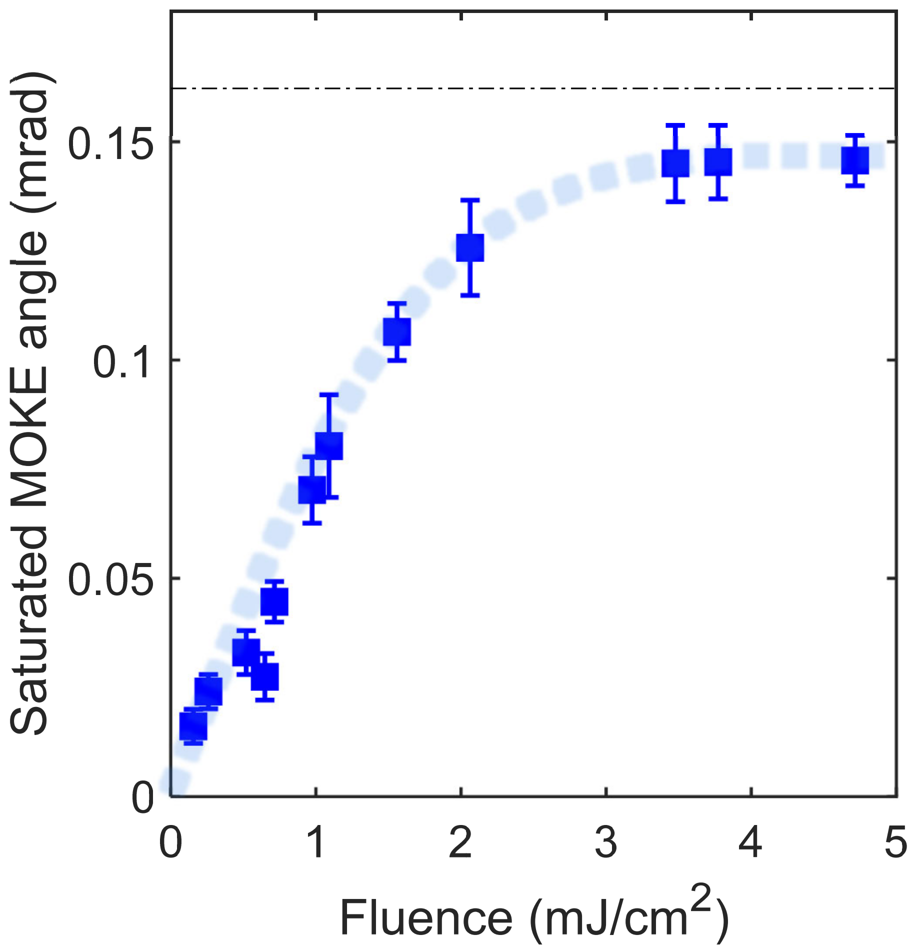
- Light-enhanced magnetization saturates close to spin-½ limit

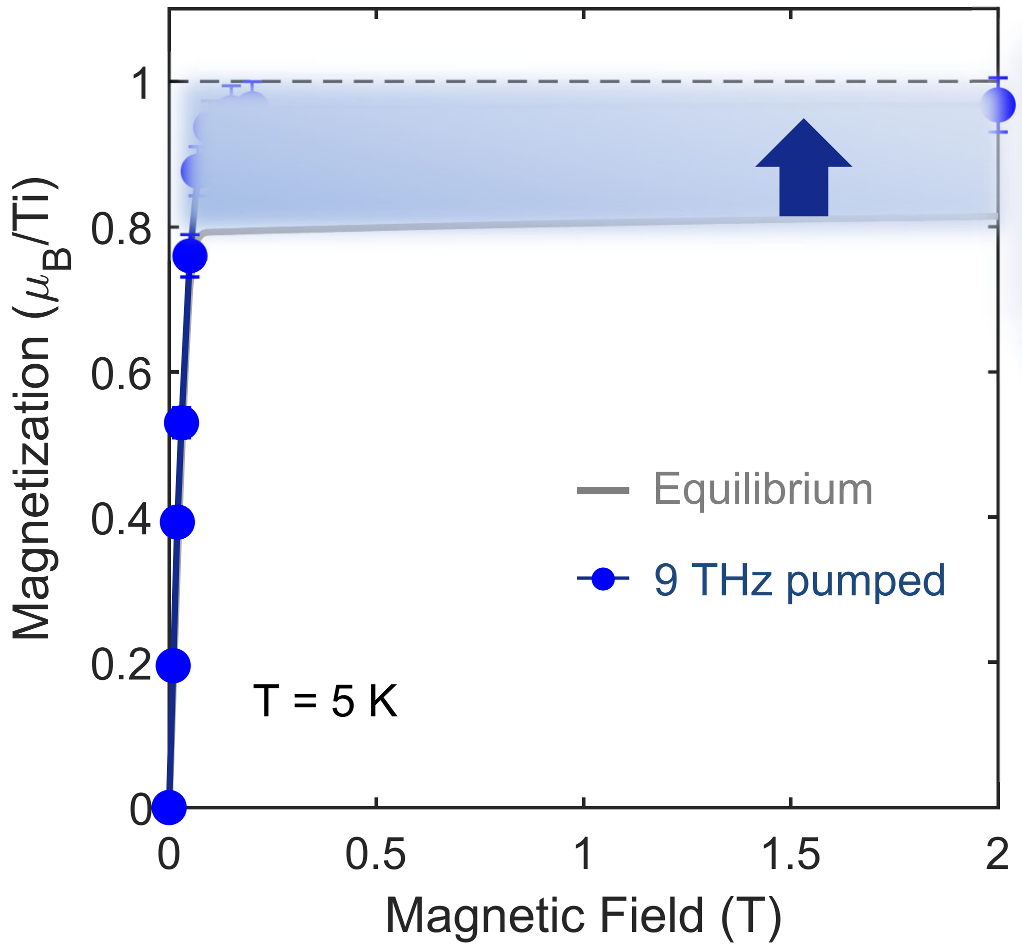
Magnetic field dependence **below** T_c

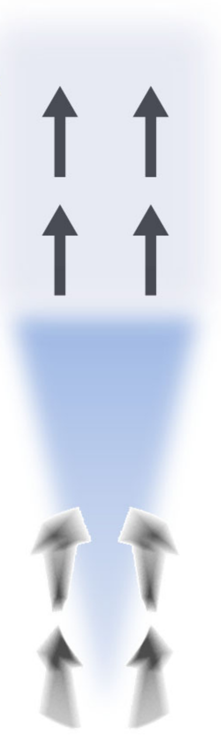
9 THz



Fluence dependence

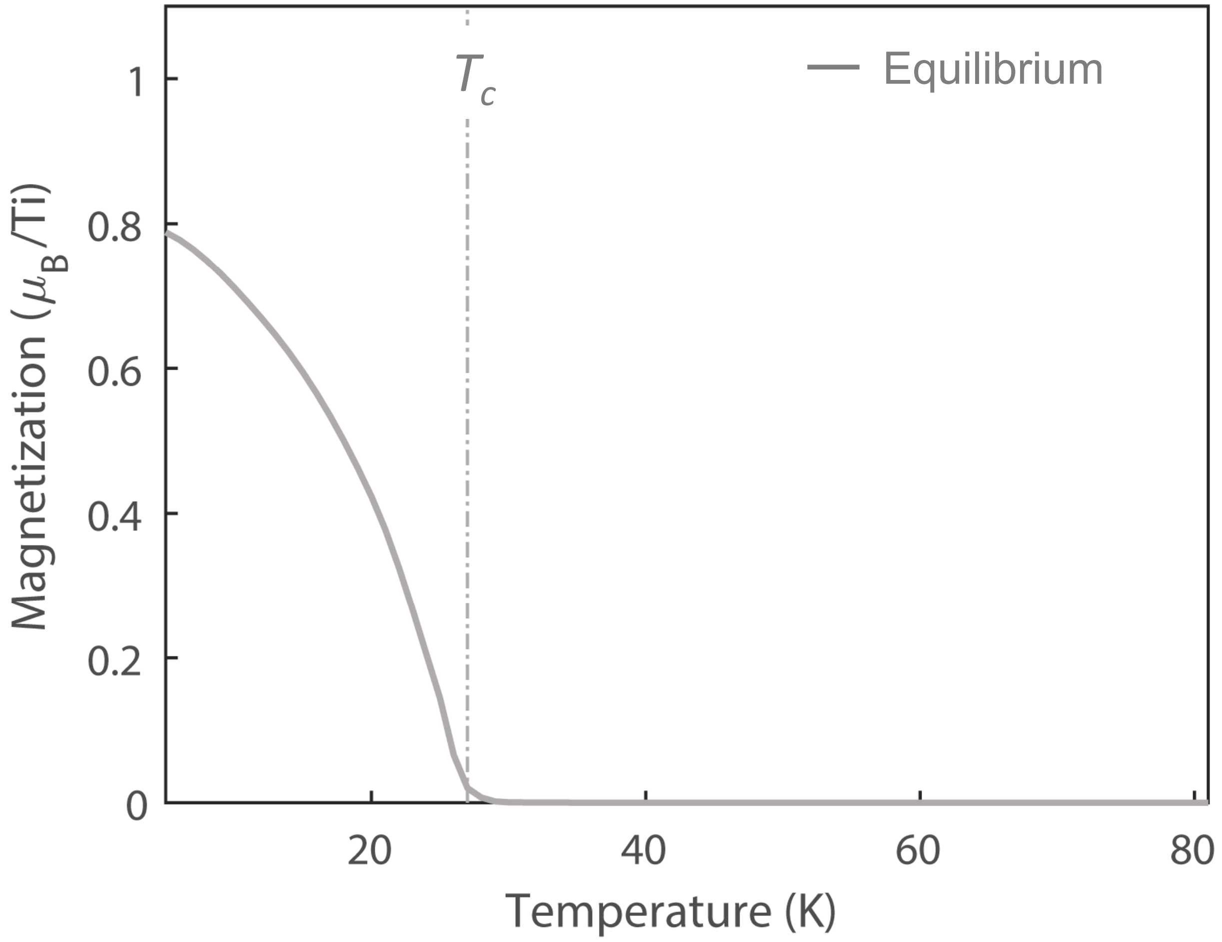




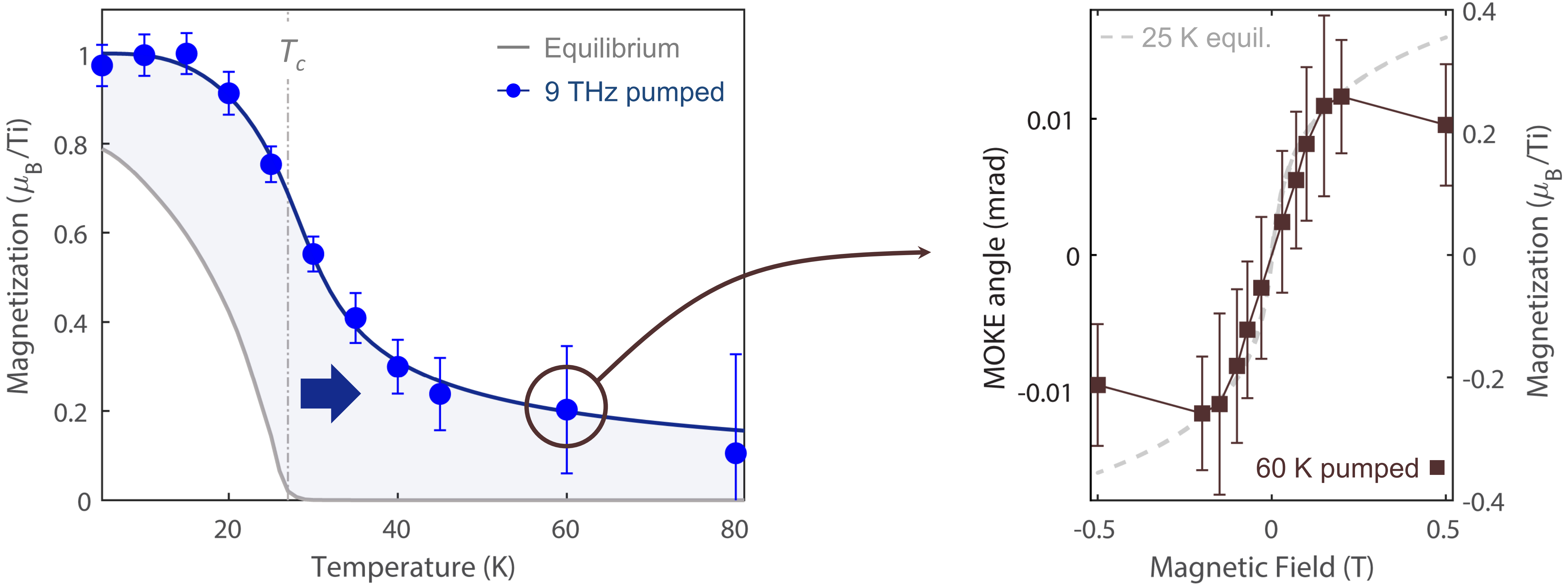


- Light-enhanced magnetization saturates close to spin-½ limit
- Saturation limit maintained with increasing fluence

Enhancement of magnetism **above** T_c

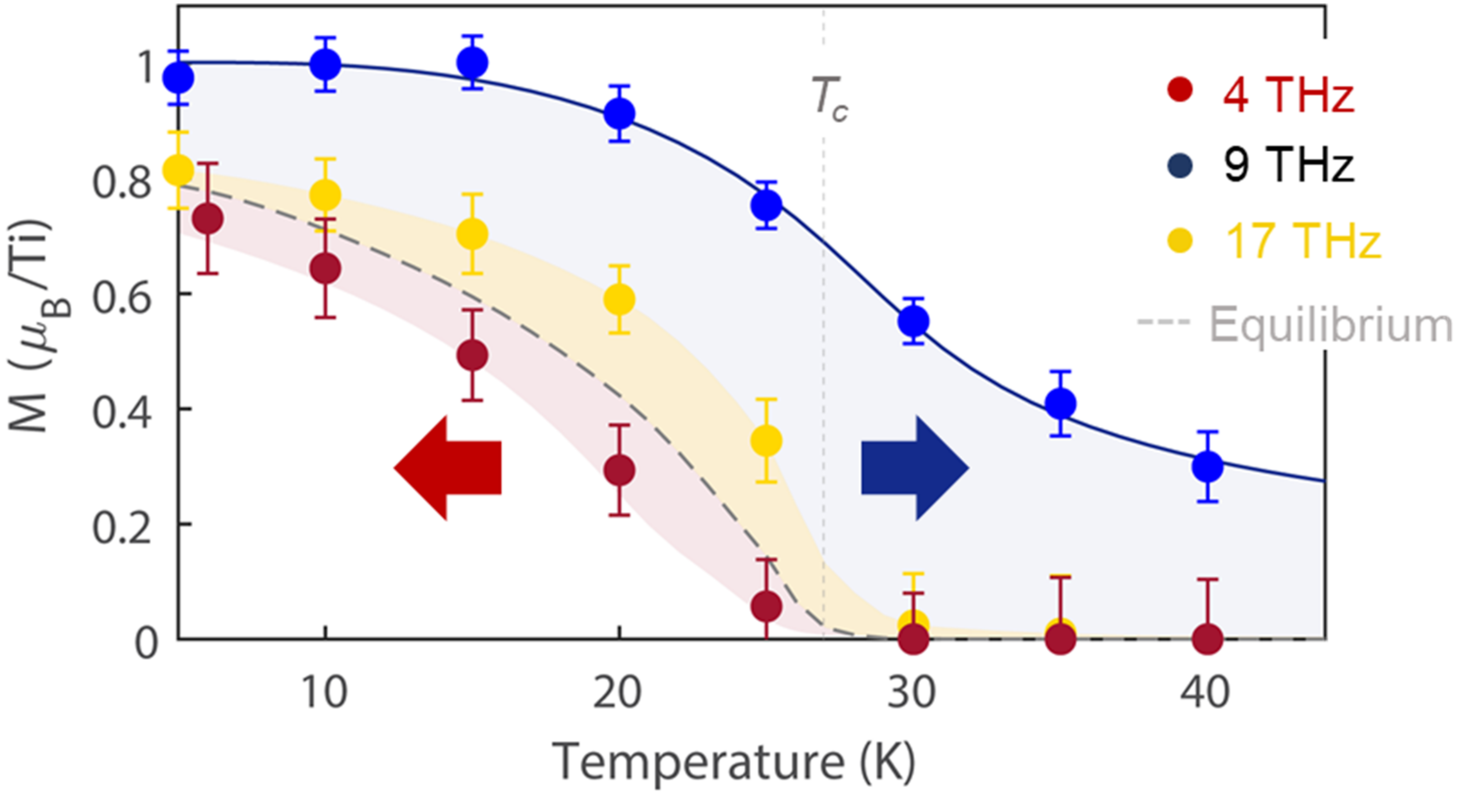


Enhancement of magnetism **above** T_c

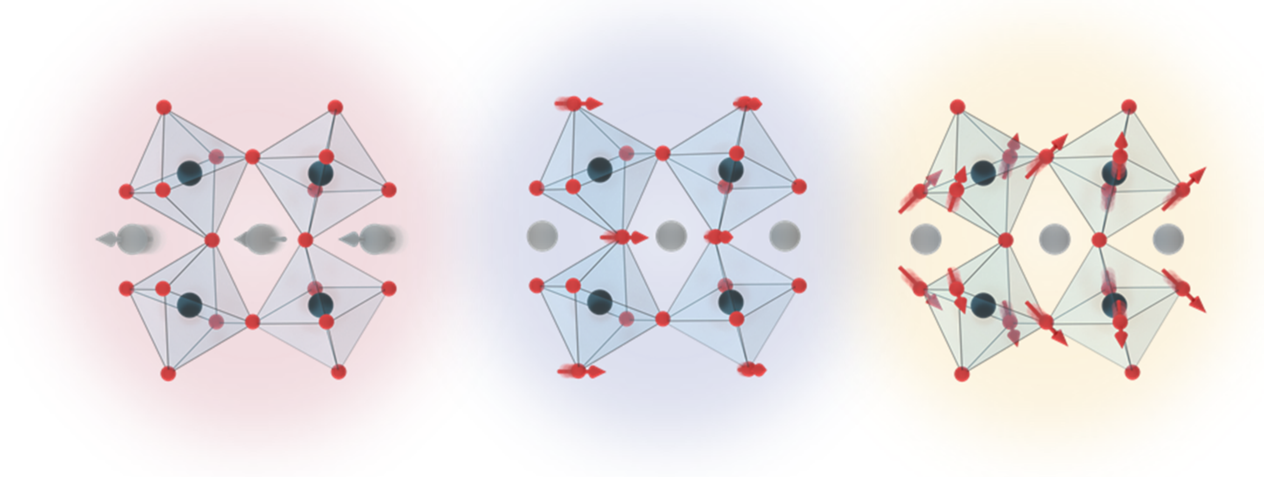


- Pump-induced magnetization up to **more than 3 × T_c**
- Non-equilibrium ferromagnetic state follows short-range spin correlations

Enhancement of magnetism **above** T_c



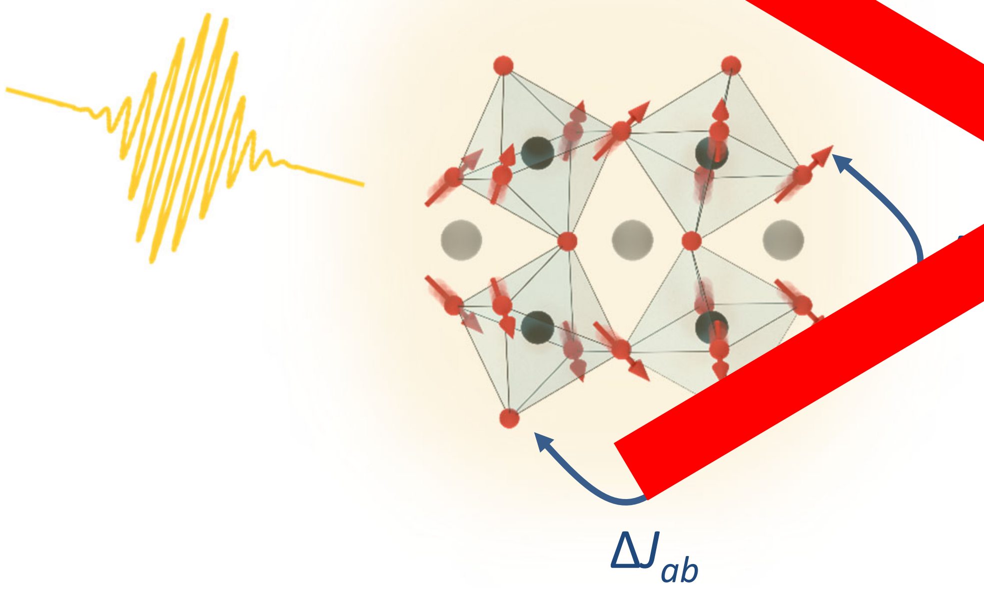
Phonon-selective enhancement/suppression



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

Origin of non-equilibrium magnetism in $YTiO_3$

Direct spin-phonon coupling?



$$E_{spin-ph} = \lambda (S_i \cdot S_j) Q^2$$

$$\Rightarrow \Delta \boldsymbol{J} = \boldsymbol{\lambda Q^2}$$

- Too small
- Wrong sign

Heating?

Optical effects? Strain?

- Enhancement
- Phonon-selective

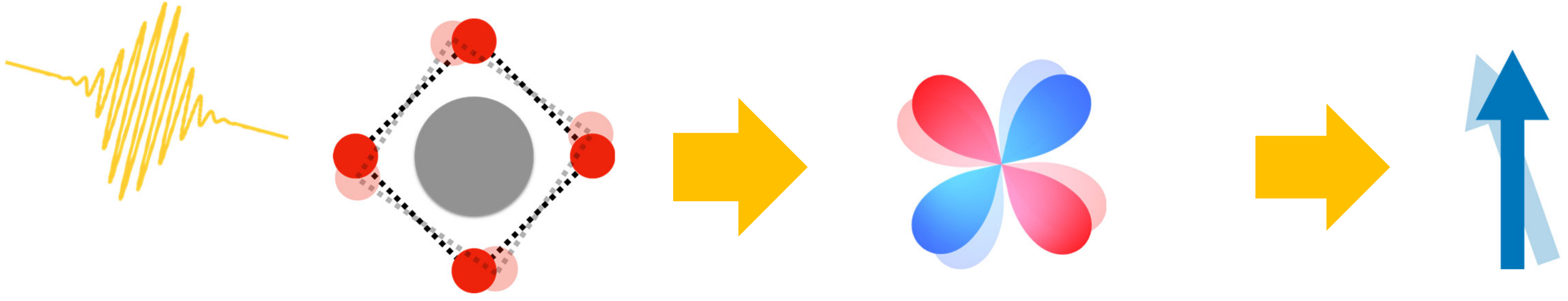
Possible model

- Consider strong correlations between lattice, orbitals, and spins

$$H = H_{CF} + \mathbb{J}_{ij}\left(S_i \cdot S_j + \frac{1}{4}\right) + \frac{1}{2}\mathbb{K}_{ij}$$

(Kugel-Khomskii type Hamiltonian)

Crystal field — Spin superexchange — Orbital superexchange

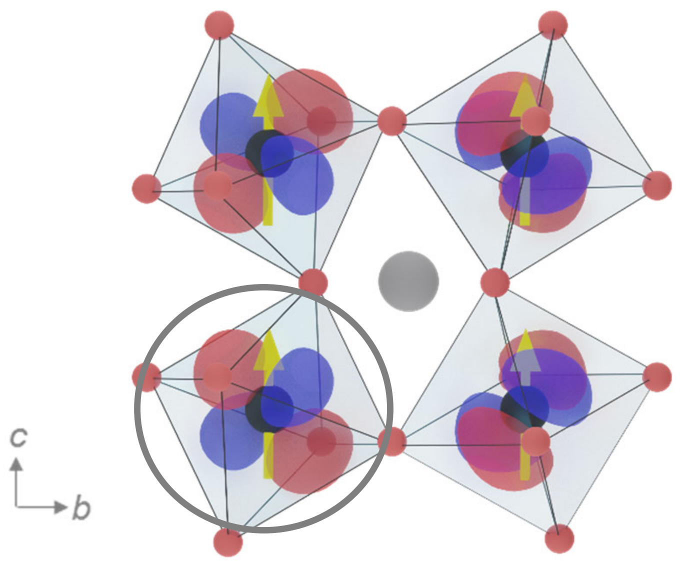


Lattice excitation — Orbital modulation — Change in magnetic order

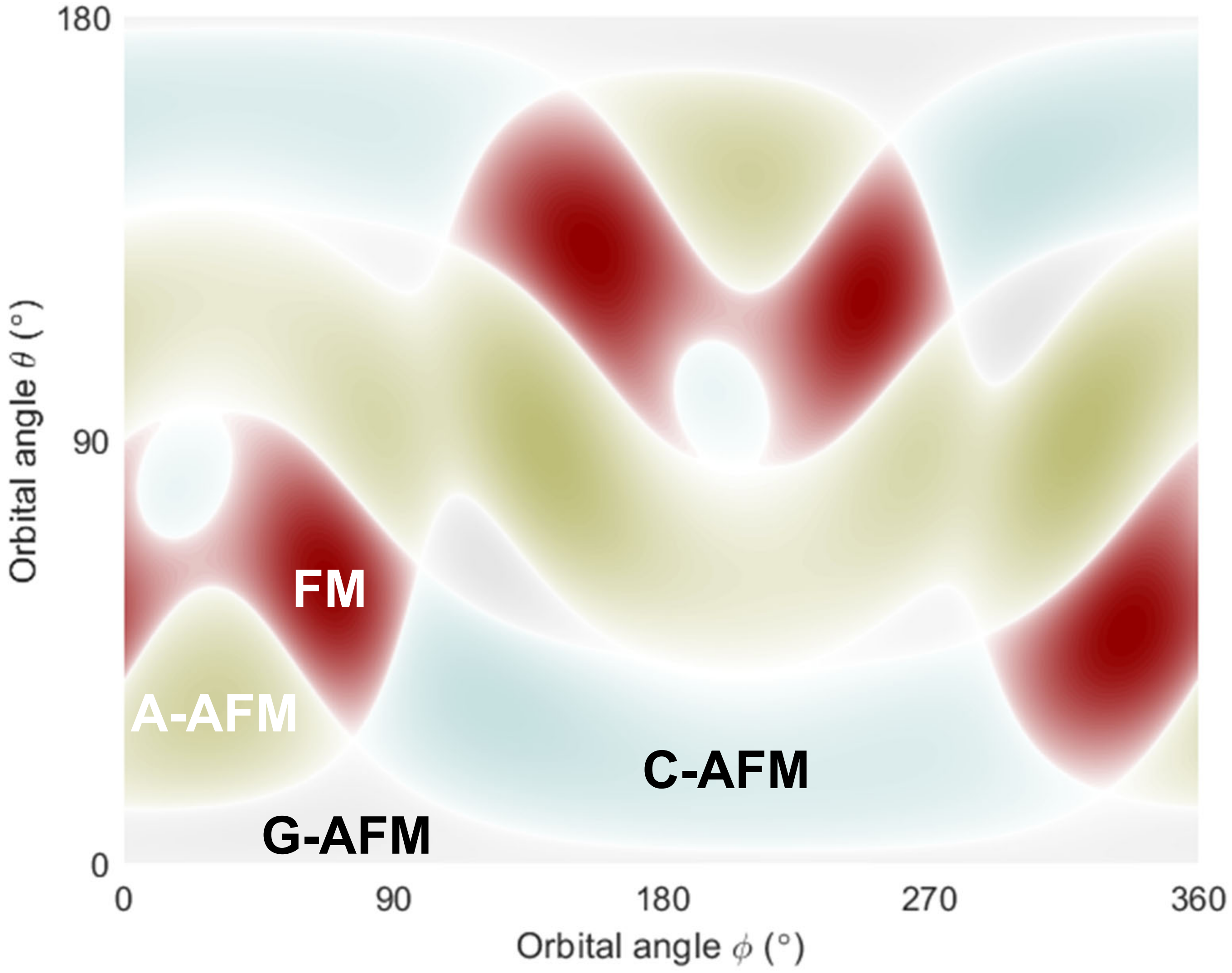
Model spin-orbital-lattice calculations

Orbital ground state

$$\psi_{GS} = \sin\theta\cos\phi\,|yz\rangle + \sin\theta\sin\phi\,|xz\rangle + \cos\theta\,|xy\rangle$$



Magnetic phase diagram



Model spin-orbital-lattice calculations

Orbital ground state

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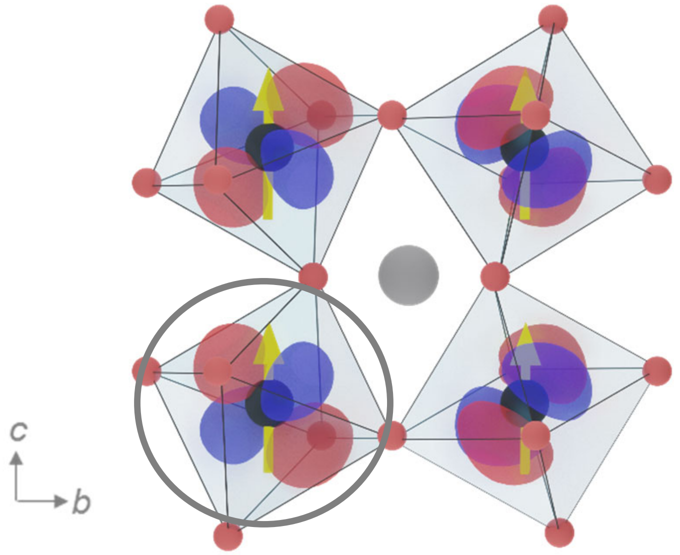
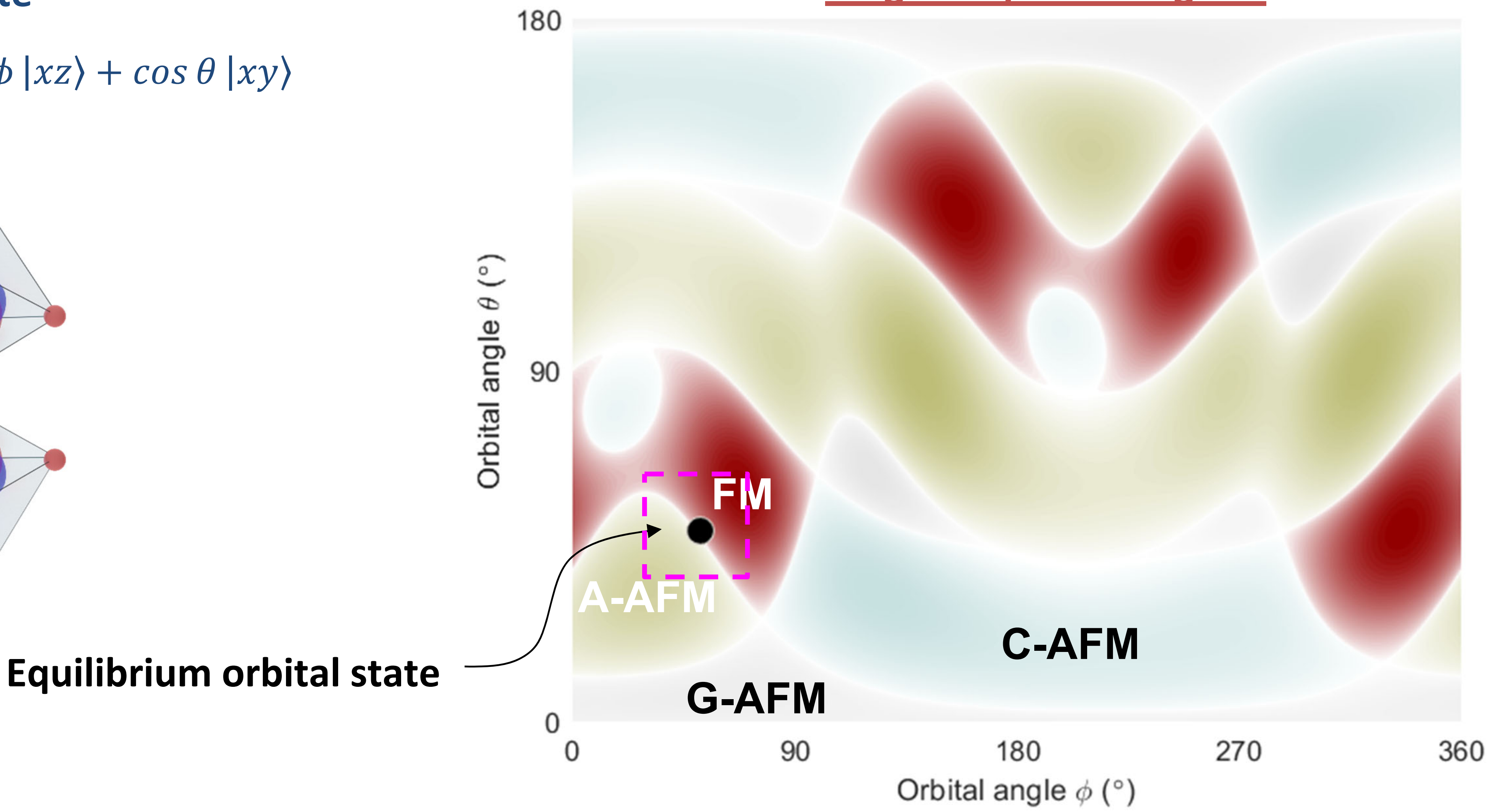


Magnetic phase diagram



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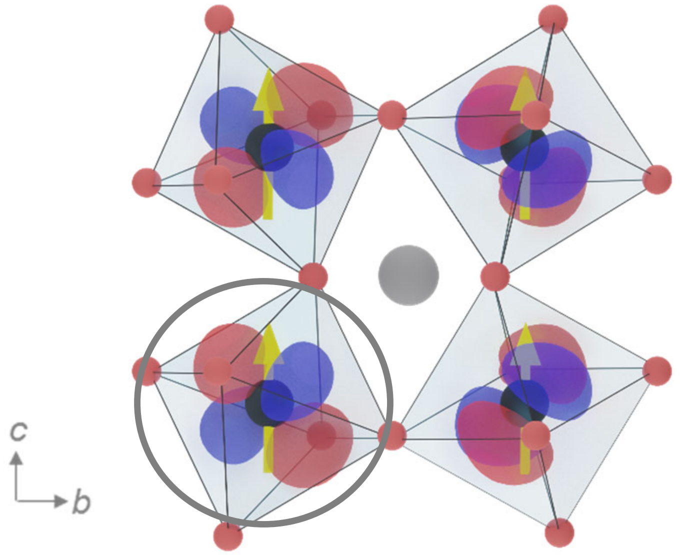
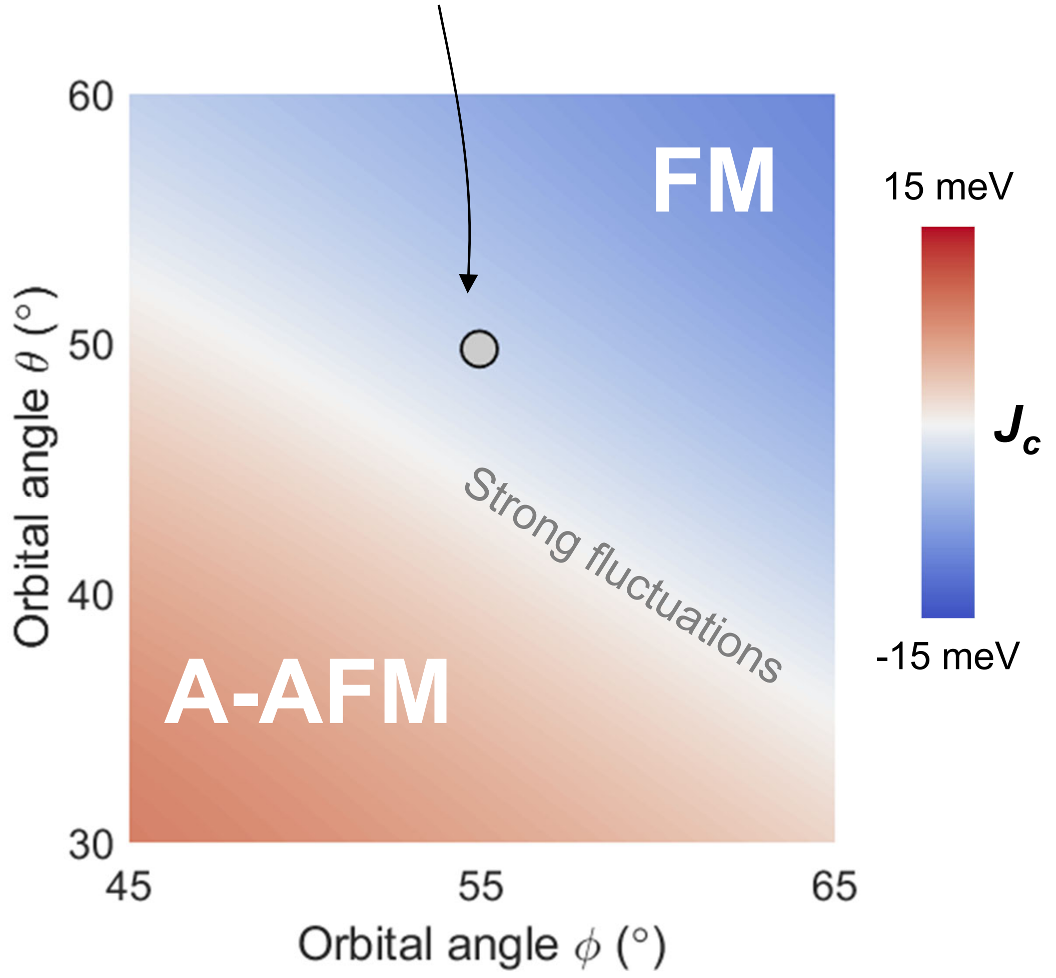
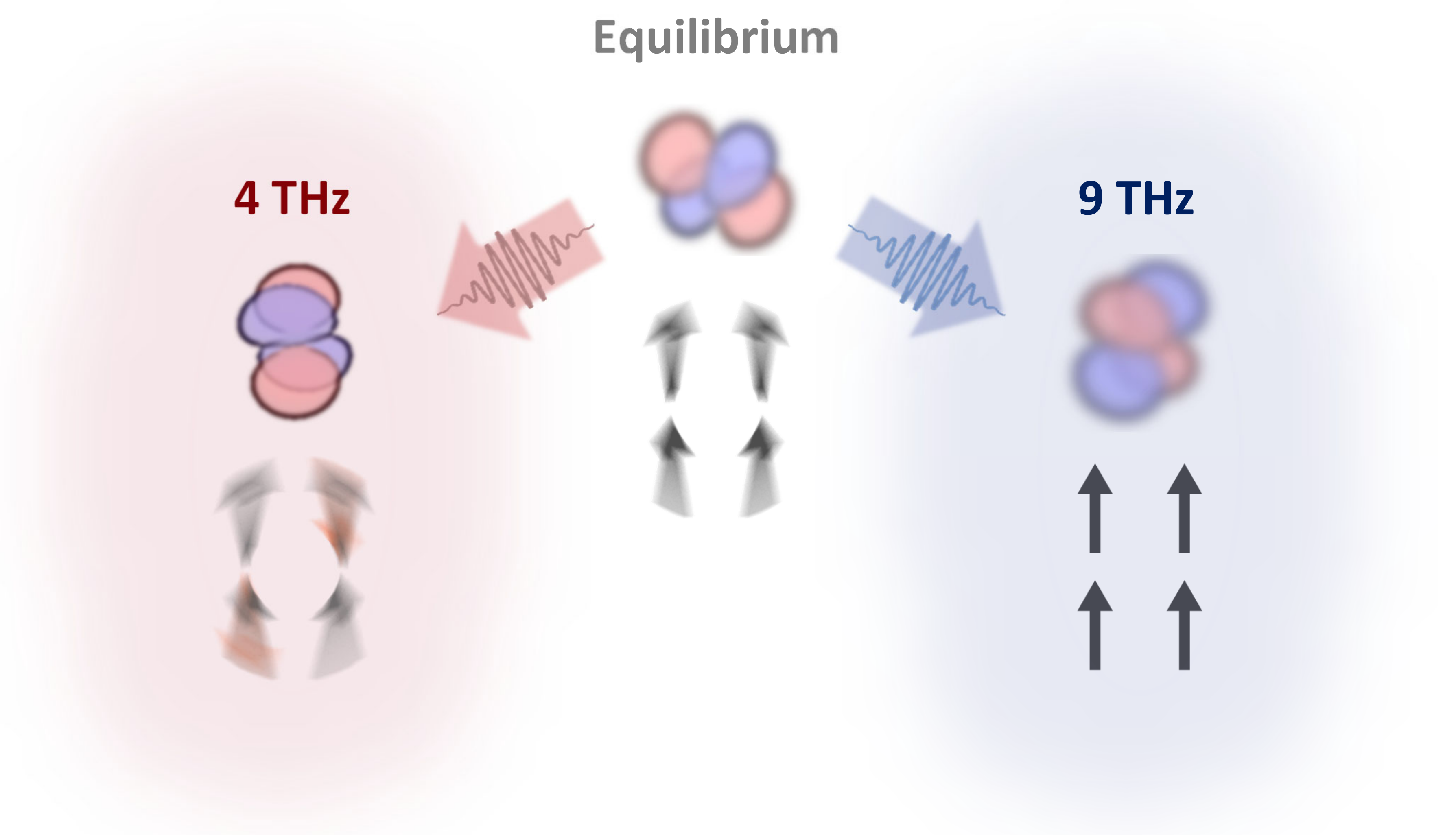


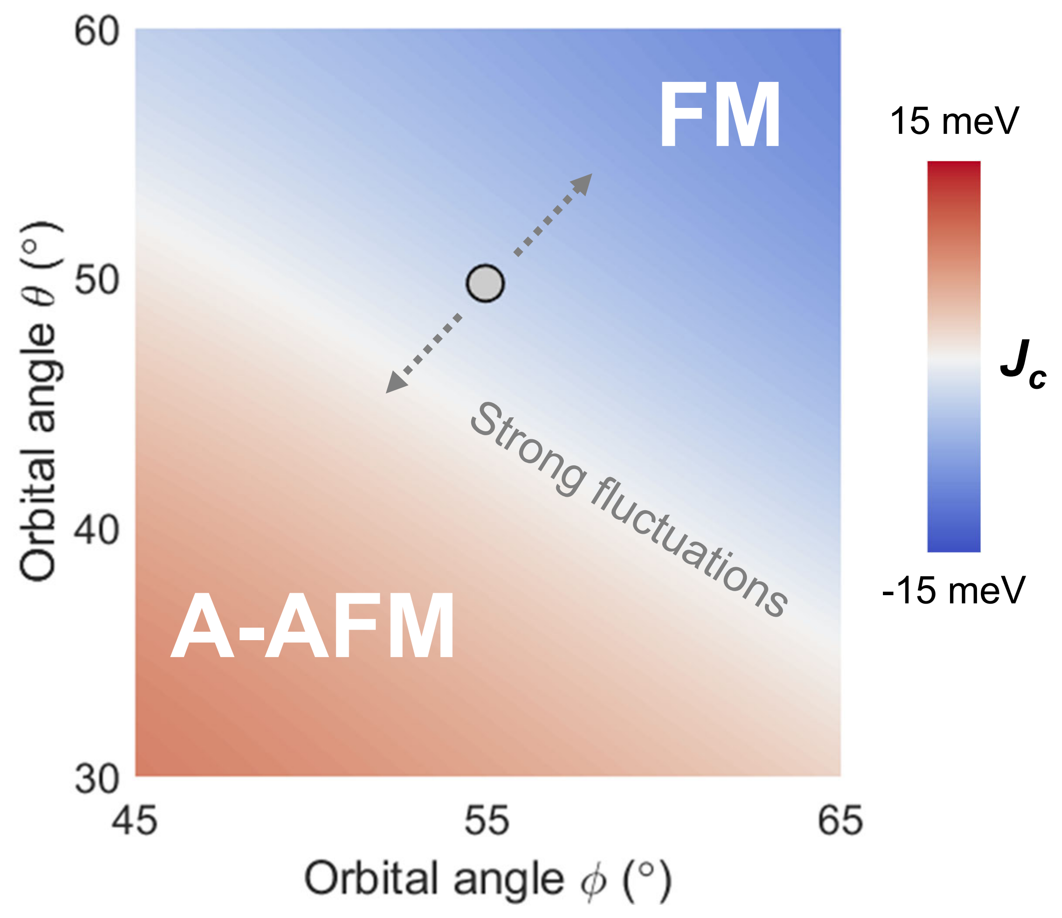
Equilibrium orbital state



Picture of non-equilibrium magnetism in $YTiO_3$

Phonon driving enhances or weakens ferromagnetism through orbital state





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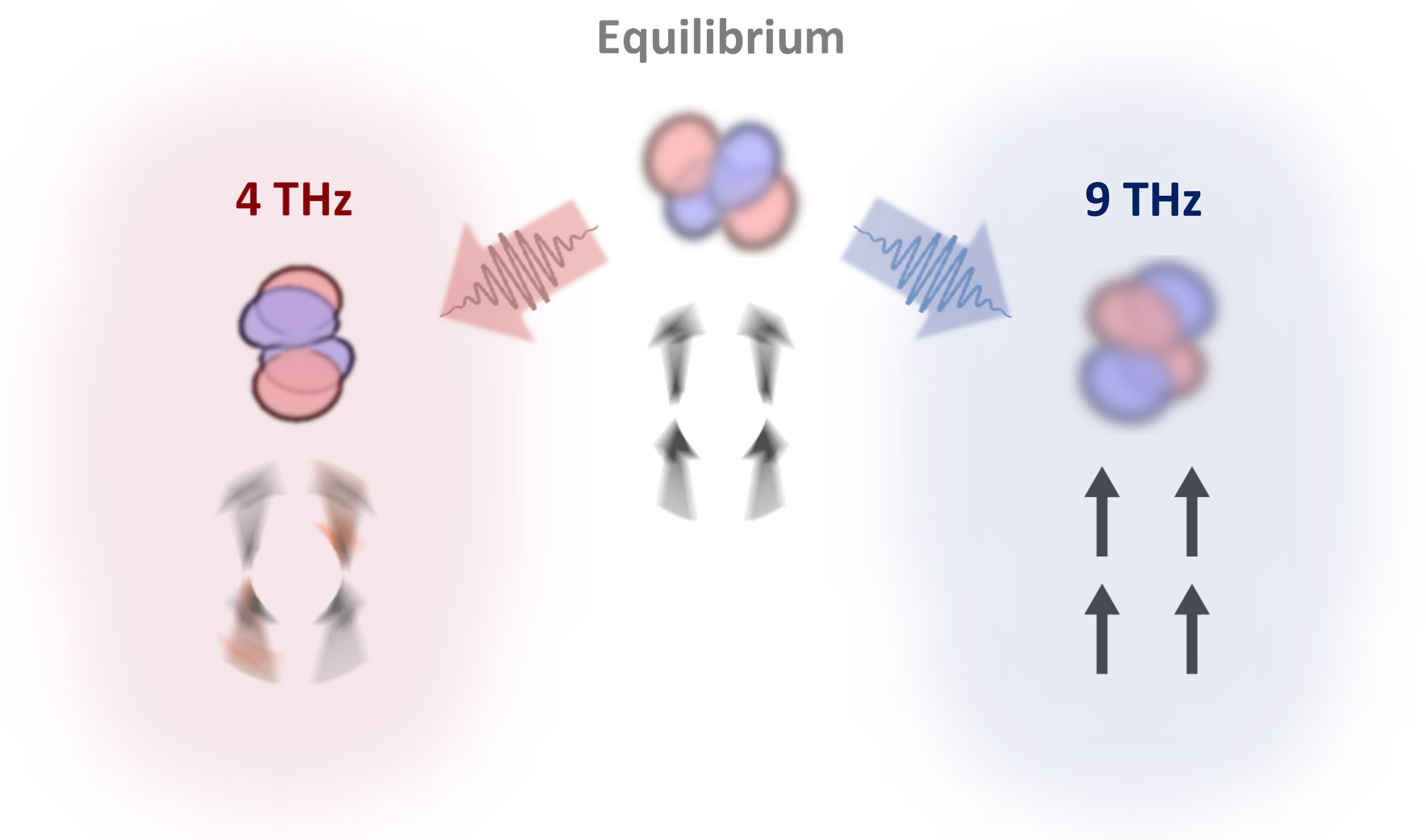
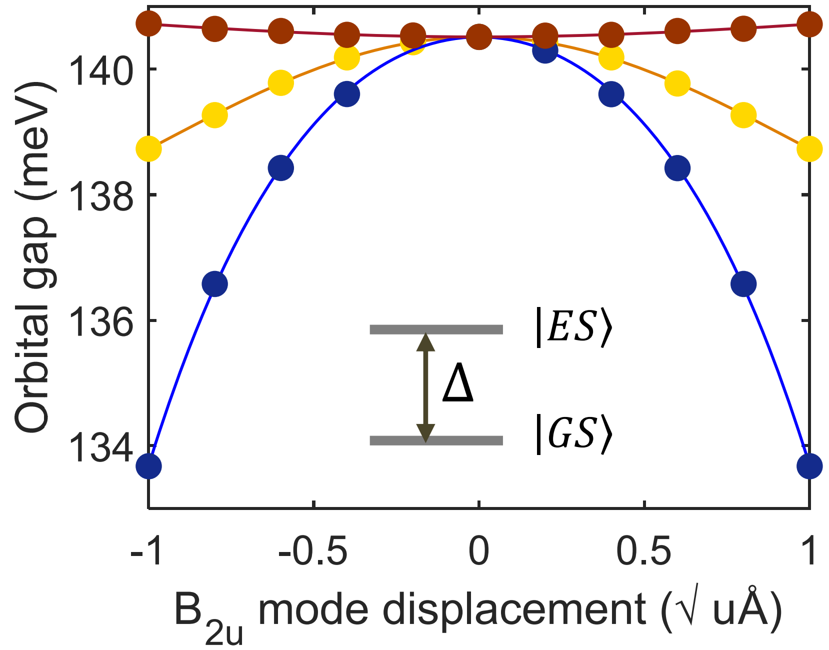


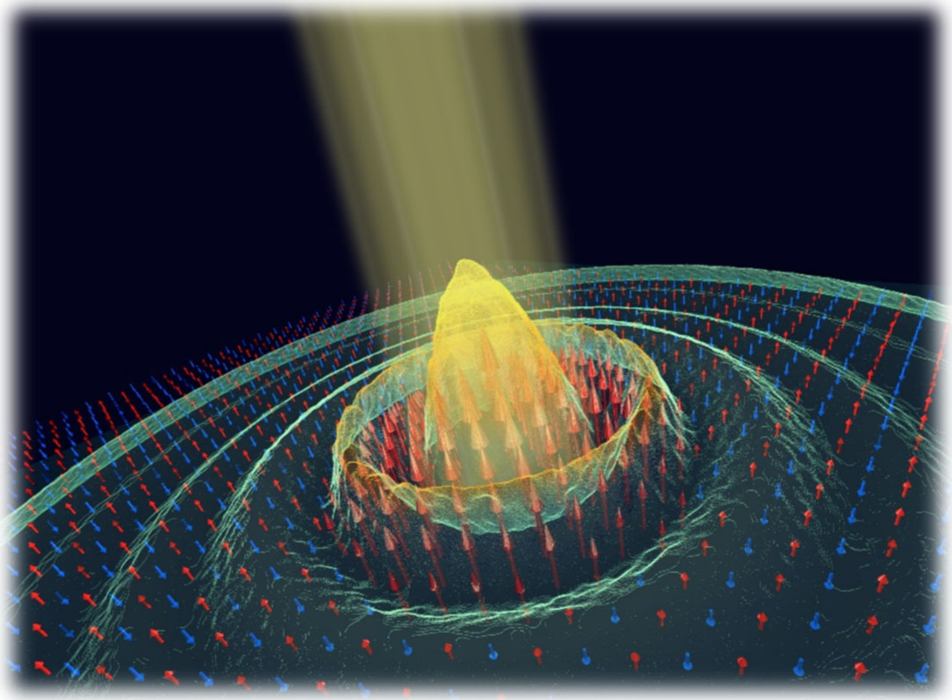
- Calculated change in orbital polarization in agreement with experiment



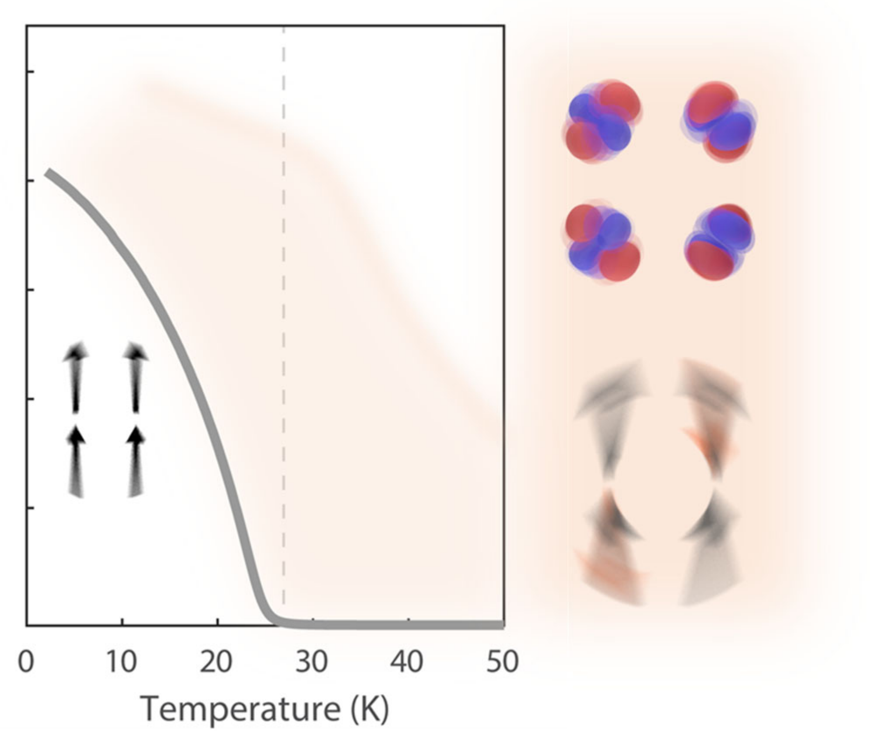
Take away message

Driving the crystal lattice with light provides a powerful means to engineer magnetic phases and induce enhanced non-equilibrium behavior

Manipulating order in an antiferromagnet



Stabilizing high-temperature ferromagnetism



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

A.S. Disa, *et al.*, arXiv:2111.13622

Advertisement!

- Moving to Cornell Applied & Engineering Physics in July 2022

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

asd47@cornell.edu
ankit.disa@mpsd.mpg.de