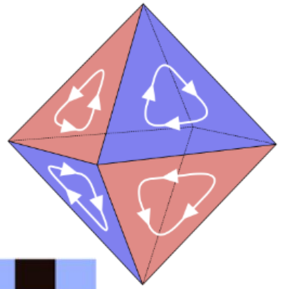
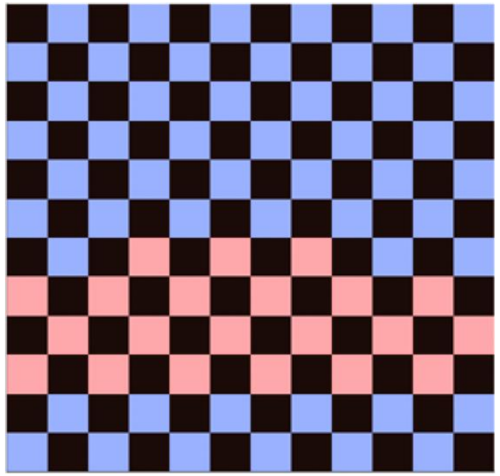
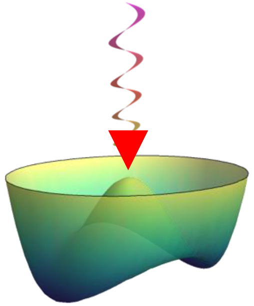
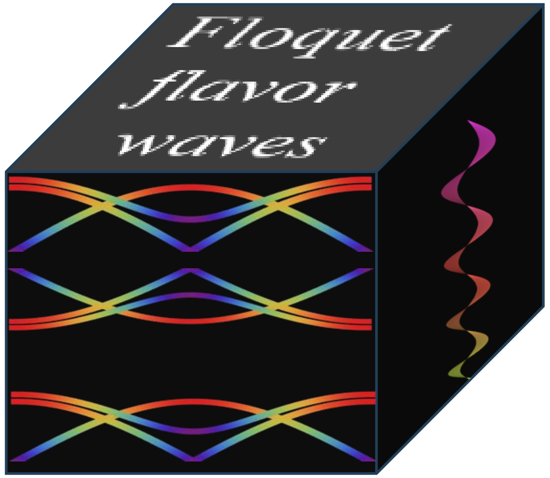
Strain and phonons as probes of hidden order in quantum materials





Arun Paramekanti
(University of Toronto)





Mainz

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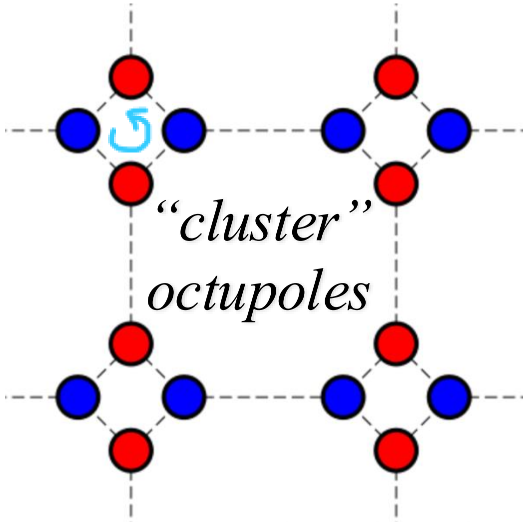
“Cluster multipoles” in quantum materials

Spin Altermagnets

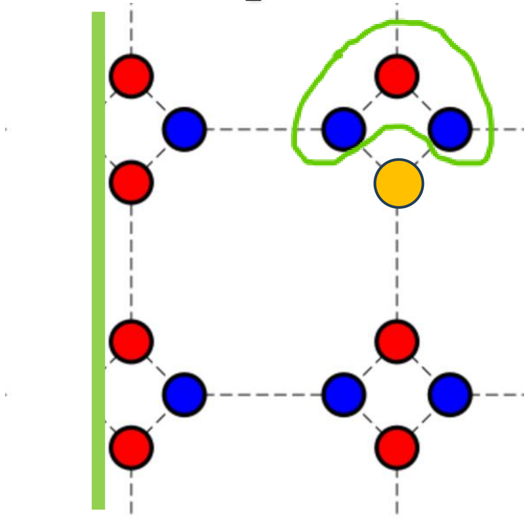
Kivelson, et al Rev. Mod. Phys. (2003)
Naka, et al, Nat Comm (2019)
Smejkal, Sinova, Jungwirth PRX (2022)

- Ferro-spin order (translationally invariant)
- Zero net magnetization (protected by point group symmetry)
- Spin-split electronic bands of interest for spintronics

d-wave ferromagnet

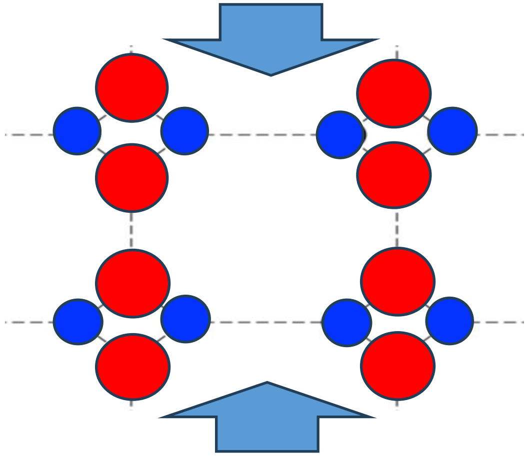


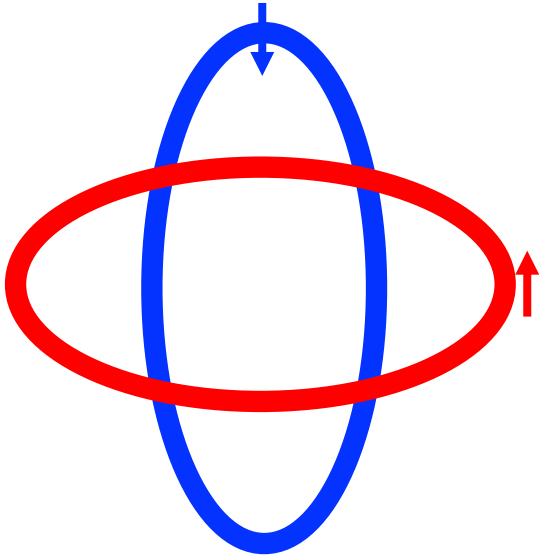
Bulk impurity dipole moments



Surface magnetization

Piezomagnetism





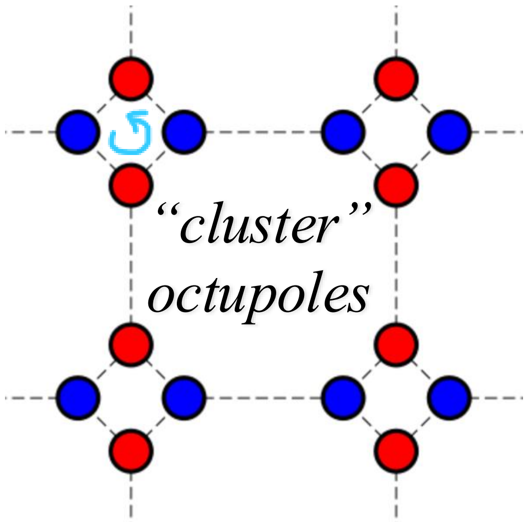
Metallic regime

“Cluster multipoles” in quantum materials

For intermediate fillings (1/4 < n < 3/4)

- Physics dominated by effective px, py “molecular orbitals”
- Quasi-1D p-bands gives rise to strong van Hove singularity near band bottom
- Weak interactions favor metallic altermagnet
- “Crossed-wires” each being Ferro but coupled AF

d-wave ferromagnet



Altermagnetism and superconductivity in a multiorbital $t - J$ model

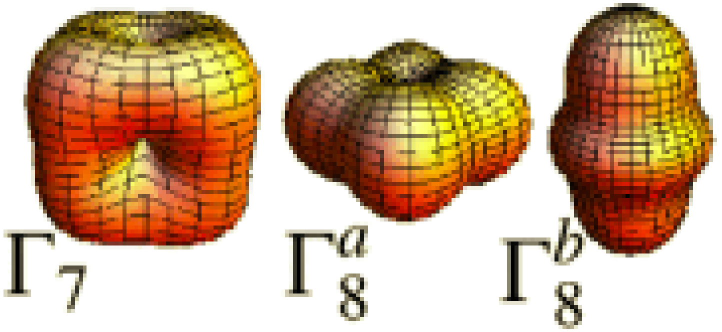
Anjishnu Bose*, Samuel Vadnais*, and Arun Paramekanti†

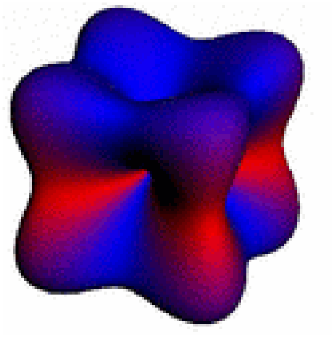
Show more

Phys. Rev. B **110**, 205120 – **Published 8 November, 2024**

“Atomic multipoles” in quantum materials

Multipolar magnetism in spin-orbit coupled materials
Strong local entanglement of spins and orbital degrees of freedom

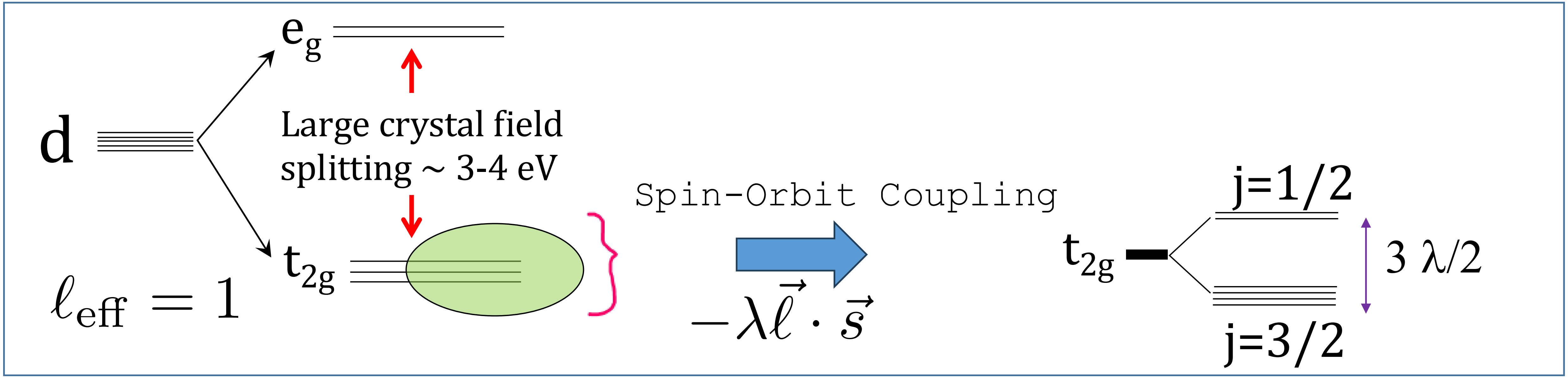




- Phase transition easily seen in thermodynamics (e.g., specific heat)
- Precise nature of hidden order: Very hard to unravel
- Well known f-electron examples: URu_2Si_2, NpO_2, PrV_2Al_{20}

- Similar difficulties arise in probing quantum spin liquids but with spatially extended entanglement – here it is “local entanglement” of many degrees of freedom (like “synthetic dimension” of cold atoms)

Multipoles in d-orbital quantum materials



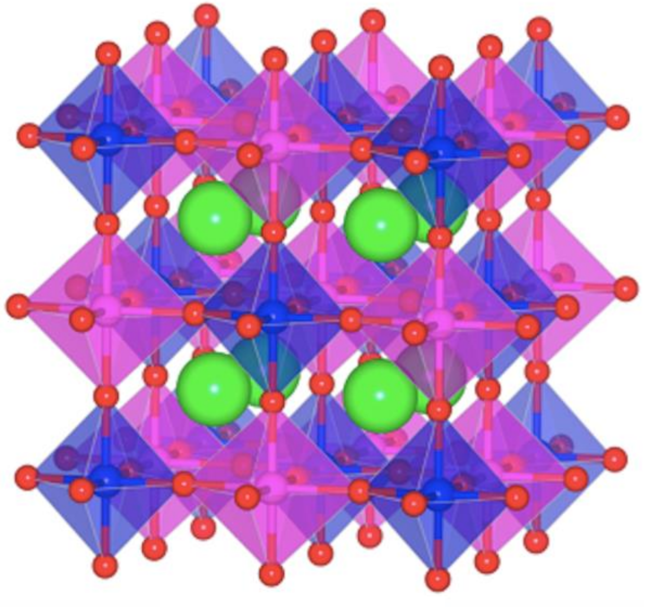
Adding local Coulomb interaction:

d^1	d^2	d^3	d^4	d^5
j=3/2	J=2	J=3/2	J=0	j=1/2

Candidates for "atomic multipolar" order: Quadrupoles, Octupoles, etc.

Can strain reveal spin-orbit coupled multipolar orders in solids?

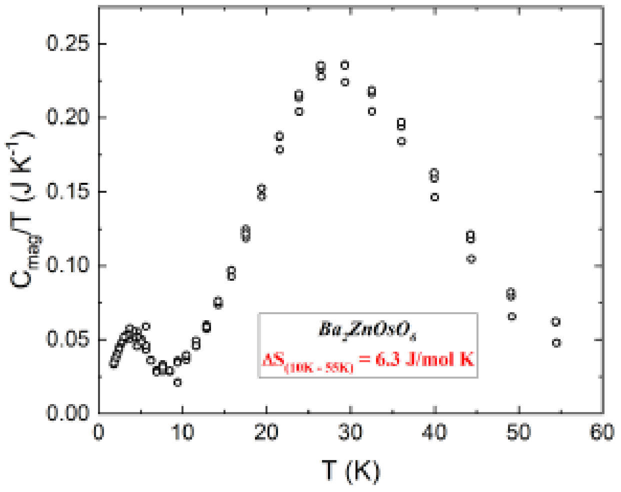
$5d^2$ Double Perovskites: Puzzles around hidden “J=2” magnetism



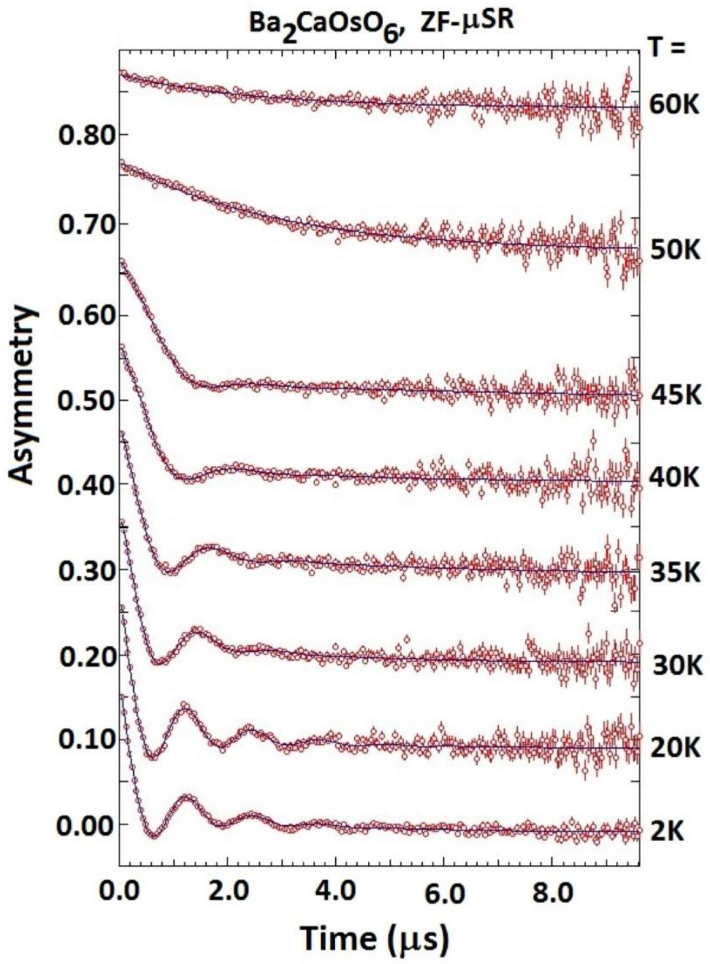
Ba_2MgOsO_6
Ba_2CaOsO_6
Ba_2ZnOsO_6

C. M. Thompson, et al, JPCM 2014
C. A. Marjerrison, et al, PRB 2016

Measured Entropy ~ R ln2
Not 2J+1=> R ln5



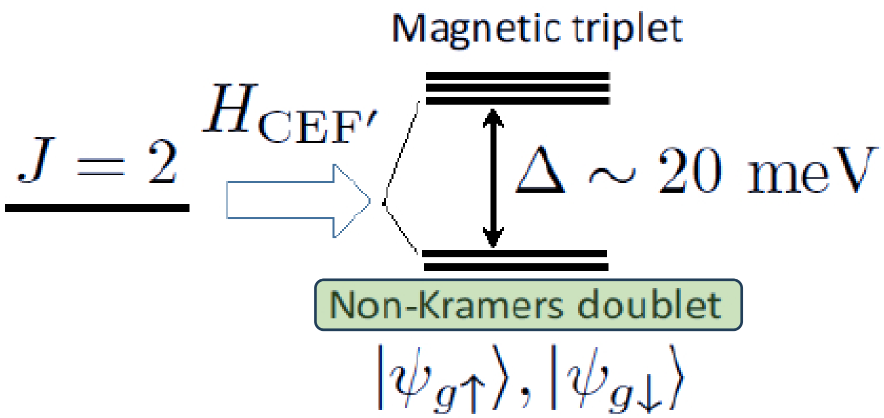
μSR: Time reversal broken
Very weak internal field



Robust Tc, yet no sign of magnetic order in neutron diffraction

System	T^*	θ_{CW}	a (Å)	Ref.	μ_{ord}
Ba_2CaOsO_6	49	–156.2(3)	8.3456	[13]	$< 0.11\mu_B$
Ba_2MgOsO_6	51	–120(1)	8.0586	[15]	$< 0.13\mu_B$
Ba_2ZnOsO_6	30	–149.0(4)	8.0786	[15]	$< 0.06\mu_B$

$5d^2$ Double Perovskites: Potential resolution of puzzles



$$|\psi_{g,\uparrow}\rangle = |0\rangle$$
$$|\psi_{g,\downarrow}\rangle = \frac{1}{\sqrt{2}}(|2\rangle + |-2\rangle)$$

$$\tau_x \propto J_x^2 - J_y^2$$
$$\tau_z \propto 3J_z^2 - J^2$$
→ Quadrupolar charge density $\sim E_g$

$$\tau_y \propto \mathrm{Sym}\,[J_x J_y J_z]$$
→ Ising magnetic octupole

- ✓ Entropy: Rln2 (unless very high T)
- ✓ No dipole order: No dipole matrix elements
- ✓ Spin gap: Doublet-Triplet gap
- ✓ T-breaking order: Hidden octupolar order!

First example of d-orbital octupolar order

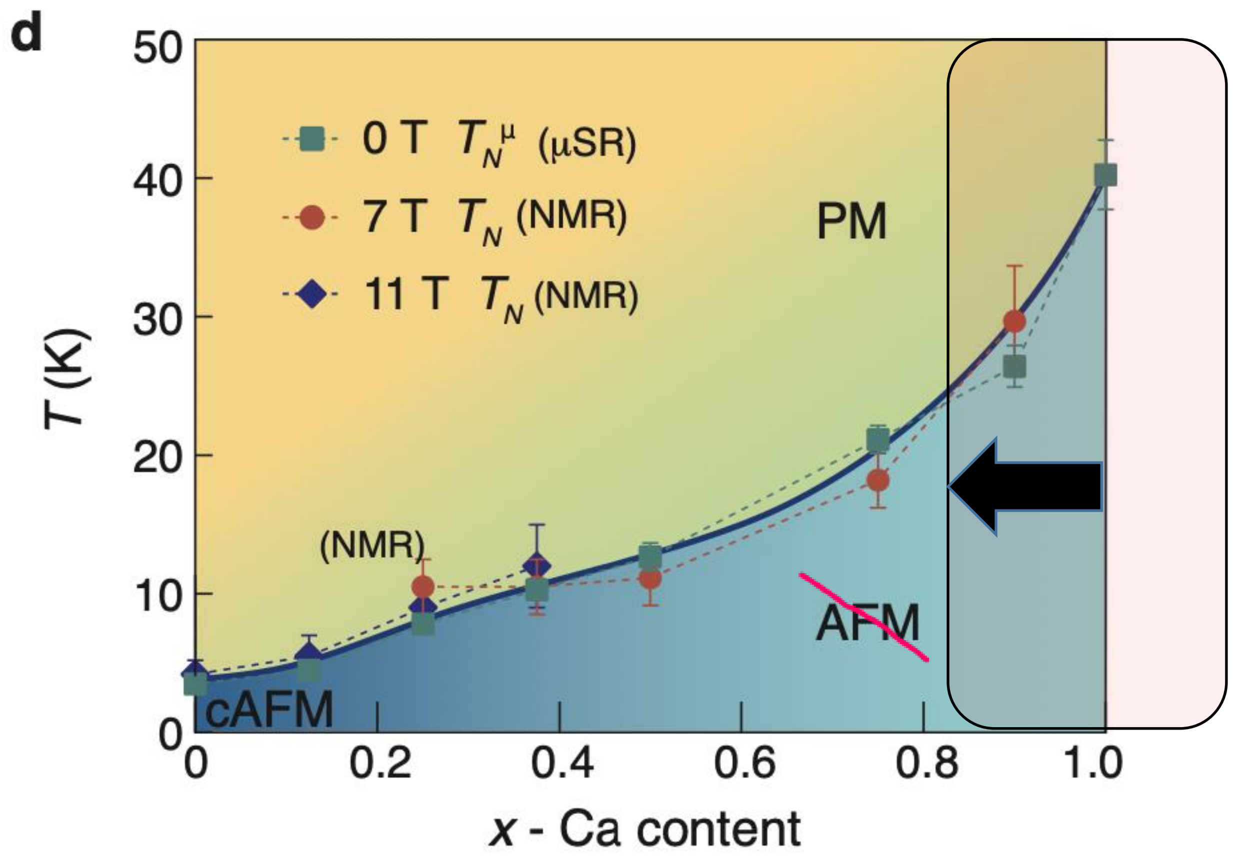
Phenomenology and microscopics

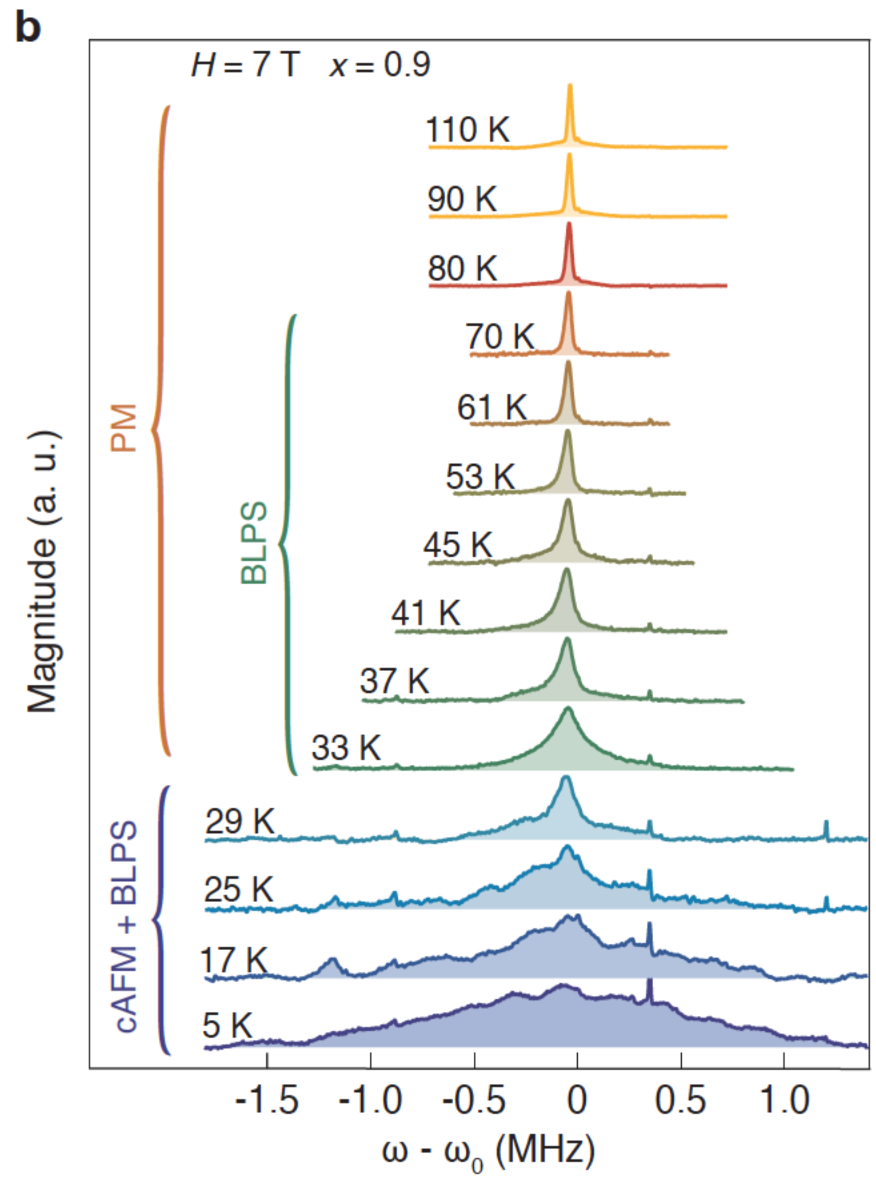
Data: D. D. Maharaj, et al PRL 124, 087206 (2020)
Proposal: Paramekanti, et al, PRB 101, 054439 (2020)
muSR: S. Voleti, et al, AP, PRB 101, 155118 (2020)
LDA+DMFT: Pourovskii, Mosca, Franchini, PRL (2021)
Tight binding + ED: Voleti, Haldar, Paramekanti, PRB (2021)
Tight binding + ED: Churchill, Kee, PRB (2022)

R. Jaschke-Ubriero, et al, 2503.10797 (“atomic altermagnetism”)

Impurity NMR

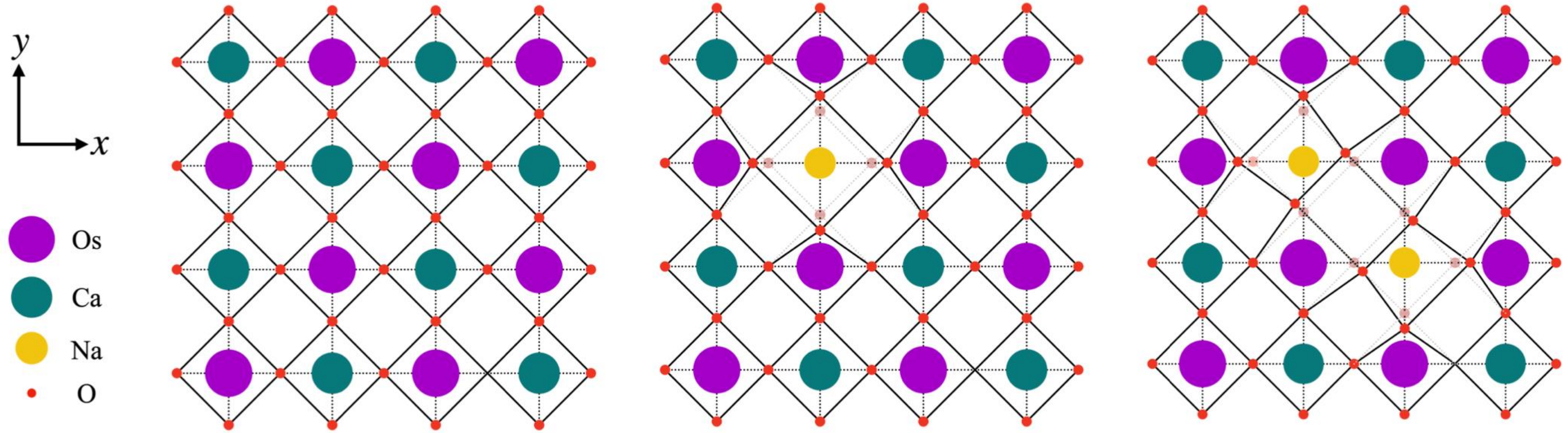
^{23}Na NMR studies on $Ba_2Na_{1-x}Ca_xOsO_6$
R. Cong et al, Phys Rev Materials (2023)



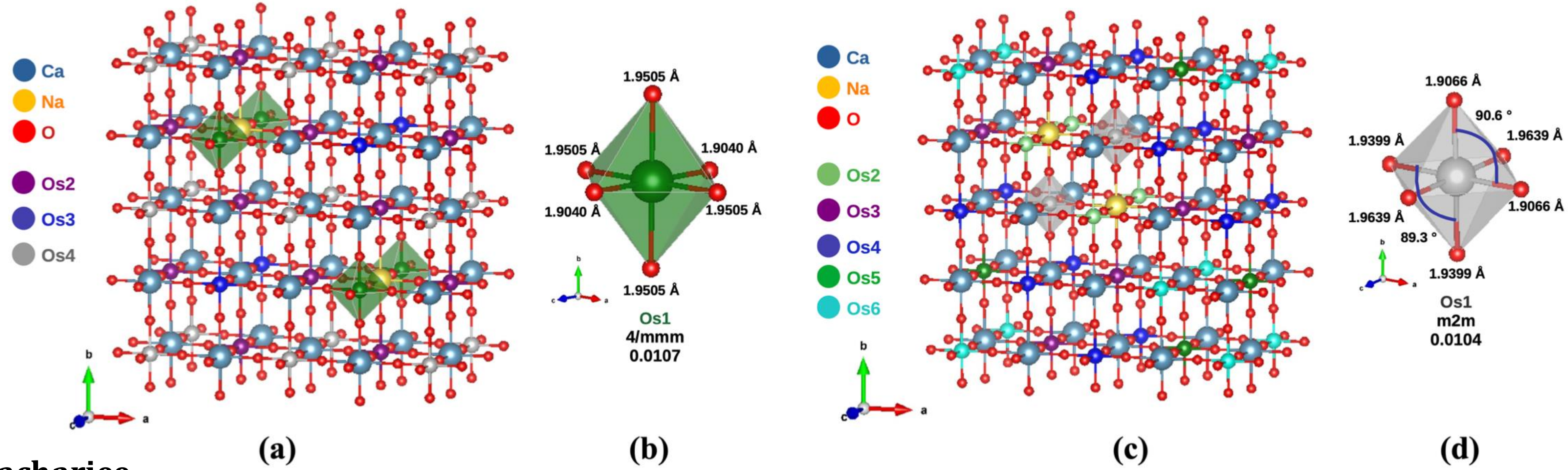


Impact of impurities: Local strain fields

Schematic



DFT study



S. Voleti, K. Pradhan, S. Bhattacharjee, T. Saha-Dasgupta, AP, npjQM (2023)

Impact of impurities: Local strain fields

Favor local quadrupole order (at all temperatures)

$$\delta H_x = -d_1 \int d^3\mathbf{r} (\varepsilon_{xx}(\mathbf{r}) - \varepsilon_{yy}(\mathbf{r}))\tau_x(\mathbf{r})$$

$$\delta H_z = -d_2 \int d^3\mathbf{r} \frac{1}{\sqrt{3}} (2\varepsilon_{zz}(\mathbf{r}) - \varepsilon_{xx}(\mathbf{r}) - \varepsilon_{yy}(\mathbf{r}))\tau_z(\mathbf{r})$$

Strain splits the non-Kramers doublet

Induce parasitic dipole order (in the octupolar ordered state, T < Tc)

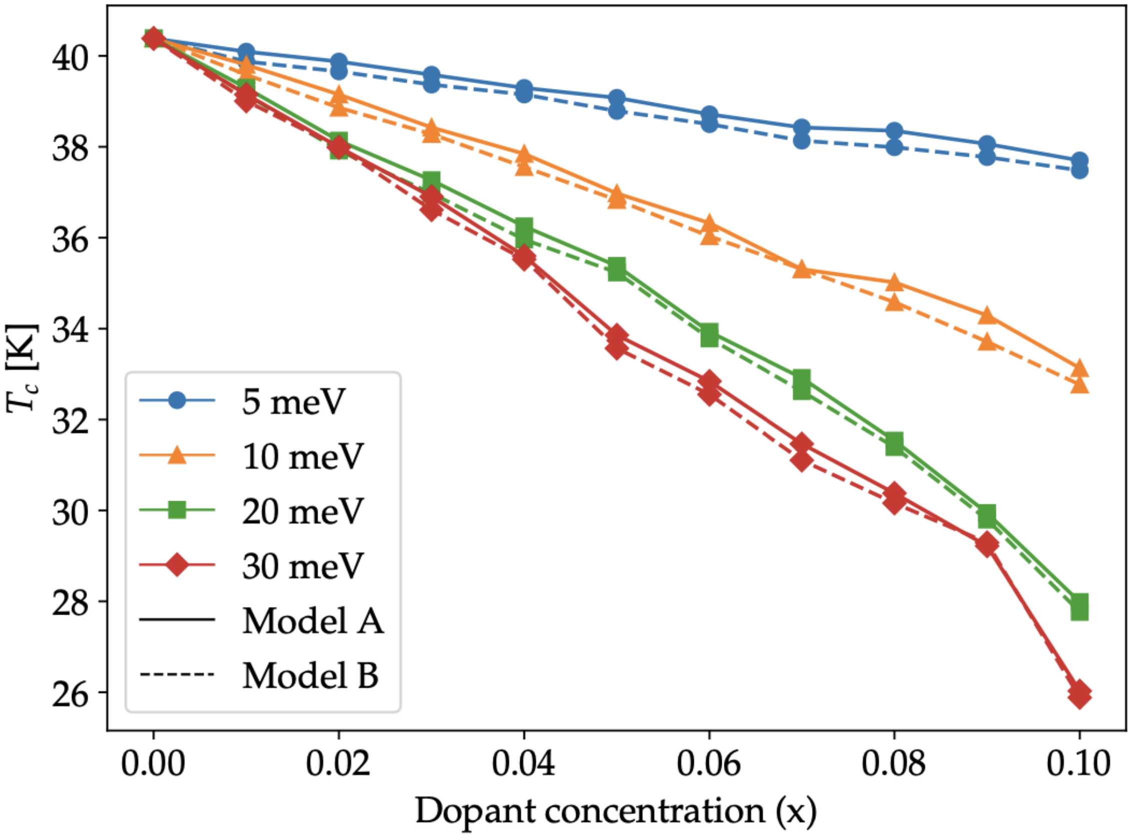
$$\delta H_{\text{od}} = -d_3 \int d^3\mathbf{r} \, [\varepsilon_{xy}(\mathbf{r}) Q_{xy}(\mathbf{r}) + \varepsilon_{yz}(\mathbf{r}) Q_{yz}(\mathbf{r}) + \varepsilon_{xz}(\mathbf{r}) Q_{xz}(\mathbf{r})]$$

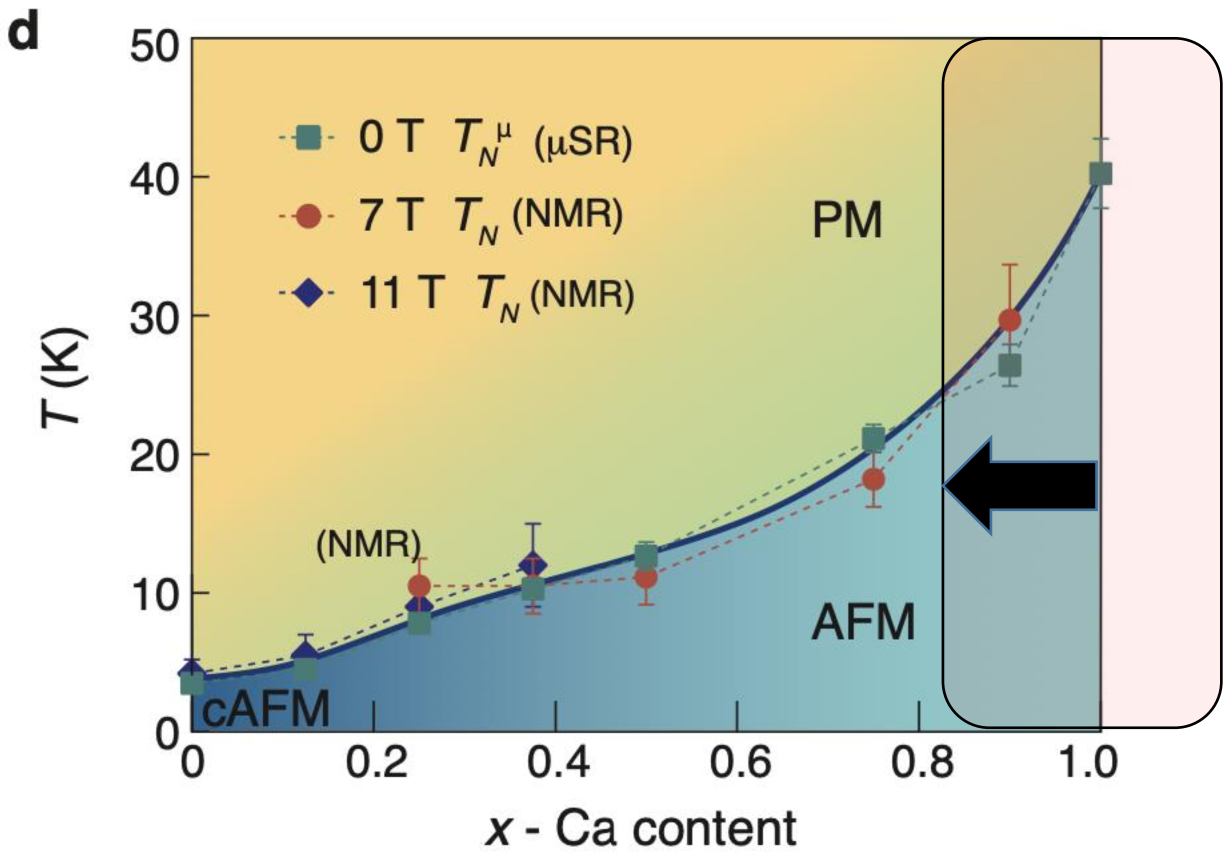
$$\langle \mathbf{J}(\mathbf{r}) \rangle = \frac{2d_3}{\Delta} \boldsymbol{\varepsilon}(\mathbf{r}) \langle \tau_y(\mathbf{r}) \rangle$$

Random strain induces random dipole moments

S. Voleti, K. Pradhan, S. Bhattacharjee, T. Saha-Dasgupta, AP, npjQM (2023)

Local pinning of quadrupoles kills octupolar Tc

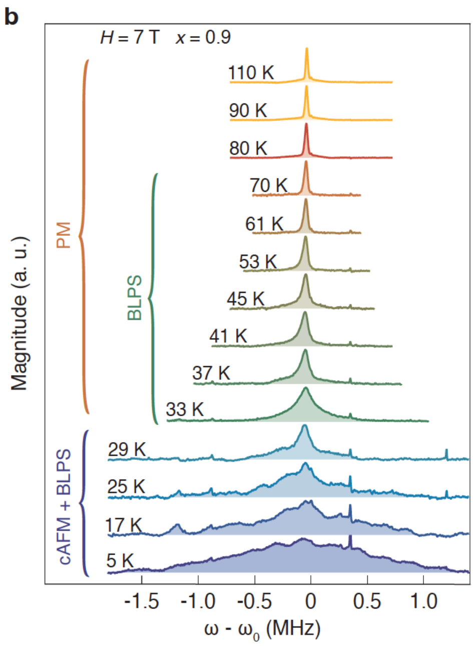


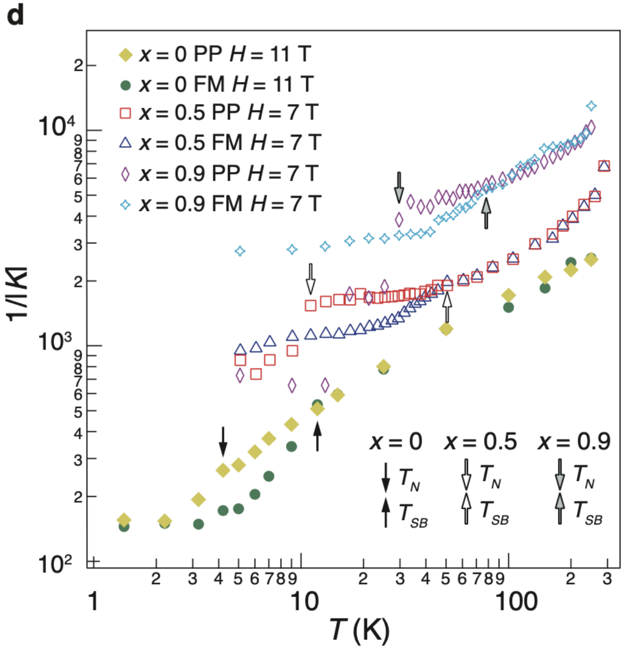


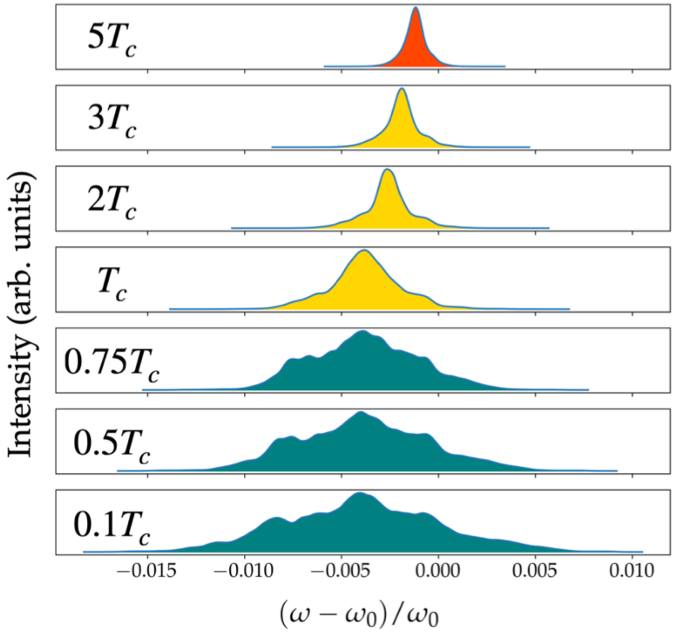
NMR: Probe of octupolar ordering via parasitic dipoles

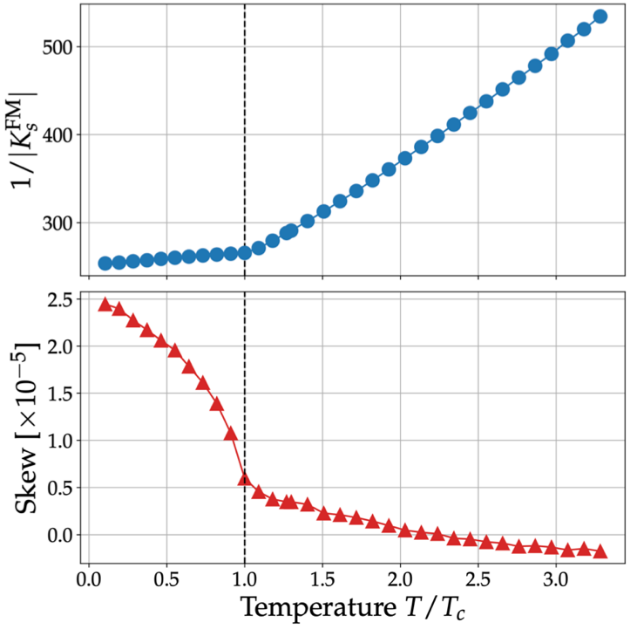
$$H_{\mathrm{Na}} = -\sum_{i} \gamma(i)\, \hat{\mathbf{I}}(i) \cdot \mathbf{B} + g_1 \sum_{\langle i,j \rangle} \hat{\mathbf{I}}(i) \cdot \tilde{\boldsymbol{\varepsilon}}_j \langle \tau_j^y \rangle + \sum_{\langle i,j \rangle} \hat{\mathbf{Q}}^{\mathrm{Na}}(i) \cdot \mathbb{A}_{ij} \cdot \langle \mathcal{Q}_j \rangle$$

$$\gamma(i) = \gamma^{\mathrm{Na}} \left(1 + g_0 \sum_{j \in \mathrm{nbr}(i)} \langle \mathcal{Q}_j^{A_{1g}} \rangle \right)$$



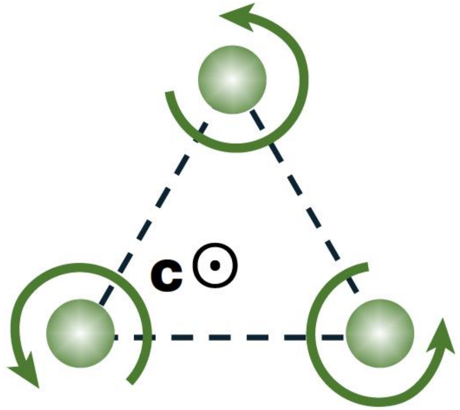


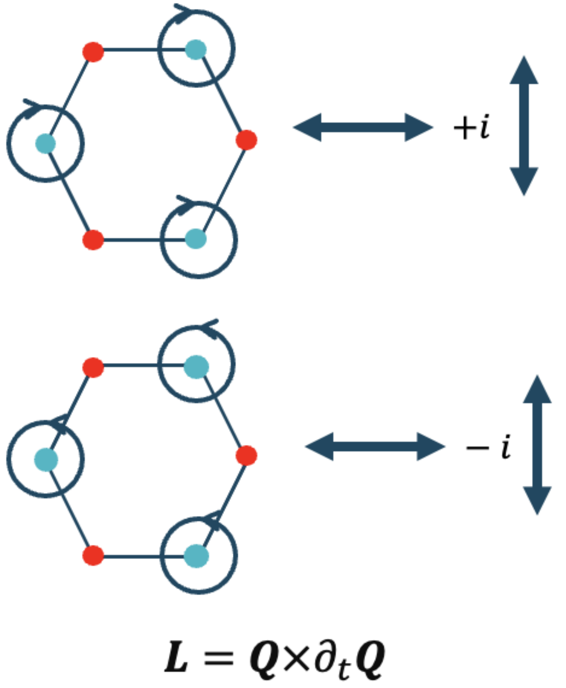




Can phonons reveal spin-orbit coupled multipolar orders in solids?

Phonon angular momentum

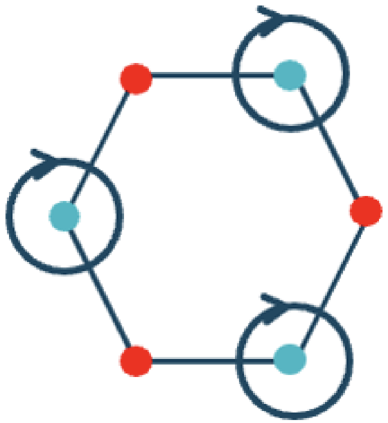




- Phonons: Quantized lattice vibrations
- Atoms in a solid can exhibit circular motion
- Phonons can carry nonzero angular momentum

L. Zhang, Q. Niu, PRL 115, 115502 (2015)
H. Zhang, et al, Nat. Phys. 21, 1387 (2025)
D. Juraschek, et al, Nat. Phys. (Perspective) 21, 1532 (2025)

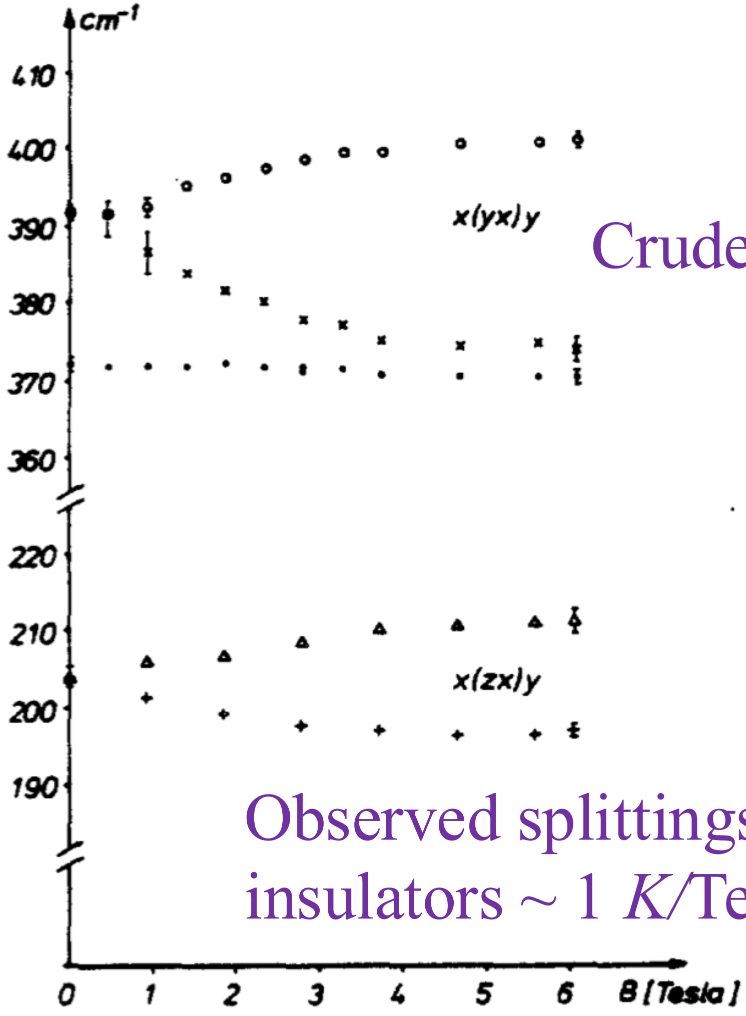
Phonon magnetic moment



Expectation:

$$\mu_{\rm ph} = \frac{Q\hbar}{2M} \sim \mu_B \frac{m_e}{M}$$

Zeeman splitting ~ 1 *mK*/Tesla?



Crudely: 1cm^{-1} ~ 1 *K*

Observed splittings in magnetic insulators ~ 1 *K*/Tesla

Raman scattering on CeF_3

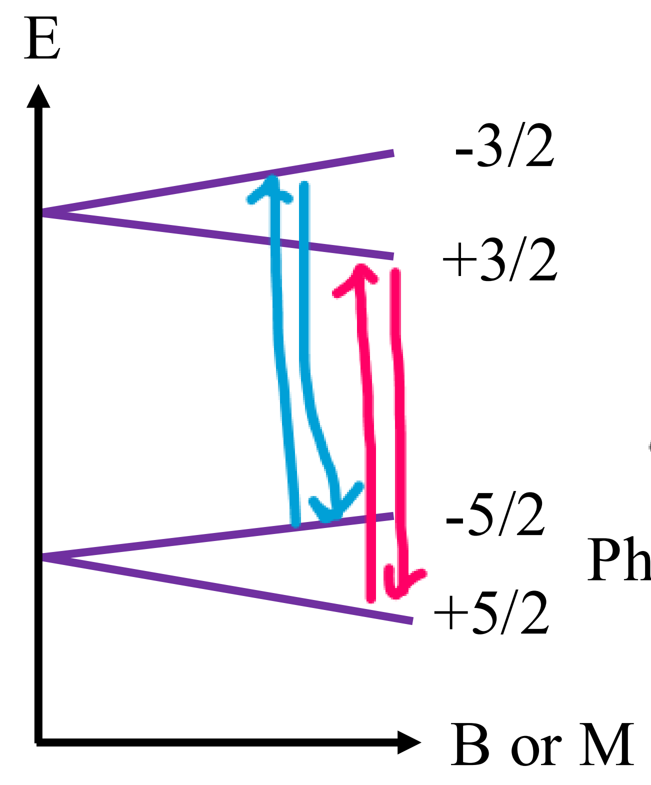
G. Schaack, Sol. State Comm. 15, 505 (1975)

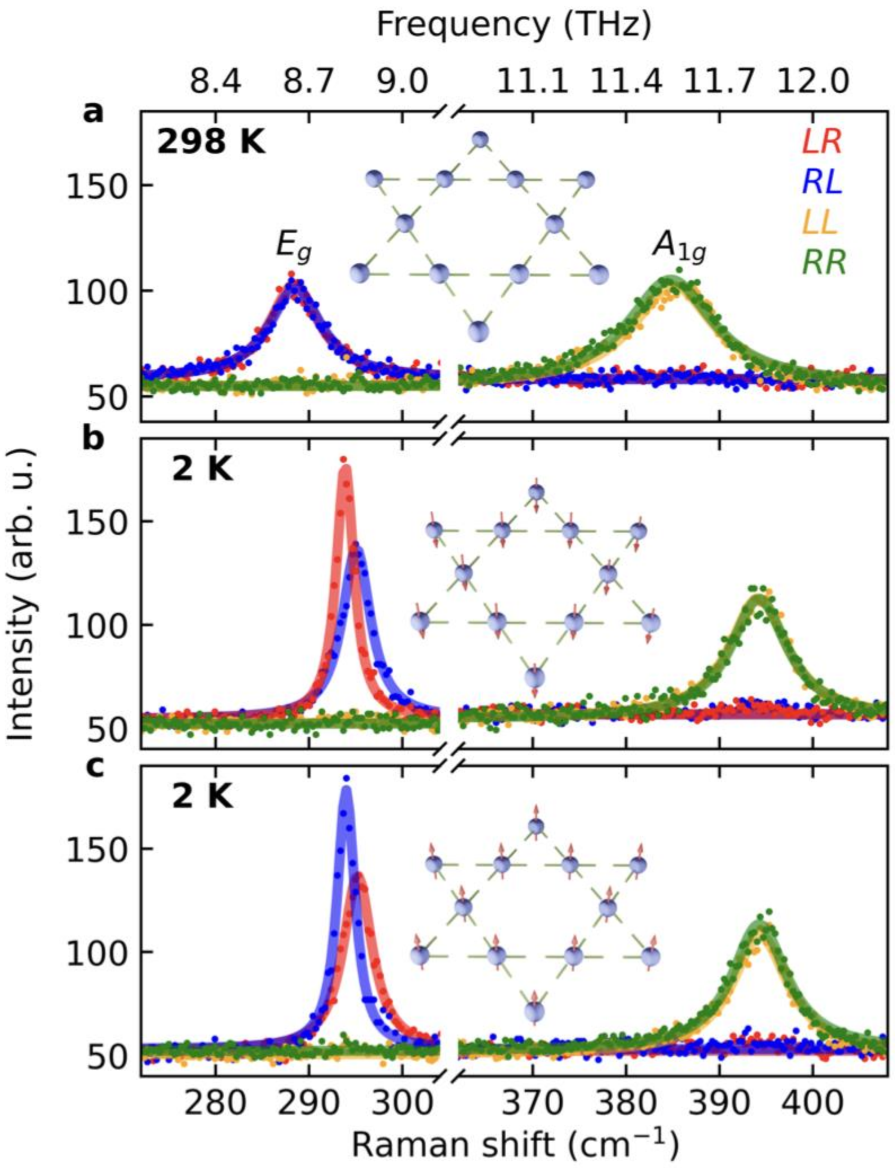
Theory:

P. Thalmeier and P. Fulde, Z. Physik B 26, 323 (1977)

S. Chaudhary, et al, PRB (2024)

Magnetic moment of phonons





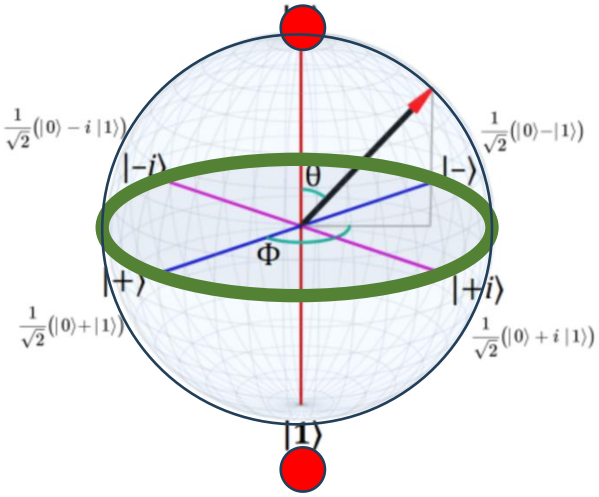
Kramers doublets

split by B-field or spontaneous magnetization

Kagome ferromagnet $Co_3Sn_2S_2$: Luyi Yang group (PRL, 2025)

What about phonons coupled to multipolar orders?

Non-Kramers doublet on the Bloch sphere



- Poles: Octupolar +/-
- Equator: Quadrupolar states

➢ Eg phonons linearly couple to quadrupole moment

$$H_{\mathrm{sp-ph}} = -\lambda \sum_{\mathbf{r}} \left[Q_x(\mathbf{r}) \tau_{\mathbf{r}x} + Q_z(\mathbf{r}) \tau_{\mathbf{r}z} \right] \qquad \begin{cases} x \equiv x^2 - y^2 \\ z \equiv 3z^2 - r^2 \end{cases}$$

➢ How do phonons react to octupolar order?

How do phonons react to octupolar ordering?

- Phonon frequency >> spin oscillations
- Phonons see “frozen” spin configuration
- Average phonon Green function over instantaneous spin configs (Monte Carlo) after integrating out spin-wave fluctuations at $\mathcal{O}(1/S)$.

How do phonons react to octupolar ordering?

$$\mathcal{G}_{xx}^{-1}(\mathbf{r}, i\nu_n) = -i\nu_n + \Omega_0 - 4S\lambda^2 \left[\frac{2h_{\mathbf{r}}\Delta_{\mathbf{r}}^x}{(i\nu_n)^2 - 4h_{\mathbf{r}}^2} \right]$$

$$\mathcal{G}_{zx}^{-1}(\mathbf{r}, i\nu_n) = \mathcal{G}_{xz}^{-1}(\mathbf{r}, -i\nu_n) = -4S\lambda^2 \left[\frac{2h_{\mathbf{r}}\chi_{\mathbf{r}} + i\nu_n\gamma_{\mathbf{r}}}{(i\nu_n)^2 - 4h_{\mathbf{r}}^2} \right]$$

$$\mathcal{G}_{zz}^{-1}(\mathbf{r}, i\nu_n) = -i\nu_n + \Omega_0 - 4S\lambda^2 \left[\frac{2h_{\mathbf{r}}\Delta_{\mathbf{r}}^z}{(i\nu_n)^2 - 4h_{\mathbf{r}}^2} \right]$$

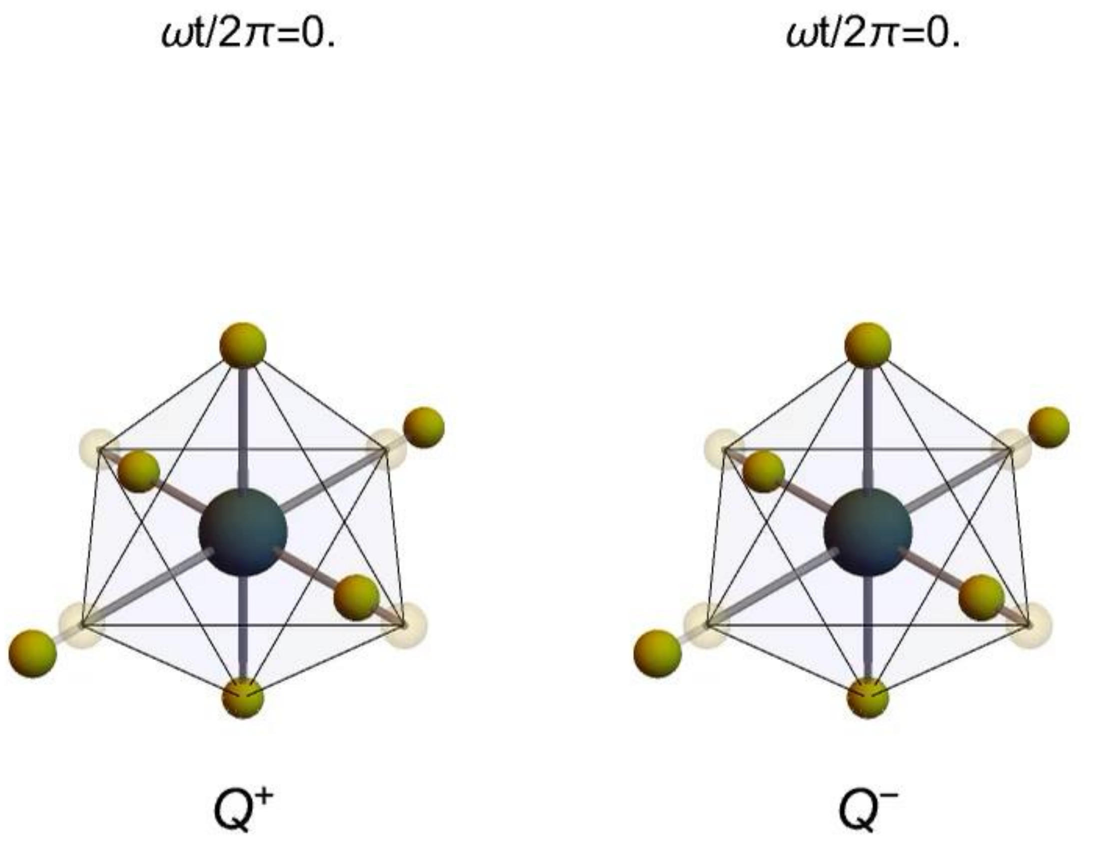
Related to octupolar order parameter

Information about local Weiss field strength and orientation

- Octupolar order leads to off-diagonal Green function coupling Eg modes ~ "Hall effect"

$$\begin{pmatrix} -i\nu_n + \Omega_0 + \delta\Omega & i\Phi \\ -i\Phi & -i\nu_n + \Omega_0 + \delta\Omega \end{pmatrix}$$

Pseudochiral phonons peering down [111] axis



$$R'(Q^+) = \begin{pmatrix} -i\,a^+ & a^+ & \sqrt{2}\,a^+ \\ a^+ & i\,a^+ & -i\,\sqrt{2}\,a^+ \\ \sqrt{2}\,a^+ & -i\,\sqrt{2}\,a^+ & 0 \end{pmatrix}$$

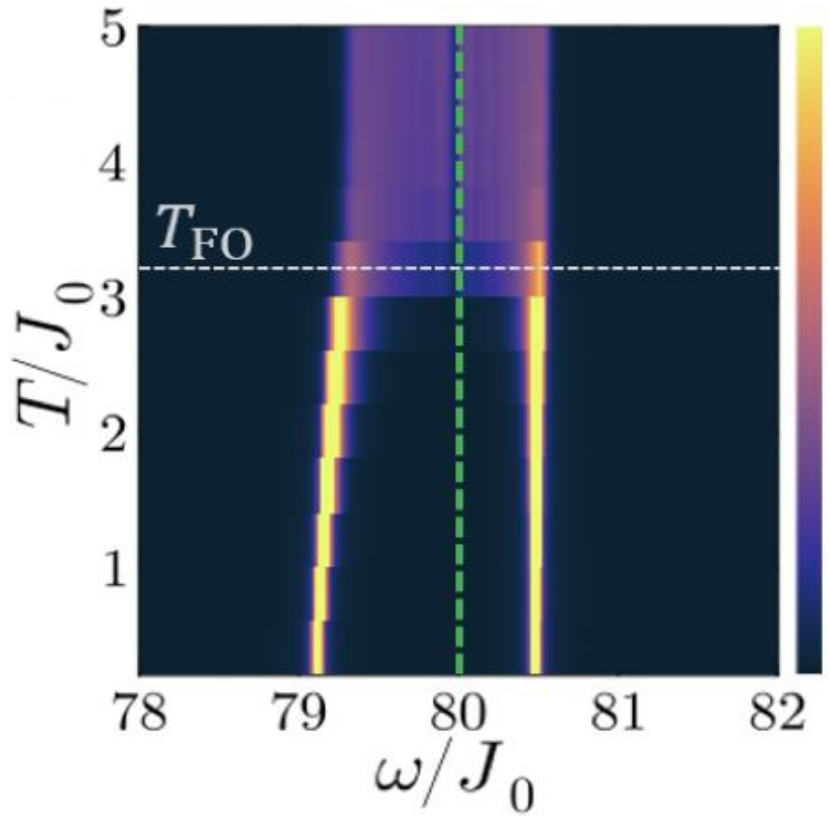
$$R'(Q^-) = \begin{pmatrix} i\,a^- & 0 & \sqrt{2}\,a^- \\ 0 & -i\,a^- & i\,\sqrt{2}\,a^- \\ \sqrt{2}\,a^- & i\,\sqrt{2}\,a^- & 0 \end{pmatrix}$$

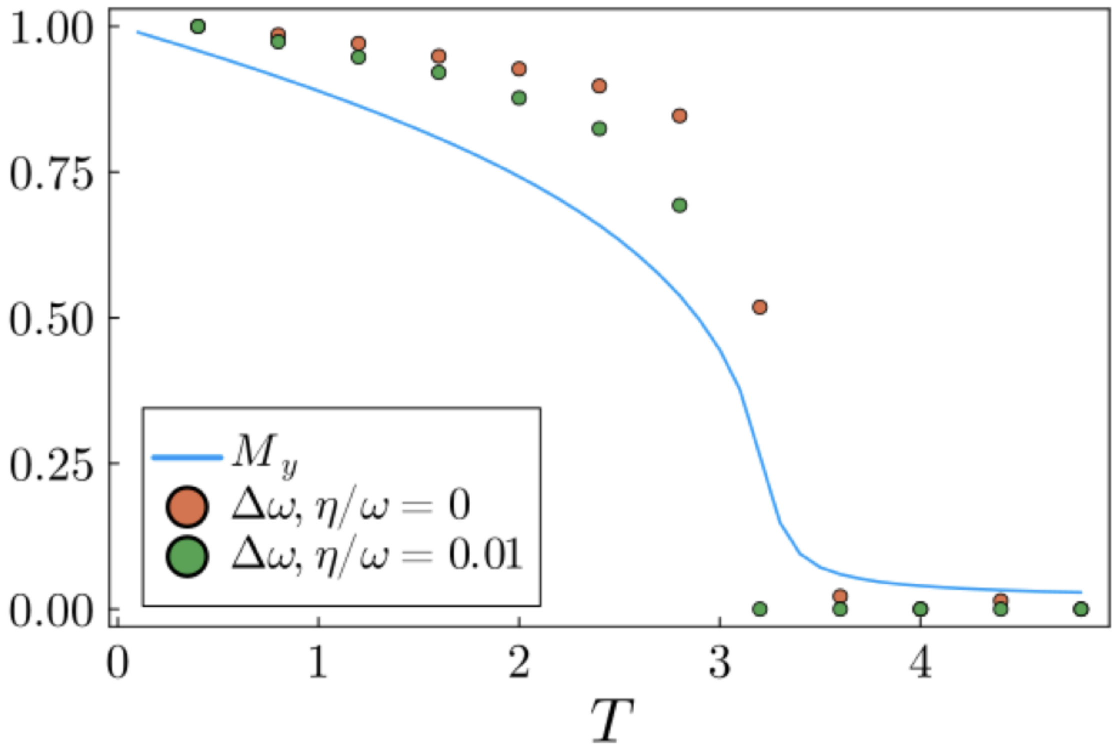
$$|\sigma^+ \cdot R'(Q^+) \cdot \sigma^-|^2 = |a^+|^2$$

$$|\sigma^- \cdot R'(Q^-) \cdot \sigma^+|^2 = |a^-|^2$$

Detect using Raman scattering
in cross-circular channel

Phonon spectral function





- Probe using cross-circular Raman scattering
- High temperature regime looks like "phonon pseudogap"

R. Sutcliffe, K. Hart, S. Chaudhary, AP (PRL 2026)

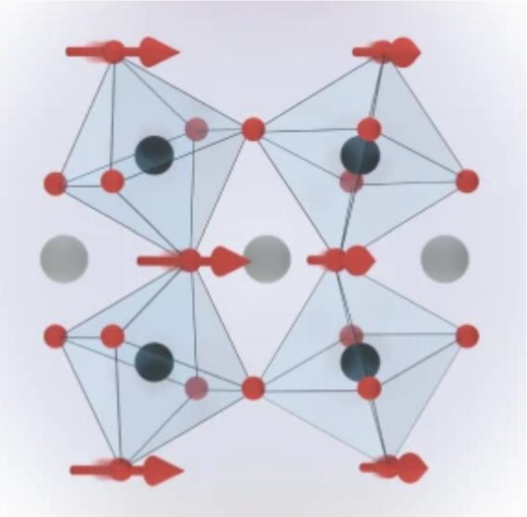
Can we use phonons to control multipolar orders in solids?

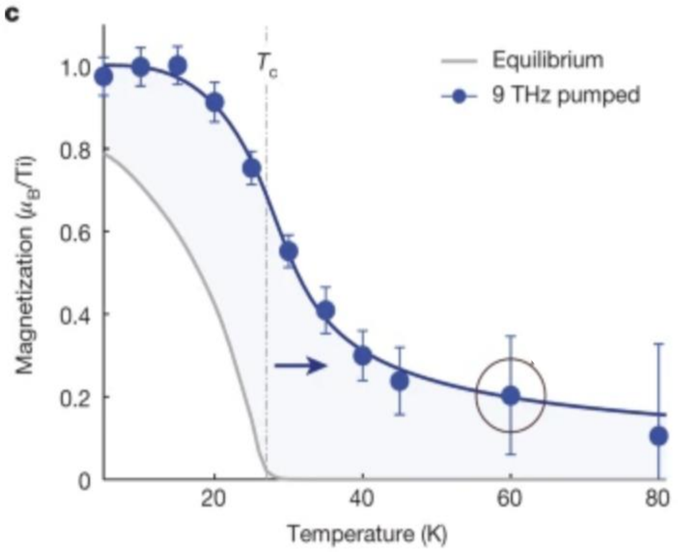
Dipolar magnetization induced by driven phonons

Photo-induced high-temperature ferromagnetism in $YTiO_3$

A. S. Disa[1,2]✉, J. Curtis[3,4], M. Fechner[1], A. Liu[1], A. von Hoegen[1], M. Först[1], T. F. Nova[1], P. Narang[3,4], A. Maljuk[5], A. V. Boris[6], B. Keimer[6] & A. Cavalleri[1,7]✉

Nature 617, 73 (2023)



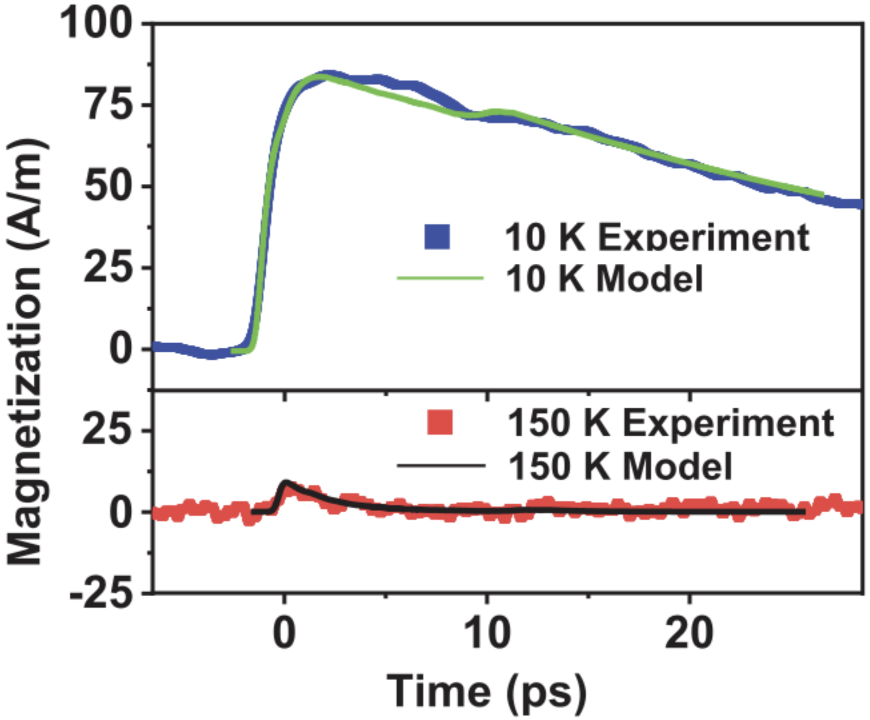


Phonon drive controls distortions in the crystal

Large effective magnetic fields from chiral phonons in rare-earth halides

Jiaming Luo[1,2], Tong Lin[1], Junjie Zhang[1], Xiaotong Chen[1], Elizabeth R. Blackert[1], Rui Xu[1], Boris I. Yakobson[1], Hanyu Zhu[1]*

Luo *et al.*, *Science* **382**, 698–702 (2023)

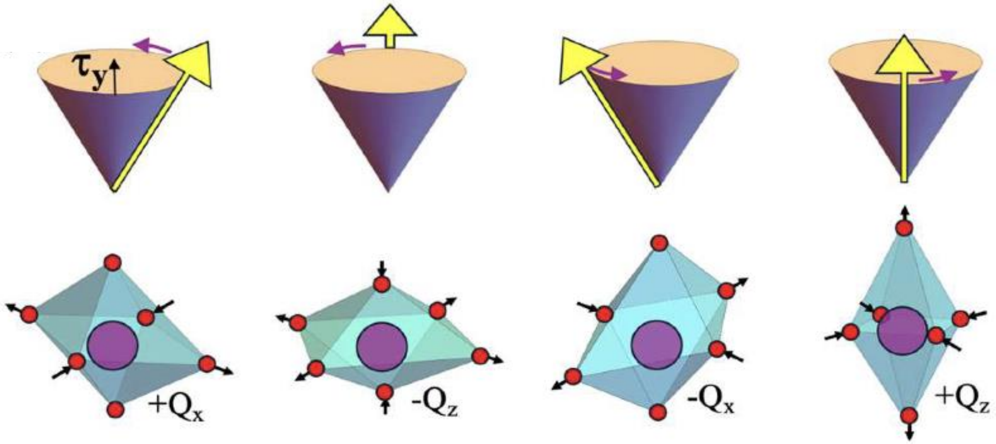


Theory: D. Juraschek, T. Neuman, P. Narang, Phys. Rev. Res. 4, 013129 (2022)

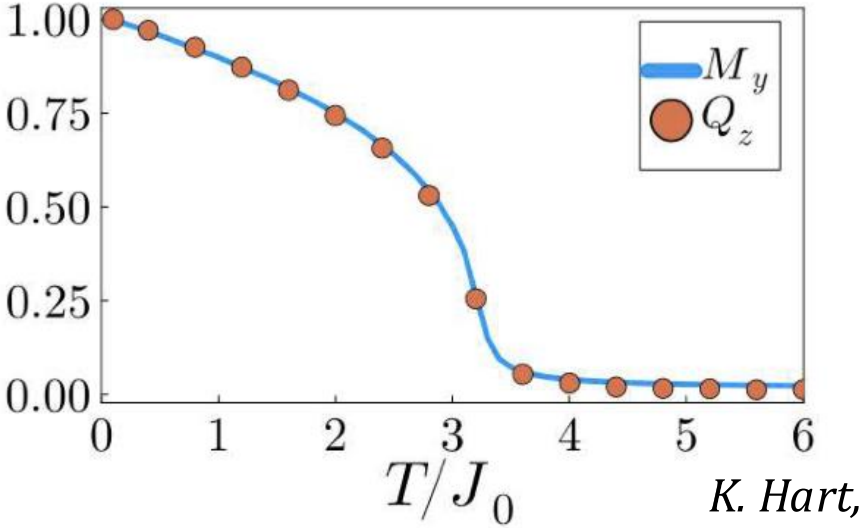
Pump-probe: Kick X-mode, track Z-mode

$$H_{\mathrm{sp-ph}} = -\lambda \sum_{\mathbf{r}} \left[Q_x(\mathbf{r})\tau_{\mathbf{r}x} + Q_z(\mathbf{r})\tau_{\mathbf{r}z} \right]$$

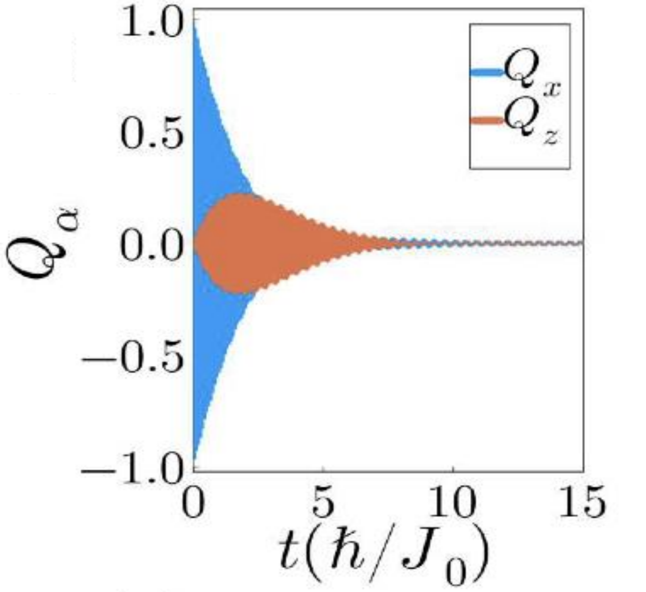
Pseudospin precession drives phonon "precession"

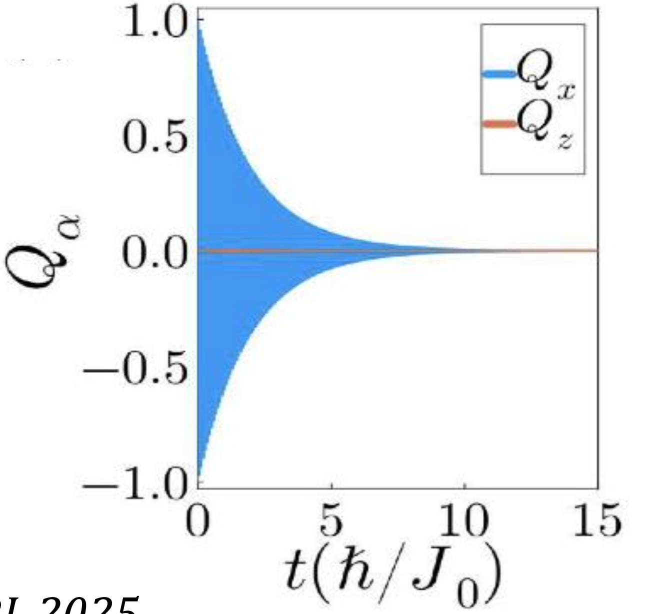


Computed cross-response tracks octupolar order



Spin-Phonon Molecular Dynamics Simulations





*K. Hart, R. Sutcliffe, G. Refael, **AP**, PRL 2025*

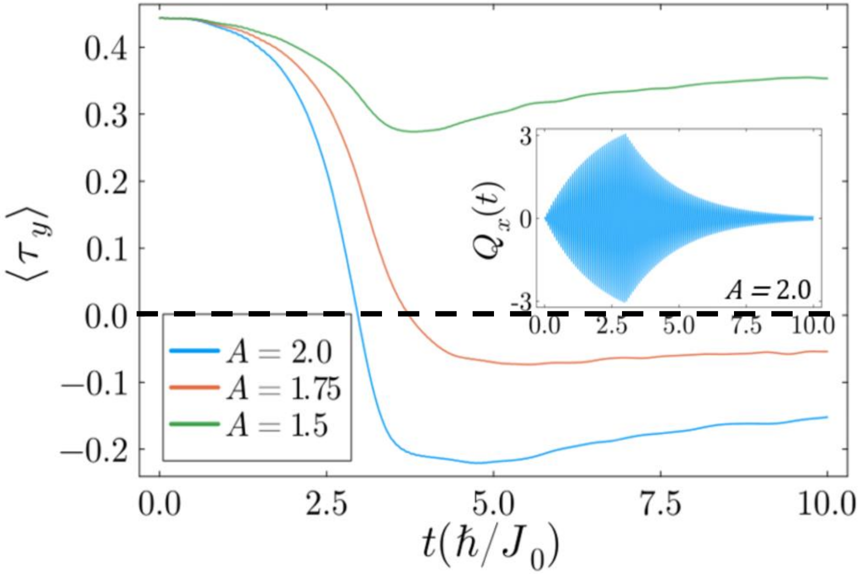
Pumping pseudochiral phonons

Chiral phonon drive $H^{\mathrm{ph}}_{\mathrm{drive}} = -A\sum_{\mathbf{r}}[Q_x(\mathbf{r})\cos(\Omega t) + Q_z(\mathbf{r})\cos(\Omega t - \phi)]$

Floquet analysis

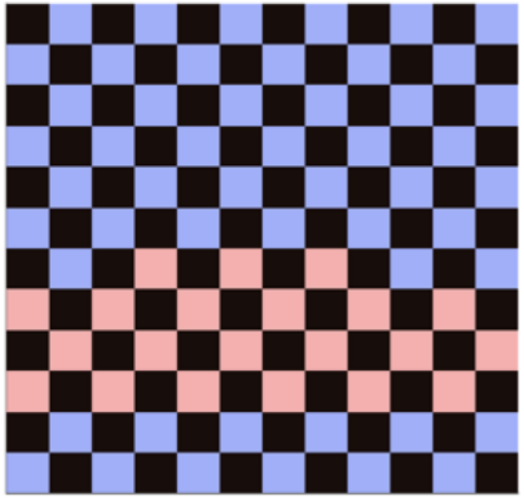
$$H^{\mathrm{sp}}_{\mathrm{drive},0} = 0; \quad H^{\mathrm{sp}}_{\mathrm{drive},\pm} = -\frac{\lambda Q_0}{2}\sum_{\mathbf{r}}(\tau_{\mathbf{r}x} + e^{\mp i\phi}\tau_{\mathbf{r}z})$$

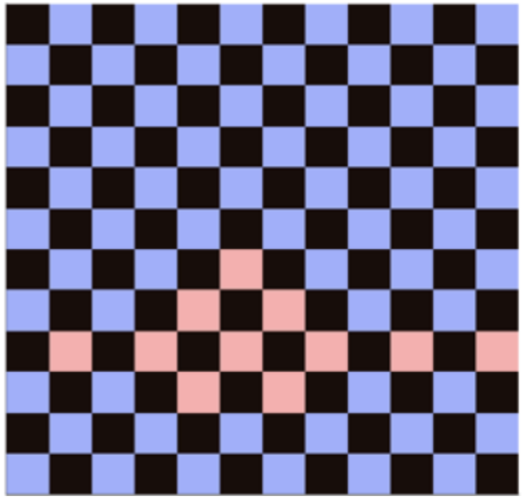
$$H^{\mathrm{sp,eff}}_{\mathrm{drive}} = \frac{1}{\Omega}[H^{\mathrm{sp}}_{\mathrm{drive},+}, H^{\mathrm{sp}}_{\mathrm{drive},-}] = \frac{\lambda^2 Q_0^2}{\Omega}\sin\phi\sum_{\mathbf{r}}\tau_{\mathbf{r}y}$$



Phonon driven switching of octupolar order

Chiral phonon driven growth of octupolar domains





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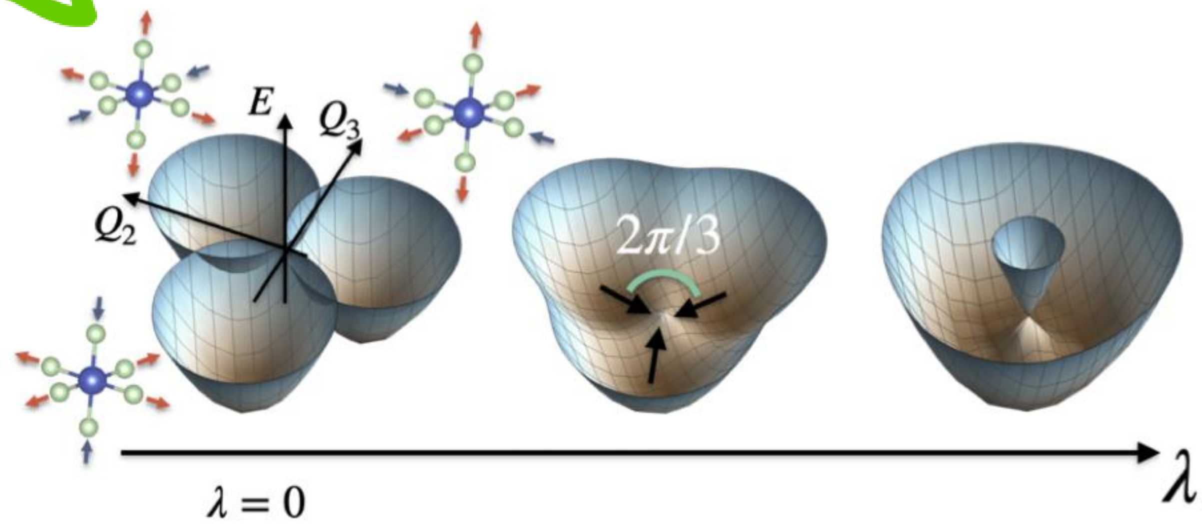
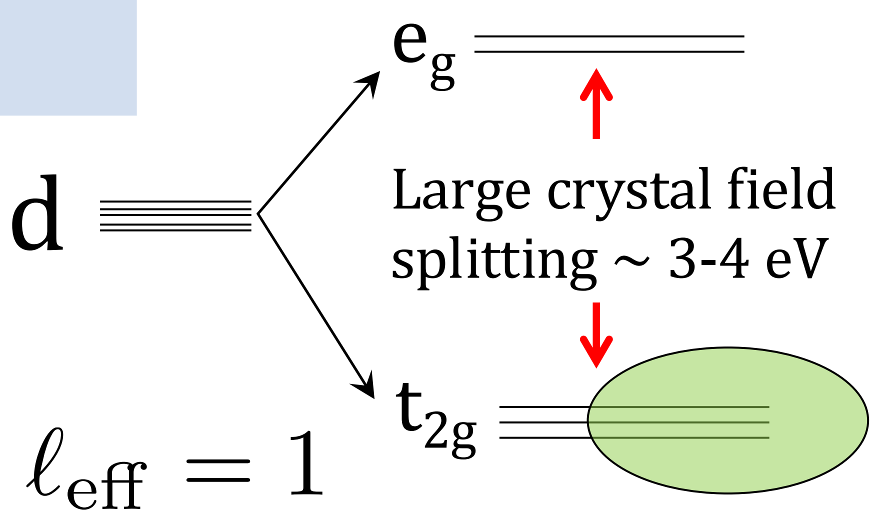
Competition between Jahn-Teller and Multipolar orders

Competition between SOC and Jahn-Teller distortions

d^1	d^2	d^3	d^4	d^5
j=3/2? Distort?	J=2? Octupolar!	J=3/2 SOC quenched?	J=0	j=1/2





Jahn-Teller Effect and Spin-Orbit Coupling: Friends or Foes?

Sergey V. Streltsov[1,2,*] and Daniel I. Khomskii[3]

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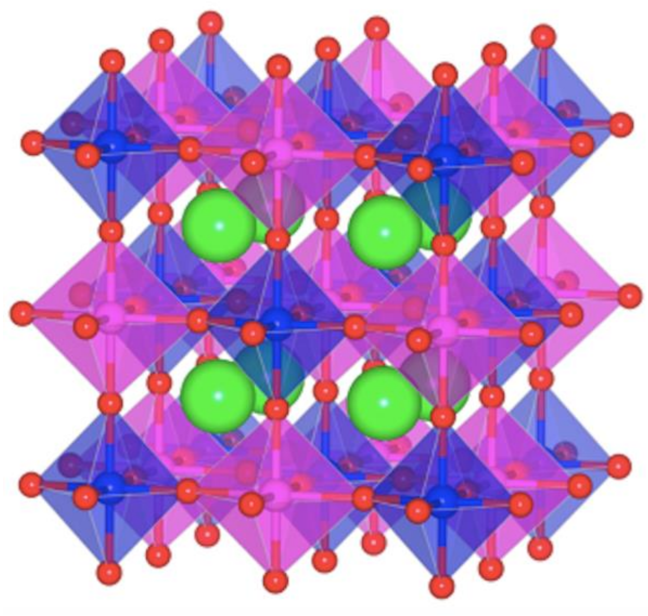
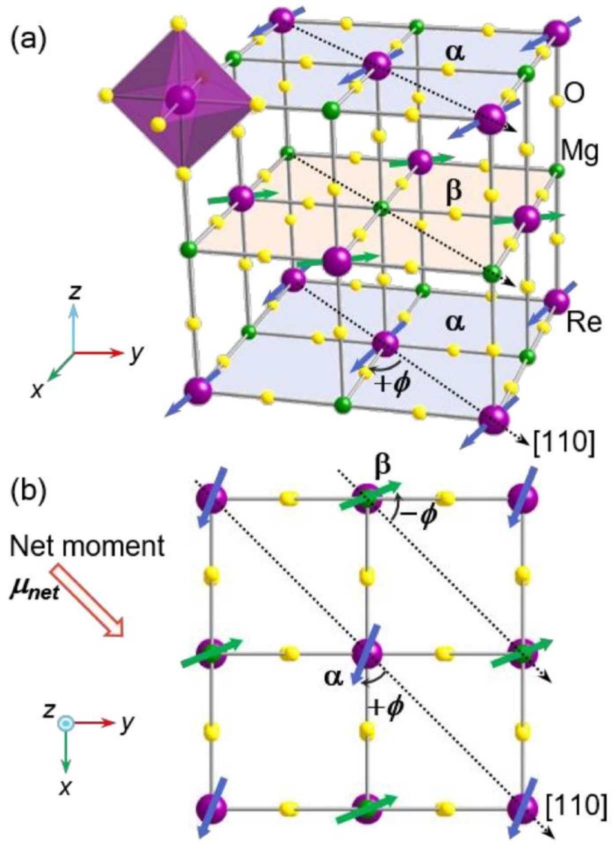
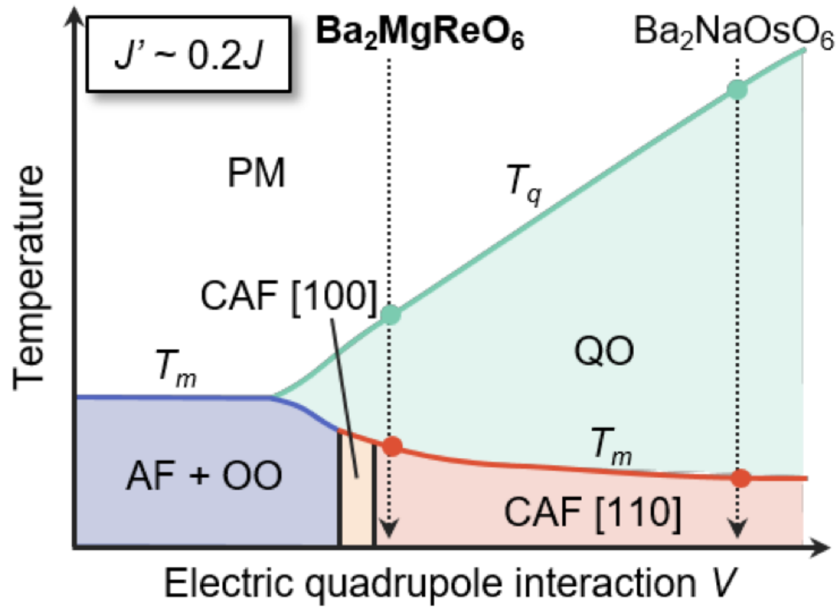
$5d^1$ Double Perovskites: j=3/2 magnetism



Ba_2MgReO_6
Ba_2YMoO_6
Ba_2NaOsO_6





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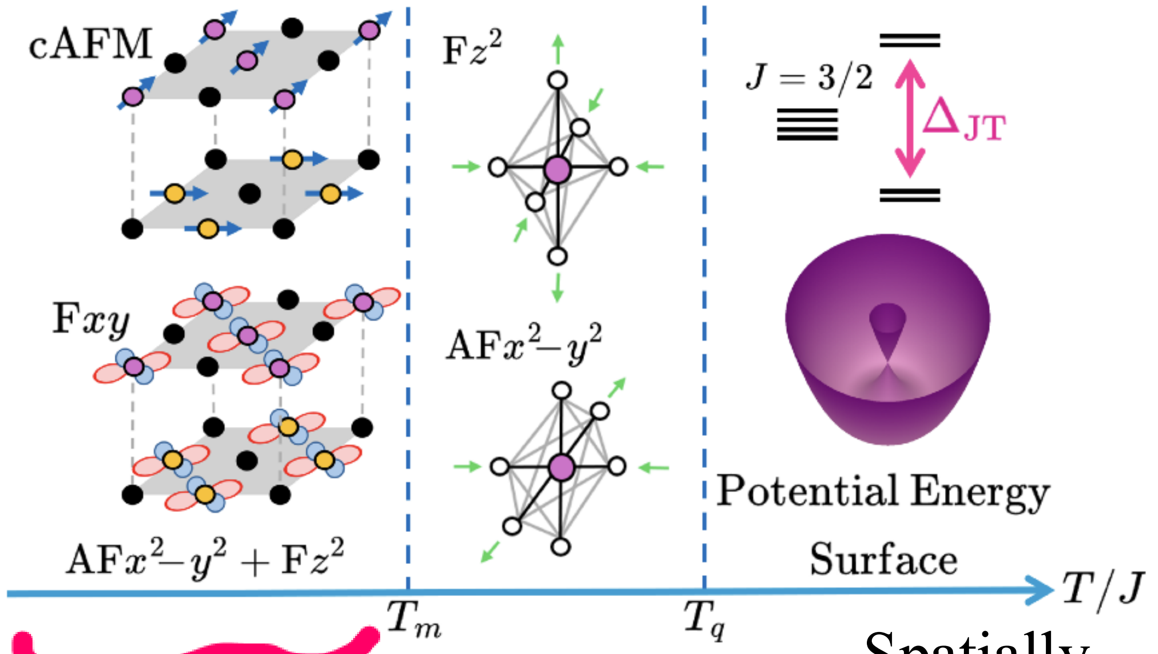
Liu et al, Physica B 2018
D. Hirai, et al, JPSJ 2019
D. Hirai, et al, PRR 2020

Puzzles:

- *Role of the lattice degrees of freedom in governing the order?*
- *How is the entropy recovered upon heating?*
- *What is the role of fluctuating local Jahn-Teller distortions?*

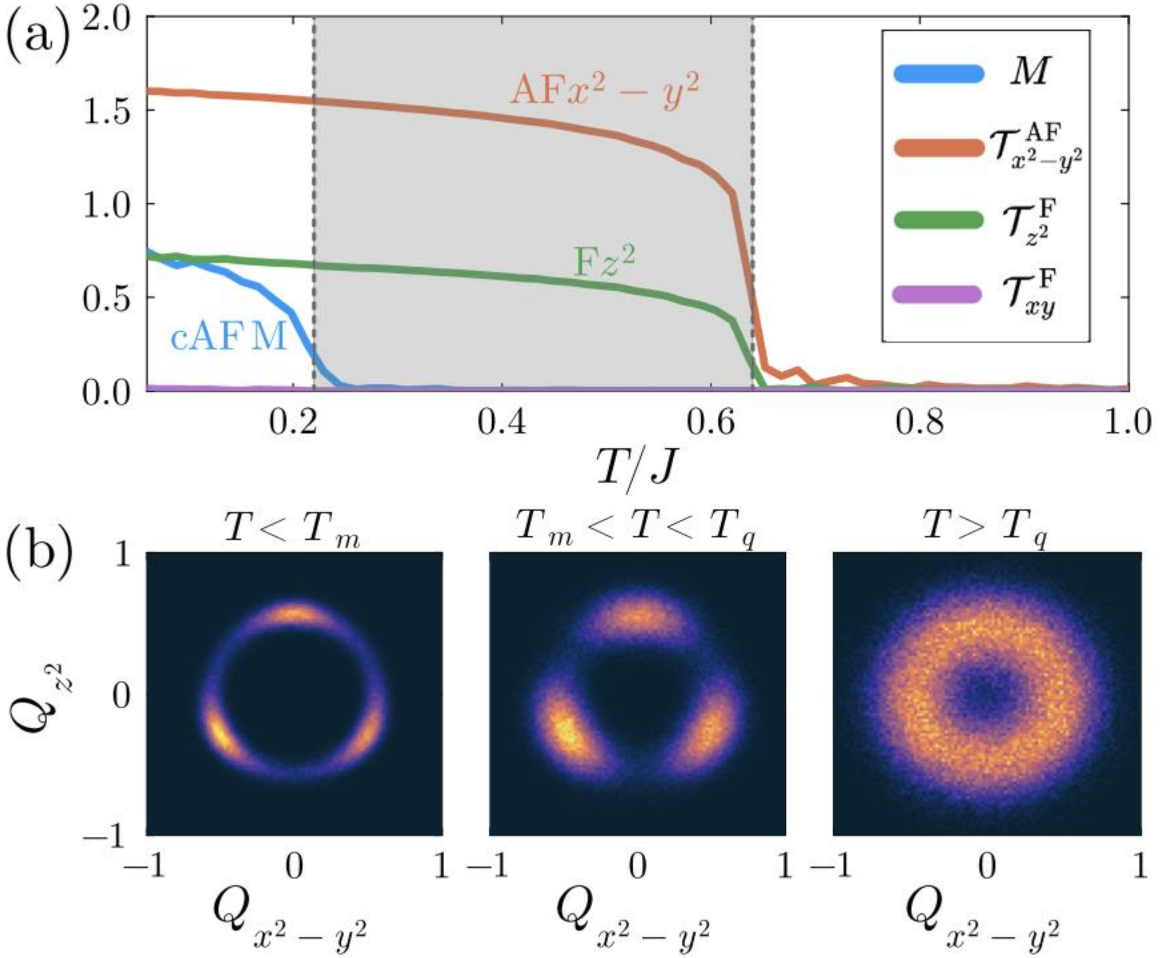
j=3/2 magnetism coupled to JT phonons



Spatially
Fluctuating
JT distortions

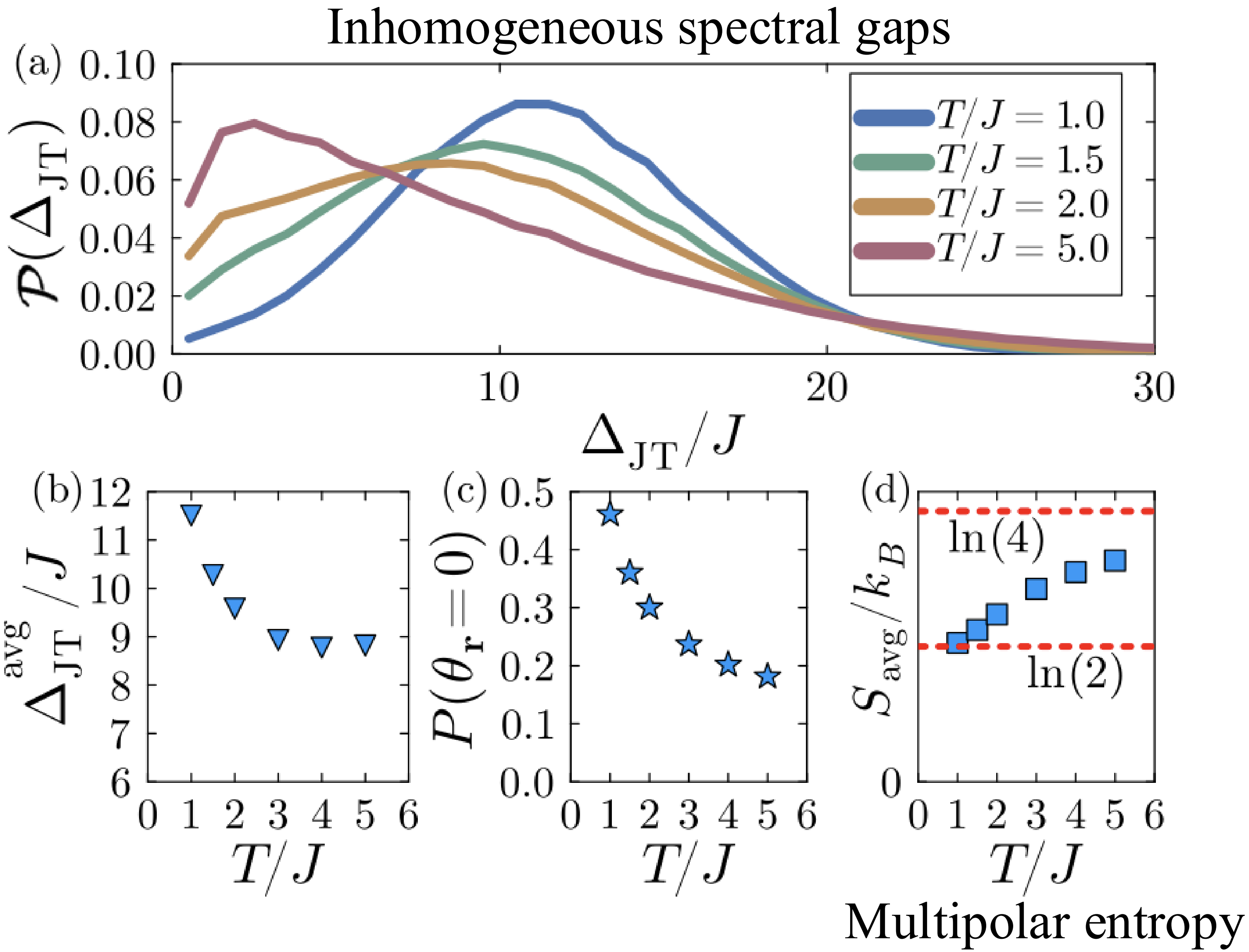
Magnetic

Quadrupolar +
JT distortions



R. Sutcliffe, K. Hart, AP (arXiv: 2026)

j=3/2 magnetism coupled to JT phonons



R. Sutcliffe, K. Hart, AP (arXiv: 2026)

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Francois Fauth, Casey Marjerrison,
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Summary

1. Phonons: Can carry angular momentum and show chiral splittings

2. Multipolar Order: Serves to synthesize various forms of hidden order
   - Spin altermagnets
   - Loop current orders
   - Octupolar order

3. Phonons can serve to probe, drive, and switch multipolar orders

4. Competition between local JT distortions and multipolar orders leads to rich crossovers