

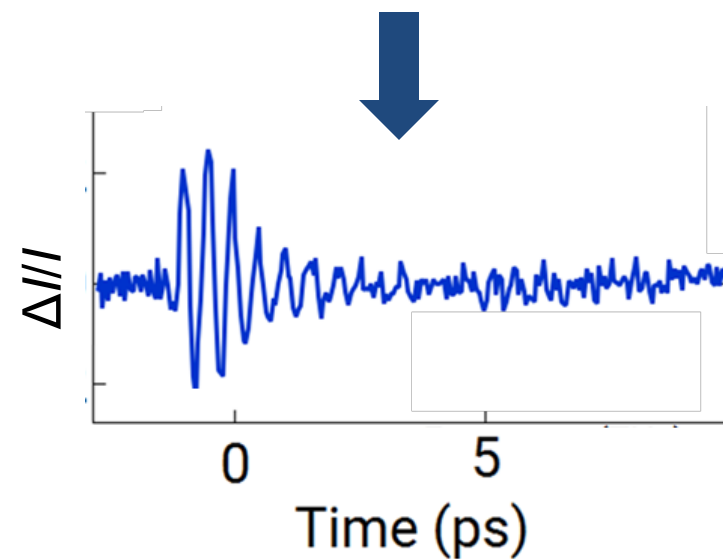
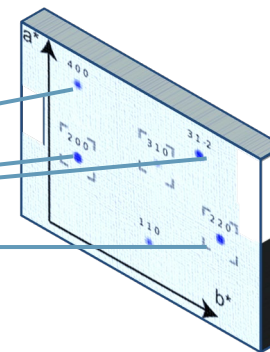
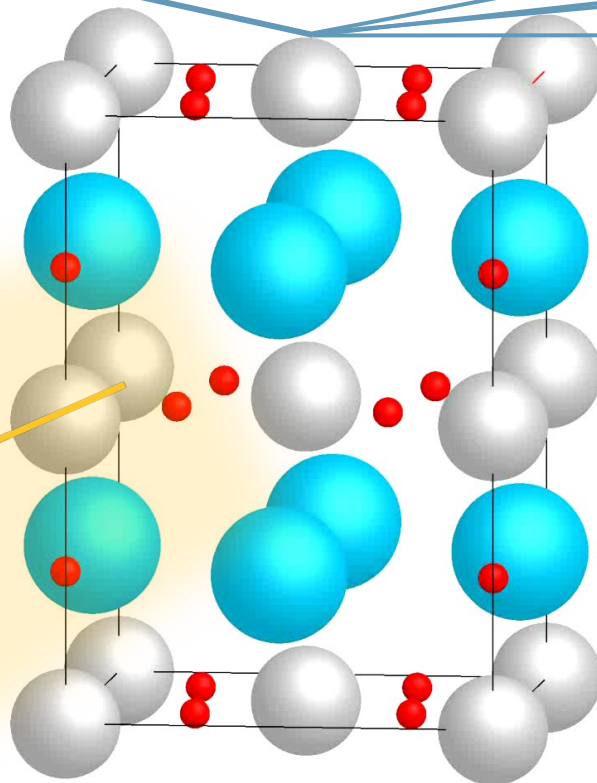
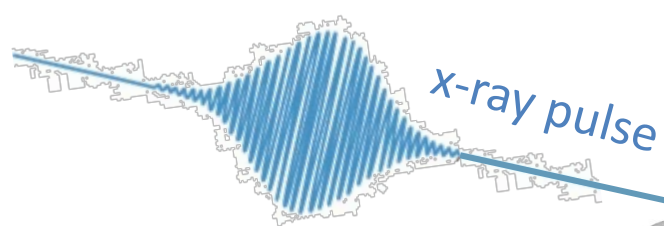
# Ultrafast X-ray Modulation with Quantum Materials

**Ankit Disa**

School of Applied & Engineering Physics, Cornell University

AFOSR + NSF Workshop, “Scientific Opportunities with Compact, High Intensity Accelerators”,  
February 24, 2026

# TLDR: Ultrafast control of materials $\rightarrow$ X-rays

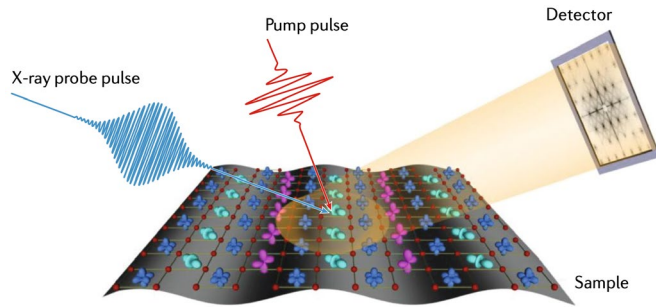


Optical control of quantum materials



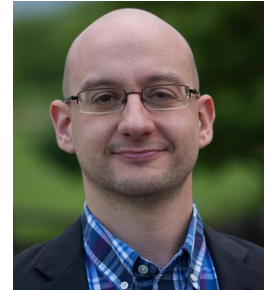
High-frequency x-ray modulation

# Potential application: x-ray communications



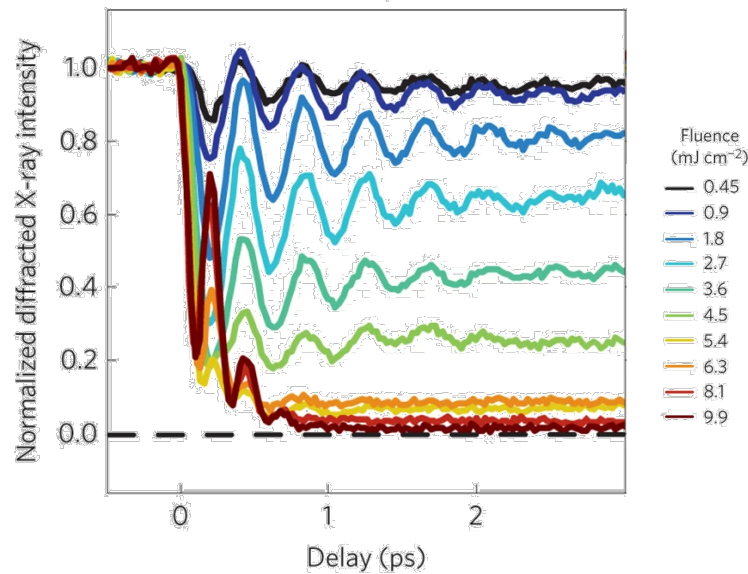
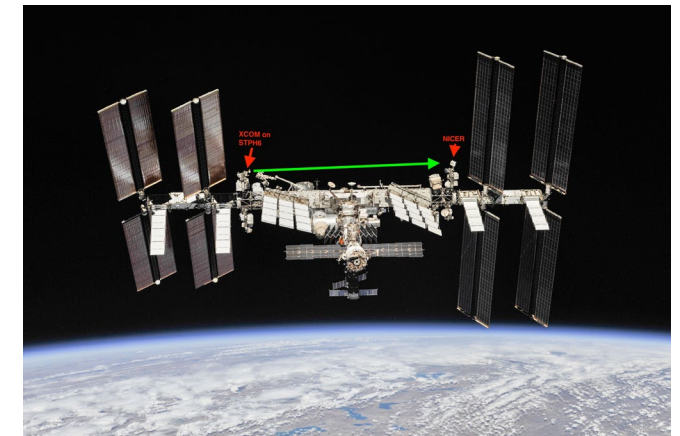
## X-ray modulation with quantum materials:

- Large amplitudes (up to 100%)
- High frequencies (THz)
- Wide tunability, highly efficient



## Potential use for XCOM:

- Long-distance, penetrative
- Secure, directional
- High bandwidth (potential)



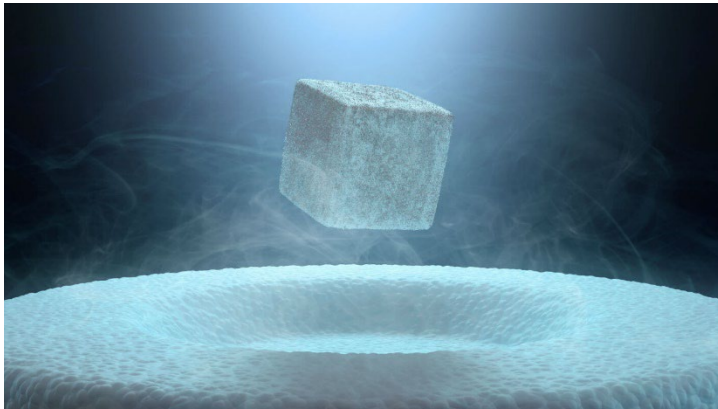
Could be a complementary approach to direct modulation of x-ray source



# The promise of quantum materials

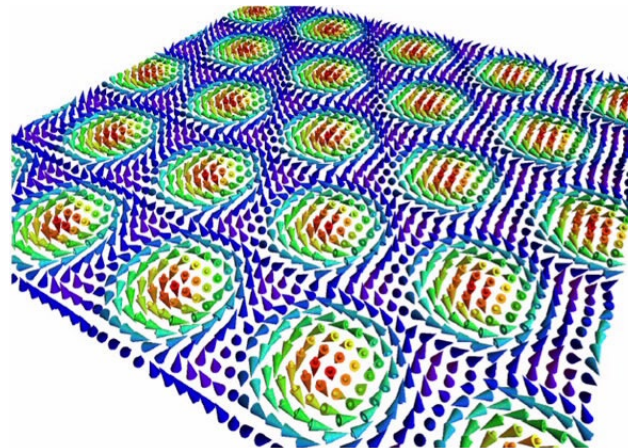
*Extraordinary electronic and magnetic phenomena*

## Superconductivity



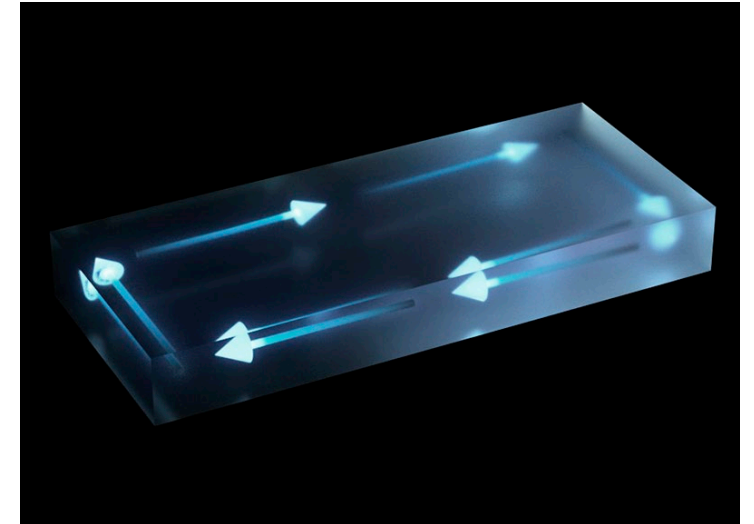
*Lossless energy transmission*

## Quantum Magnetism



*Energy-efficient information processing*

## Topology

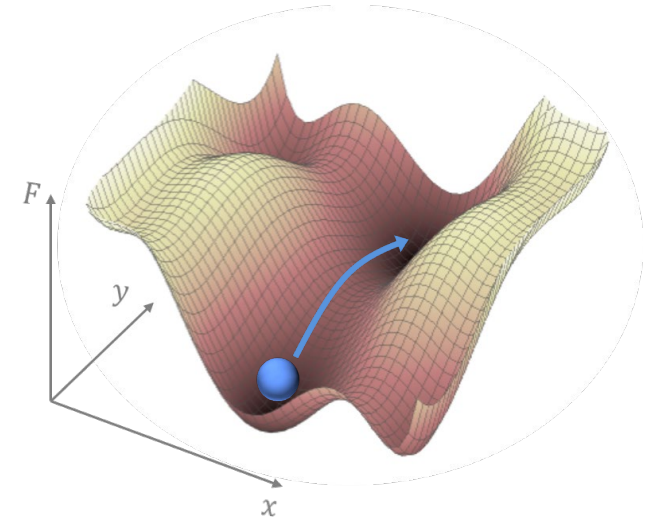
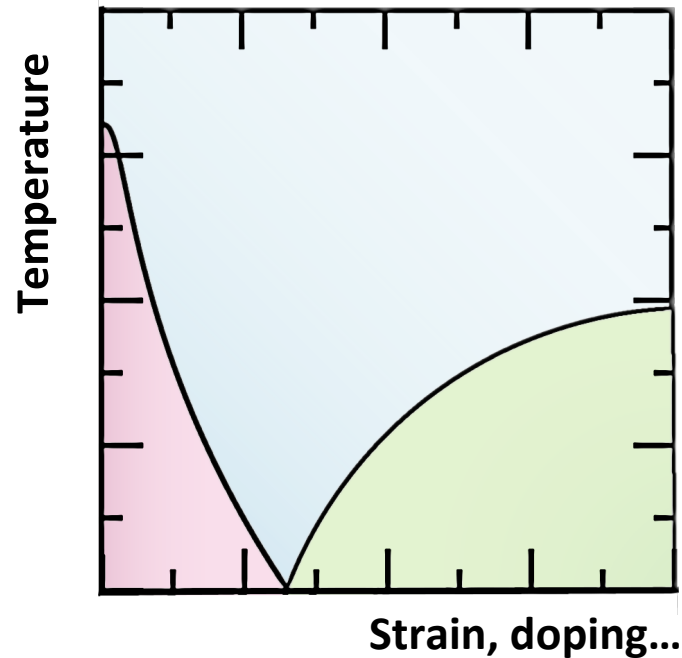


*Quantum computing*

**How can we harness their properties?**

# Controlling functionality in quantum materials

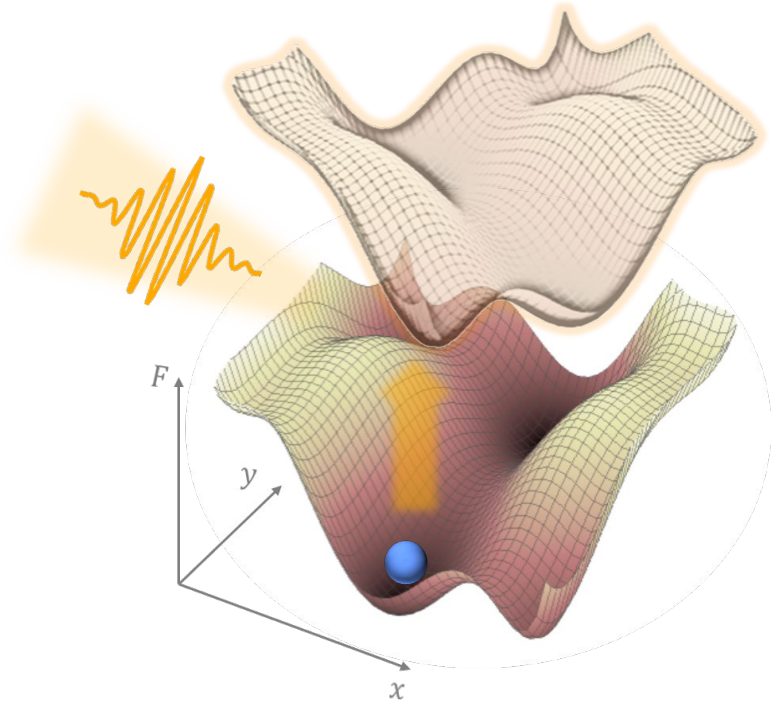
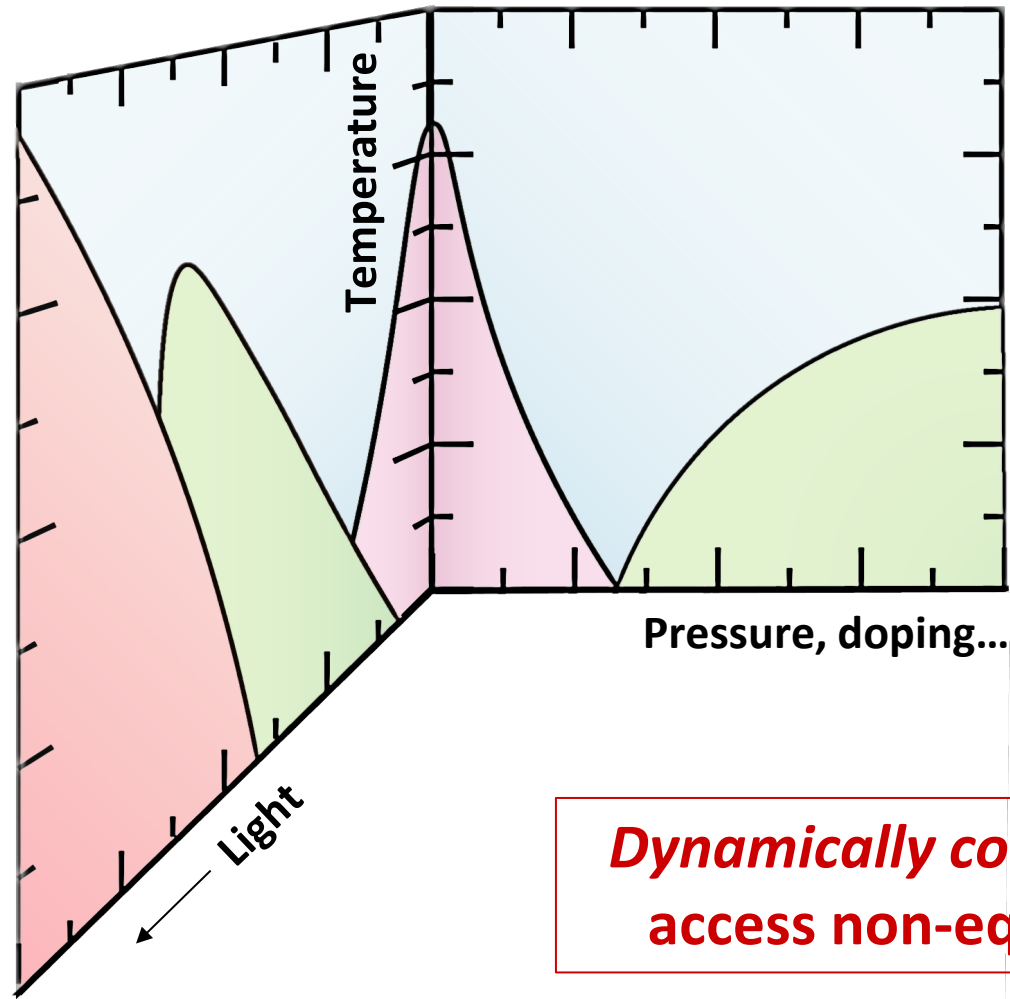
Generic phase diagram of a quantum material



Can we change phase boundaries and ordering temperatures?

Can we induce new phases and functionalities?

# Controlling functionality in quantum materials

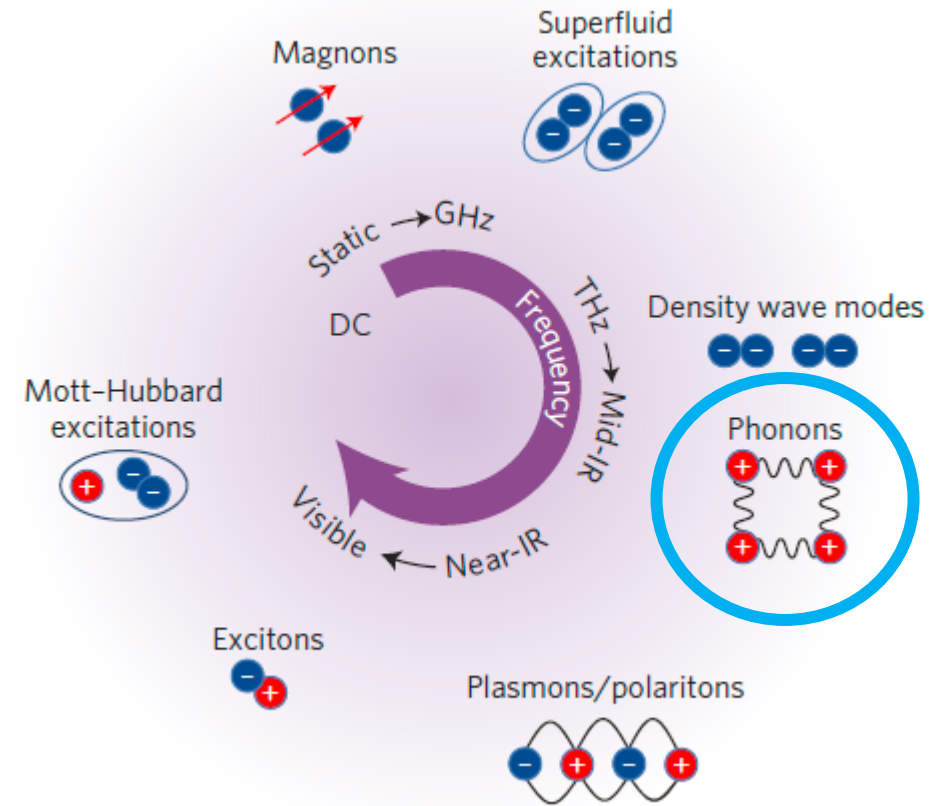


***Dynamically control materials properties to access non-equilibrium phases with light***

# Harnessing light-matter interactions

## Mode-selective control:

Specifically excite targeted degrees of freedom by driving collective modes in solids at their natural energy scales



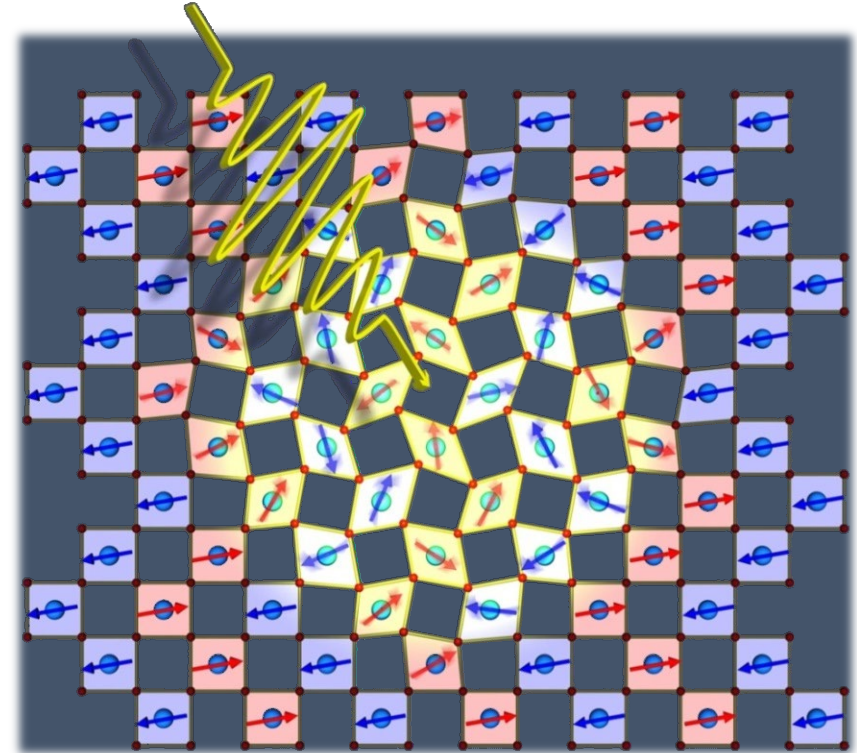
# Engineering structure with light

**Drive large amplitude structural distortions with laser pulses**

→ Resonantly excite optical phonons  
( $\sim 2-200$  meV,  $\sim 0.5-50$  THz)

$\sim$  MV/cm electric fields  $\rightarrow$  5-10% atomic displacements

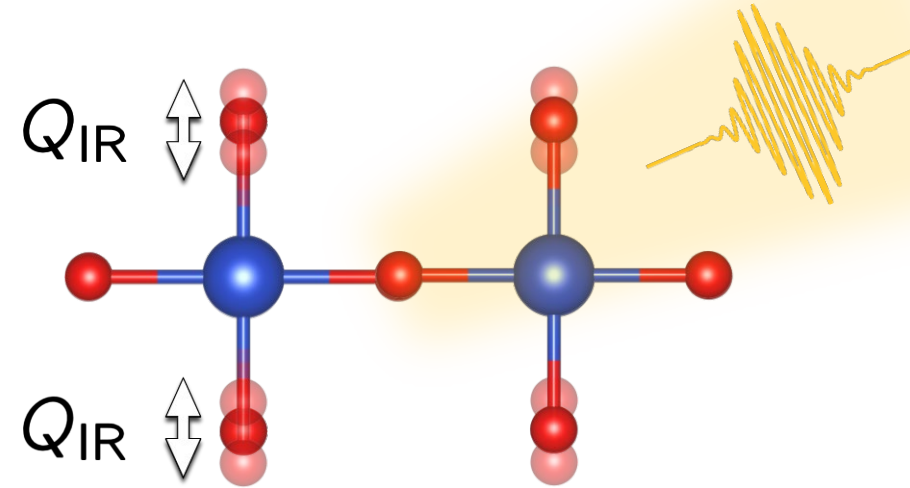
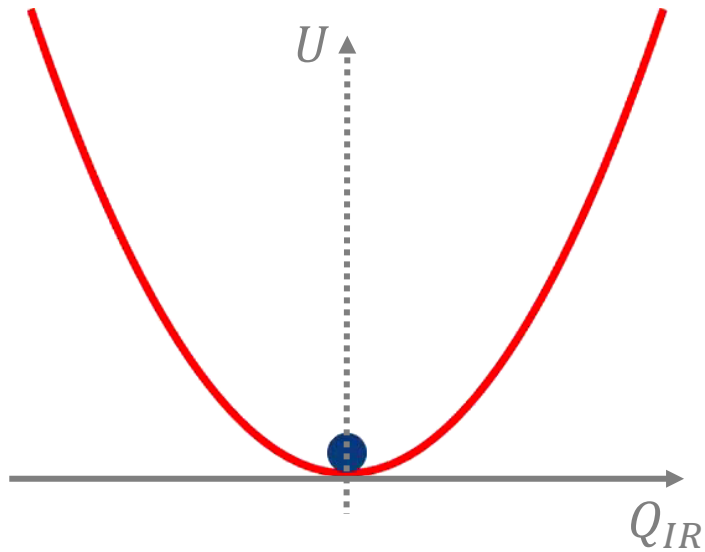
**Leads to highly nonlinear response of crystal lattice to access new functionalities**



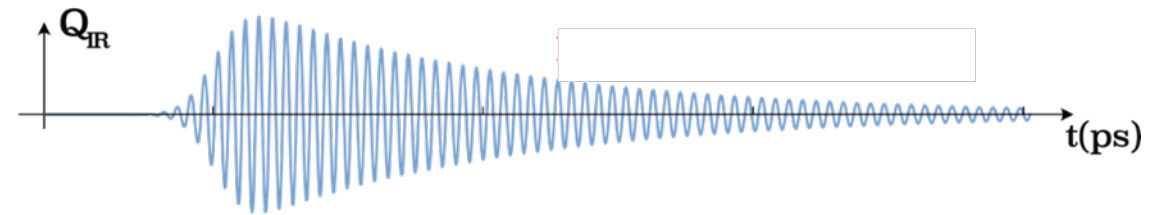
# "Linear phononics"

- Light drives infrared-active modes:

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



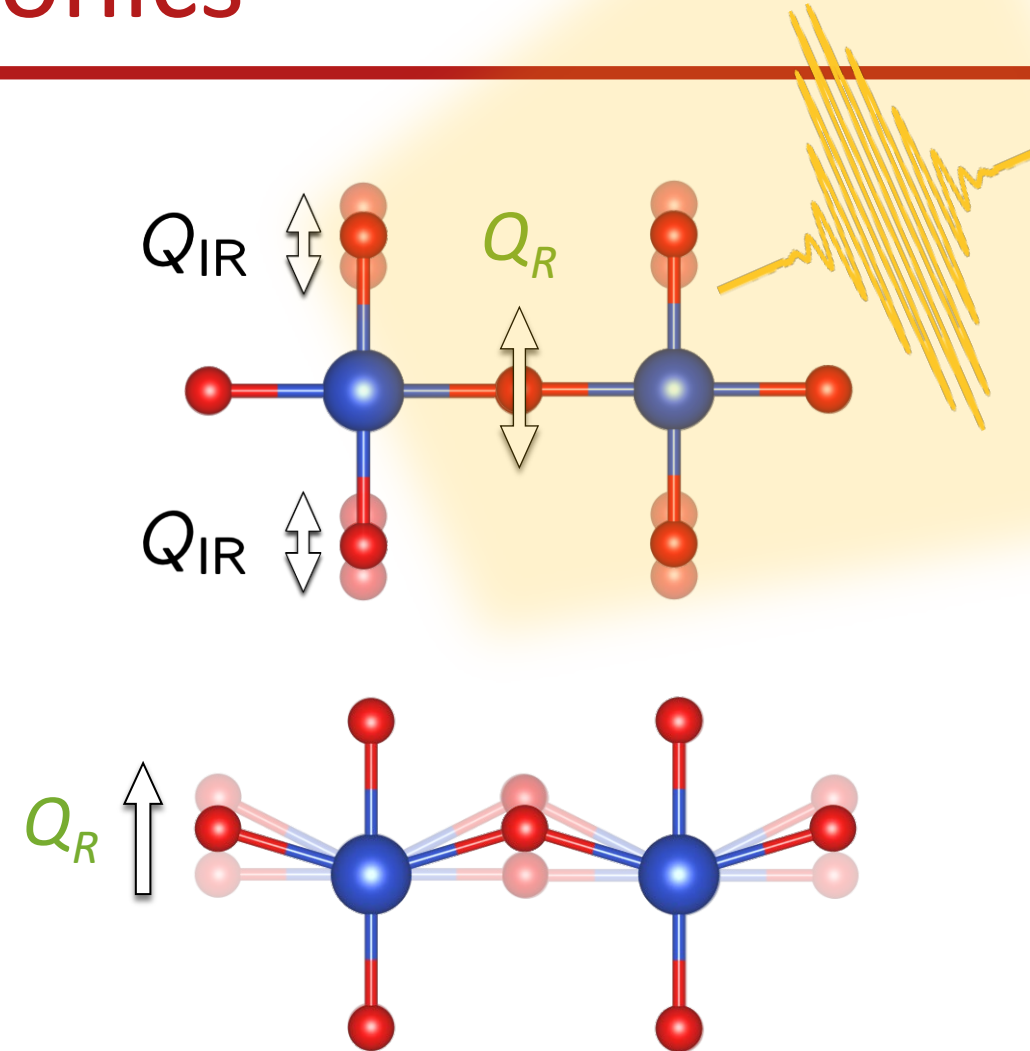
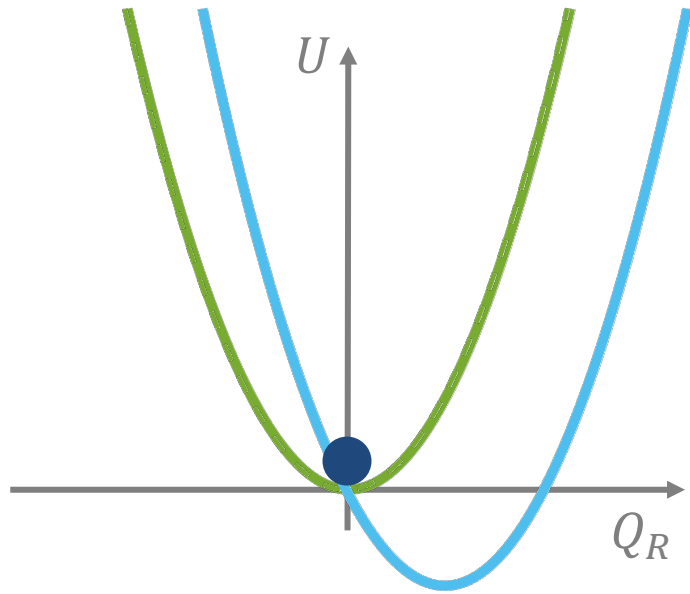
No average change to the lattice



# “Nonlinear phononics”

- Large IR motions can couple to other modes:

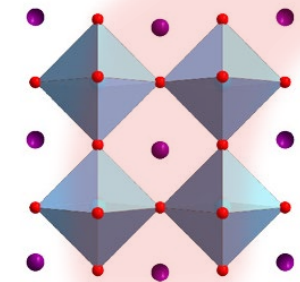
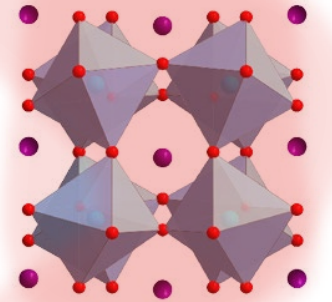
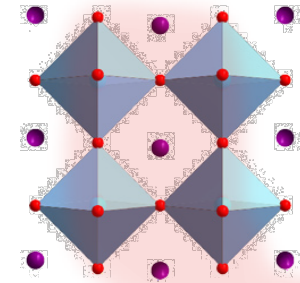
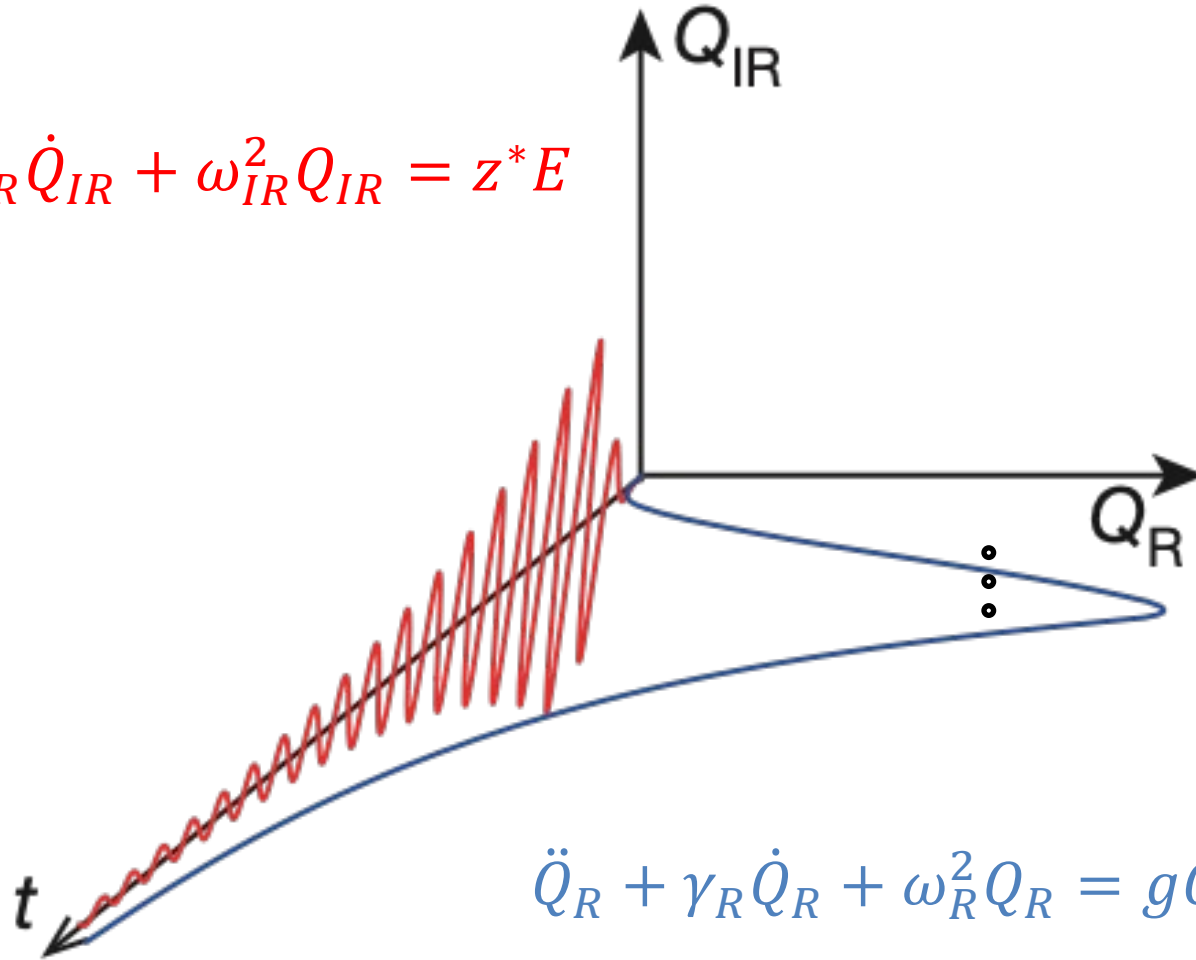
$$U_{lattice} = \frac{1}{2} \omega_{IR}^2 Q_{IR}^2 + \frac{1}{2} \omega_R^2 Q_R^2 - g Q_{IR}^2 Q_R + \dots$$



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 does this affect x-rays?

**X-ray scattering structure factor for crystals:**

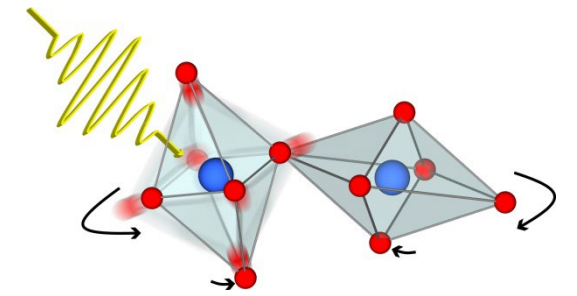
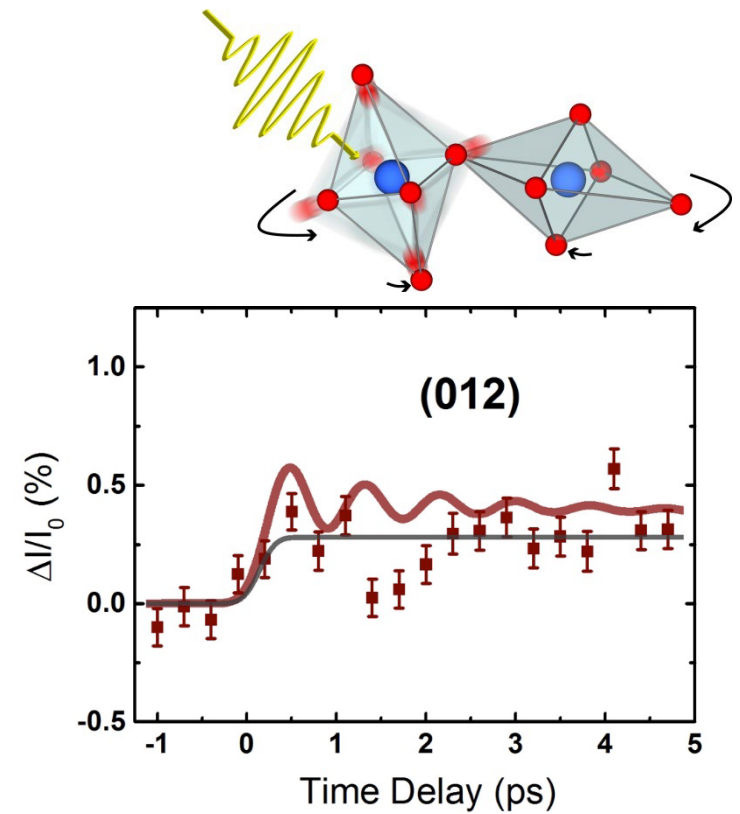
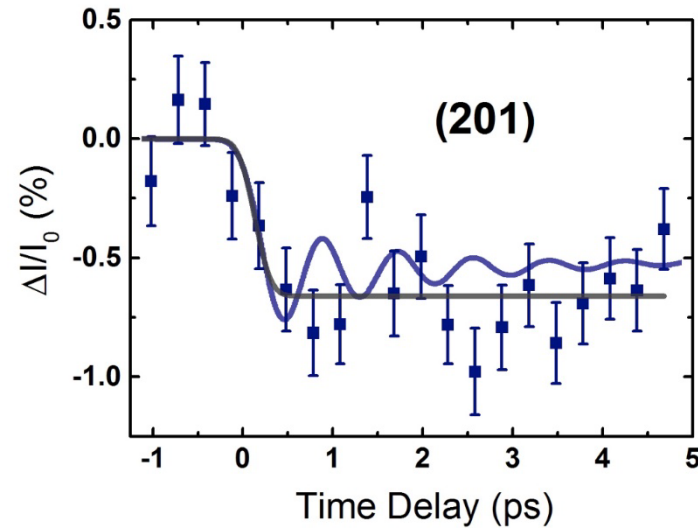
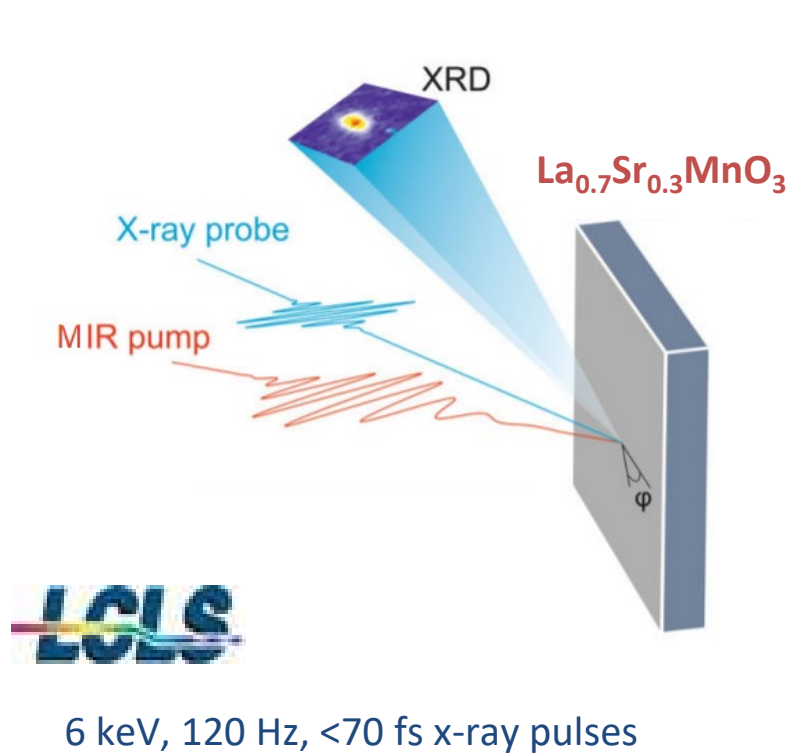
$$F(\vec{q}) = \sum_j f_j(\vec{q}) e^{i\vec{q} \cdot \vec{r}_j(t)}$$

Momentum transfer =  $\frac{4\pi \sin \theta}{\lambda}$

atomic positions  
(time-dependent)

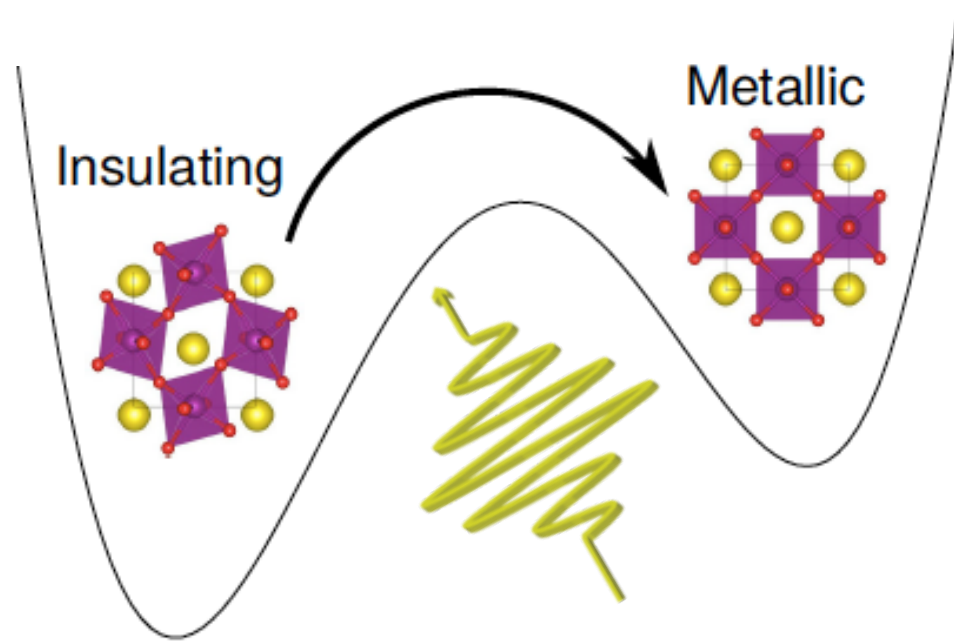


# Time-resolved x-ray diffraction

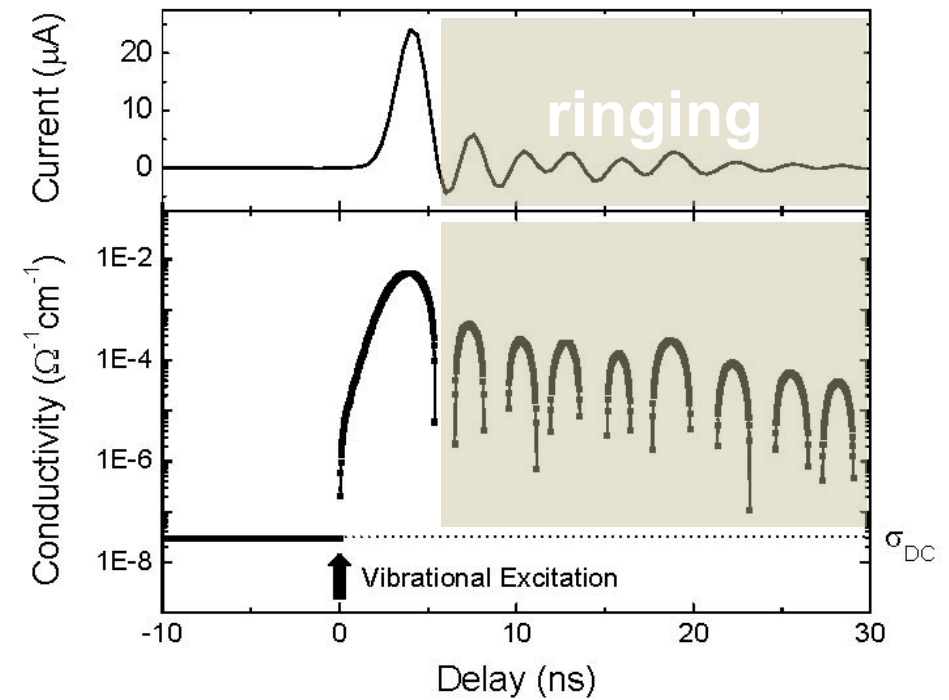


- Displacive excitation of Raman mode confirmed by time-resolved structural measurements

# Phonon-driven metal-insulator transition



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



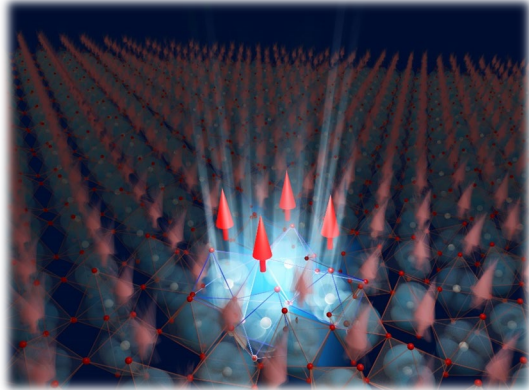
# Suite of nonlinear phononic processes

$$U = \sum_i \frac{1}{2} \omega_i^2 Q_i^2 + \underbrace{V_{NL}}$$

Nonlinearity	Effect
$Q_{\text{driven}}^2 \cdot Q_c$ , $Q_{\text{driven},1} \cdot Q_{\text{driven},2} \cdot Q_c$	Quasi-static lattice distortion <small>(6, 8-11, 19, 21)</small>
$(Q_{\text{driven}} \times Q_{\text{driven}}) \cdot M$ , $Q_{\text{driven}}^2 S_i \cdot S_j$	Change of magnetism <small>(8, 30, 33, 34, 36)</small>
$Q_{\text{driven}}^2 \cdot U$ , $Q_{\text{driven}}^2 \cdot t$	Change of electronic structure <small>(65, 66)</small>
$Q_{\text{driven}}^2 \cdot \varepsilon$	Strain <small>(7, 37)</small>
$Q_{\text{driven}}^4$ , $Q_{\text{driven}}^2 \cdot Q_c^2$ , ...	Phonon amplification, phonon squeezing, ... <small>(23, 25, 26)</small>

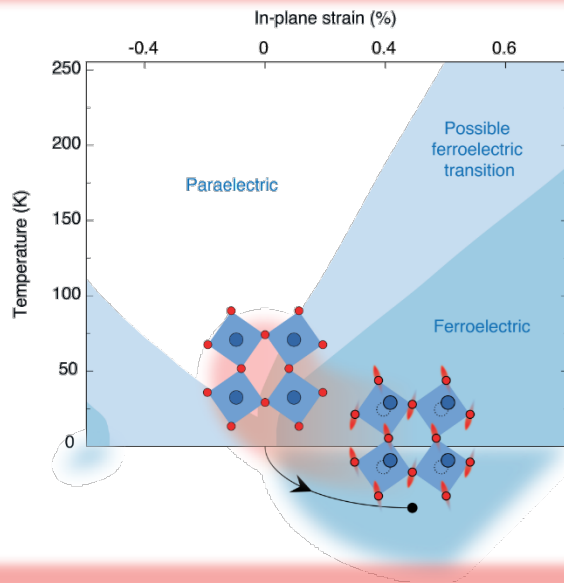


# Many examples of optical control through lattice



## Non-equilibrium magnetism

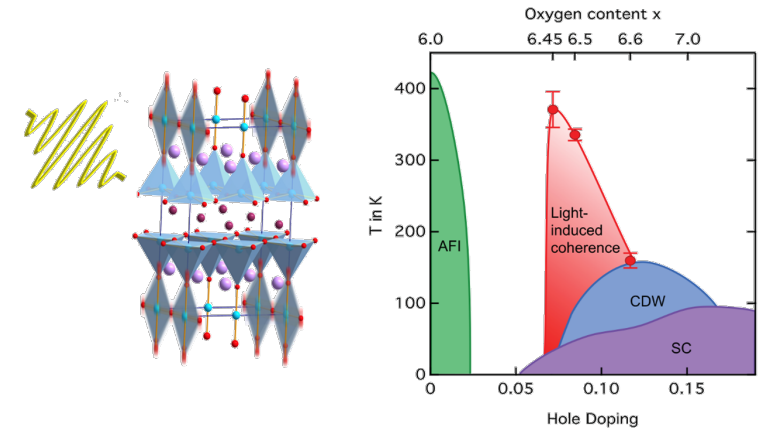
- Disa, *et al.*, *Nature* **617**, 73 (2023)
- Disa *et al.*, *Nature Phys.* **16**, 937 (2020)
- Nova *et al.*, *Nature Phys.* **13**, 132 (2017)
- Maehrlein *et al.*, *Sci. Adv.* **4**, 7 (2018)
- Afanasiev *et al.*, *Nature Mater.* **20**, 607 (2021)



## Inducing and switching ferroelectricity

- Nova, Disa *et al.*, *Science* **364**, 1075 (2019)
- Li *et al.*, *Science* **364**, 1079 (2019)
- Mankowsky *et al.*, *Phys. Rev. Lett.* **449**, 72 (2007)
- Henstridge *et al.*, *Nature Phys.* **18**, 457 (2022)

## Light-induced superconductivity



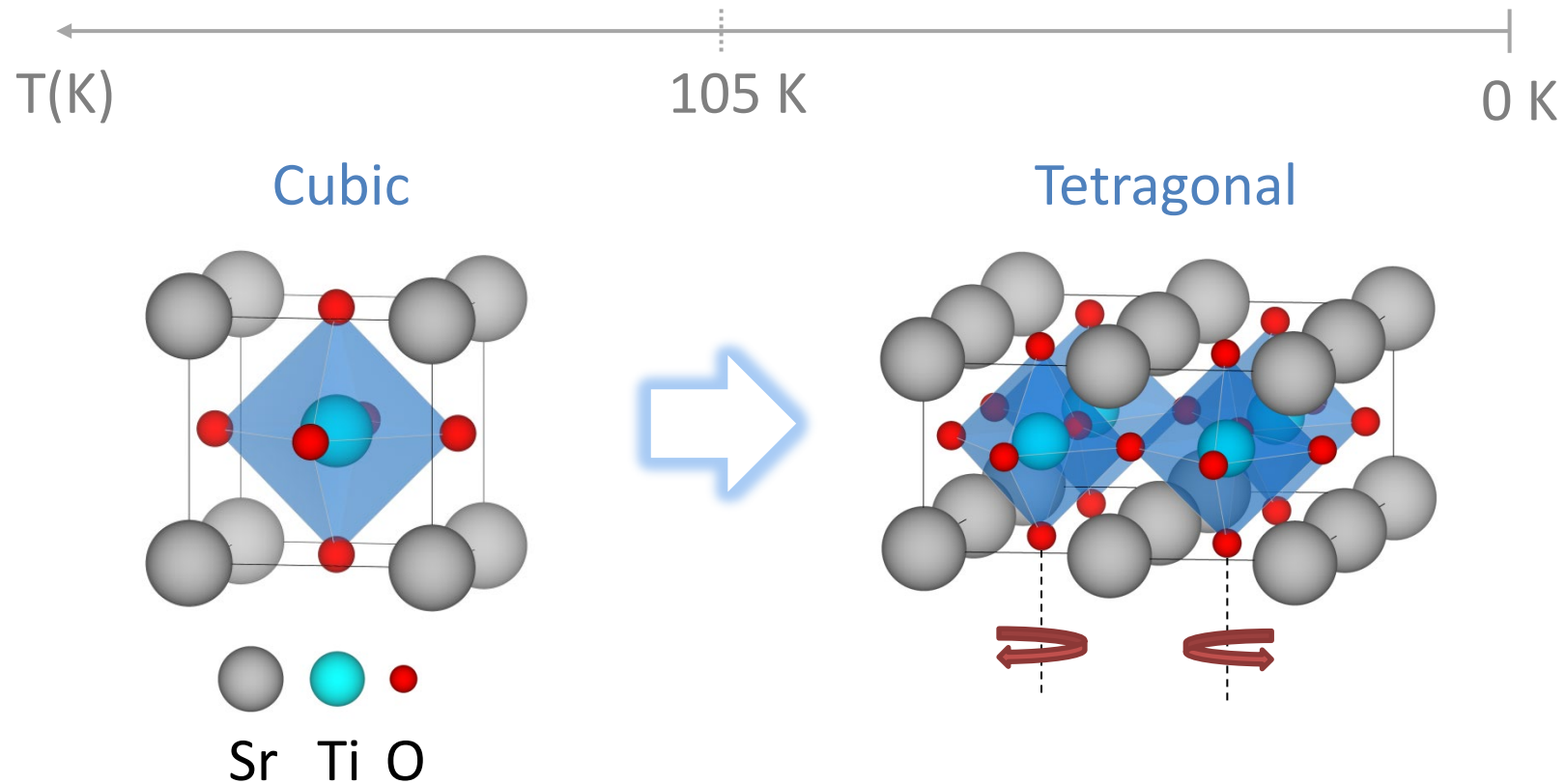
- Mitrano *et al.*, *Nature* **530**, 461 (2016)
- Mankowsky *et al.*, *Nature* **516**, 71 (2014)
- Fausti *et al.*, *Science* **331**, 189 (2011)

## Metal-insulator transitions

- Rini *et al.*, *Nature* **449**, 72 (2007)
- Hu *et al.*, *Phys. Rev. B* **93**, 161107 (2016)
- Cavaglia *et al.*, *Phys. Rev. Lett.* **108**, 136801 (2012)

⋮

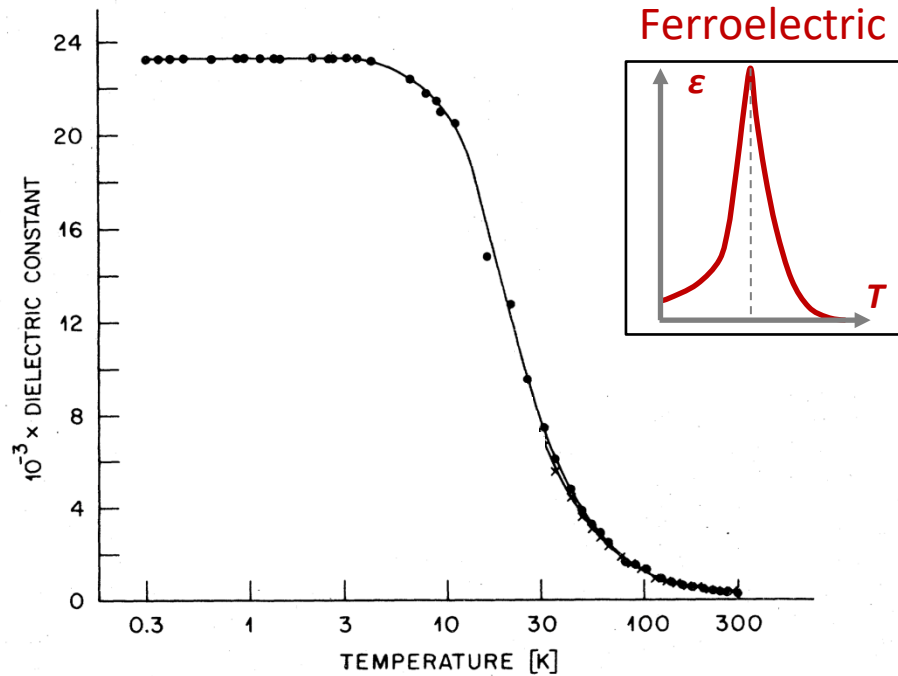
# Our fruit fly: Strontium Titanate ( $\text{SrTiO}_3$ )



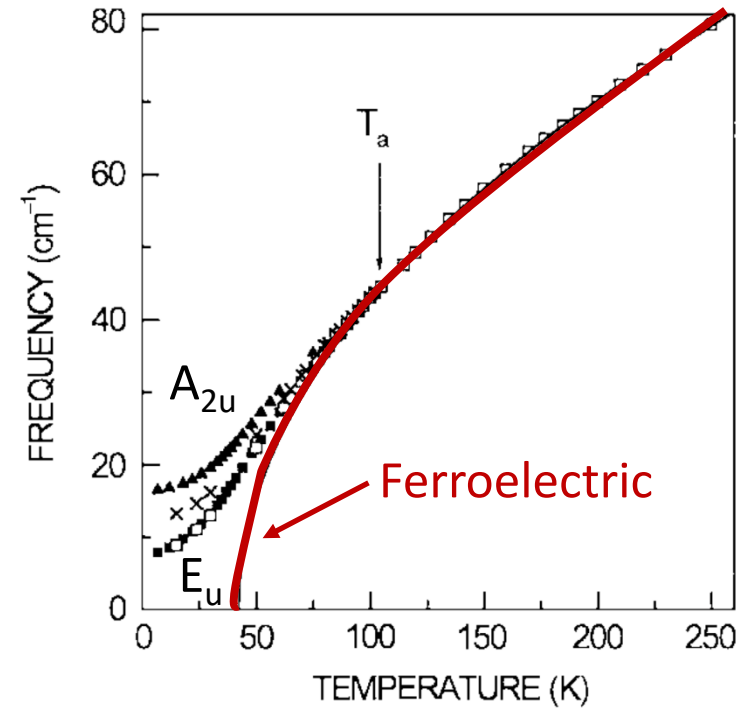
- **Centrosymmetric and paraelectric** at any finite temperature

# Incipient ferroelectricity in SrTiO<sub>3</sub>

$\epsilon$  saturates at low  $T$

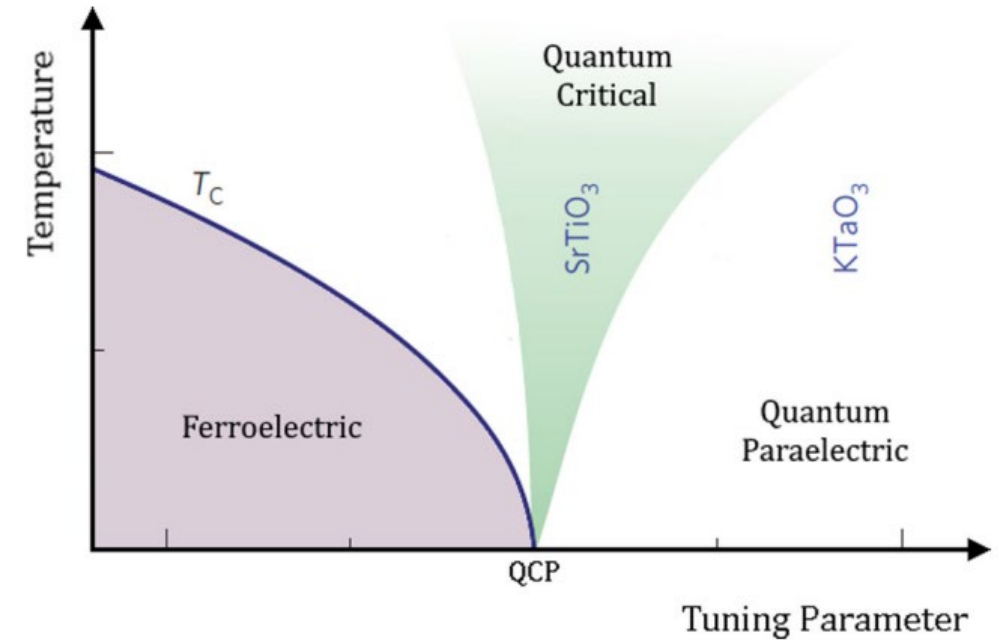
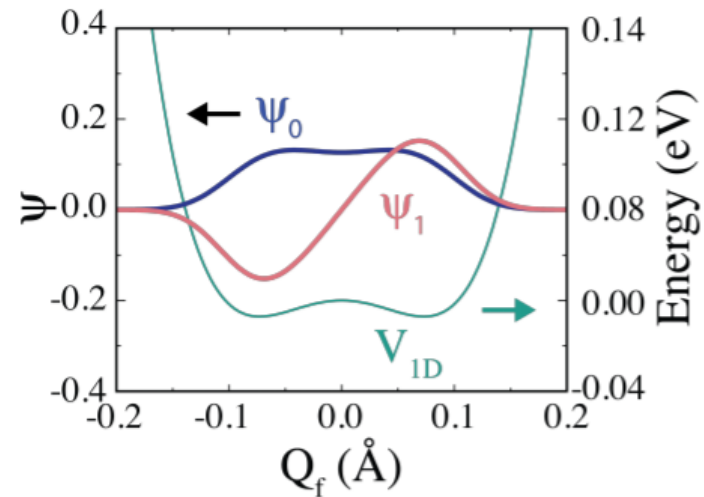
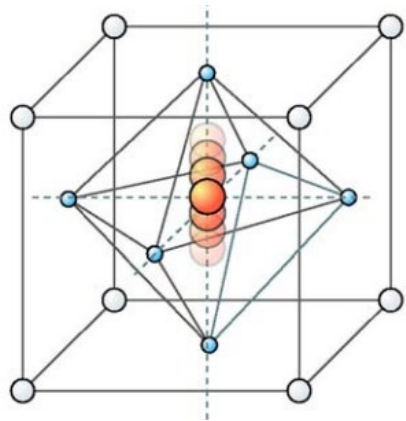


Incomplete softening of polar phonon



- Exhibits signs of proximity to ferroelectric state

# Quantum paraelectricity and fluctuations

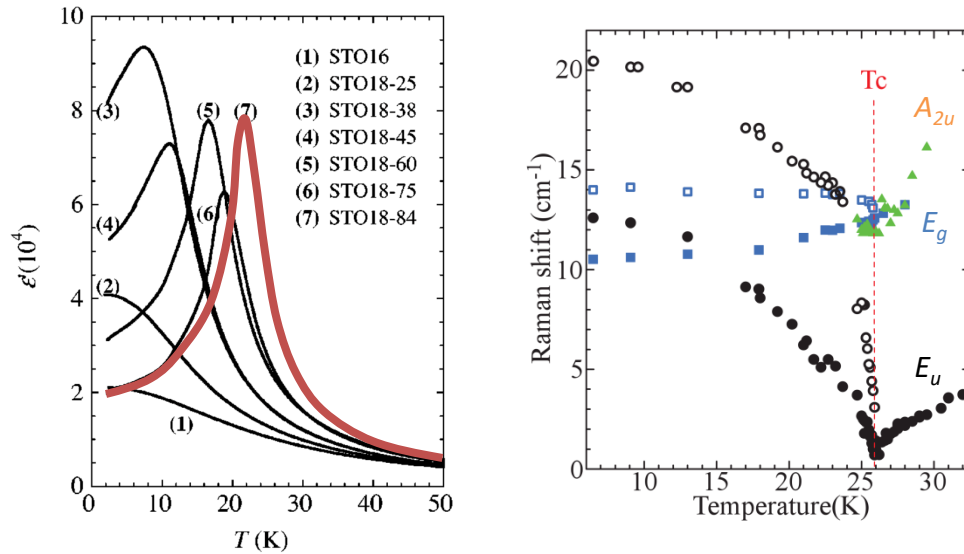


- Zero-point fluctuations of the dipole moment suppress ferroelectricity

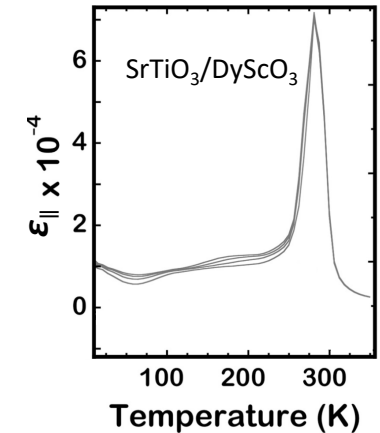
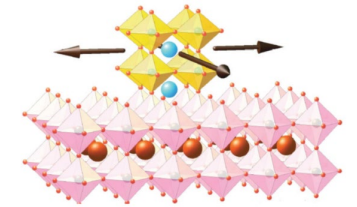
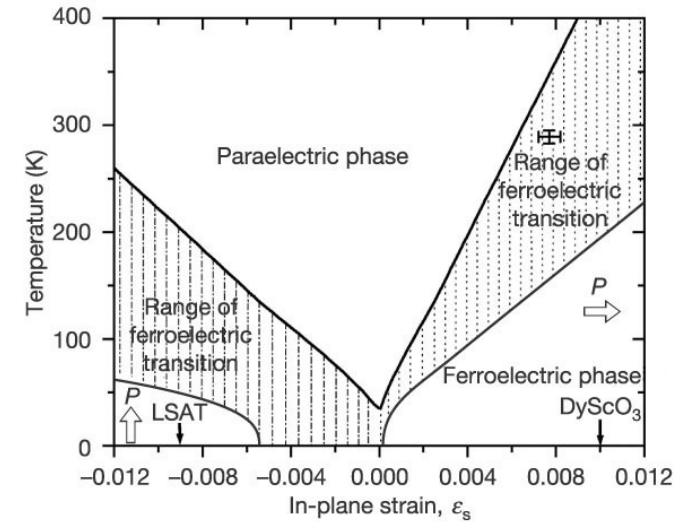


# Susceptibility to perturbations

## Isotope substitution ( $^{18}\text{O}$ )



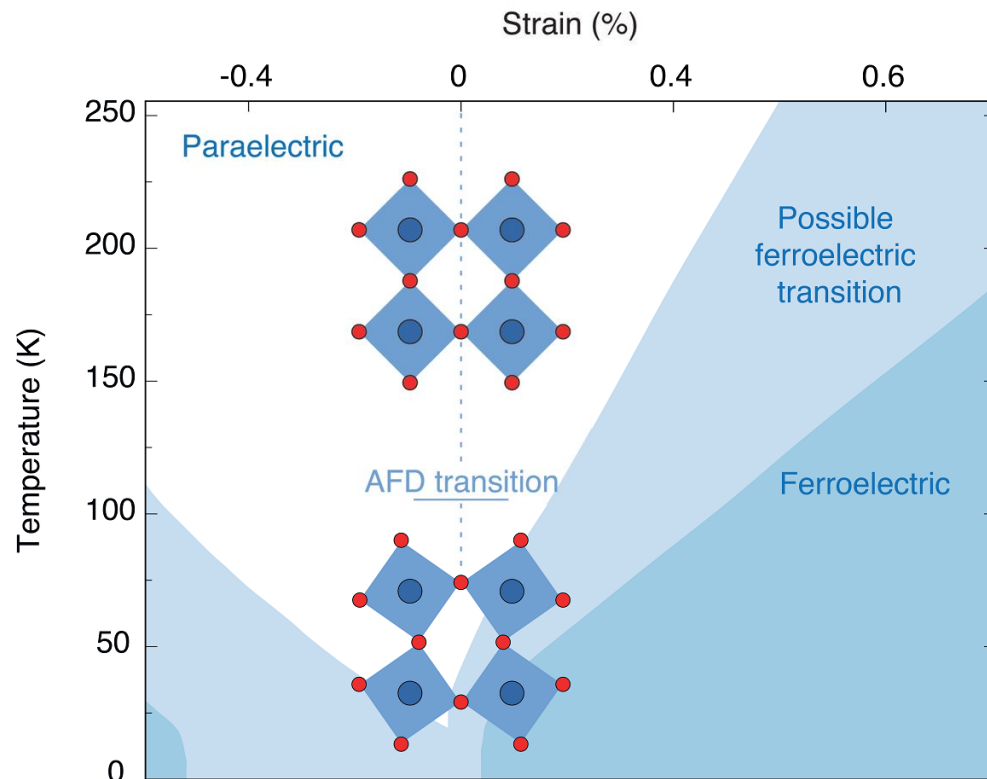
## Epitaxial strain



- Ferroelectricity up to  $T_c \approx 25$  K with “textbook” transition

- Ferroelectricity observed up to nearly room temperature with epitaxial strain

# Light-driven ferroelectricity in SrTiO<sub>3</sub>



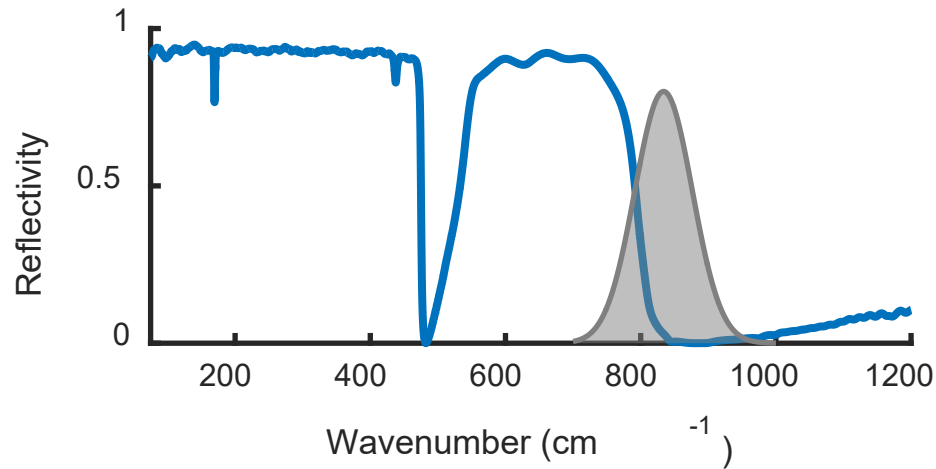
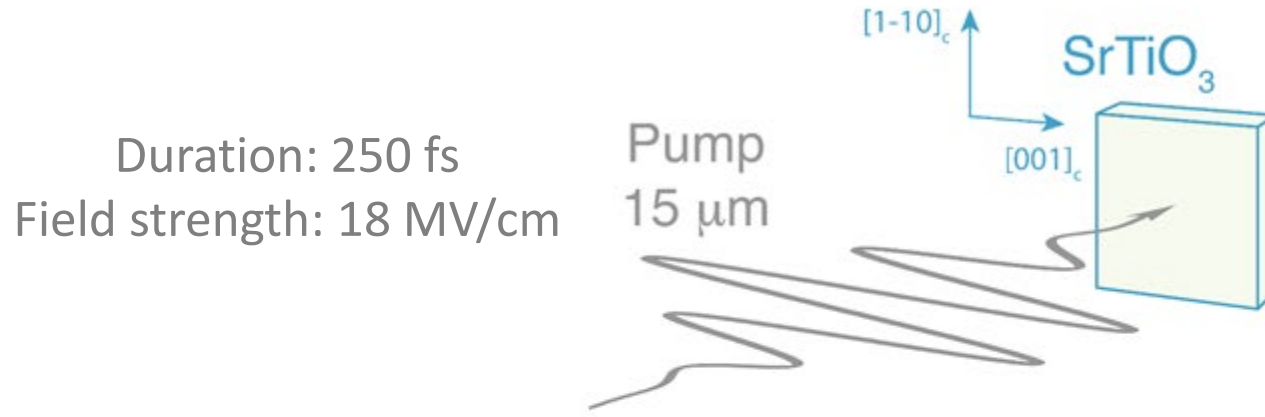
- Use resonant phonon excitation to induce ferroelectric phase

$$U = \sum_i \frac{1}{2} \omega_i^2 Q_i^2 + V_{NL}$$

IR-phonon coupling to strain:

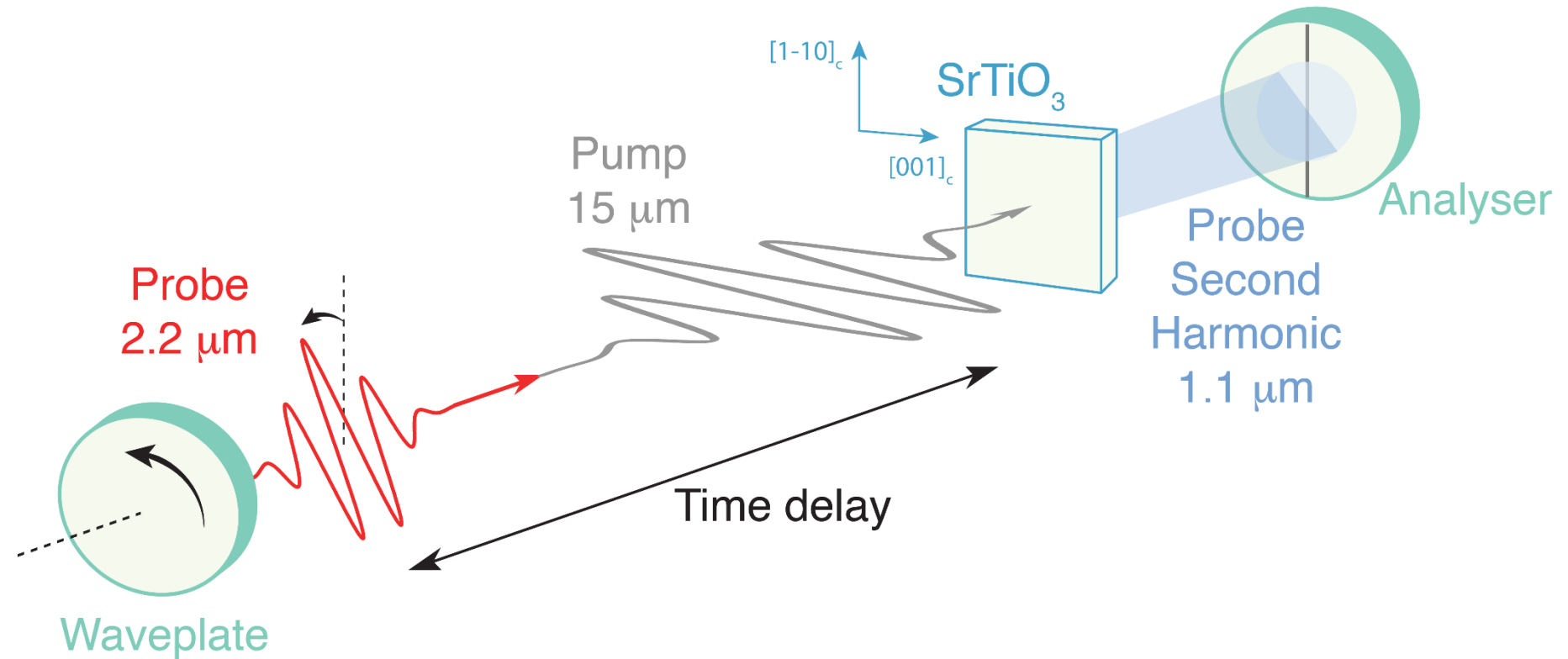
$$Q_{\text{driven}}^2 \cdot \epsilon$$

# Experimental setup

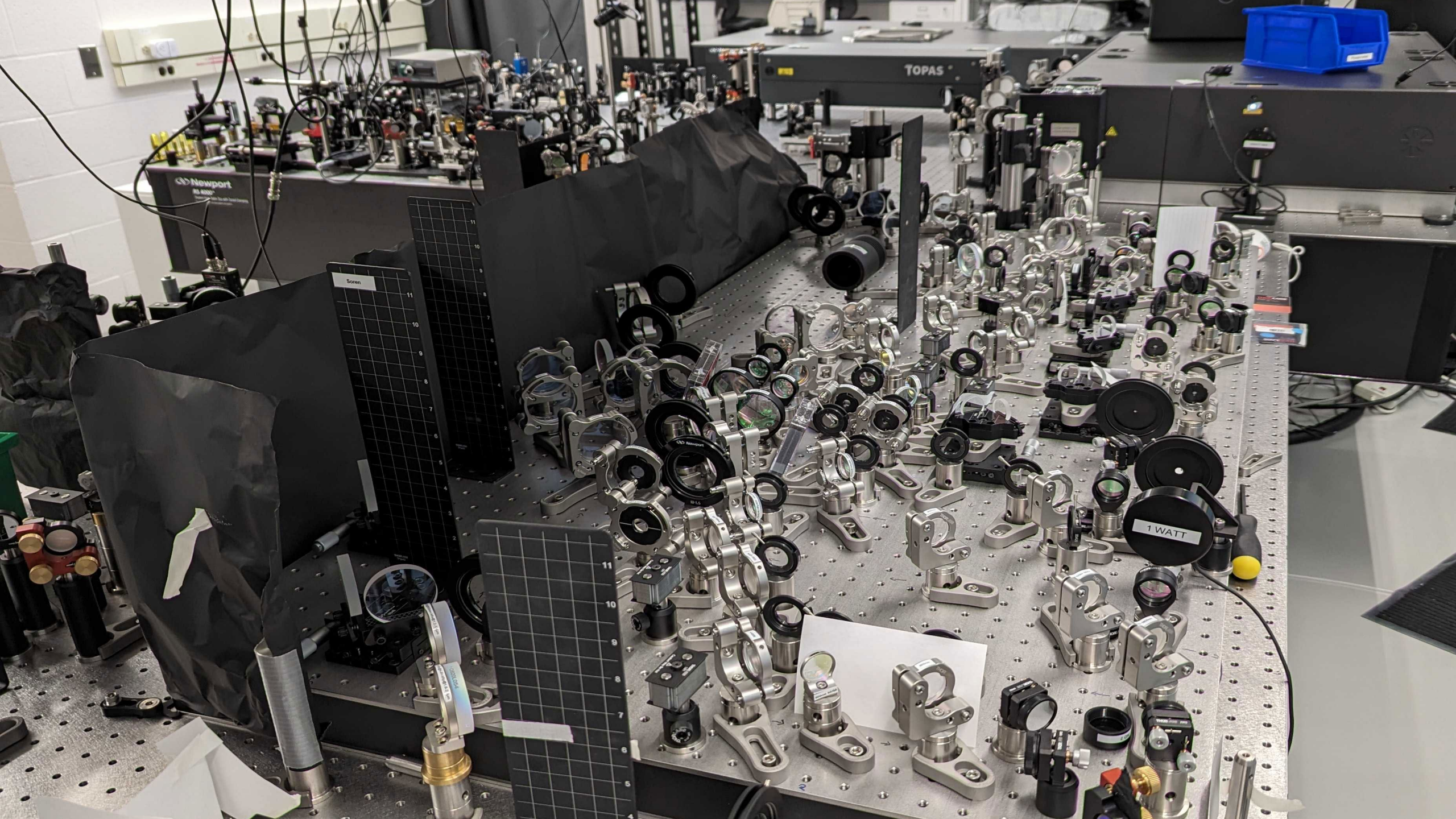


Pump resonant to highest frequency IR phonon

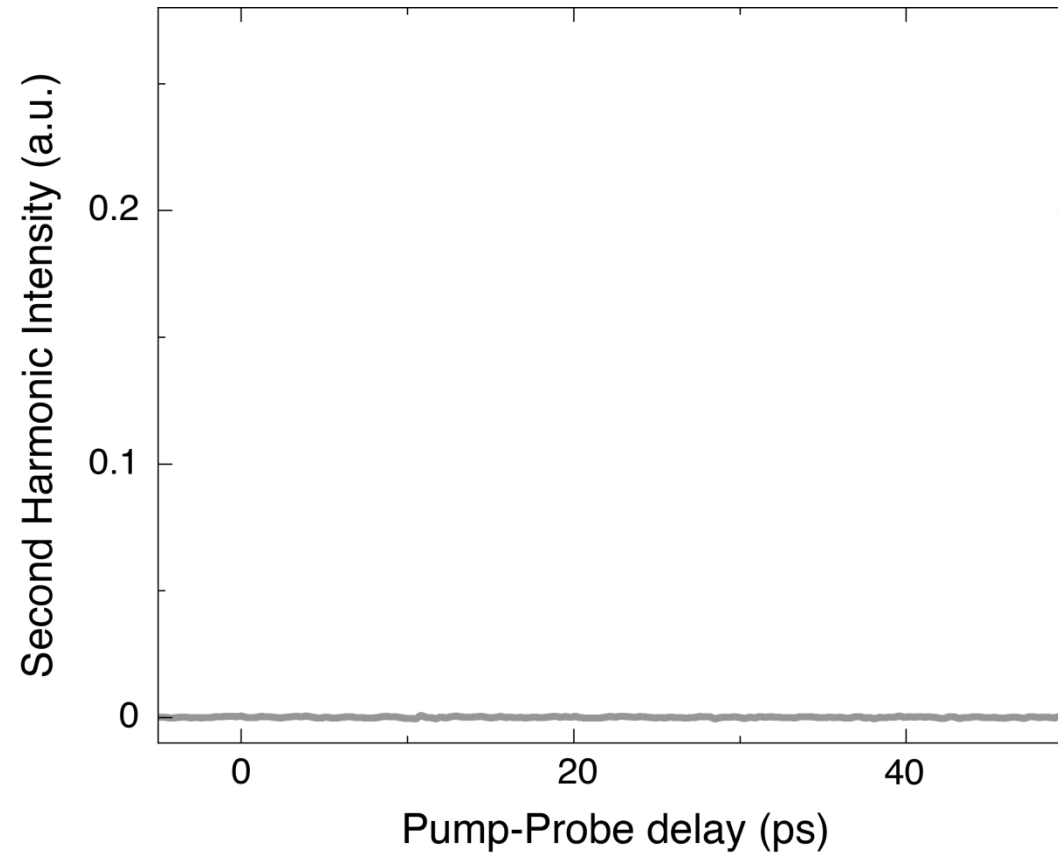
# Experimental setup



Second harmonic generation sensitive to **breaking of inversion symmetry**



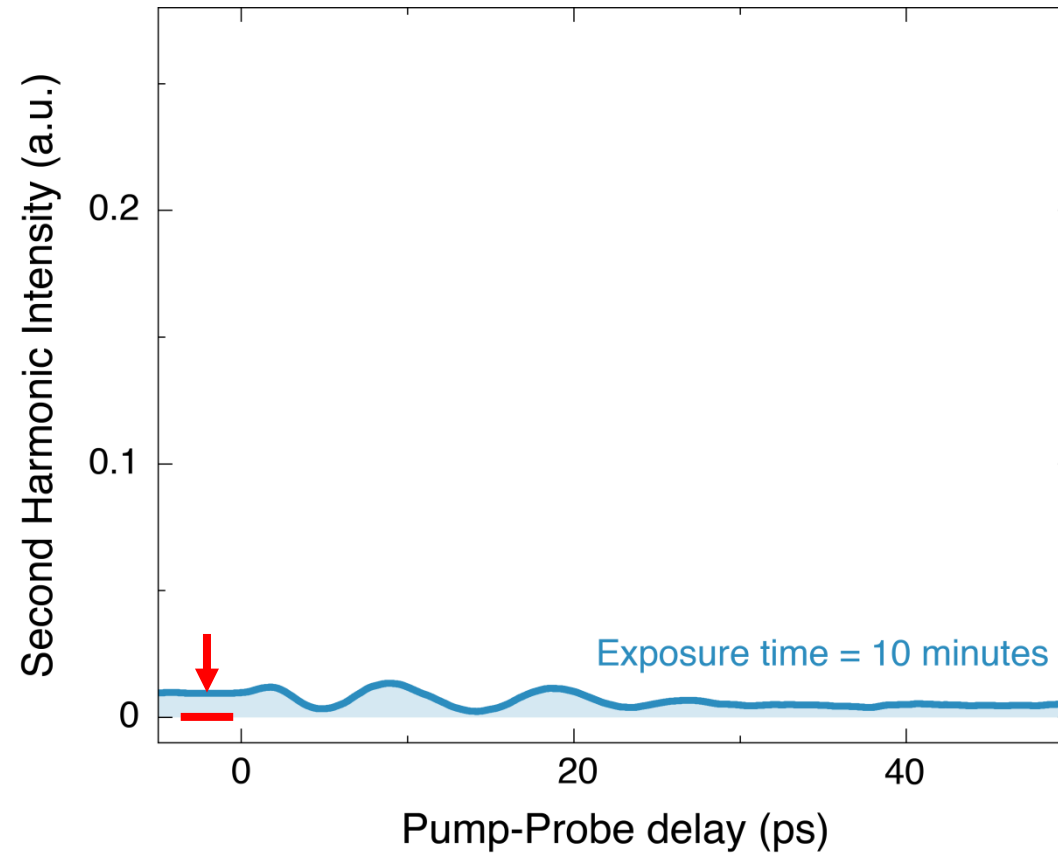
# Pump-probe signal



- No initial second harmonic signal



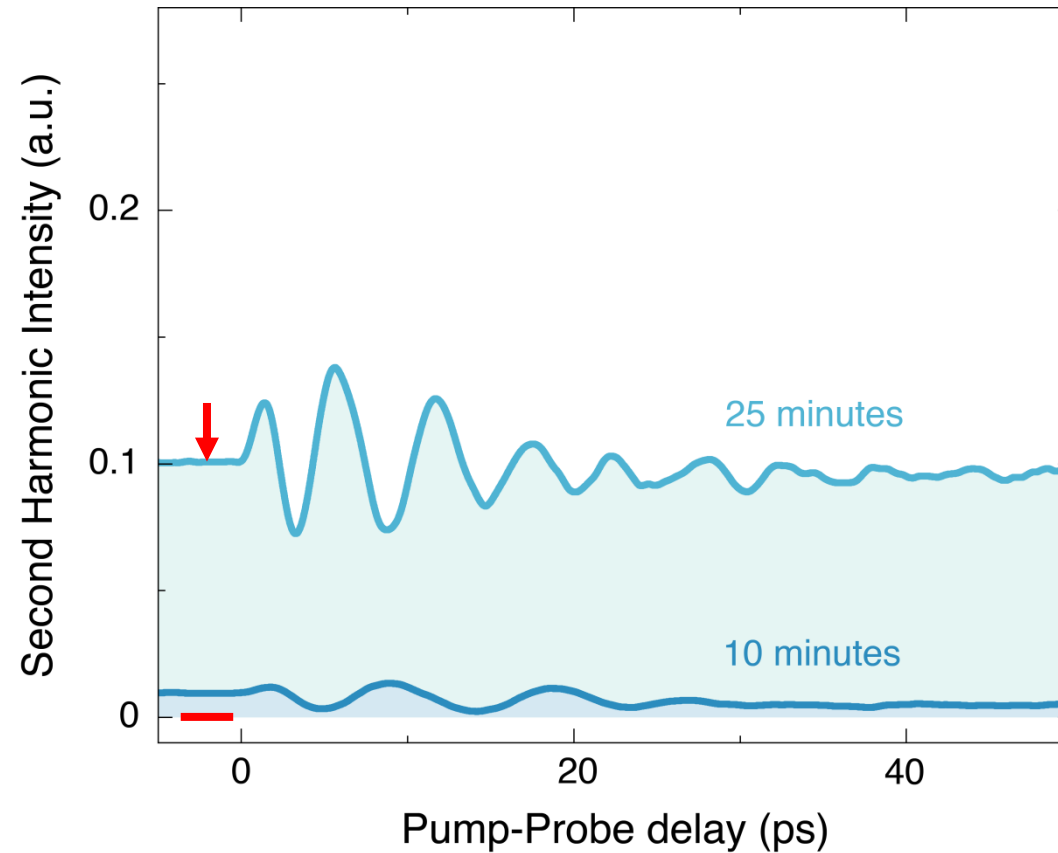
# Pump-probe signal



- Non-zero second harmonic signal
- Pump-induced coherent oscillations



# Pump-probe signal

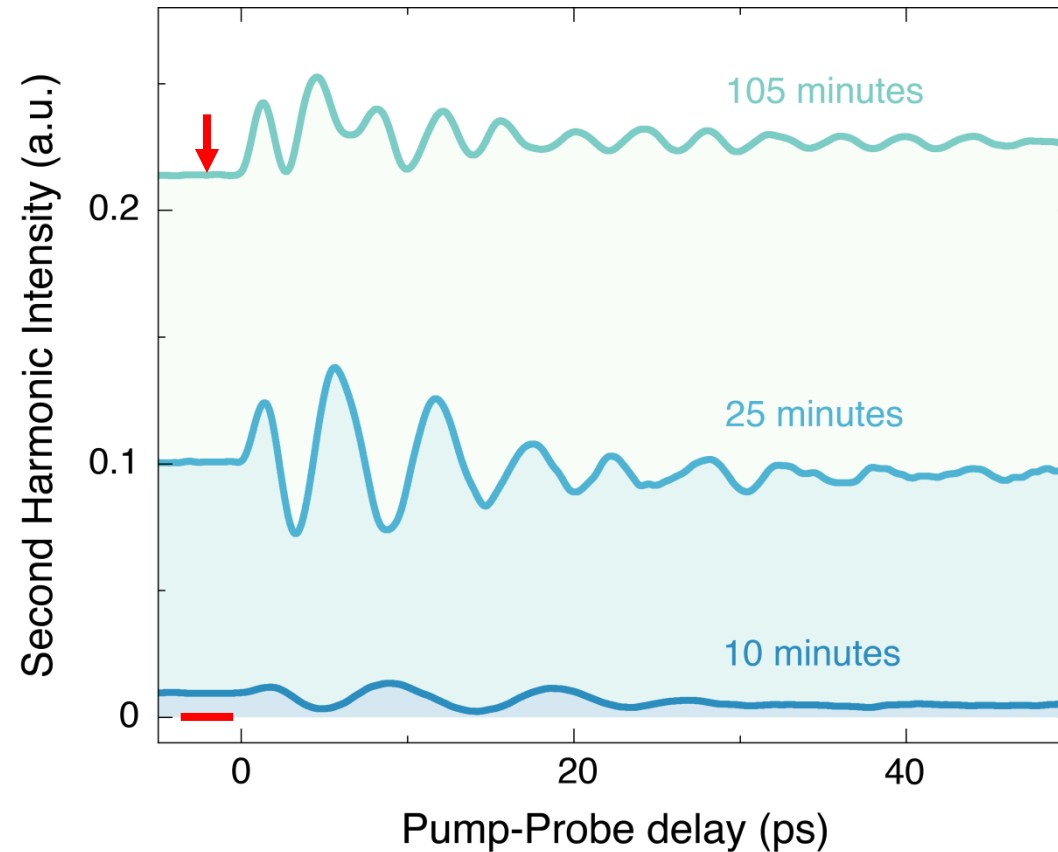


- Non-zero second harmonic signal

- Pump-induced coherent oscillations



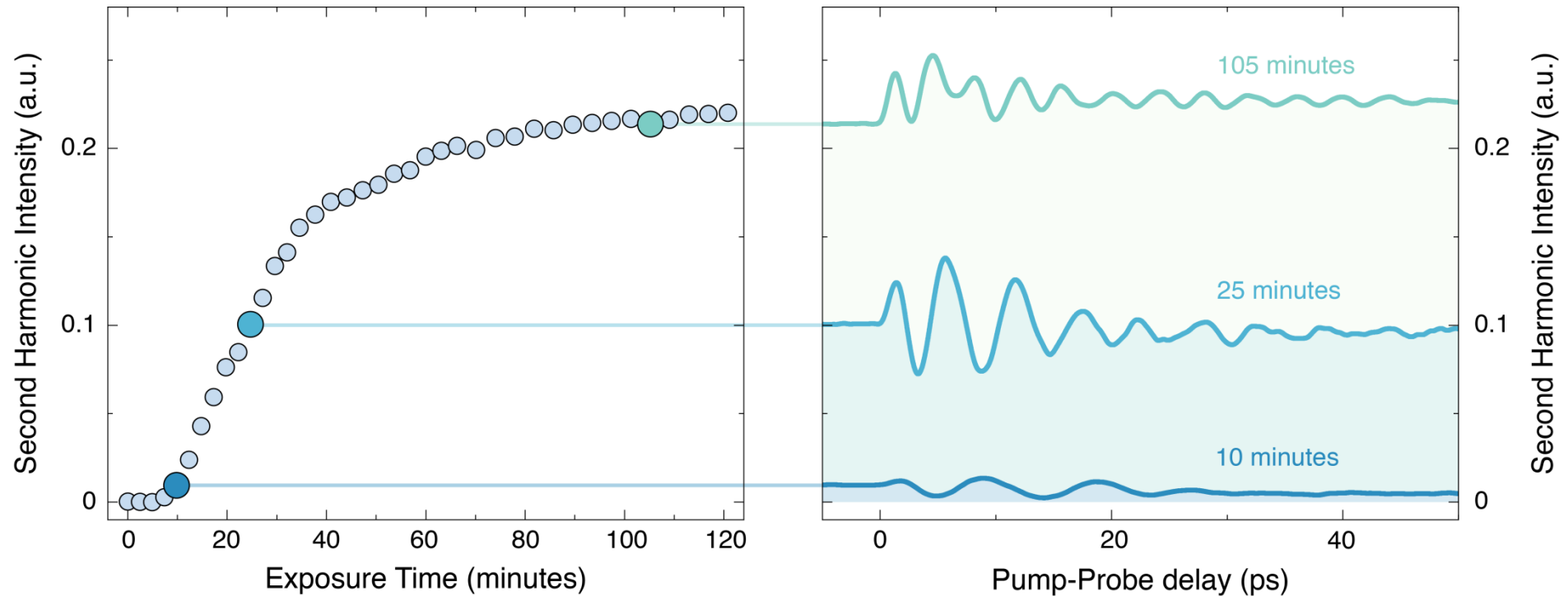
# Pump-probe signal



- Non-zero second harmonic signal
- **SHG signal grows with pump exposure**
- Pump-induced coherent oscillations
- Oscillations change with exposure



# Second Harmonic Growth/Saturation

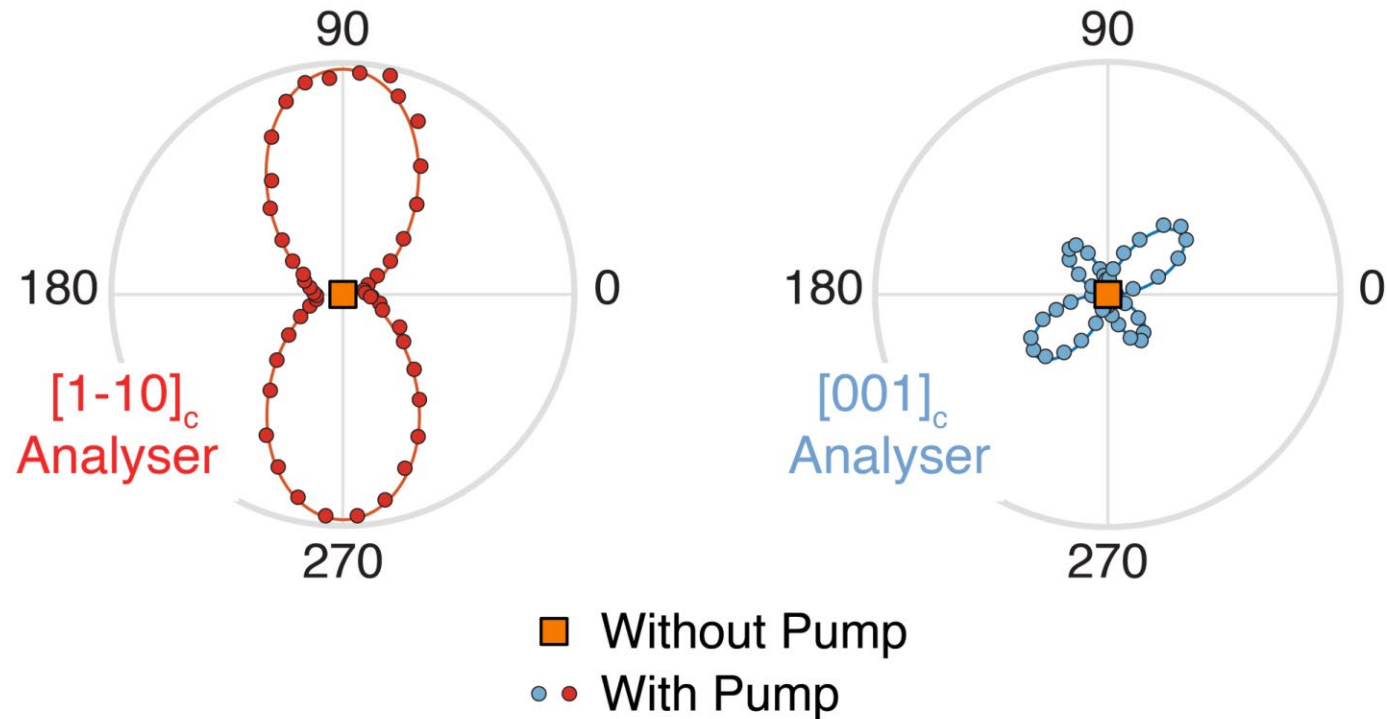


- Non-zero second harmonic signal
- **SHG signal grows with pump exposure**
- Pump-induced coherent oscillations
- Oscillations change with exposure



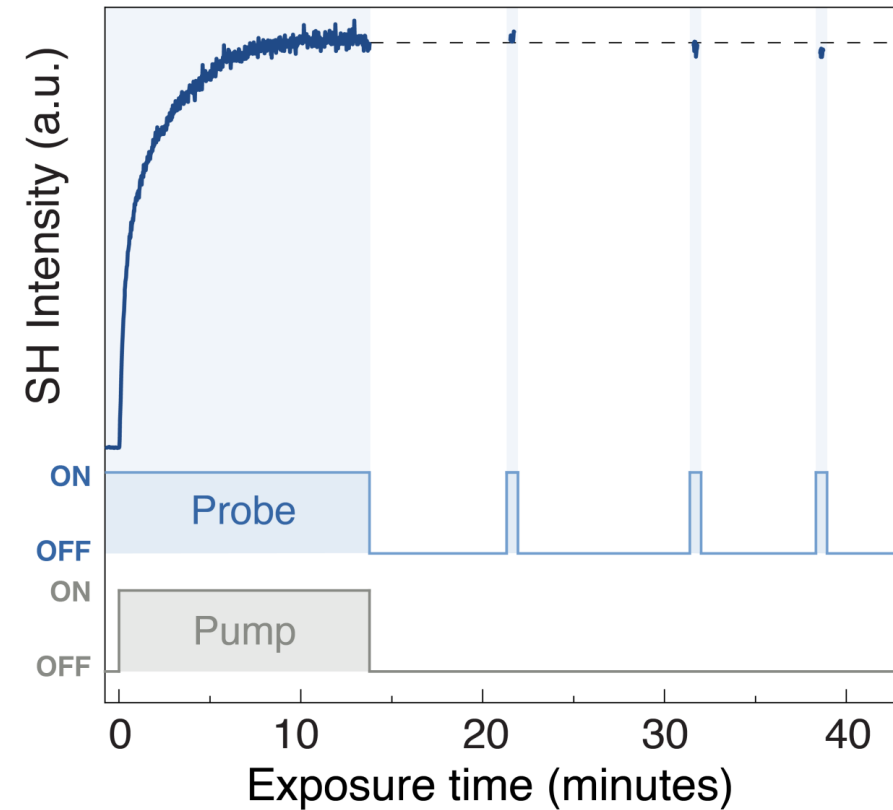
# Photo-induced polar order

## Incident probe polarization dependence



SHG angular dependence consistent with  $C_{2v}$  **polar symmetry** oriented along  $[1\bar{1}0]_c$

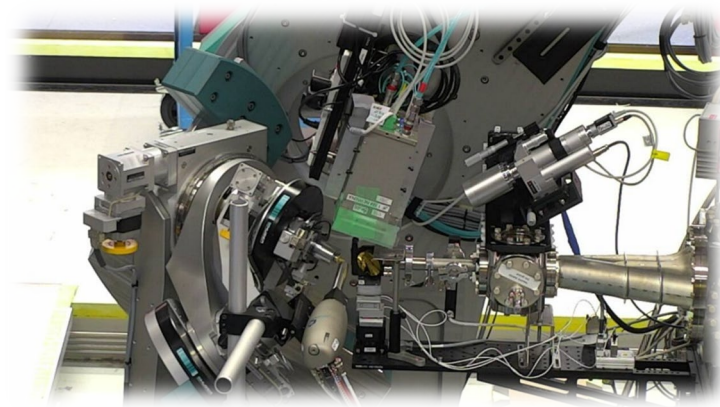
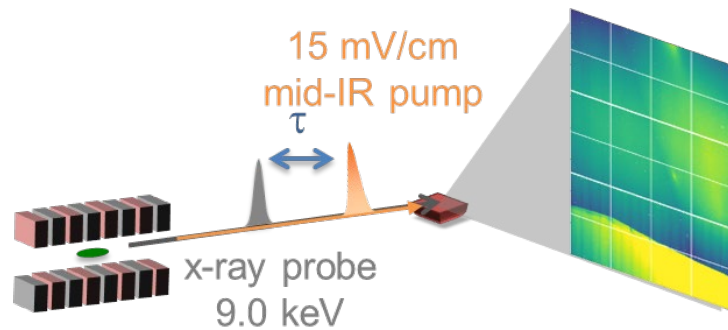
# Metastable ferroelectricity



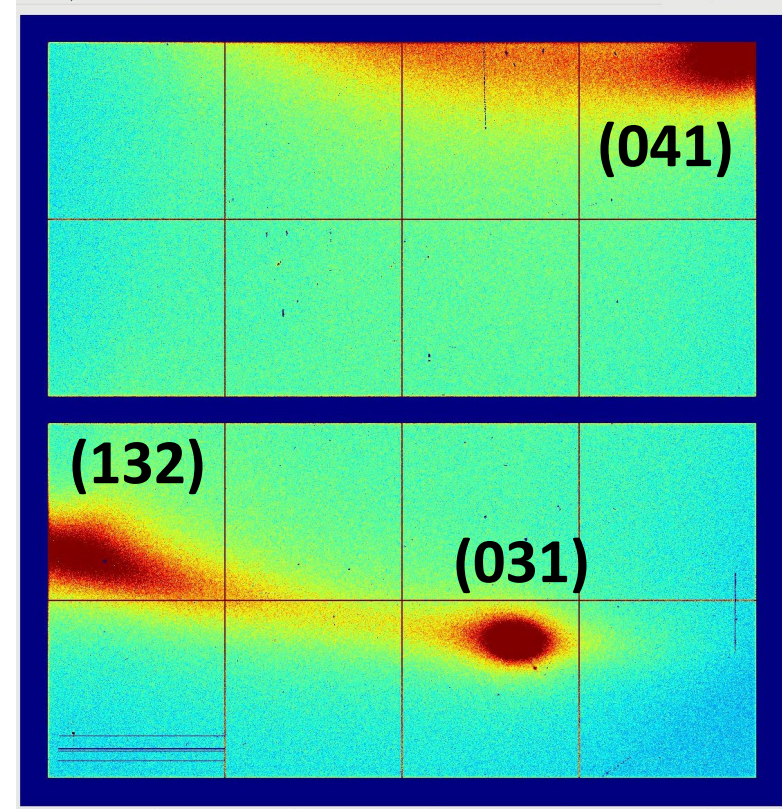
- Lifetime of **several hours** after pump exposure

# What is the origin of the effect?

- Time-resolved x-ray diffraction at SwissFEL to directly measure structural dynamics

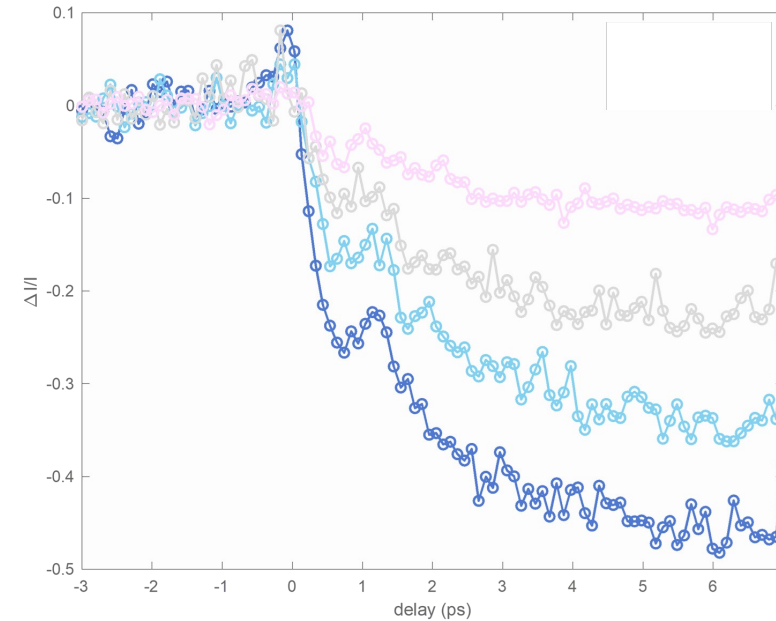
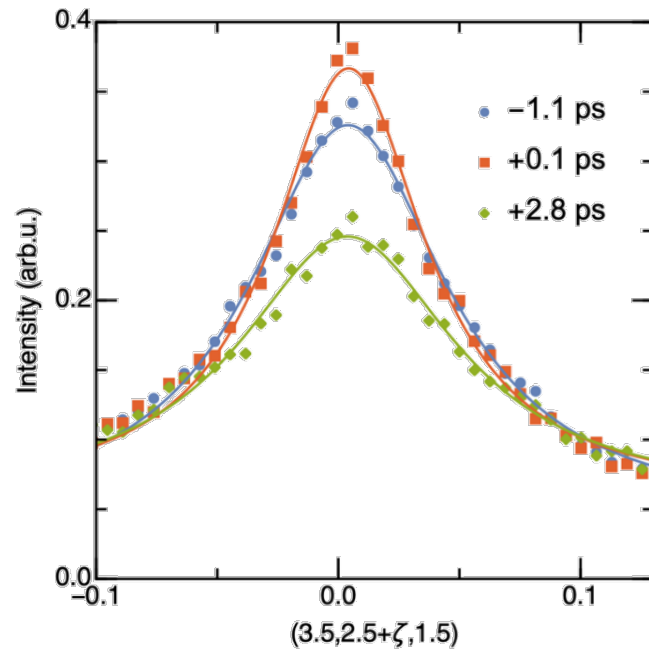
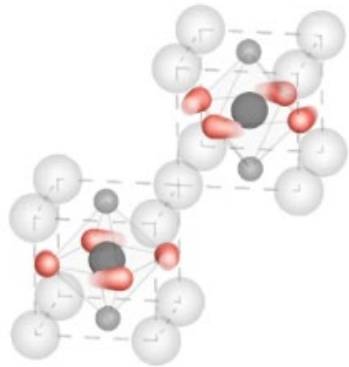


Static diffraction image



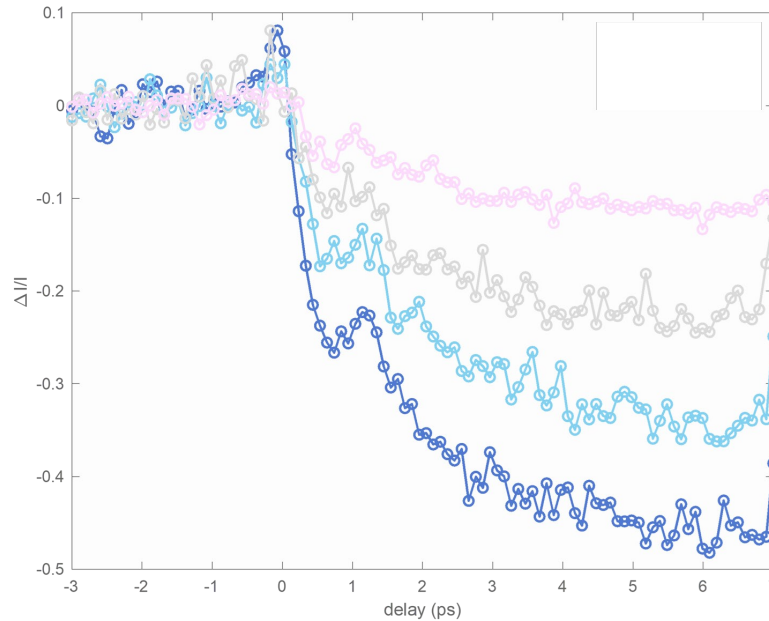
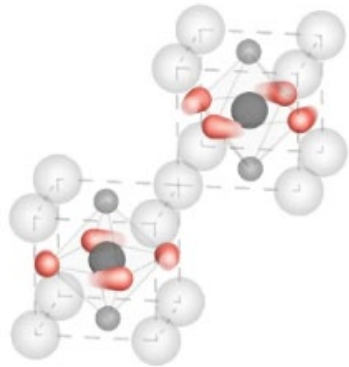
# What is the origin of the effect?

- (3.5, 2.5, 1.5) Bragg peak  $\rightarrow$  Sensitive to rotational mode and strain

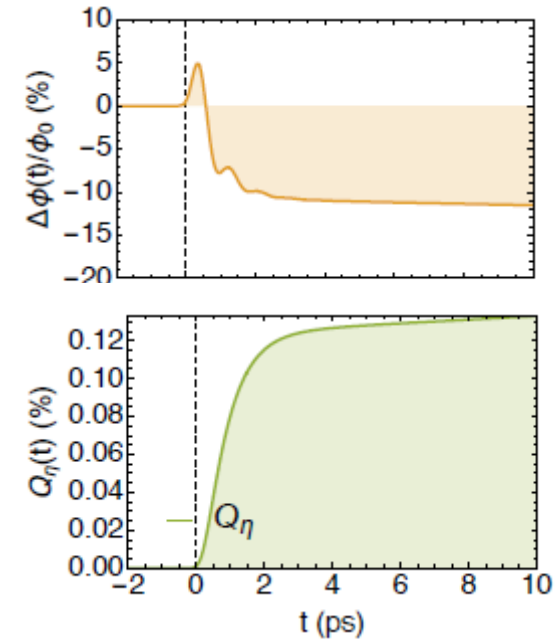


# What is the origin of the effect?

- (3.5, 2.5, 1.5) Bragg peak → Sensitive to rotational mode and strain



Density functional theory simulation

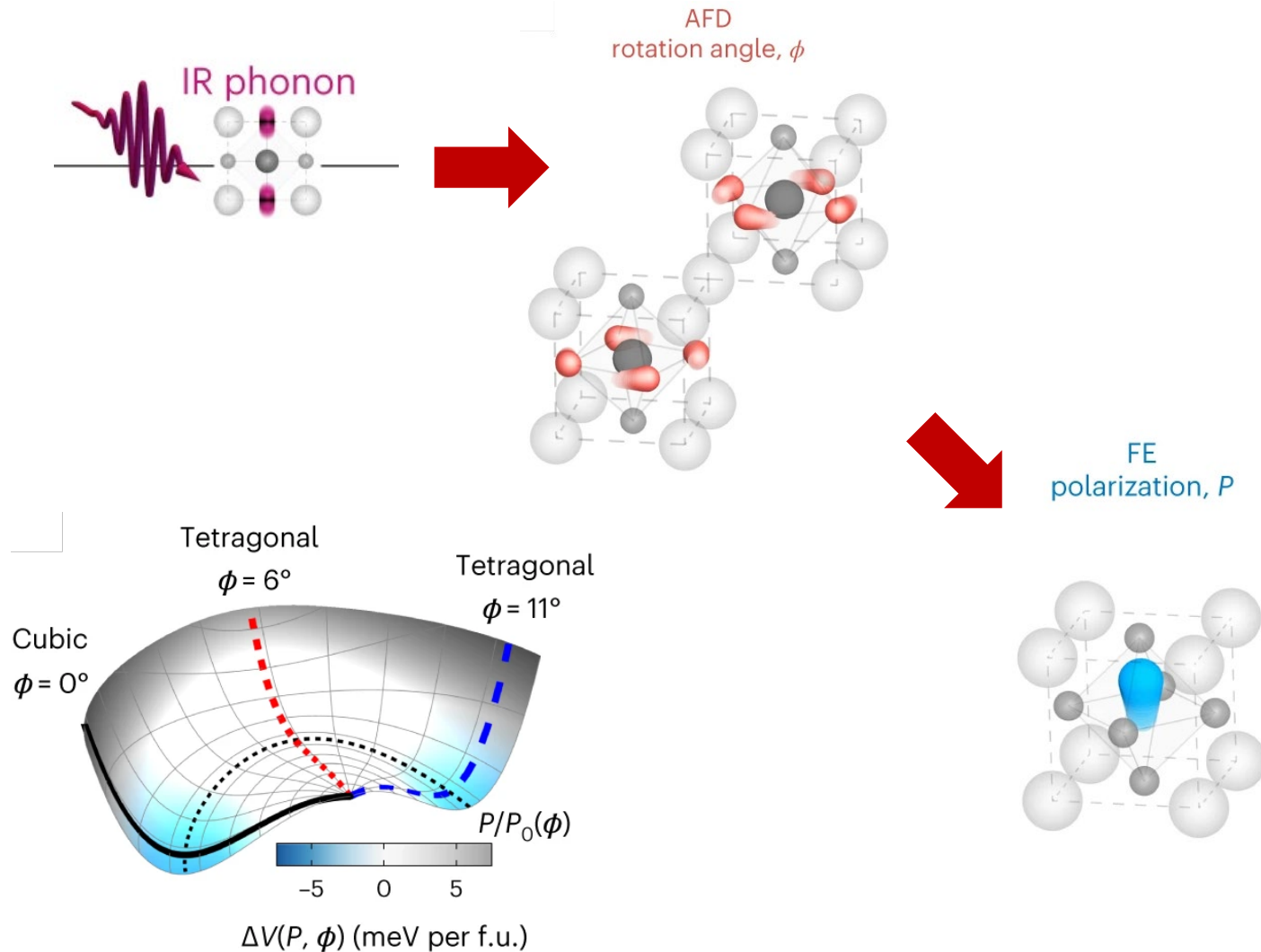


Driven mode: 
$$\frac{\partial^2}{\partial t^2} Q_{\text{IR}}[t] + 2\gamma_{\text{IR}} \frac{\partial}{\partial t} Q_{\text{IR}}[t] + \omega_{\text{IR}}^2 Q_{\text{IR}}[t] = Z^* E[t]$$

Strain mode: 
$$\frac{\partial^2}{\partial t^2} Q_{\eta}[t] + 2\gamma_{\eta} \frac{\partial}{\partial t} Q_{\eta}[t] + \omega_{\eta}^2 Q_{\eta}[t] = g_{\text{IR},\eta} Q_{\text{IR}}^2$$

Rotational mode: 
$$\begin{aligned} & \frac{\partial^3}{\partial t^3} \langle u_{\text{AFD}}^2[t] \rangle + 2\gamma_{\text{AFD}} \frac{\partial^2}{\partial t^2} \langle u_{\text{AFD}}^2[t] \rangle \\ & + 4(\omega_{\text{AFD}}^2 + g_{\text{IR,AFD}} Q_{\text{IR}}^2[t] + g_{\eta,\text{AFD}} Q_{\eta}[t]) \frac{\partial}{\partial t} \langle u_{\text{AFD}}^2[t] \rangle \\ & = -2 \langle u_{\text{AFD}}^2[t] \rangle \left( 2g_{\text{IR,AFD}} Q_{\text{IR}}[t] \frac{\partial Q_{\text{IR}}[t]}{\partial t} + g_{\eta,\text{AFD}} \frac{\partial Q_{\eta}[t]}{\partial t} \right) \end{aligned}$$

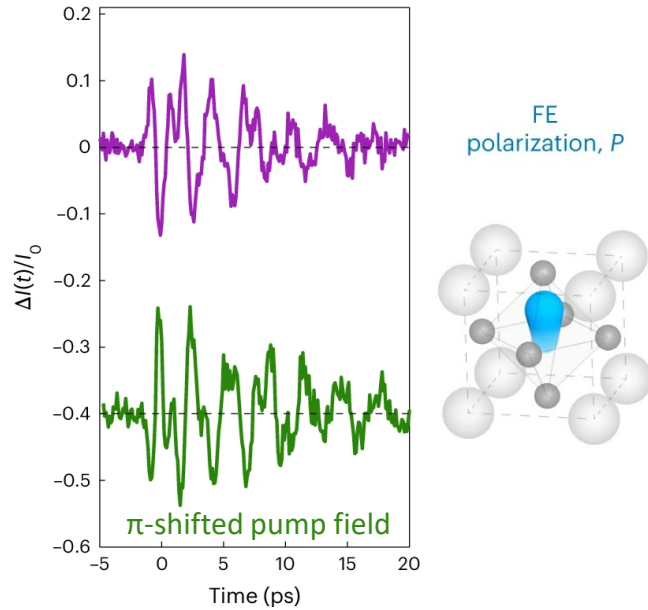
# Physical picture of light-induced ferroelectricity



1. Resonantly driving IR mode reduces rotation angle
2. Leads to lowering of energy of ferroelectric state
3. Ferroelectricity stabilized on long times by strain

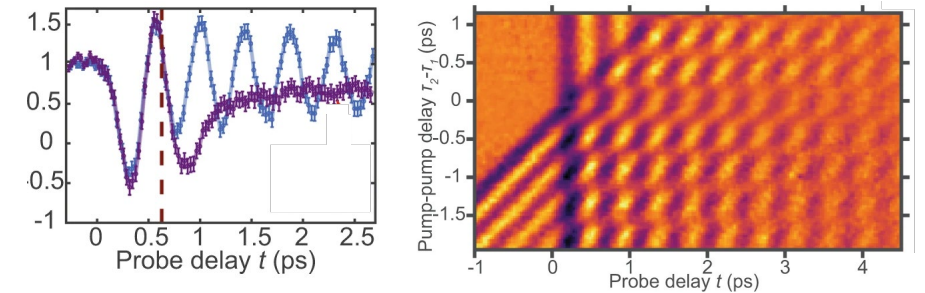
# More examples of THz x-ray modulation

- Direct soft mode stimulation (0.5 THz)



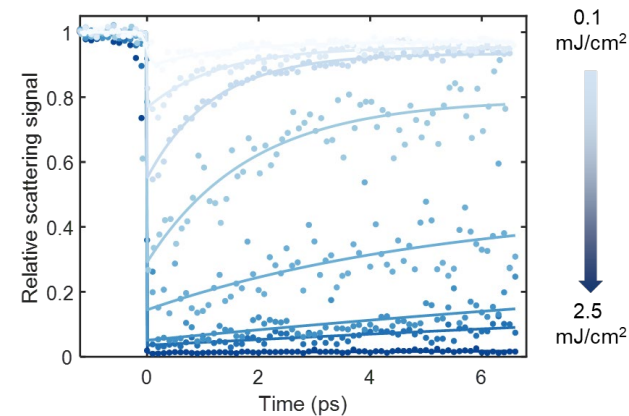
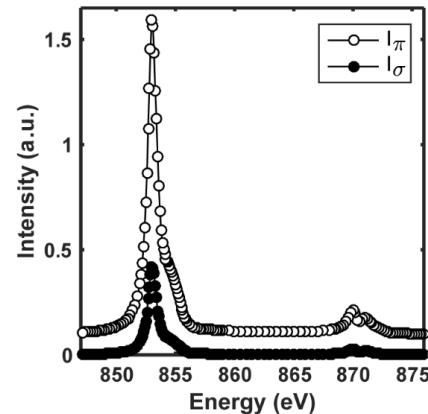
G. Orenstein *et al.* Nat. Phys. **21**, 961 (2025)

- Coherent control of x-ray intensity



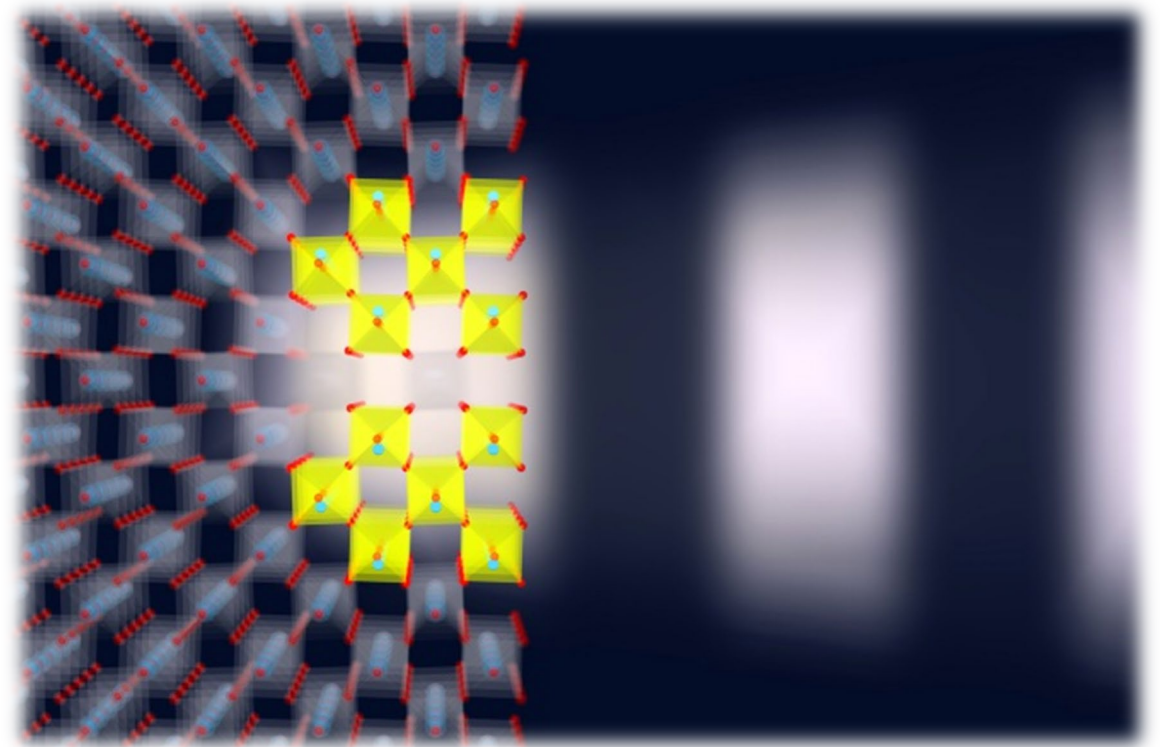
G. Gorobtsov *et al.* Nat. Comm. **12**, 2865 (2021)

- Resonant soft x-ray quenching



# Take away messages

- **Driving the crystal lattice with light provides a powerful means to engineer functionalities quantum materials**
- **Resonant vibrational control can also lead to strong, ultrafast modulation of x-ray intensities**



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

Nova, Disa, *et al.* Science 364, 1075 (2019)

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