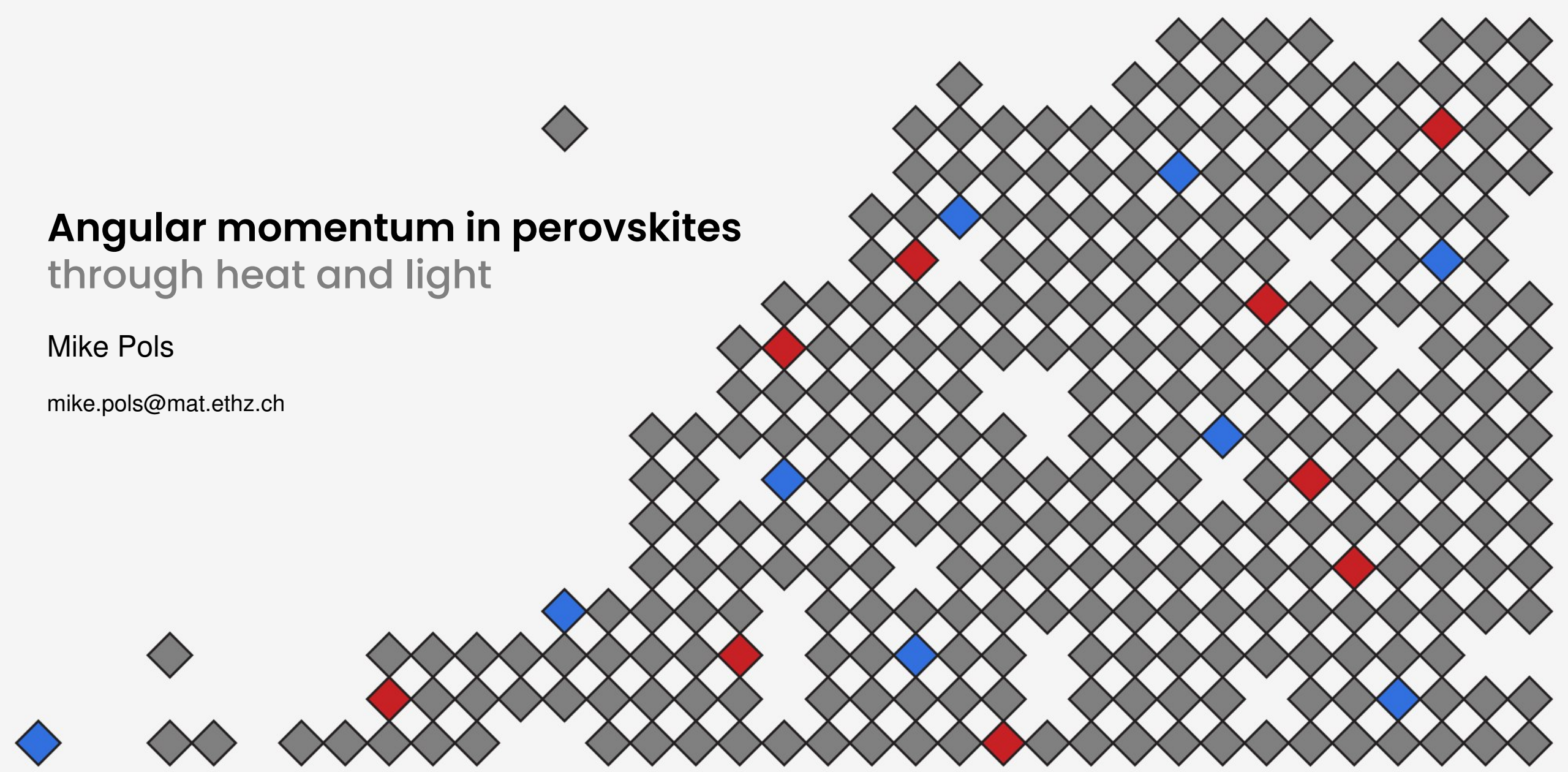


Angular momentum in perovskites through heat and light

Mike Pols

mike.pols@mat.ethz.ch

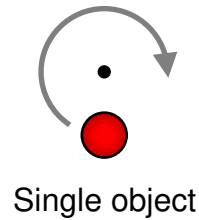


Angular momentum: an additional degree of freedom

Definition

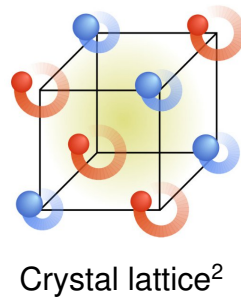
Classical definition of angular momentum

$$\mathbf{J} = \mathbf{r} \times \mathbf{p} = \mathbf{r} \times m\mathbf{v}$$

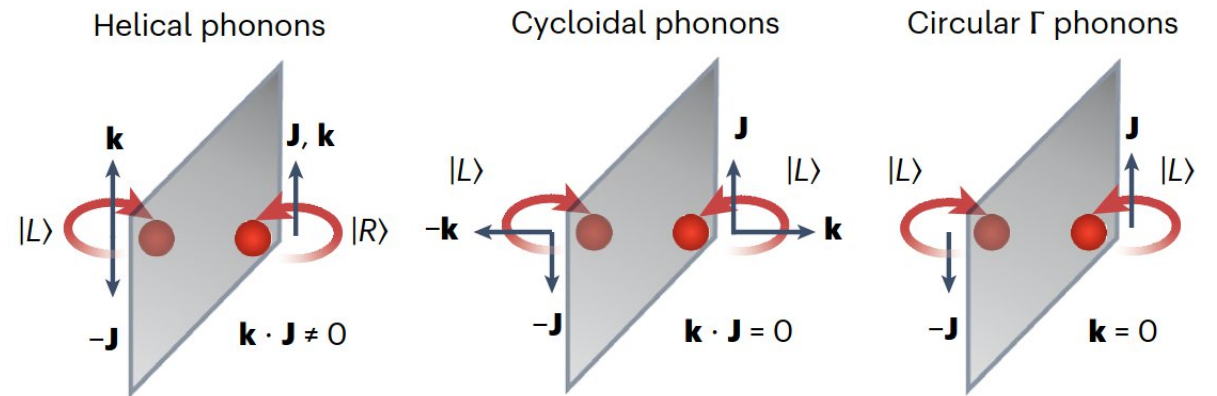


Angular momentum of phonons¹

$$\mathbf{J}^{\text{ph}} = \sum_{\kappa} m_{\kappa} \mathbf{u}_{\kappa} \times \dot{\mathbf{u}}_{\kappa}$$



Classification of phonons³



Effects: magnetic moments, Zeeman effect, magnetization switching etc.

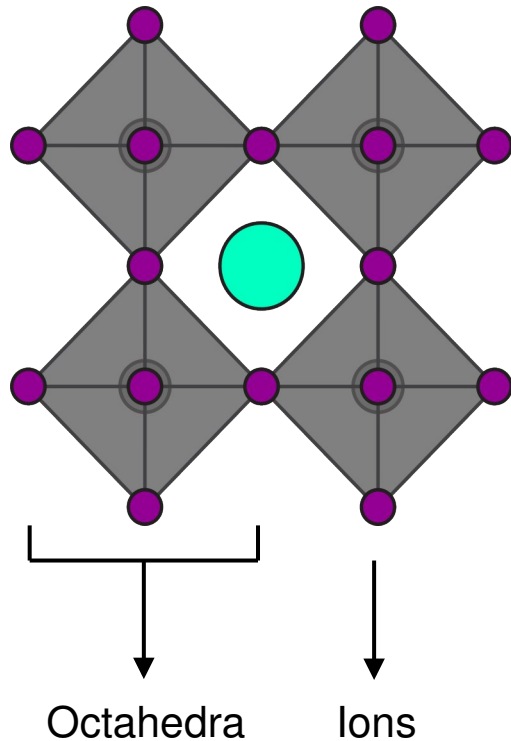
¹ Zhang, L. & Niu, Q. Phys. Rev. Lett., 112, 085503 (2014).

² Juraschek, D.M. et al. Phys. Rev. Mater., 1, 014401 (2017).

³ Juraschek, D.M. et al. Nat. Phys., 21, 1532–1540 (2025).

Perovskites: a versatile class of materials

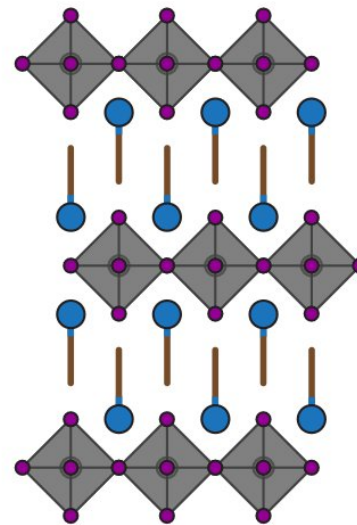
Perovskite
 ABX_3 crystal



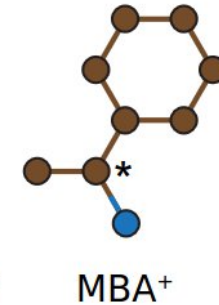
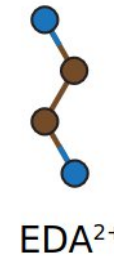
Broad chemical complexity including:

1. **Oxide perovskites:** LiNbO_3 (piezoelectric, waveguide)
2. **Halide perovskites:** CsPbI_3 , $(\text{S-MBA})_2\text{PbI}_4$ (optoelectronics)

Various morphologies: A_2BX_4



Organic cations



Machine-learning potentials for material dynamics

Material dynamics require a potential energy surface (PES)

$$m_{\kappa} \ddot{\mathbf{r}}_{\kappa} = \mathbf{F}_{\kappa} = -\nabla E(\{\mathbf{r}_{\kappa}\})$$

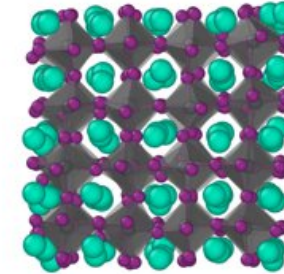


First-principles methods
density functional theory (DFT)

$$\hat{H}\Psi = E\Psi$$



Approximated by machine-learning interatomic potentials
GAP¹ and MACE²



Dynamical snapshots

Relevant data:

1. Energy
2. Forces
3. Stresses

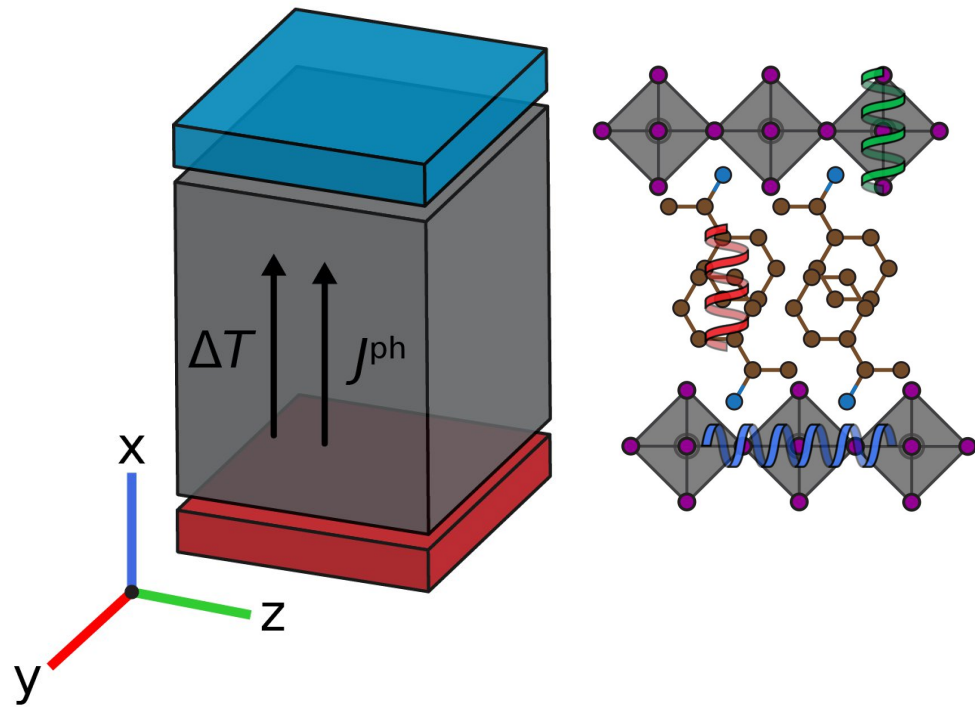
$$\mathcal{L} = \sum_{i=1}^{N_T} \left[\frac{y_{i,\text{ref}} - \tilde{y}_{i,\text{model}}}{\sigma_i} \right]^2$$

Data point weight

¹ Jinnouchi, R. et al. Phys. Rev. B 100, 014105 (2019).

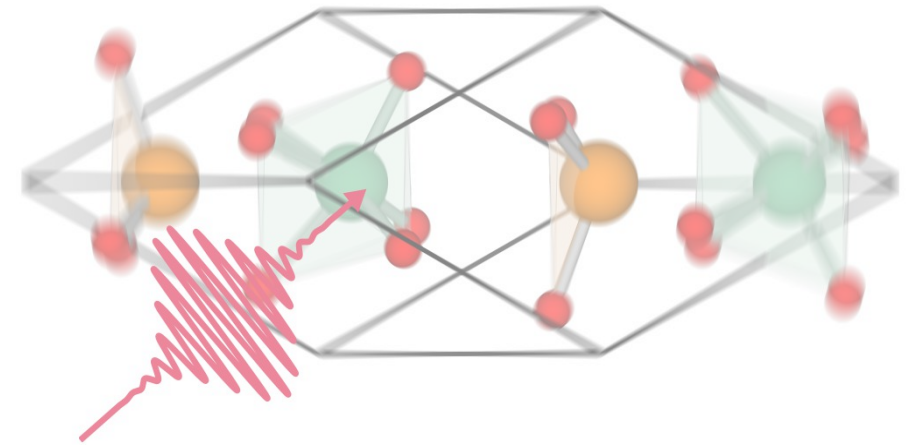
² Batatia, I. et al. NeurIPS, (2022).

Part 1:



Angular momentum from heat
in 2D halide perovskites

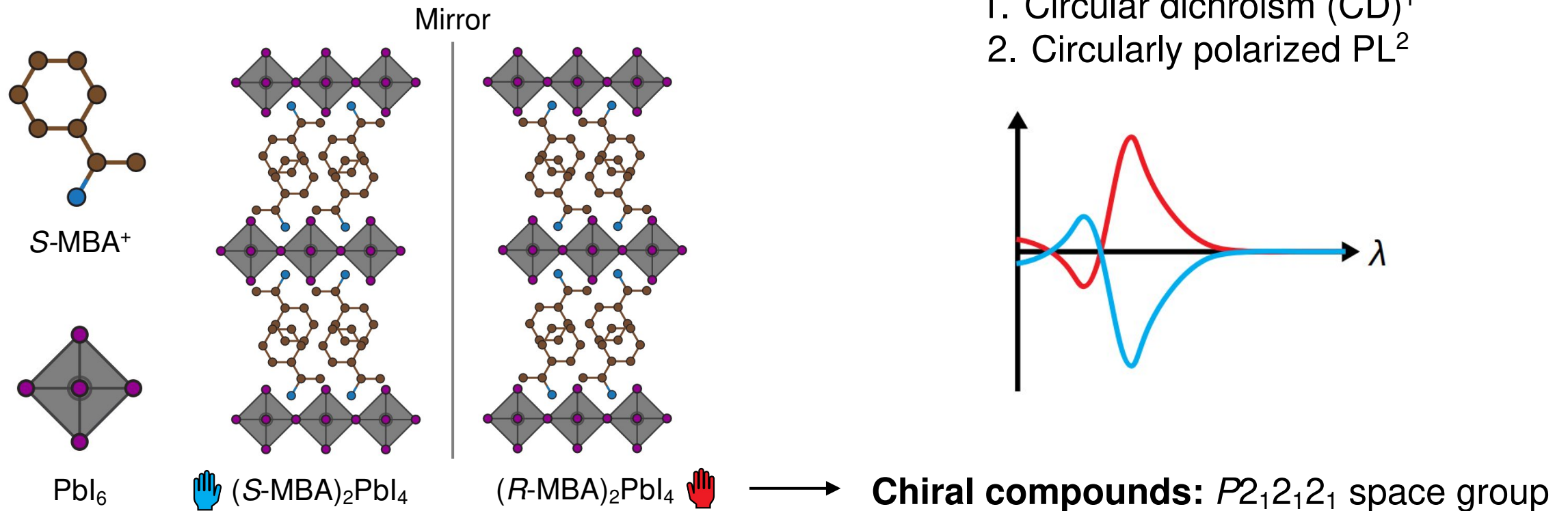
Part 2:



Laser-induced angular
momentum in LiNbO_3

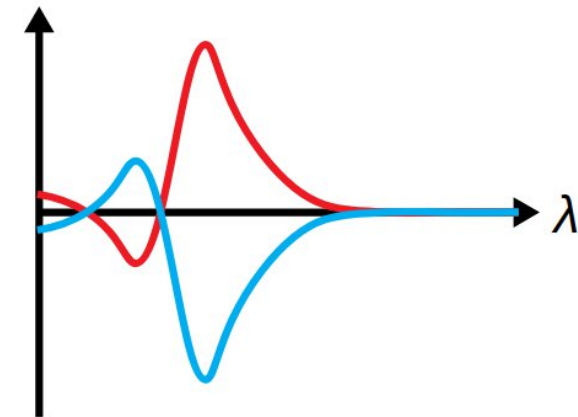
Engineering chiral materials

Two-dimensional perovskites adding functionality into lattice



Chiral material properties from inorganic framework:

1. Circular dichroism (CD)¹
2. Circularly polarized PL²

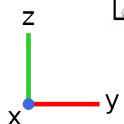
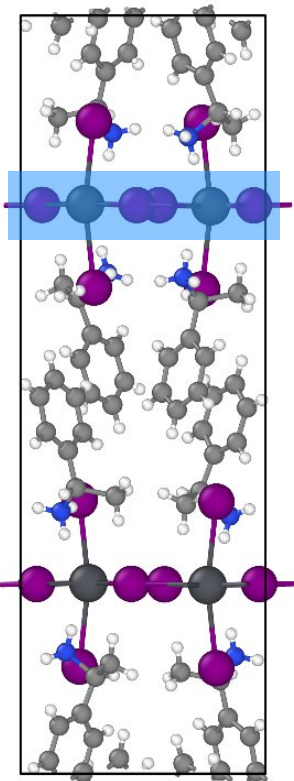


¹ Ahn, J. et al. Mater. Horiz. 4, 851 (2017).

² Ma, J. et al. ACS Nano 13, 3659-3665 (2019).

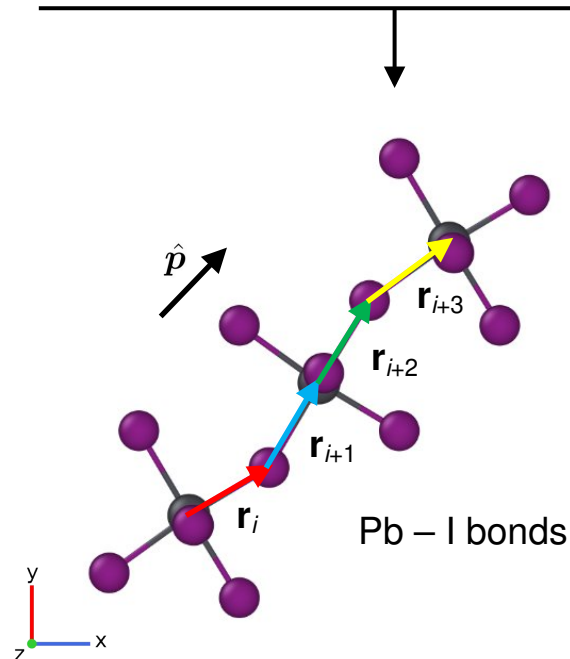
What parts of the crystal lattice are chiral?

(S-MBA)₂PbI₄



Different components of the crystal lattice¹

1. Arrangement of cations
2. Out-of-plane distortions octahedra
3. **In-plane** distortions octahedra



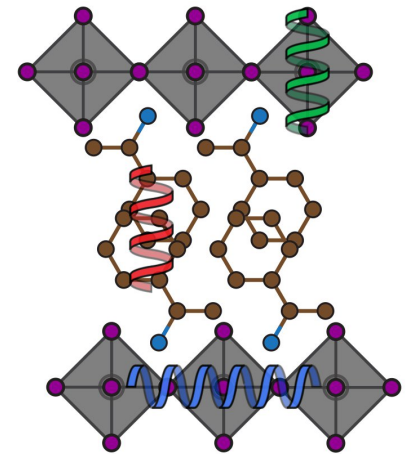
Structural chirality¹ (helicity)

$$\epsilon = \frac{1}{N} \sum_{i=1}^N \hat{\mathbf{u}}_i \times \hat{\mathbf{u}}_{i+1}$$

with $\hat{\mathbf{u}}_i = \mathbf{r}_i / |\mathbf{r}_i|$

$$\epsilon = \epsilon \cdot \hat{\mathbf{p}} \longrightarrow$$

All components chiral!



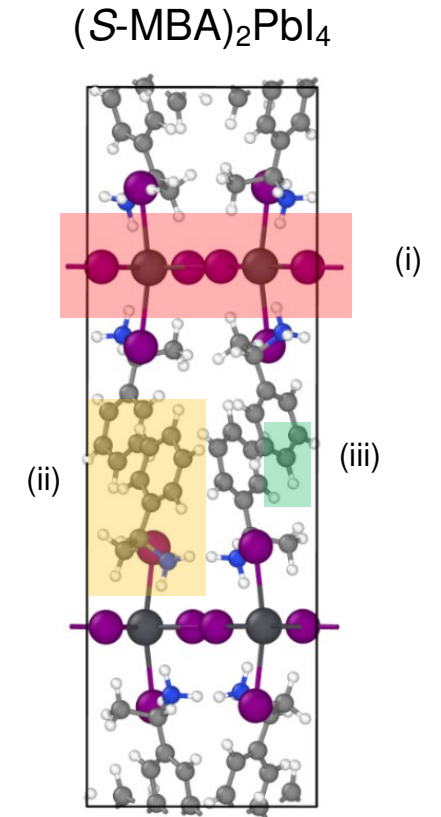
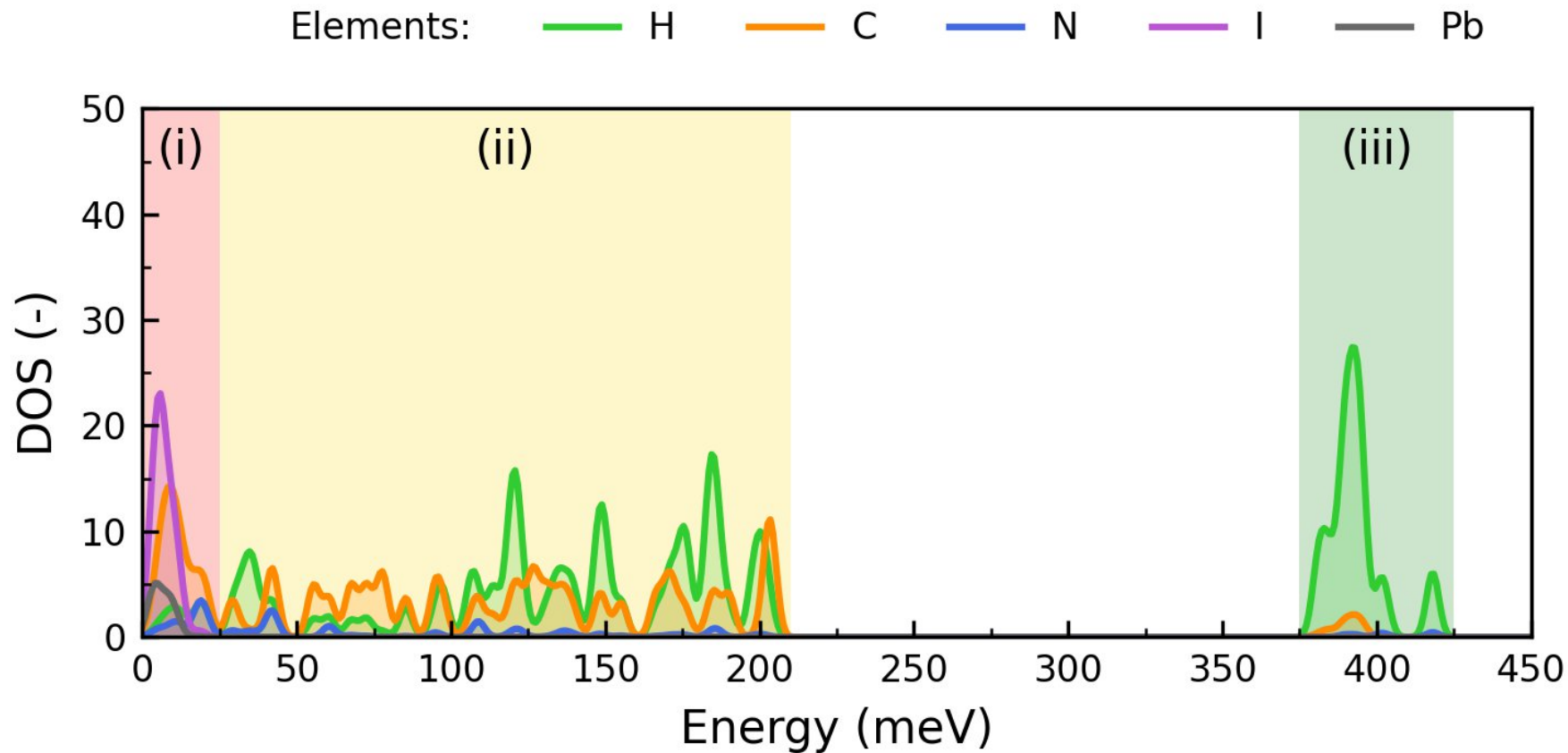
Even at finite temperatures¹
using MLIPs in MD simulations

Left-/right-handed ($\epsilon \neq 0$)

Non-chiral ($\epsilon = 0$)

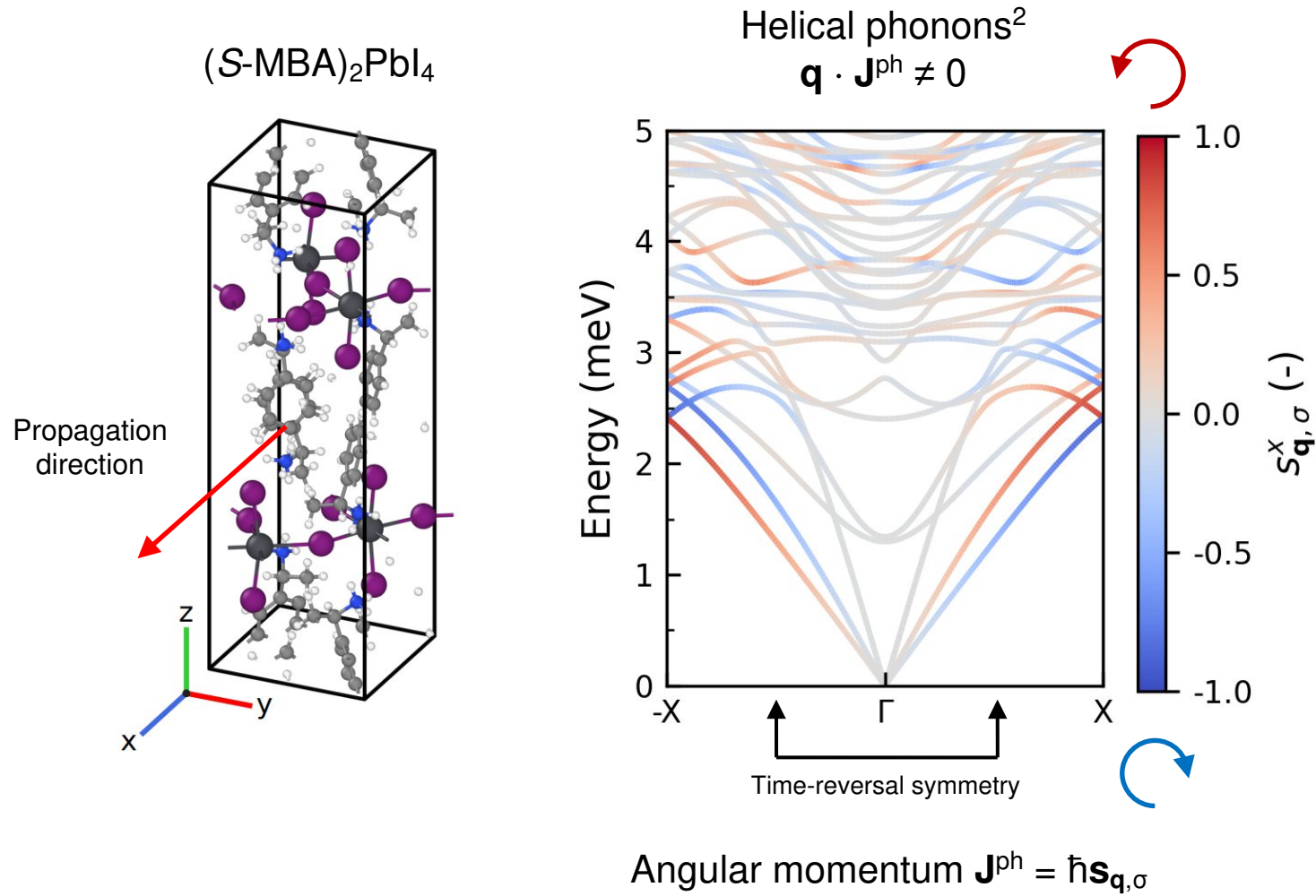
¹ Pols, M. et al. J. Phys. Chem. Lett. 15, 8057-8064 (2024).

Phonons in chiral 2D perovskites



¹ Pols, M. et al. Nano Lett. 25, 10003-10009 (2025).

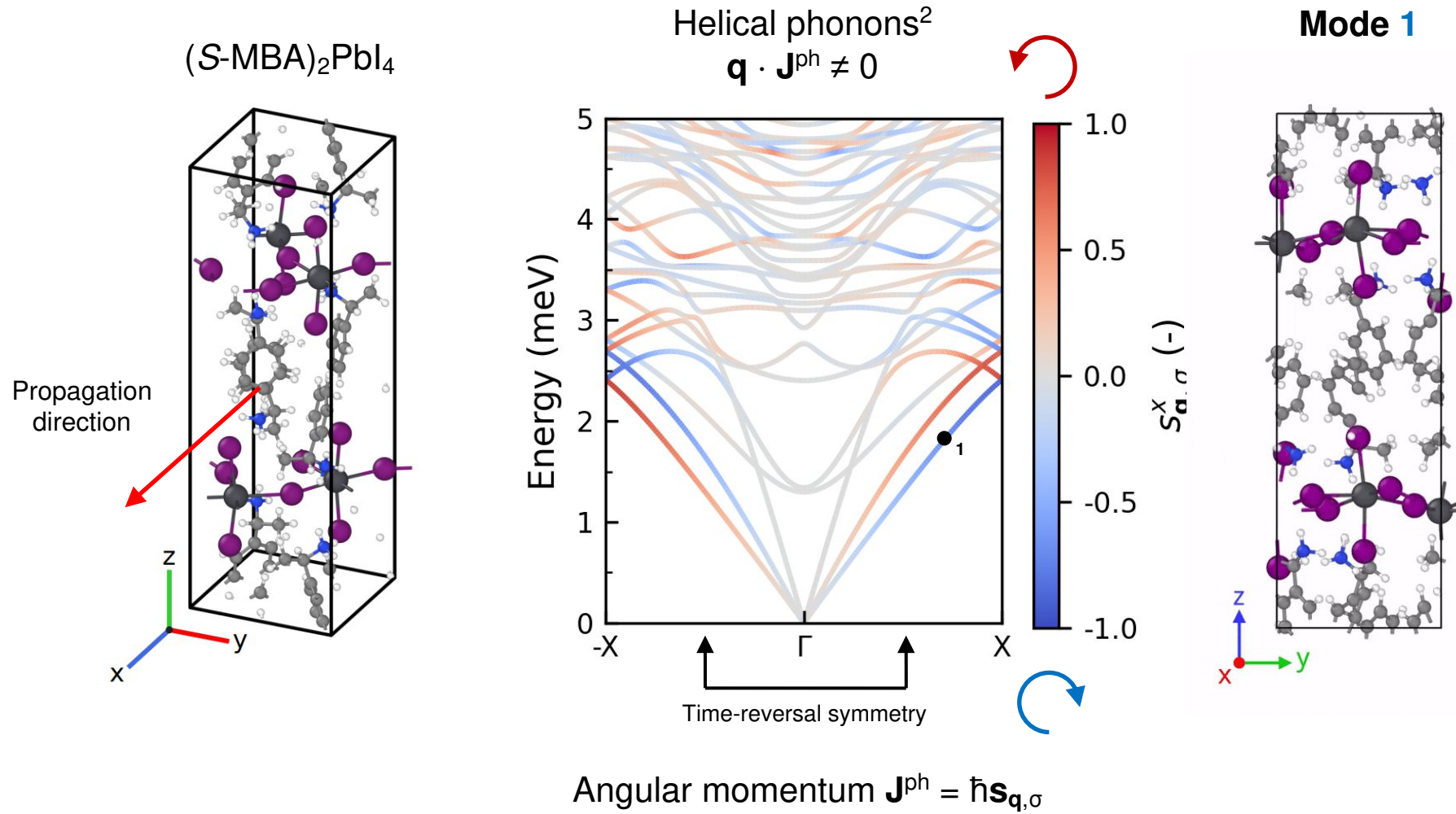
And, are they chiral?



¹ Pols, M. et al. Nano Lett. 25, 10003-10009 (2025).

² Juraschek, D.M. et al. Nat. Phys., 21, 1532-1540 (2025).

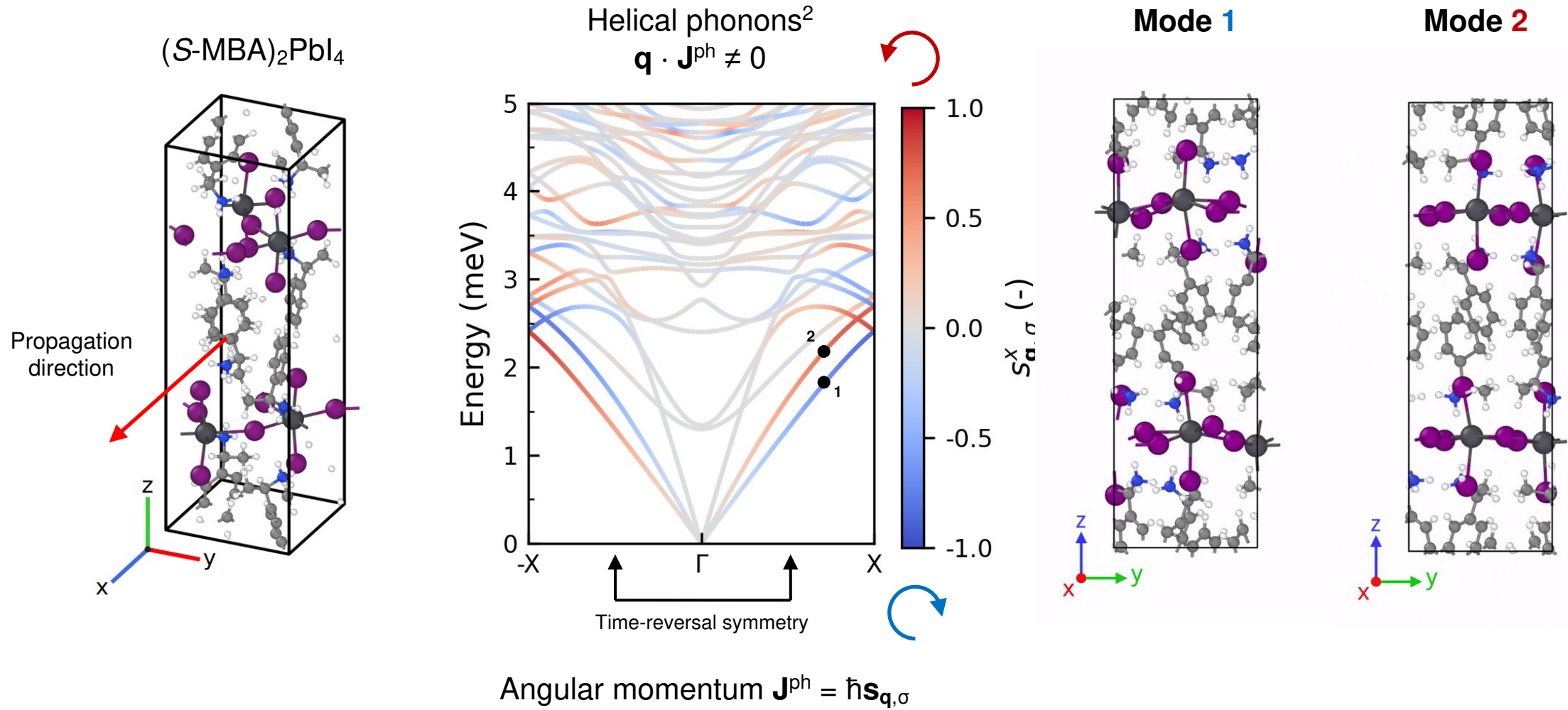
And, are they chiral?



¹ Pols, M. et al. Nano Lett. 25, 10003-10009 (2025).

² Juraschek, D.M. et al. Nat. Phys., 21, 1532-1540 (2025).

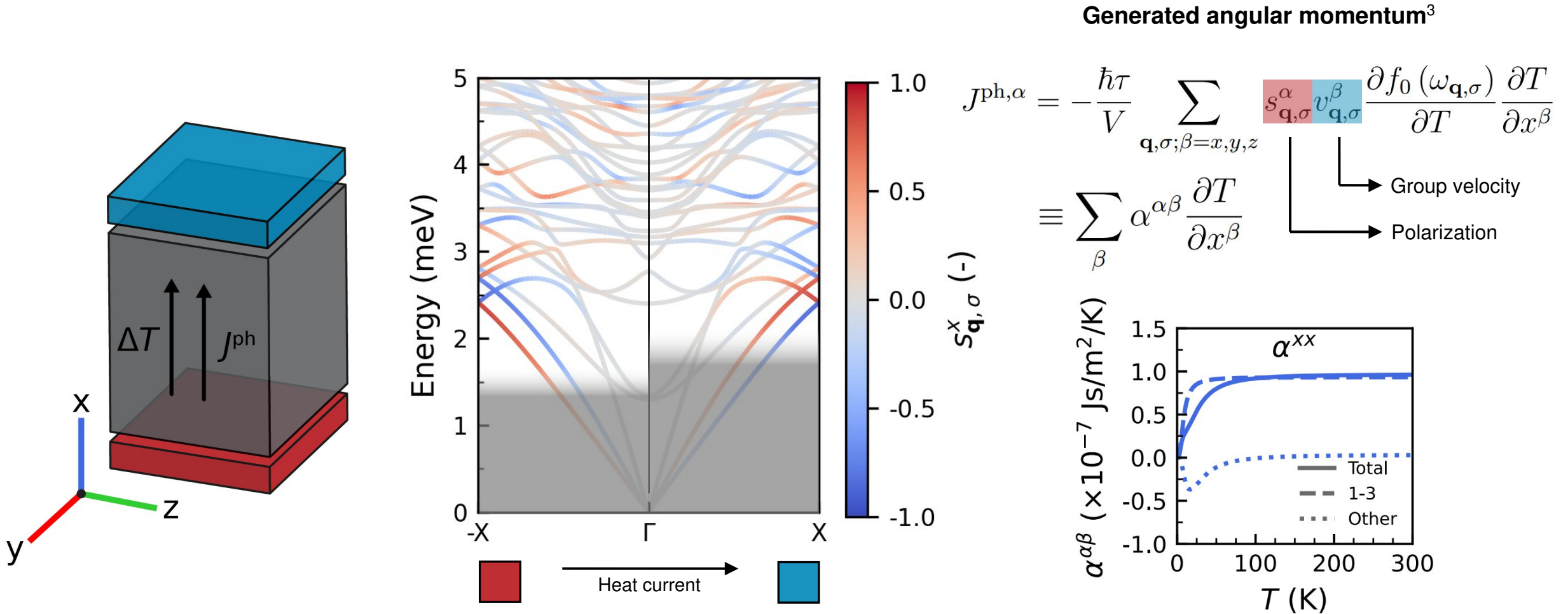
And, are they chiral?



¹ Pols, M. et al. Nano Lett. 25, 10003-10009 (2025).

² Juraschek, D.M. et al. Nat. Phys., 21, 1532-1540 (2025).

As a result: Einstein-de Haas effect!²



¹ Pols, M. et al. Nano Lett. 25, 10003-10009 (2025).

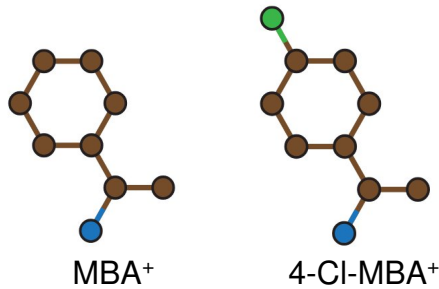
² Zhang, L. and Niu, Q. Phys. Rev. Lett. 112, 085503 (2014).

³ Hamada, M. et al. Phys. Rev. Lett. 121, 175301 (2018).

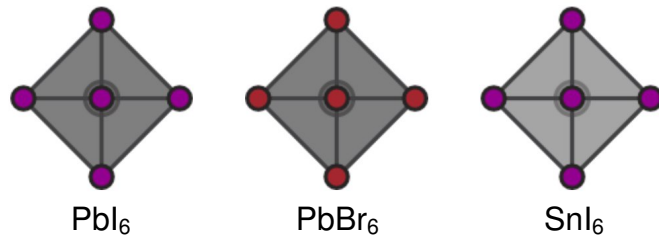
What makes these materials unique?

Tunable chiral properties

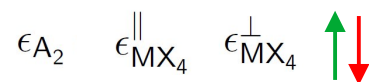
Organic cations¹



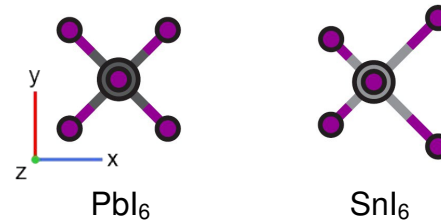
Metal halide octahedra²



Crystal chirality



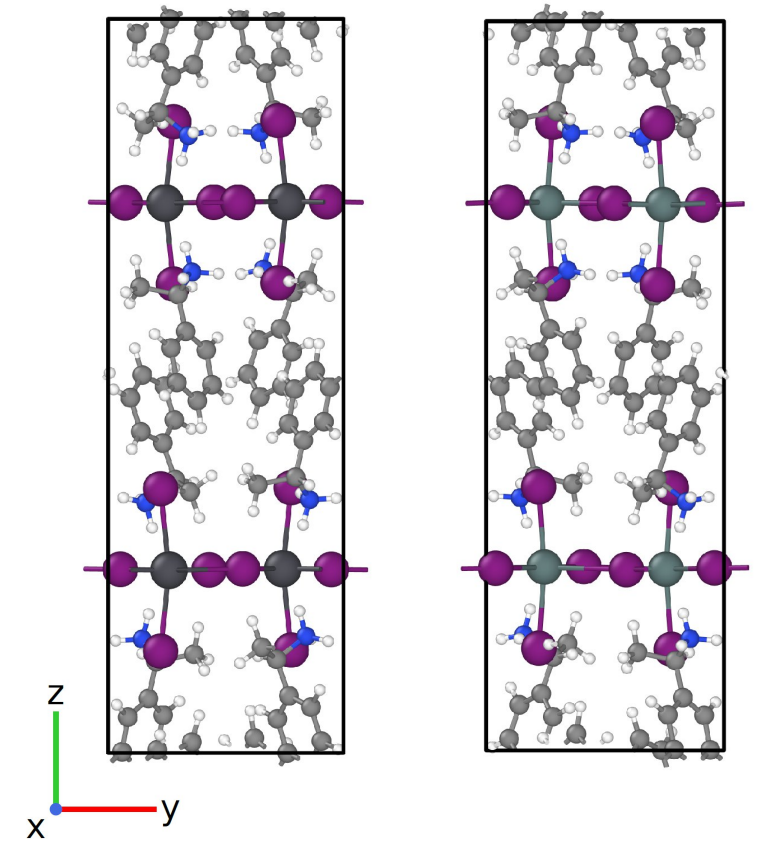
Octahedral distortions
due to lone-pair
around metal ion



Equivalent crystal structures

(S-MBA)₂PbI₄

(S-MBA)₂SnI₄



¹ Pols, M. et al. J. Phys. Chem. Lett. 15, 8057-8064 (2024).

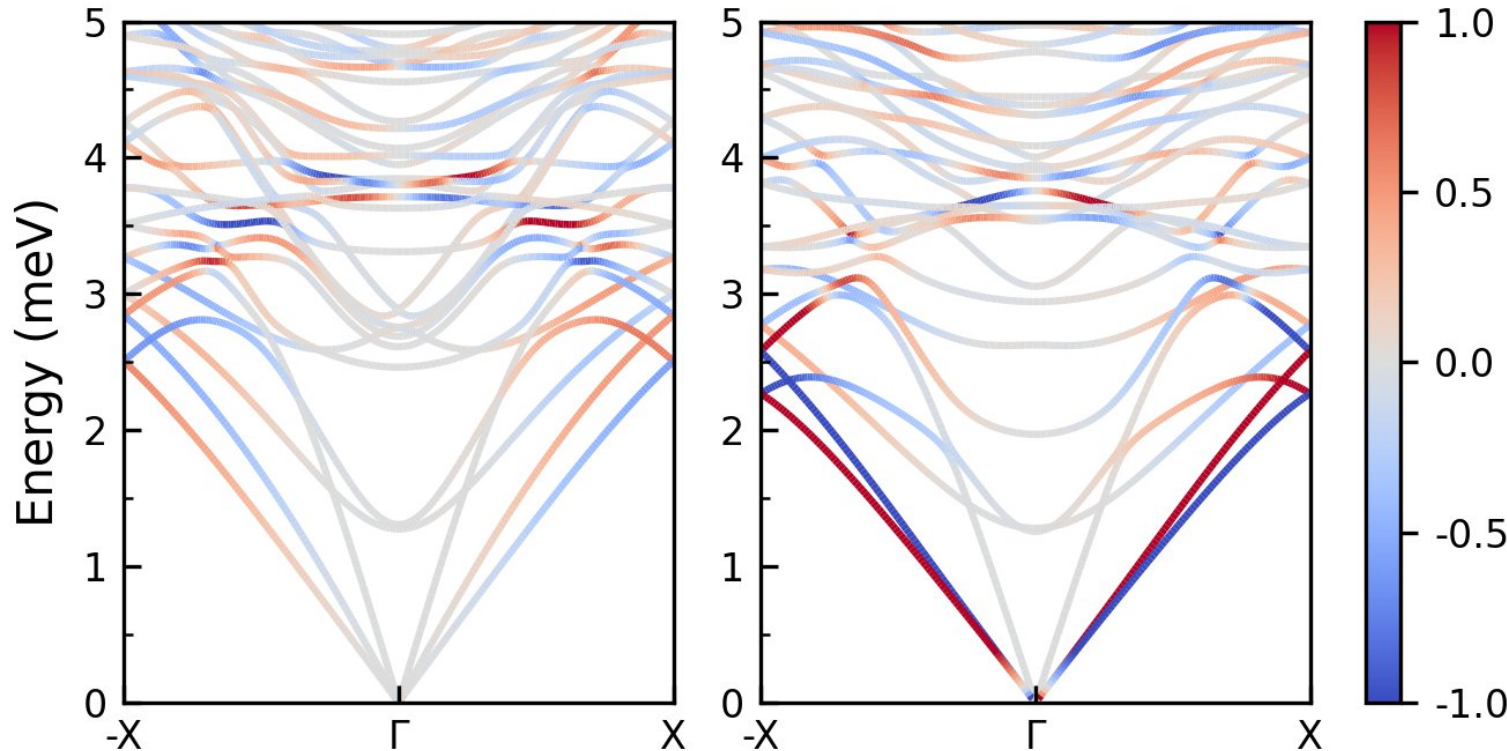
² Pols, M. et al. Phys. Rev. Mater. 9, 113601 (2025).

Chiral properties can also be tuned!

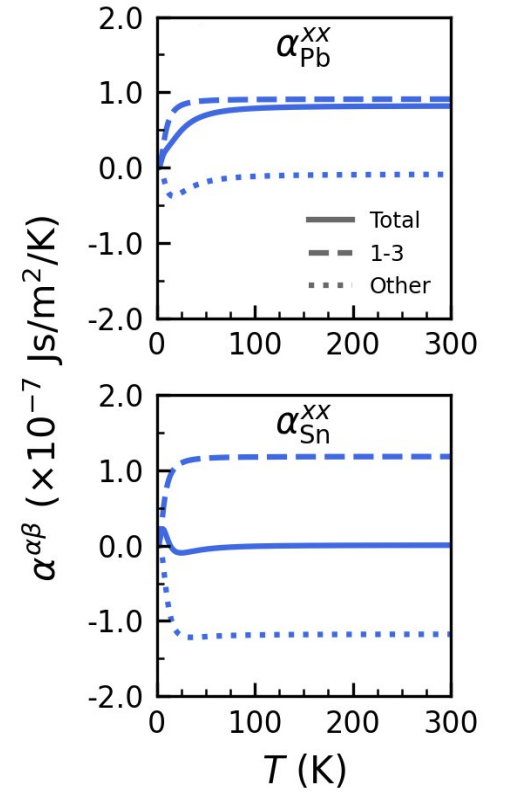
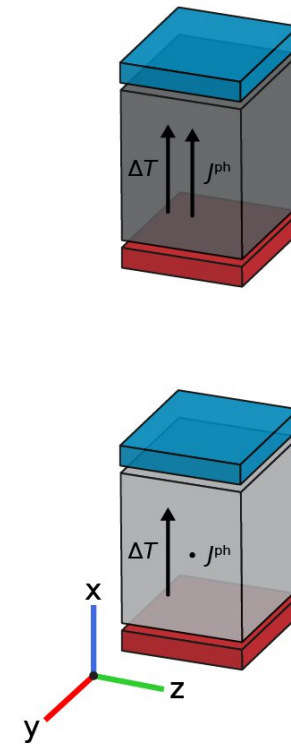
Phonon dispersions¹

(S-MBA)₂PbI₄

(S-MBA)₂SnI₄

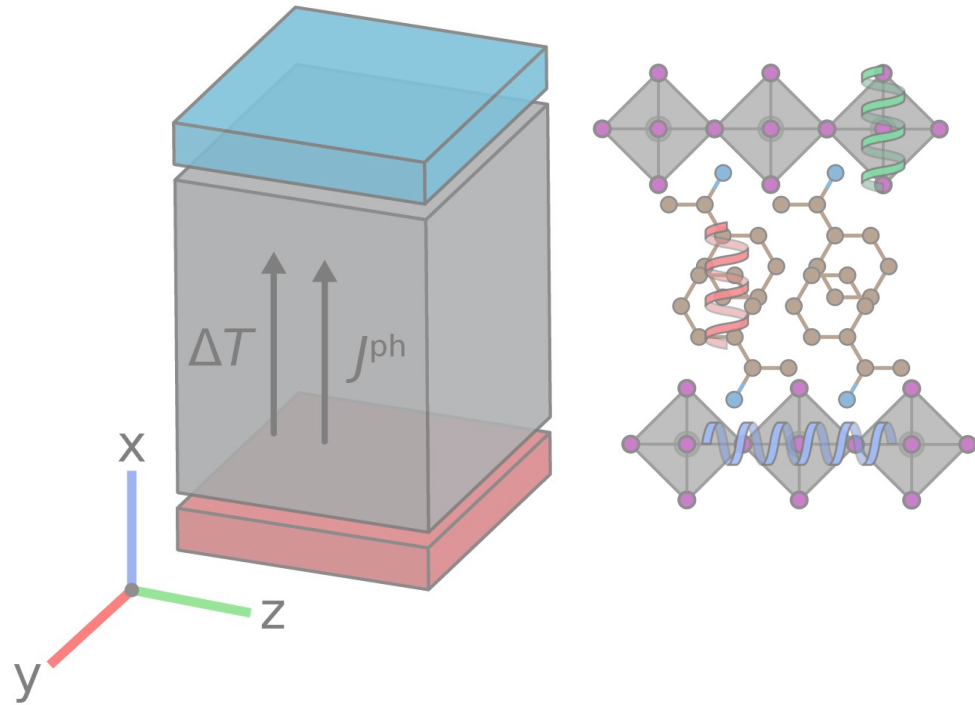


Thermal effects!



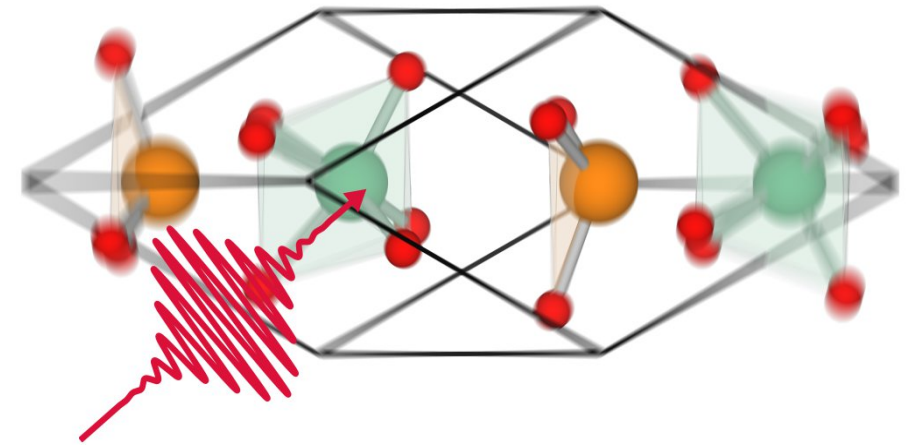
¹ Pols, M. et al. Phys. Rev. Mater. 9, 113601 (2025).

Part 1:



Angular momentum from heat
in 2D halide perovskites

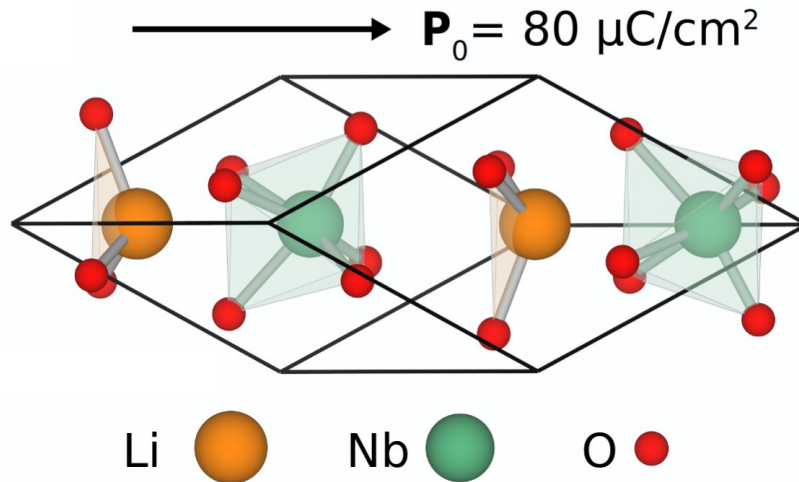
Part 2:



Laser-induced angular
momentum in LiNbO₃

Exciting phonon dynamics with a laser

Ferroelectric LiNbO₃



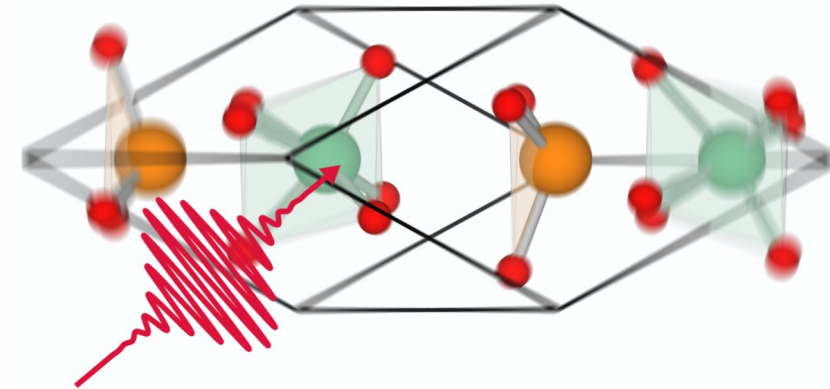
Crystal structure: $R3c$ space group

Phonon modes: A_1, A_2, E modes

Laser-driven functionality:

superconductivity¹, ferroelectricity², magnetism³

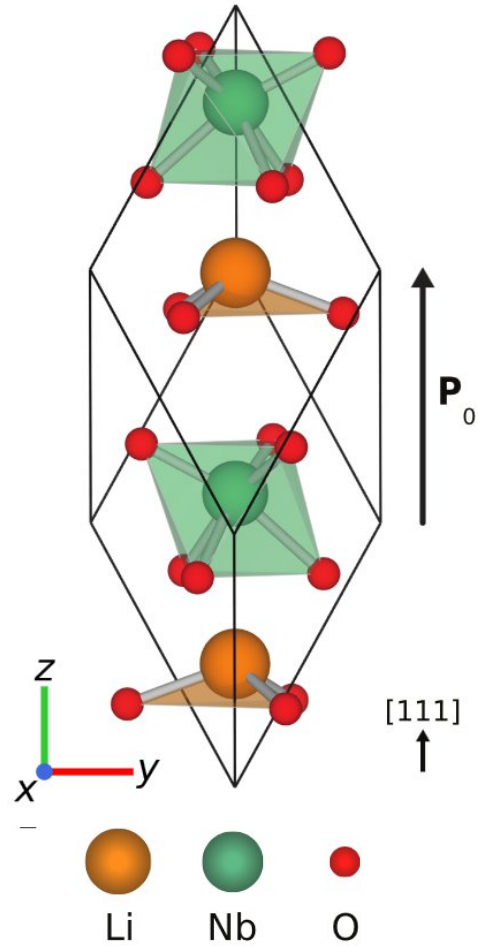
Laser-driven LiNbO₃



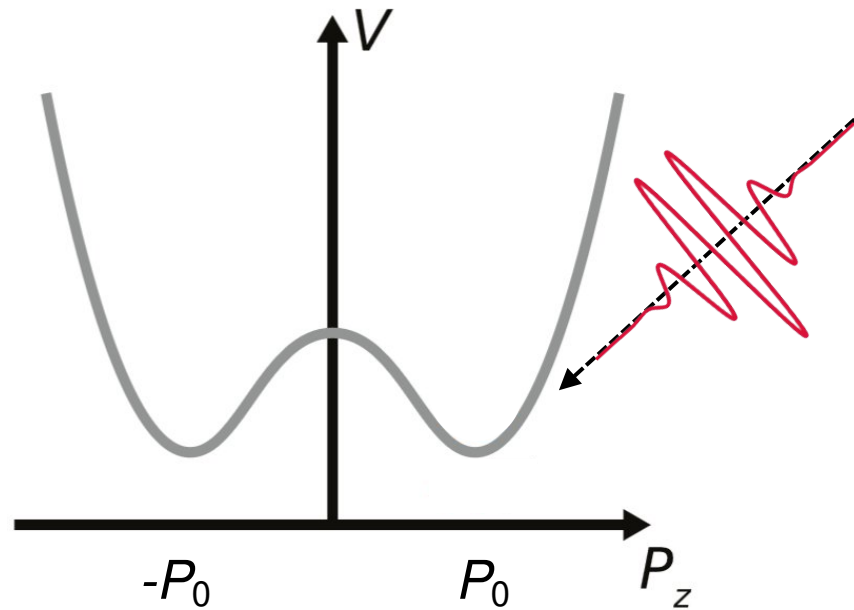
Excitation of resonant modes through
infrared (IR) or **Raman** driving!

Active modes: A_1, E modes

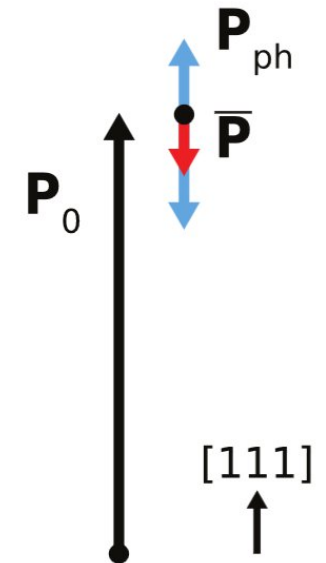
Using the anharmonicity of the phonon modes



Ferroelectric mode
 A_1 mode



Polarization
Magnitude change

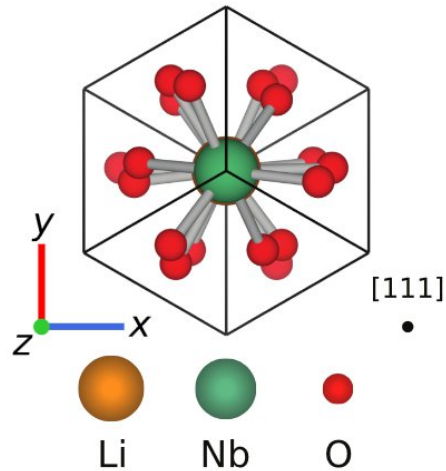


¹ Pols, M. et al. Phys. Rev. Lett. 136, 166801 (2026).

Additionally: making use of in-plane anharmonicity

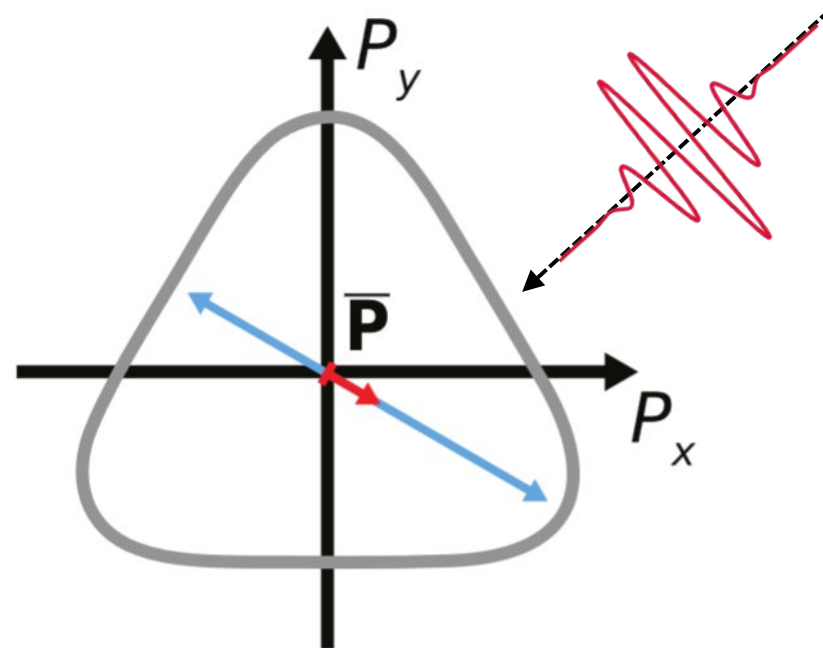
LiNbO₃ (*R3c*)

3-fold rotational symmetry



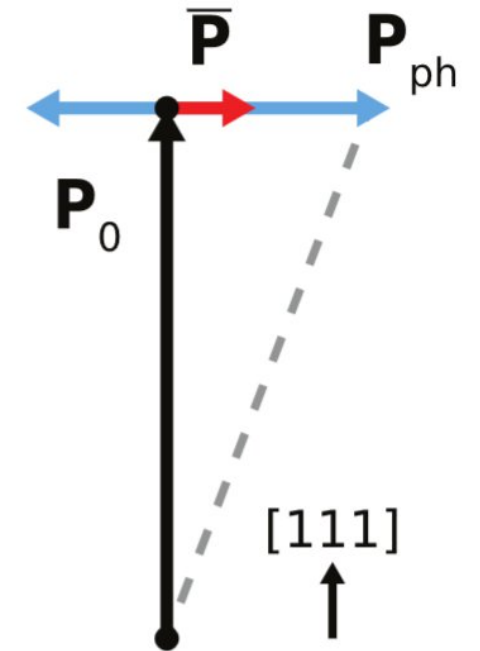
Perpendicular modes

E modes (degen.)



Polarization

Tilting

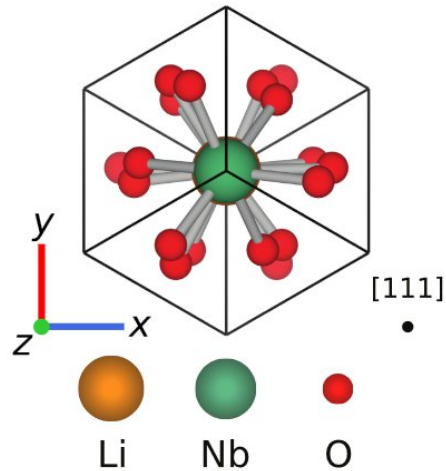


¹ Pols, M. et al. Phys. Rev. Lett. 136, 166801 (2026).

Angular momentum yields magnetization!

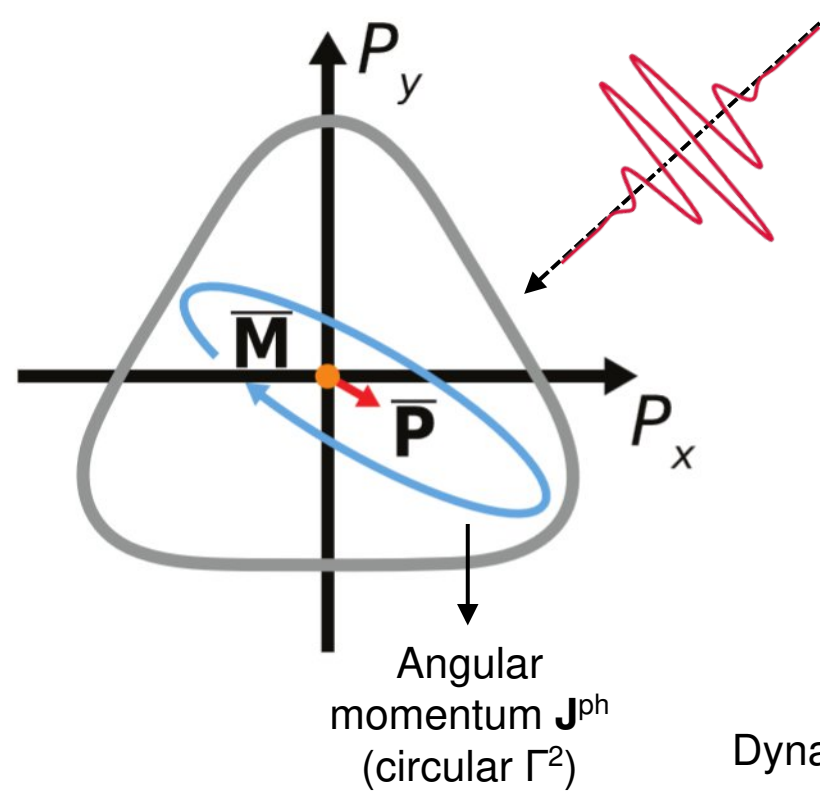
LiNbO₃ (*R3c*)

3-fold rotational symmetry



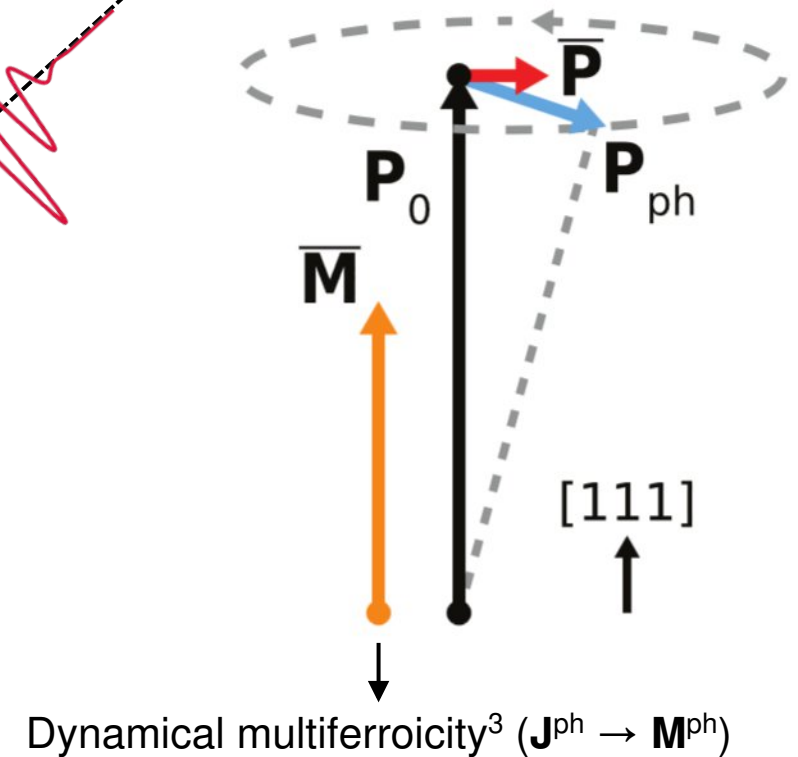
Perpendicular modes

E modes (degen.)



Polarization

Tilting and precessing!

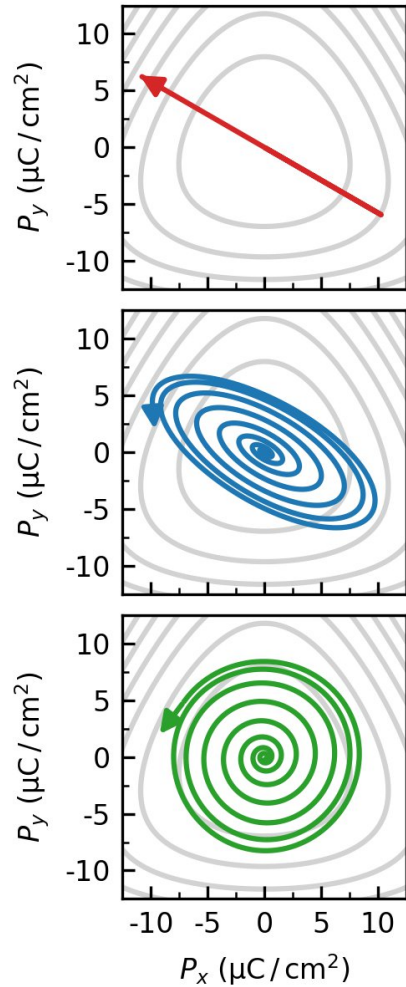


¹ Pols, M. et al. Phys. Rev. Lett. 136, 166801 (2026).

² Juraschek, D.M. et al. Nat. Phys., 21, 1532–1540 (2025).

³ Juraschek, D.M. et al. Phys. Rev. Mater. 1, 014401 (2017).

Polarization and magnetization dynamics

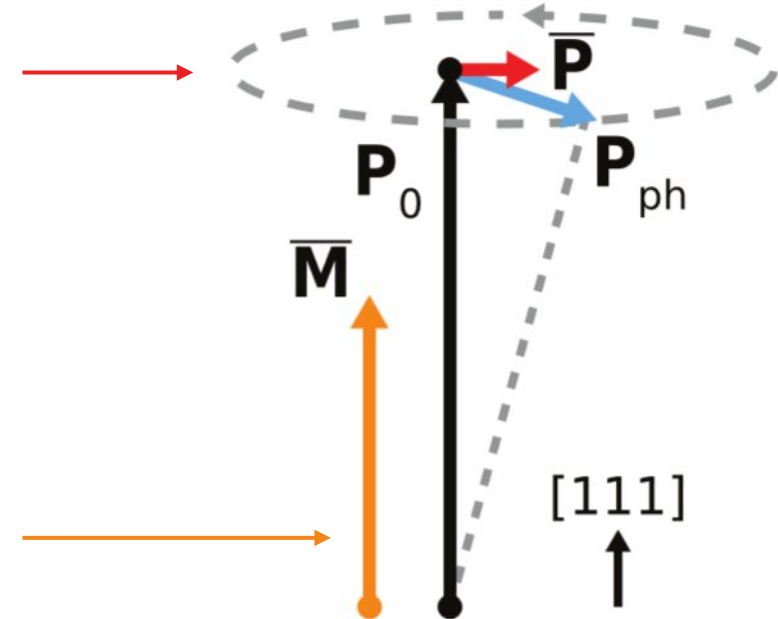


Relative magnitudes

$$\mathbf{P}_0 = 80.0 \mu\text{C}/\text{cm}^2$$

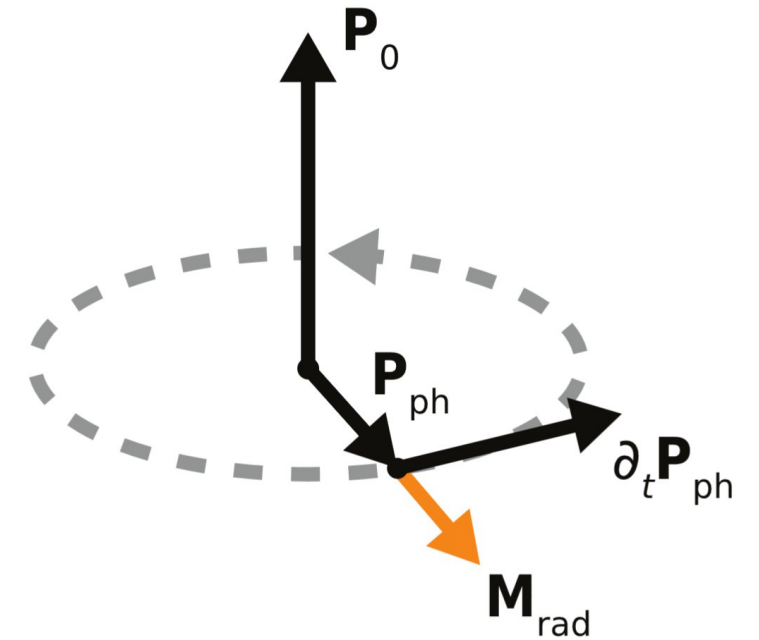
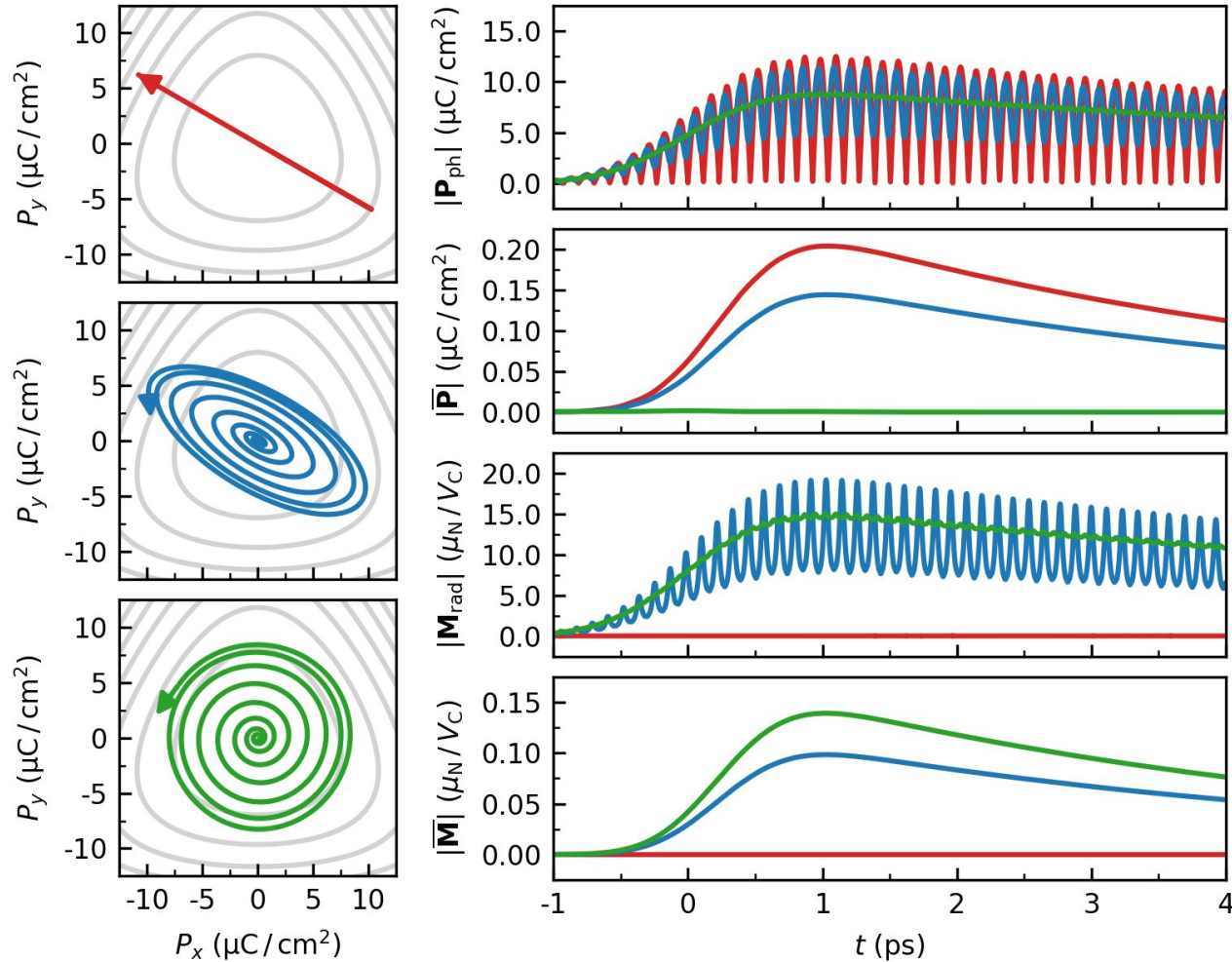
$$\bar{\mathbf{P}} = 0.15 \mu\text{C}/\text{cm}^2$$

$$\bar{\mathbf{M}} = 0.10 \mu\text{N}/V_c$$



¹ Pols, M. et al. Phys. Rev. Lett. 136, 166801 (2026).

Even multipolar order!



Radial magnetization with multipolar order¹

¹ Pols, M. et al. Phys. Rev. Lett. 136, 166801 (2026).

Acknowledgements

Shuxia Tao, Geert Brocks, Sofia Calero (TU/e)

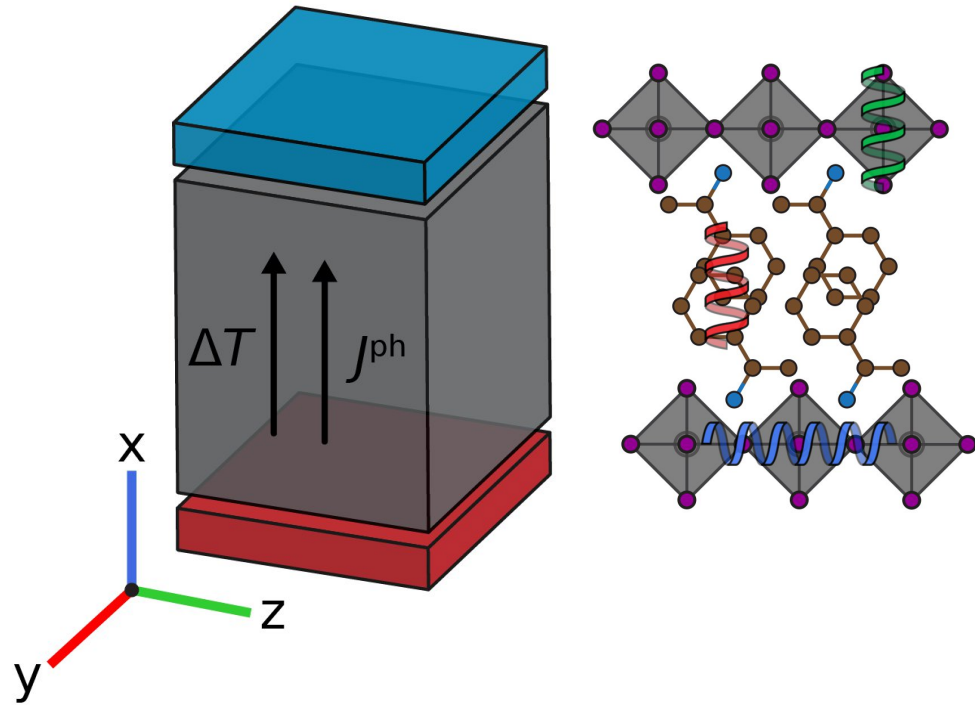


Dominik Juraschek (Juraschek Lab, TU/e)

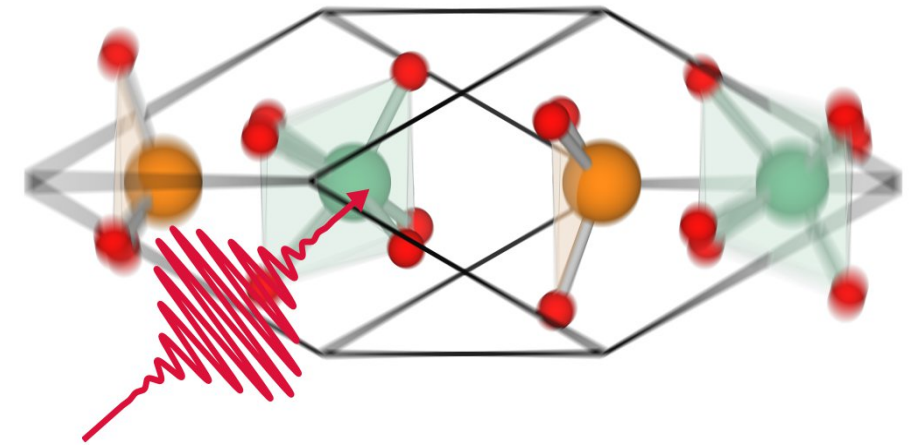
Nicola Spaldin (Materials Theory, ETH Zurich)



Heat and light-driven angular momentum



2D halide perovskites with tunable angular momentum arising from heat!



Excitations with a net polarization and magnetization in LiNbO_3 !

Angular momentum in perovskites through heat and light

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