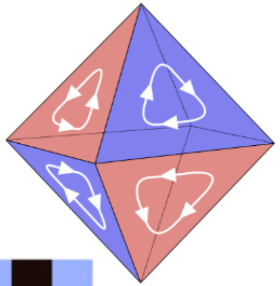
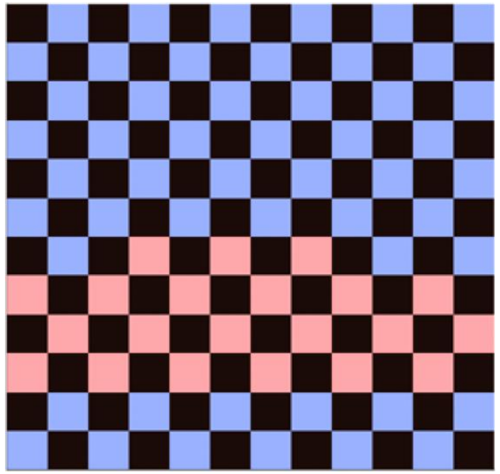
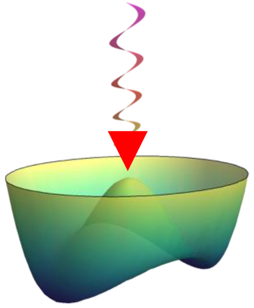
Phonons as a probe of hidden order in quantum materials



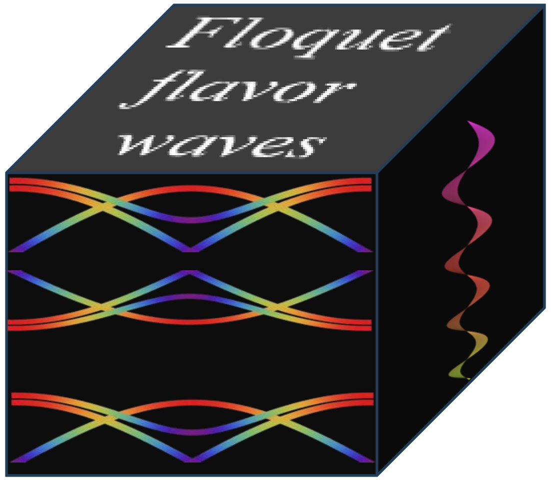




Arun Paramekanti
(University of Toronto)

Advanced School and Conference on Quantum Matter, ICTP Trieste

Dec 01-12, 2025



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Spin liquid story: Quick summary but not the focus of this talk

Proximate Dirac spin liquid in the honeycomb lattice J_1–J_3 XXZ model: Numerical study and application to cobaltates

Anjishnu Bose[1,*], Manodip Routh[2,*], Sreekar Voleti[1,*], Sudip Kumar Saha[2], Manoranjan Kumar[2], Tanusri Saha-Dasgupta[2], and Arun Paramekanti[1,2,3]

Show more

Phys. Rev. B **108**, 174422 – **Published 16 November, 2023**

- Study "XY"-type frustrated honeycomb magnets
- Find candidate easy-plane Dirac spin liquid wfn
- Explains THz spectroscopy as function of (B,T)
- Explains "universal" metallic thermal conductivity

Modified large-N approach to gapless spin liquids, magnetic orders, and dynamics: Application to triangular lattice antiferromagnets

Anjishnu Bose[*], Kathleen Hart, Ruairidh Sutcliffe, and Arun Paramekanti[†]

Show more

Phys. Rev. B **111**, 214410 – **Published 4 June, 2025**

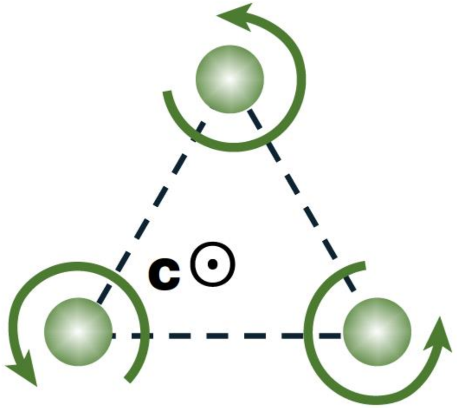
DOI: https://doi.org/10.1103/PhysRevB.111.214410

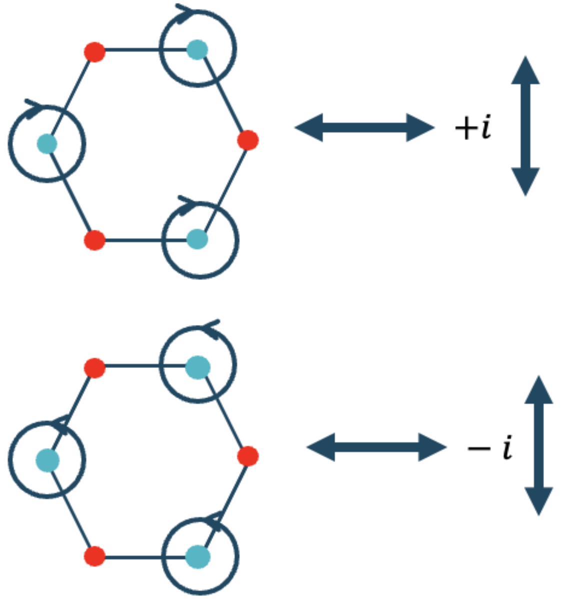
- Formulate a distinct large-N approach to QSLs
- Allows access to QSL-Magnetic order transitions
- Parton-parton interactions capture S(Q,w)
- Successfully applied to triangular J1-J2 AFM

- A. Bose, K. Hart, R. Sutcliffe, AP, arXiv:2503.09695
- Willsher, J. Knolle, arXiv:2503.13831

Phonons in solids

Phonon angular momentum



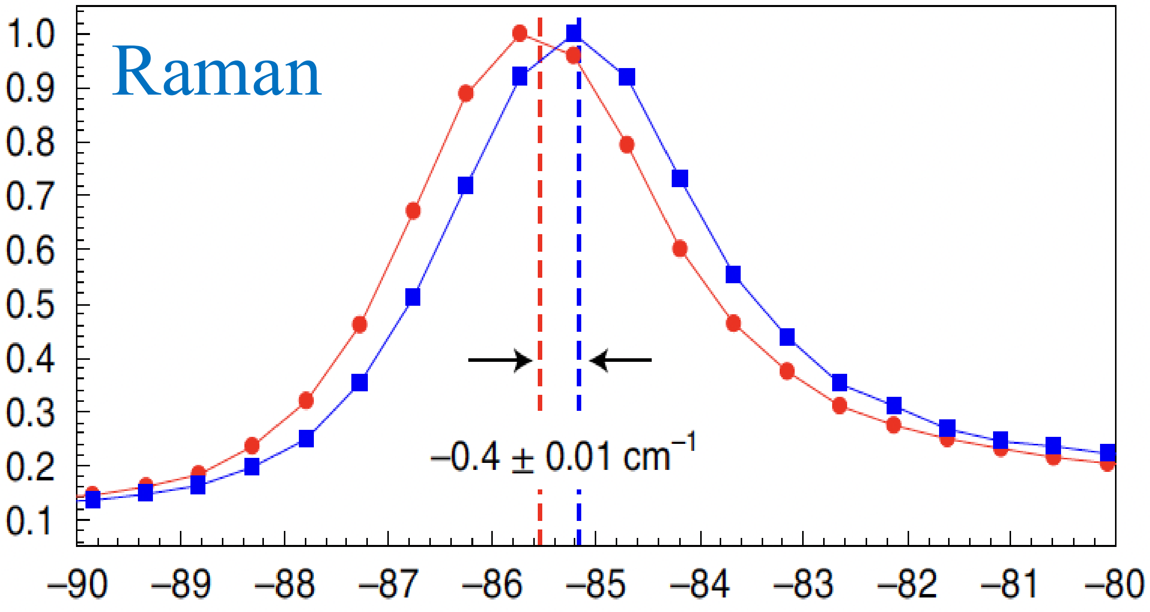


$\boldsymbol{L} = \boldsymbol{Q} \times \partial_t \boldsymbol{Q}$

- 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 angular momentum in non-centrosymmetric insulators



Phonon mode splitting in a chiral crystal
α-HgS: Ishito, et al, Nat. Phys. 19, 35 (2023)

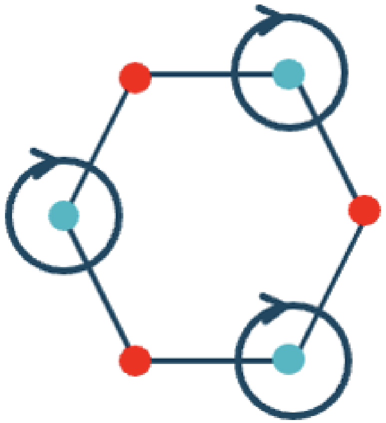
Allowed term: $\vec{k} \cdot \vec{L}$

- Breaks inversion
- Preserves time-reversal symmetry
- Like a "spin-orbit coupling" for phonons
- Really L ~ "crystal angular momentum"

Zone center phonon: k=0 (not split)
Actually probing small nonzero $k=2\pi/\lambda$

Light wavelength $\lambda \sim 7000$A
Lattice spacing a ~ 10A
k.a ~ 1/100

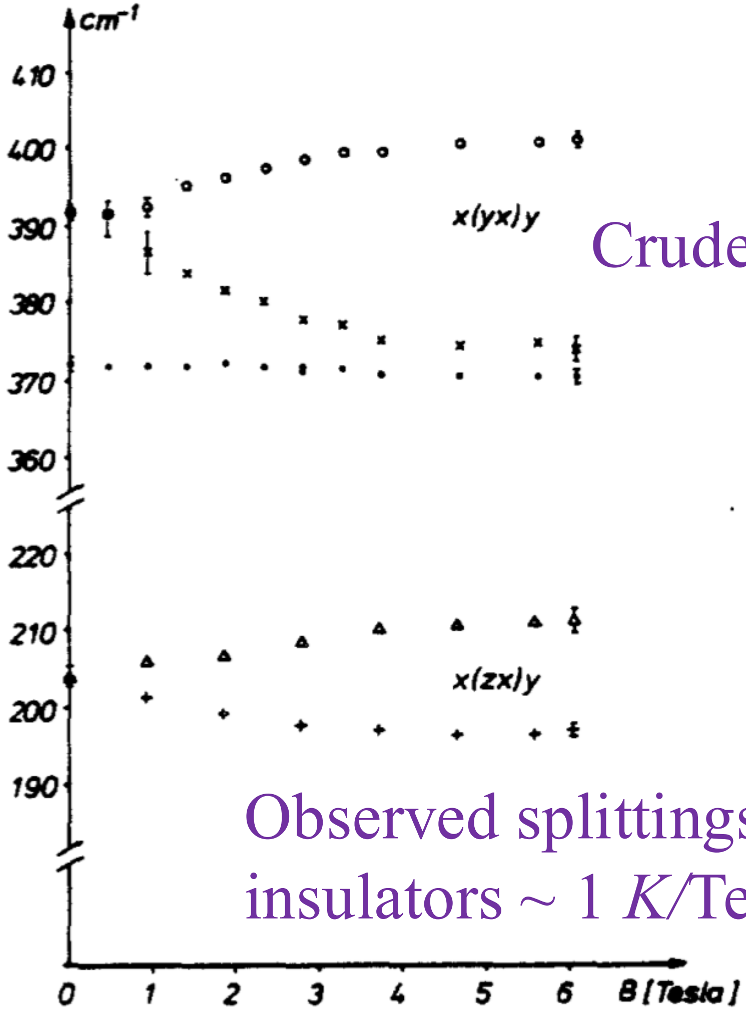
Magnetic moment of phonons



Expectation:

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

Zeeman splitting ~ 1 *mK*/Tesla



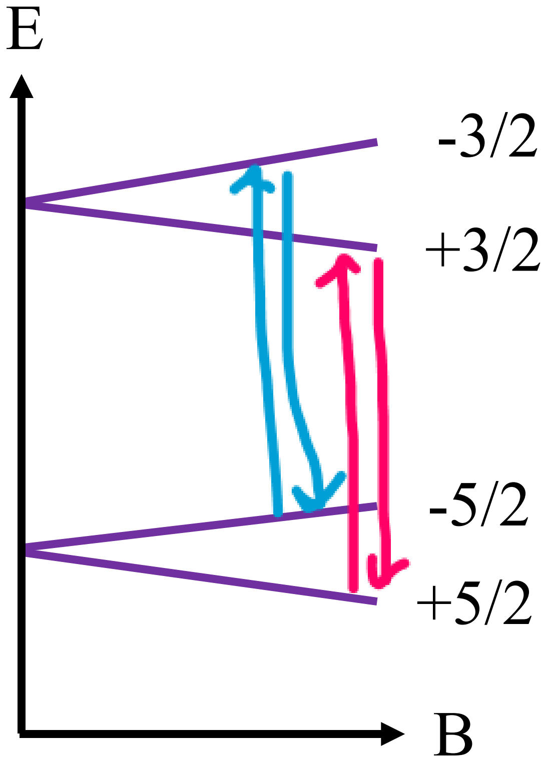
Crudely: $1\text{cm}^{-1} \sim 1\ K$

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

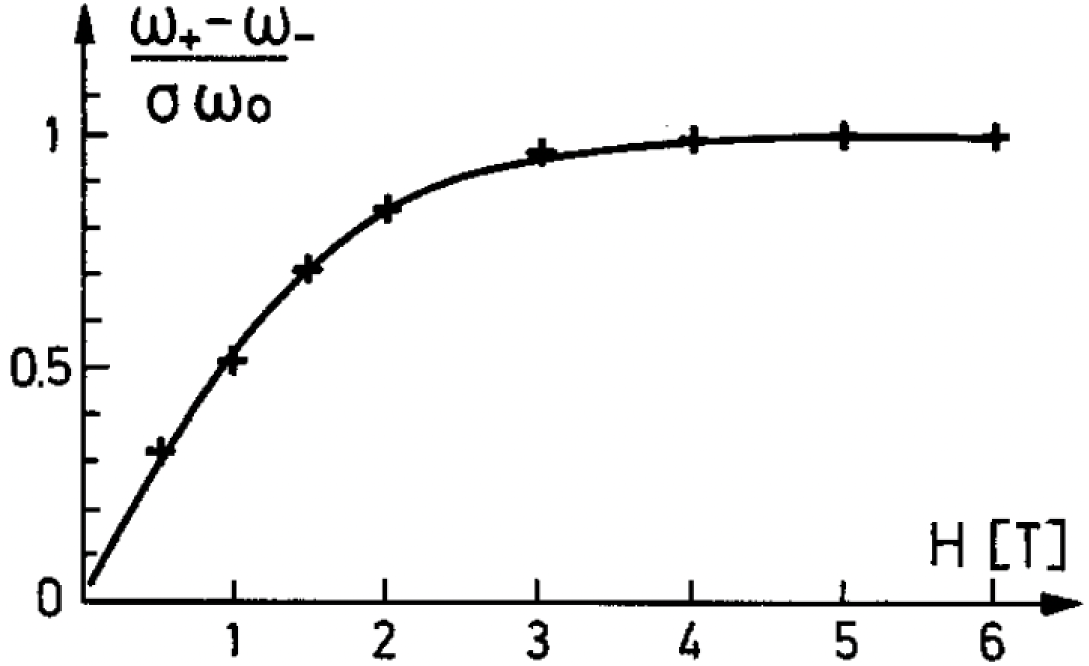
Raman scattering on CeF_3

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

Magnetic moment of phonons



Phonon self energy



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

Crystal field Kramers doublets split by B-field

Fully microscopic theory + d-orbital materials

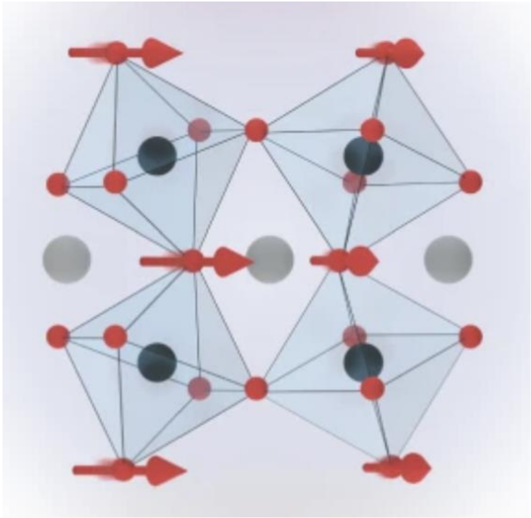
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, G. Fiete, PRB (2024)

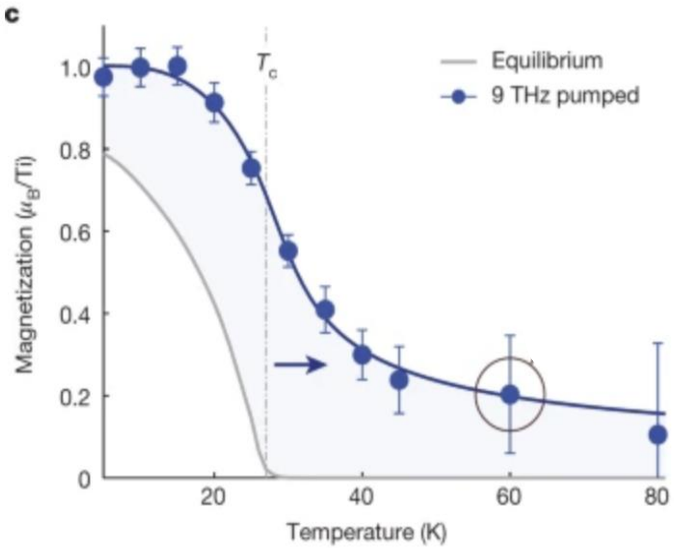
Magnetization induced by driven phonons

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)



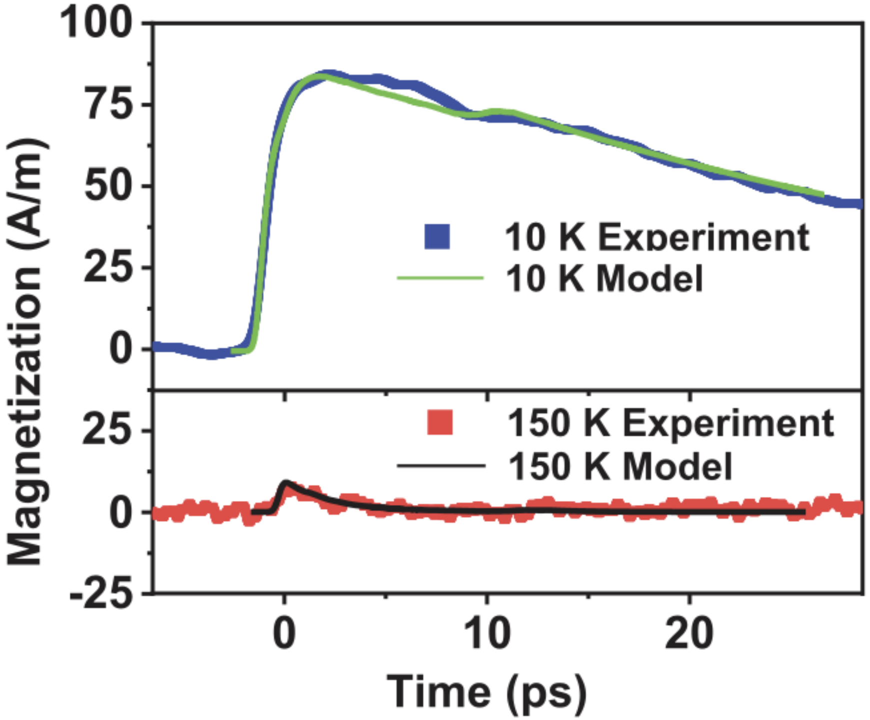


Phonon drive controls distortions in the crystal

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*]

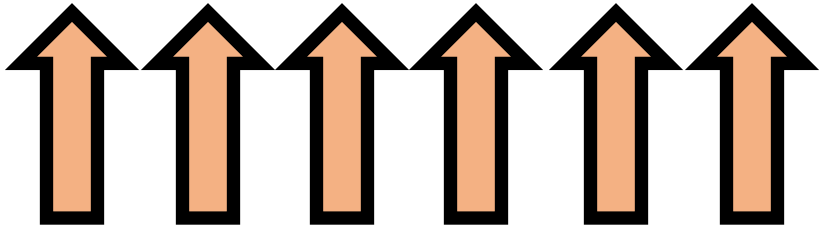
Luo *et al.*, *Science* **382**, 698–702 (2023)



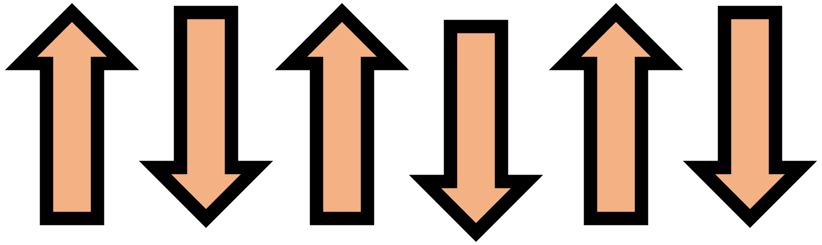
Theory: D. Juraschek, T. Neuman, P. Narang, Phys. Rev. Res. 4, 013129 (2022)

Hidden orders in solids

Quantum materials: Simple orders



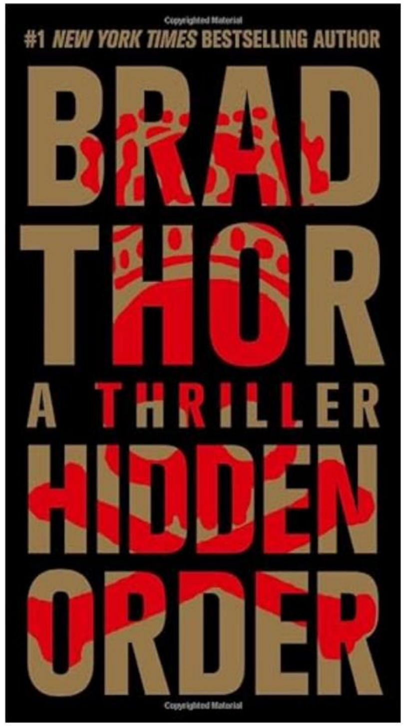
Ferromagnetism ~ 600 BC
Easily observable
(lodestone, fridge magnets)



Antiferromagnetism
Neutron diffraction (1940s)

- Simple patterns of symmetry breaking
- Well developed experimental probes
- Even AFM is “hidden” until we can “see” the pattern

What is hidden order?



Hidden Order: A Thriller (12) (The Scot Harvath Series)

Mass Market Paperback – May 20, 2014

by Brad Thor (Author)

4.4 9,858 ratings   4.2 on Goodreads 17,203 ratings

Book 12 of 23: The Scot Harvath   See all formats and editions

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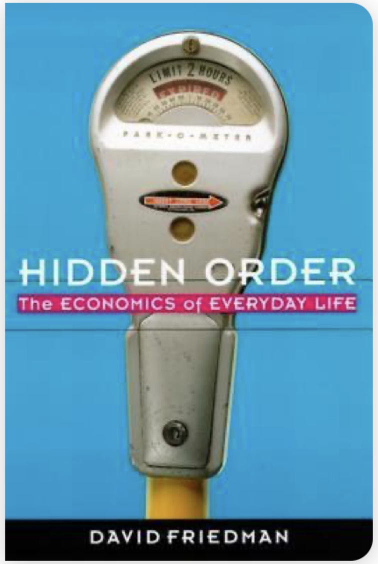
The most secretive organization in America operates without accountability to the American people. Hiding in the shadows, pretending to be part of the United States Government, its power is beyond measure.

Control of this organization has just been lost and the future of the nation has been thrust into peril.



Hidden Order: The Economics of Everyday Life

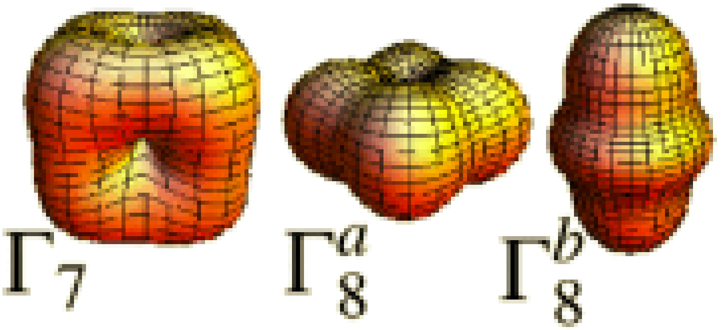
David D. Friedman

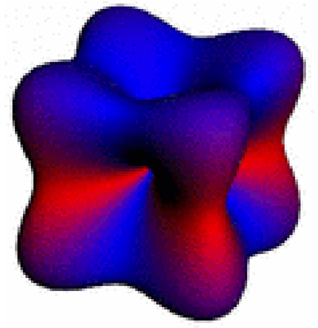
3.74 505 ratings · 32 reviews

David Friedman has never taken an economics class in his life. Sure, he's taught economics at UCLA. Chicago, Tulane, Cornell, and Santa Clara, but don't hold that against him. After all, everyone's an economist. We all make daily decisions that rely, consciously or not, on an acute understanding of economic theory--from picking the fastest checkout tine at the supermarket to voting or not voting, from negotiating the best job offer to finding the right person to marry. Hidden Order is an essential guide to rational living, revealing all you need to know to get through each day

Hidden order in quantum materials

Multipolar magnetism in spin-orbit coupled materials

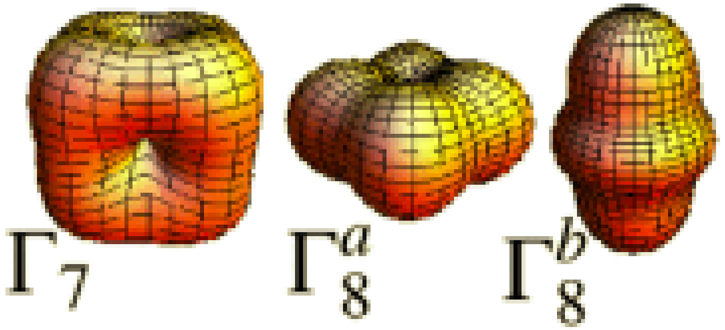


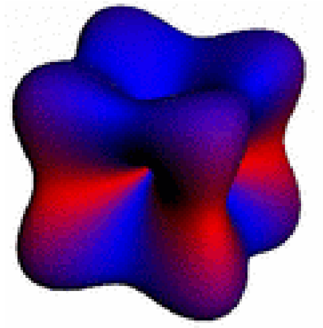


Strong local entanglement of spins and orbital degrees of freedom

Hidden order in quantum materials

Multipolar magnetism in spin-orbit coupled materials





- Phase transition easily seen in thermodynamics (e.g., specific heat)
- Precise nature of hidden order: Very hard to unravel
- Many examples: URu_2Si_2, NpO_2, PrV_2Al_{20}, 5d transition metal oxides

- 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”)

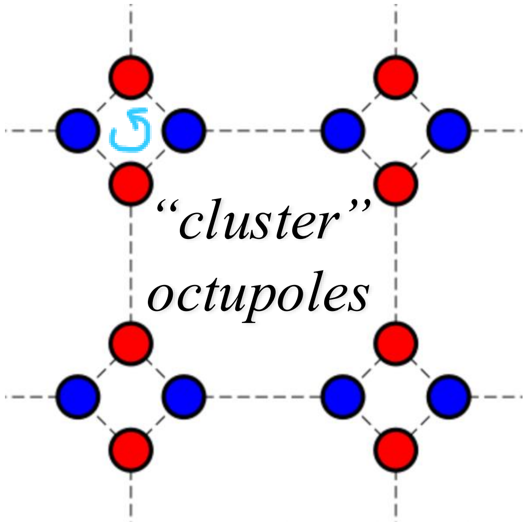
“Cluster multipoles” in quantum materials

Spin Altermagnets

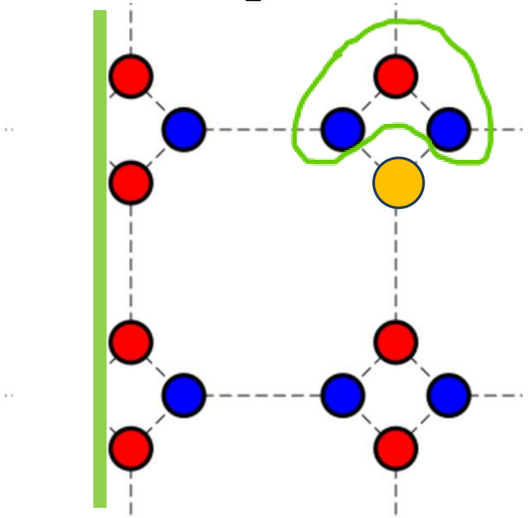
Kivelson, et al Rev. Mod. Phys. (2003)
Naka, et al, Nat Comm (2019)
Smejkal, Sinova, Jungwirth PRX (2022)

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

d-wave ferromagnet

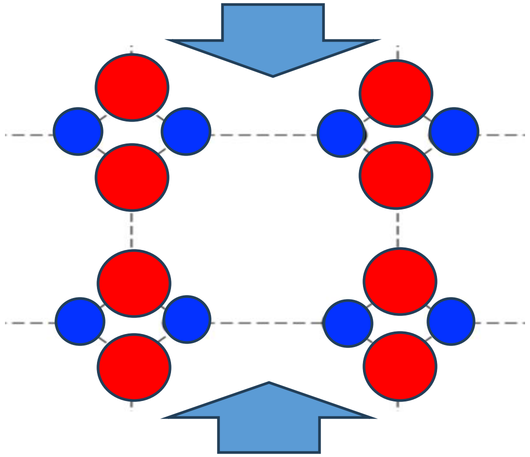


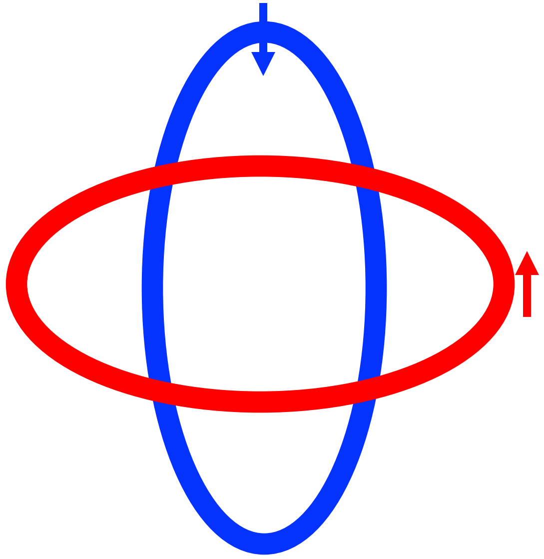
Bulk impurity dipole moments



Surface magnetization

Piezomagnetism





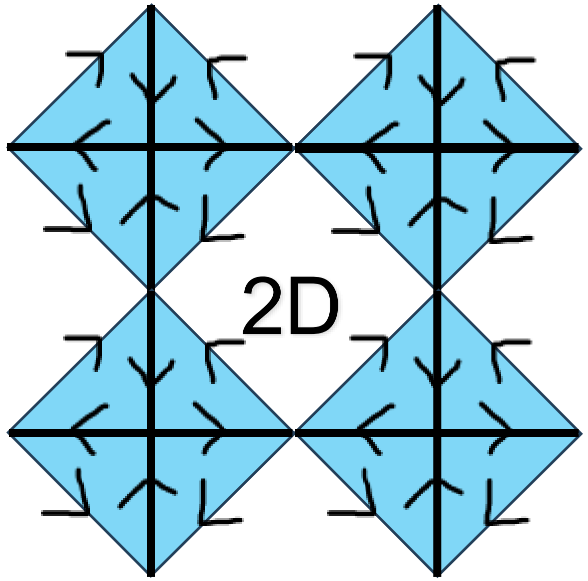
Metallic regime

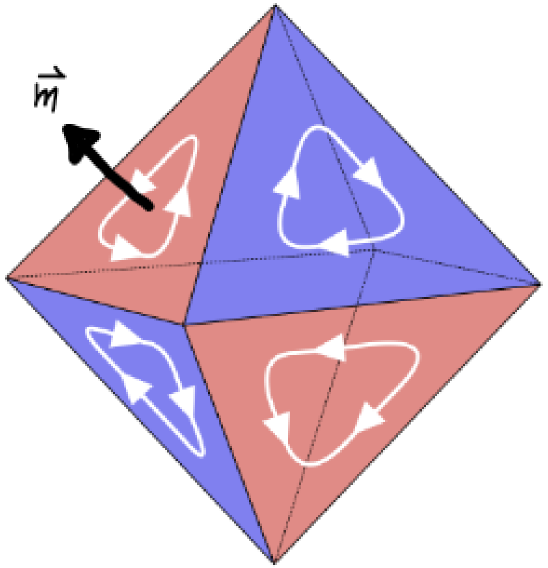
"Cluster multipoles" in quantum materials

"Orbital" Altermagnets

Loop current orders

Q=0 orbital magnetism





3D

(111) Surface

- orbital magnetization
- Kerr effect?

e.g., Loop currents in cuprates

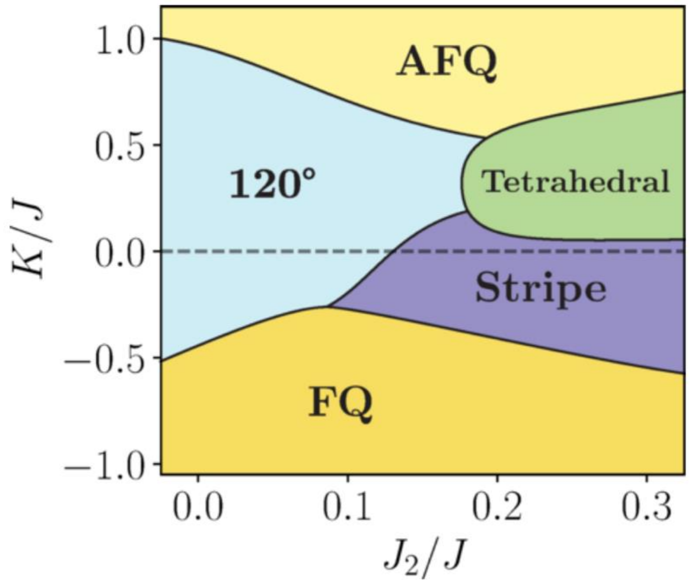
Simon and Varma, PRL 2002
Varma, PRB 2006
Bourges, Bounoua, Sidis, Comptes Rendus 2021

Toy example of "atomic multipoles"

Consider spin S=1 at each site: three-level system

Local 3x3 Hermitian operators: 9 observables

- Identity
- 3 Dipoles: Sx, Sy, Sz
- 5 Quadrupoles: $Q_{ab} = S_a S_b + S_b S_a$



Triangular lattice
A. Szasz, C. Wang, Y.C. He (PRB 2022)

$$H = \sum_{i,j} \left[J_{ij}\ \vec{S}_i \cdot \vec{S}_j + K_{ij} (\vec{S}_i \cdot \vec{S}_j)^2 \right]$$

Moral:

Bigger angular momentum => More multipoles (s=3/2 allows octupoles, etc)

(2S+1) = N => Nontrivial Hermitian matrices = N^2-1 [SU(N) generators]

“Atomic multipoles” in rare-earth quantum materials

$PrTi_2Al_{20}$, PrV_2Al_{20}
Diamond Lattice

$$Pr^{3+}: \quad J = 4$$

T_d crystal field splitting

$$|\Gamma_3^{(1)}\rangle = \frac{1}{2}\sqrt{\frac{7}{6}}\,|4\rangle - \frac{1}{2}\sqrt{\frac{5}{3}}\,|0\rangle + \frac{1}{2}\sqrt{\frac{7}{6}}\,|-4\rangle$$

$$|\Gamma_3^{(2)}\rangle = \frac{1}{\sqrt{2}}\,|2\rangle + \frac{1}{\sqrt{2}}\,|-2\rangle\,.$$

$$\tau_x \quad \propto \quad J_x^2 - J_y^2$$

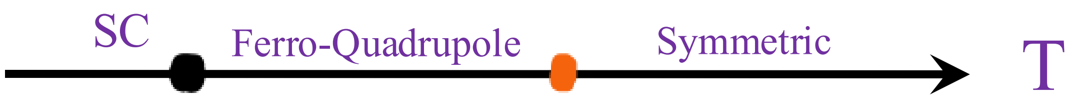
$$\tau_z \quad \propto \quad 3J_z^2 - J^2$$

Quadrupolar charge density

$$\tau_y \quad \propto \quad \mathrm{Sym}\,[J_x J_y J_z]$$

Ising magnetic octupole

$PrTi_2Al_{20}$



Recent work: A. Sakai, et al, Nat. Comm 16:2114 (2025)

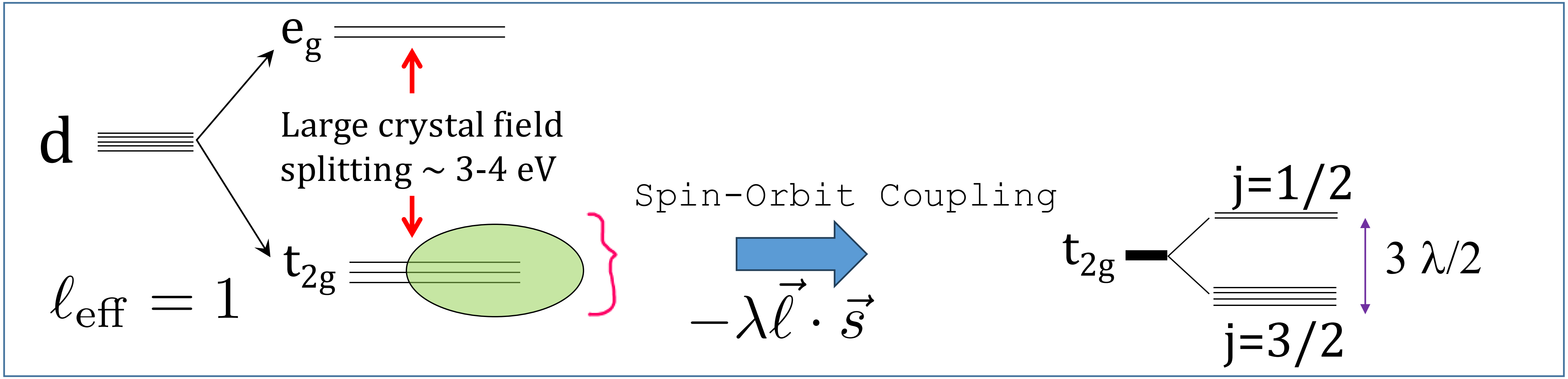
PrV_2Al_{20}

M. Tsujimoto, et al, PRL 113, 267001 (2014)

Multipolar orders

F. Freyer, J. Attig, S.B. Lee, **AP,** S. Trebst, Y.B. Kim (PRB 2017)
S. B. Lee, S. Trebst, Y. B. Kim, **AP** (PRB 2018)
A.Patri, A. Sakai, S B Lee, **AP** S. Nakatsuji, Y B Kim, Nat. Comm. (2019)
F. Freyer, S.B. Lee, Y B Kim, S Trebst, **AP,** PRR (2020)

“Atomic multipoles” in d-orbital quantum materials

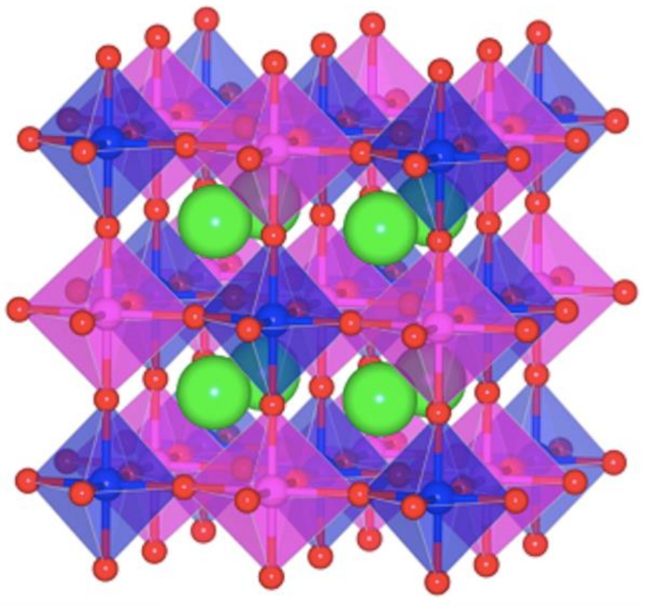


Adding local Coulomb interaction:

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.

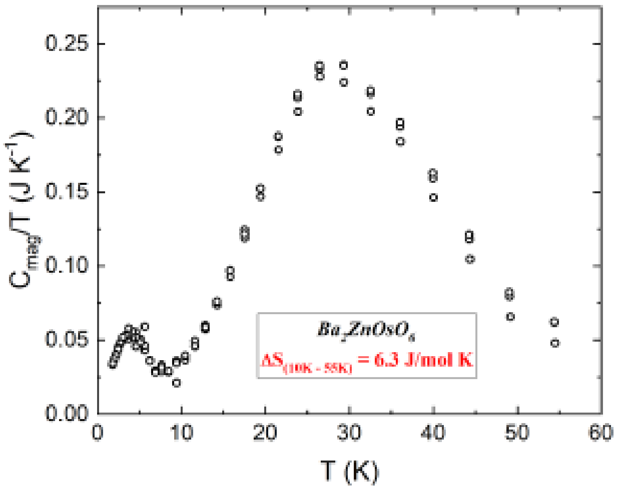
$5d^2$ Double Perovskites: Puzzles around hidden J=2 magnetism



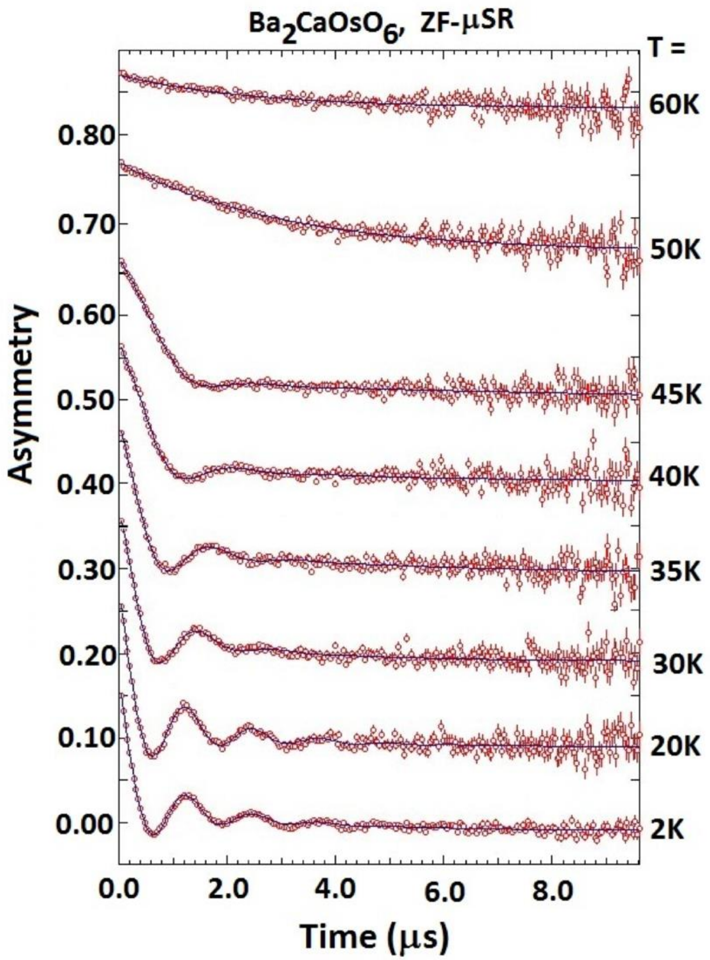
Ba_2MgOsO_6
Ba_2CaOsO_6
Ba_2ZnOsO_6

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

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



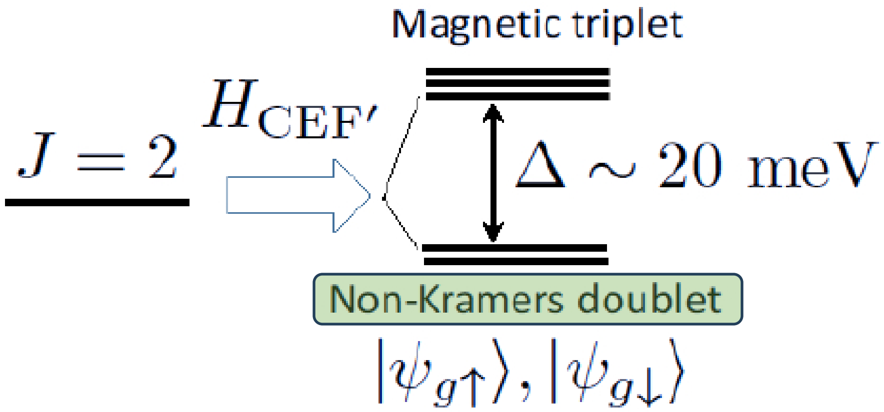
μSR: Time reversal broken
Very weak internal field



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!

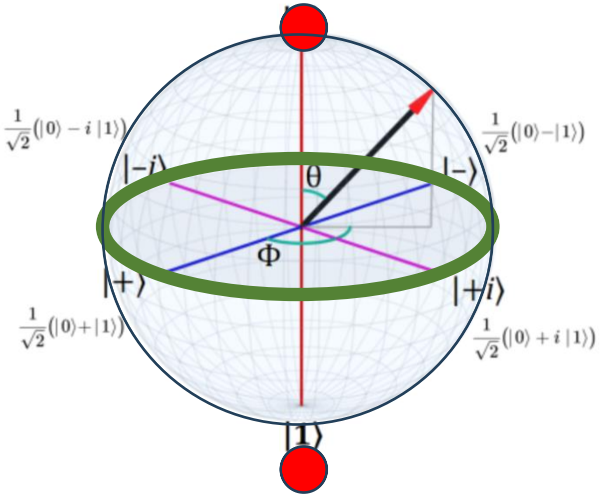
First example of d-orbital octupolar order

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)
NMR: S. Voleti, et al, AP, npjQM (2023)

Can we use phonons to probe and control hidden orders?

Non-Kramers doublet on the Bloch sphere



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

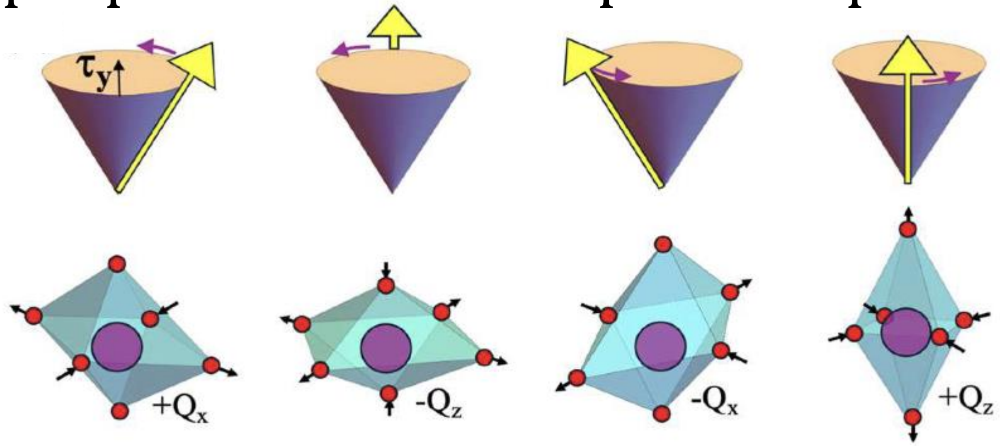
- Eg phonons linearly couple to quadrupole moment

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

- Can we use phonons as a fictitious "field" to probe and manipulate the state (like NMR)?

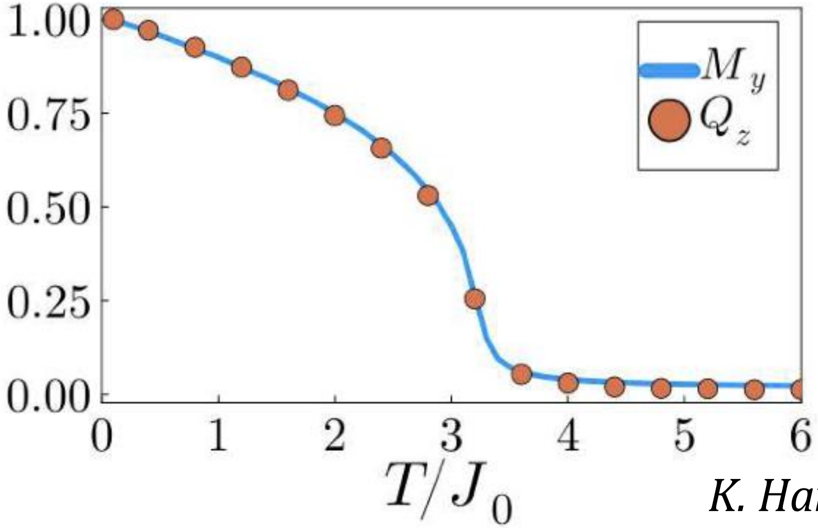
Pump-probe: Kick X-mode, track Z-mode

Pseudospin precession drives phonon "precession"

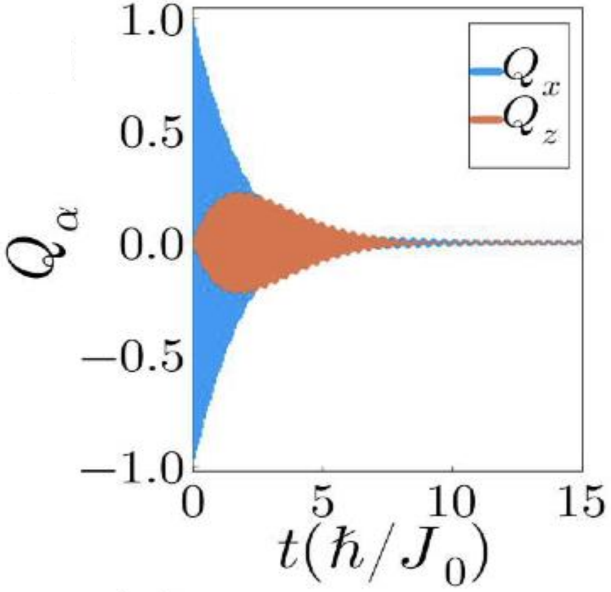


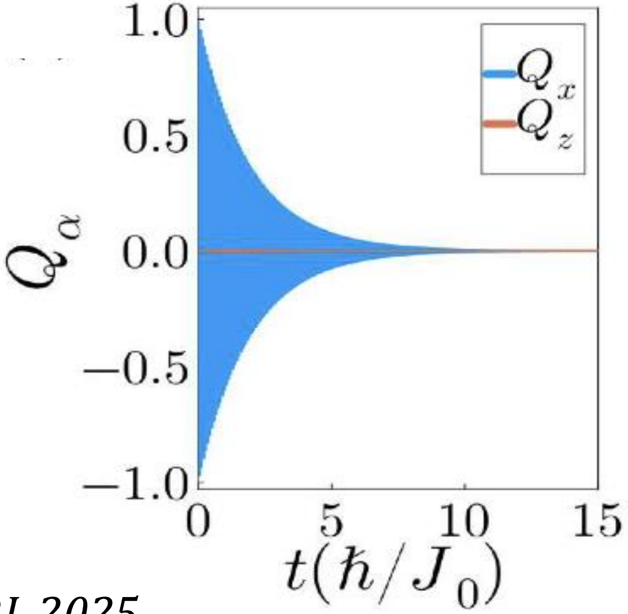
$$H_{\mathrm{sp-ph}} = -\lambda \sum_{\mathbf{r}} \left[Q_x(\mathbf{r})\tau_{\mathbf{r}x} + Q_z(\mathbf{r})\tau_{\mathbf{r}z} \right]$$

Computed cross-response tracks octupolar order



Spin-Phonon Molecular Dynamics Simulations





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

Phonon Green function

- Eg 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 *$\mathcal{O}(1/S)$*.

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

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

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

Phonon Green function

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Information about local Weiss field strength and orientation

Phonon Green function

- Eg 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 $\mathcal{O}(1/S)$.

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

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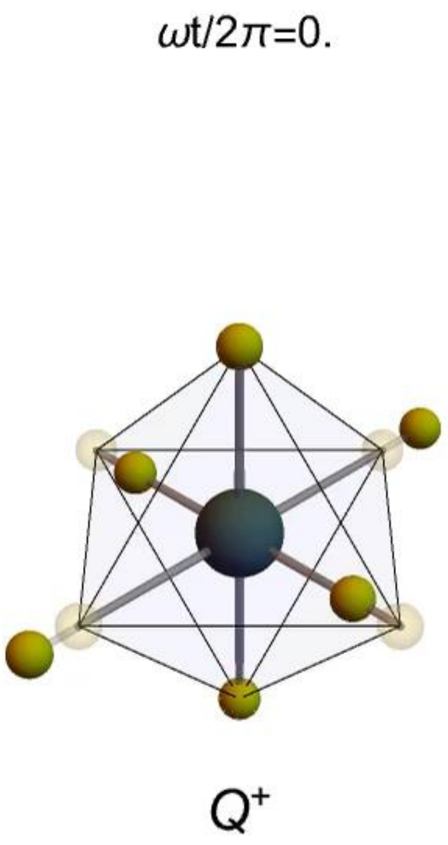
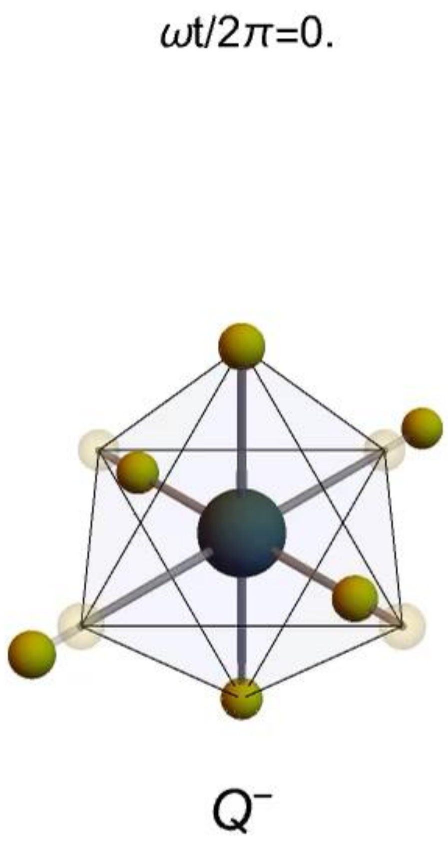
$$\mathcal{G}_{zz}^{-1}(\mathbf{r}, i\nu_n) = -i\nu_n + \Omega_0 - 4S\lambda^2 \left[\frac{2h_\mathbf{r}\Delta_\mathbf{r}^z}{(i\nu_n)^2 - 4h_\mathbf{r}^2}\right]$$

- 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





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

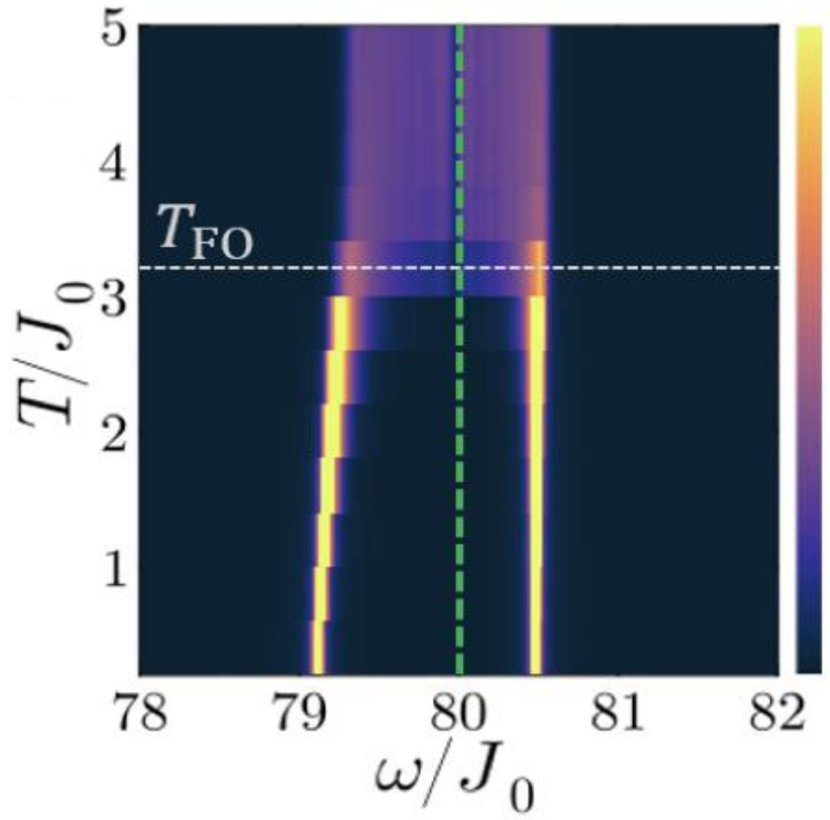
$$R'(Q^-) = \begin{pmatrix} i\,a^- & 0 & \sqrt{2}\,a^- \\ 0 & -i\,a^- & i\,\sqrt{2}\,a^- \\ \sqrt{2}\,a^- & i\,\sqrt{2}\,a^- & 0 \end{pmatrix}$$

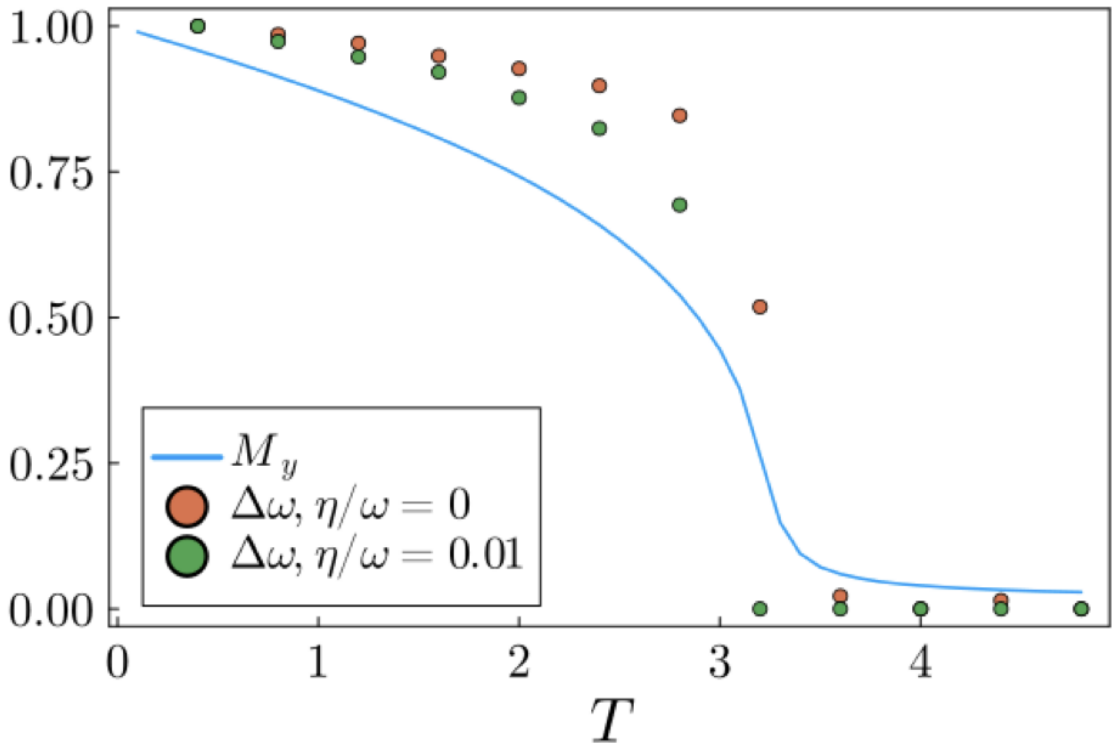
$$|\sigma^+ \cdot R'(Q^+) \cdot \sigma^-|^2 = |a^+|^2$$

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

Detect using Raman scattering
in cross-circular channel

Phonon spectral function





Probe using cross-circular Raman scattering

R. Sutcliffe, K. Hart, S. Chaudhary, AP (PRL: under review)

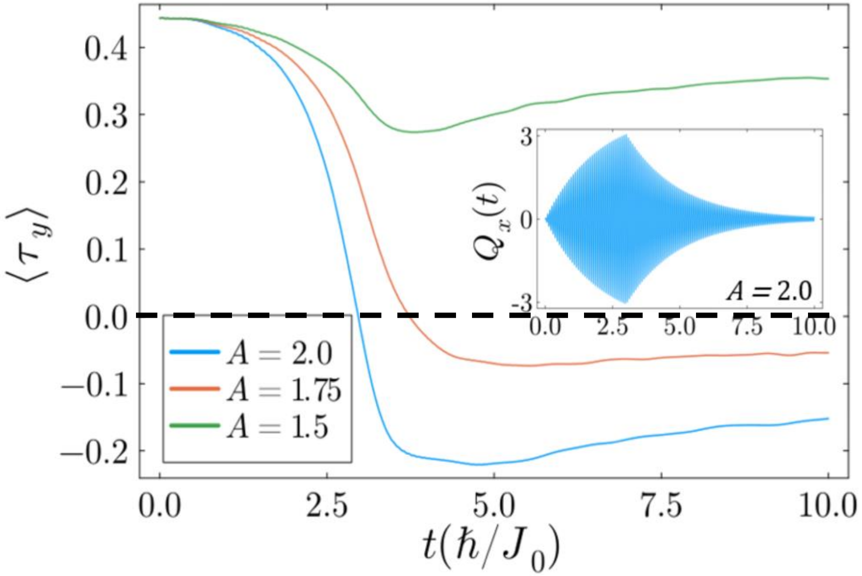
Controlling hidden order via phonons

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

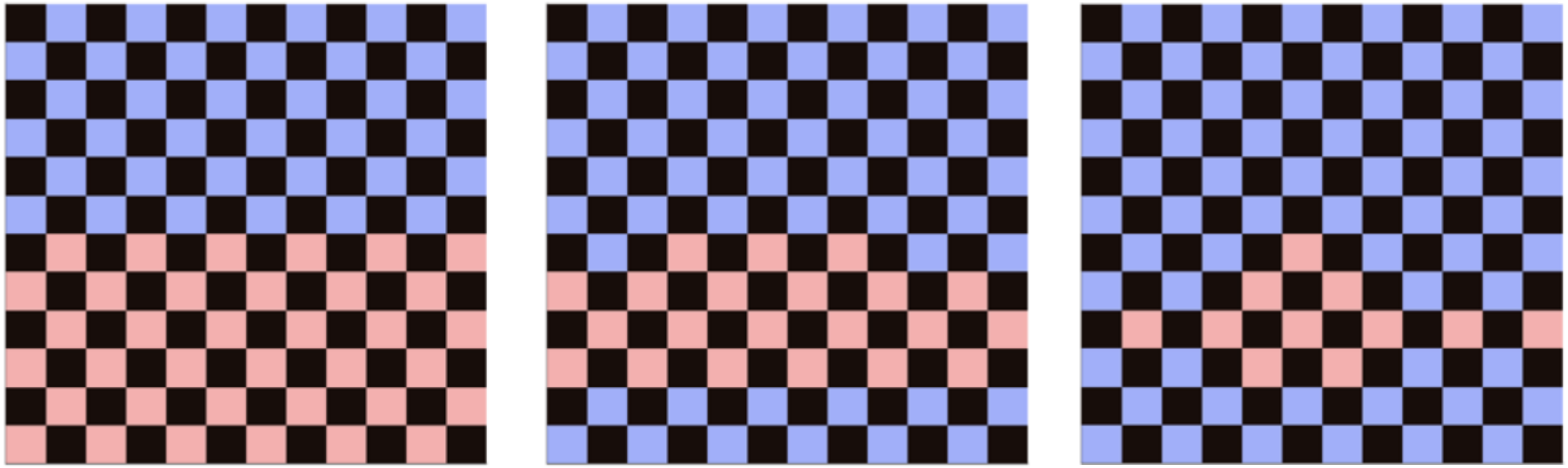
$$H^{\mathrm{sp}}_{\mathrm{drive},0} = 0; \quad H^{\mathrm{sp}}_{\mathrm{drive},\pm} = -\frac{\lambda Q_0}{2}\sum_{\mathbf{r}}(\tau_{\mathbf{r}x} + e^{\mp i\phi}\tau_{\mathbf{r}z})$$

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



Phonon driven switching of octupolar order

Chiral phonon driven growth of octupolar domains



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

Spin-1 coupled to phonons: SU(3) Monte Carlo and Dynamics

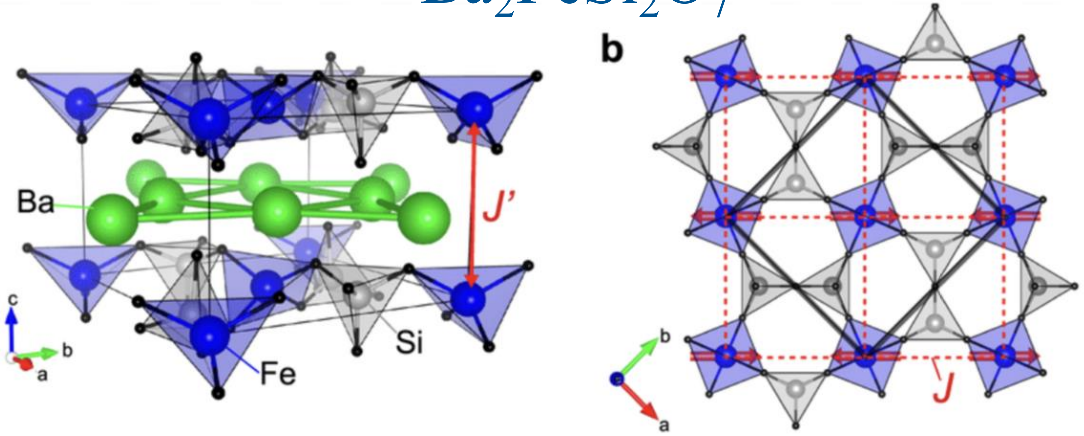
$S_z = \pm 1$ Exciton

D

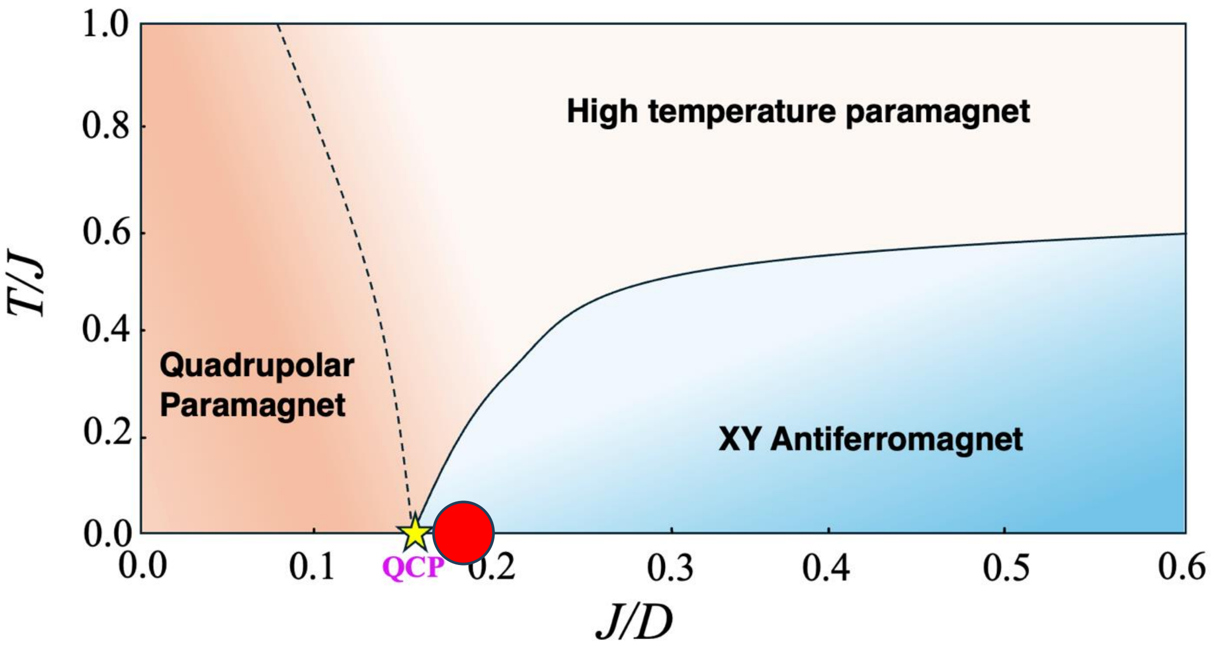
$S_z = 0$ Ground state

$$H = DS_z^2$$

$Ba_2FeSi_2O_7$



$$H_{\rm sp} = \sum_{\langle \mathbf{r},\mathbf{r}' \rangle} J_{\mathbf{r}\mathbf{r}'} \mathbf{S}(\mathbf{r}) \cdot \mathbf{S}(\mathbf{r}') + D \sum_{\mathbf{r}} S_z^2(\mathbf{r})$$



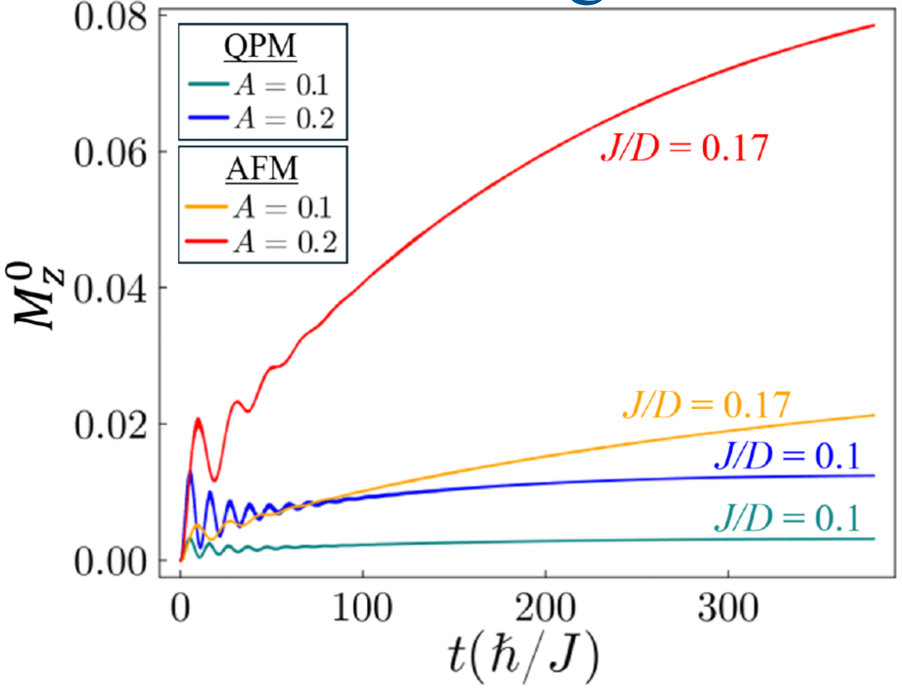
Consider phonons: $(Q_{xz},\ Q_{yz})$

Couple to $\begin{cases} T_{xz} = S_x S_z + S_z S_x \\ T_{yz} = S_y S_z + S_z S_y \end{cases}$

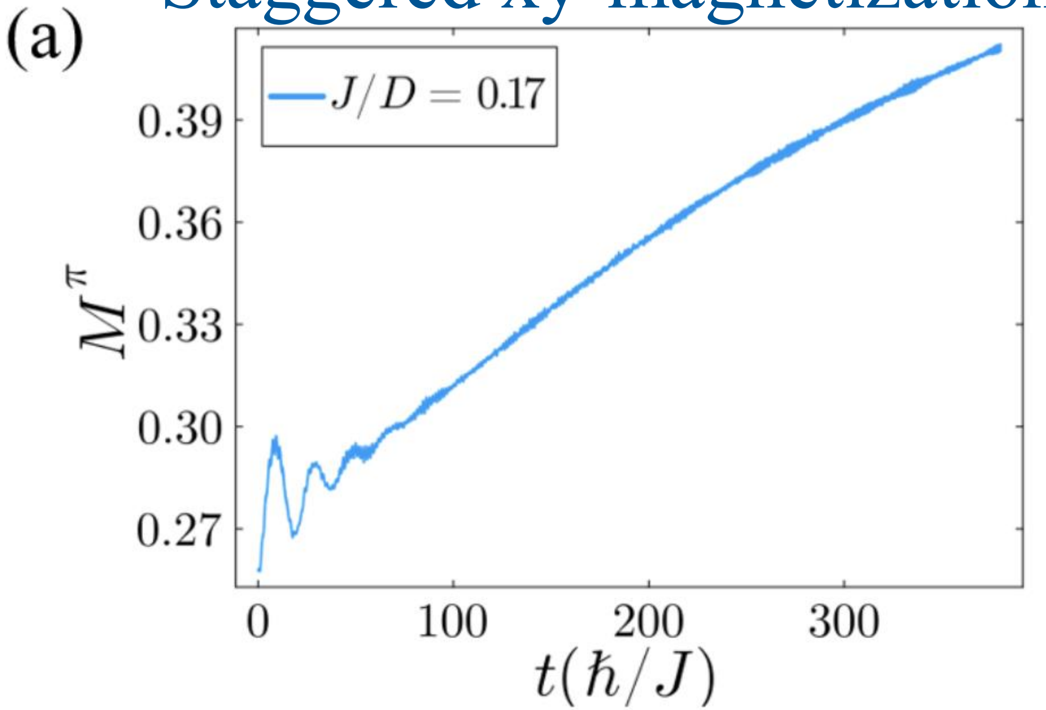
Impact of Chiral Phonon Drive

$$H_{\text{drive}}^{\text{ph}} = -A \sum_{\mathbf{r}} \left(Q_{xz}(\mathbf{r}) \cos \Omega t + Q_{yz}(\mathbf{r}) \sin \Omega t \right)$$

Uniform z-magnetization



Staggered xy-magnetization



Creates a nonzero Sz magnetization, strengthens AFM order

Phonon induced <Sz> could be potentially detected via Kerr rotation

Chiral Phonon Drive: High Frequency Magnus Expansion

$$\begin{bmatrix} H_0 - \Omega & H_{\text{drive}} & \\ H_{\text{drive}} & H_0 & H_{\text{drive}} \\ & H_{\text{drive}} & H_0 + \Omega \end{bmatrix}$$

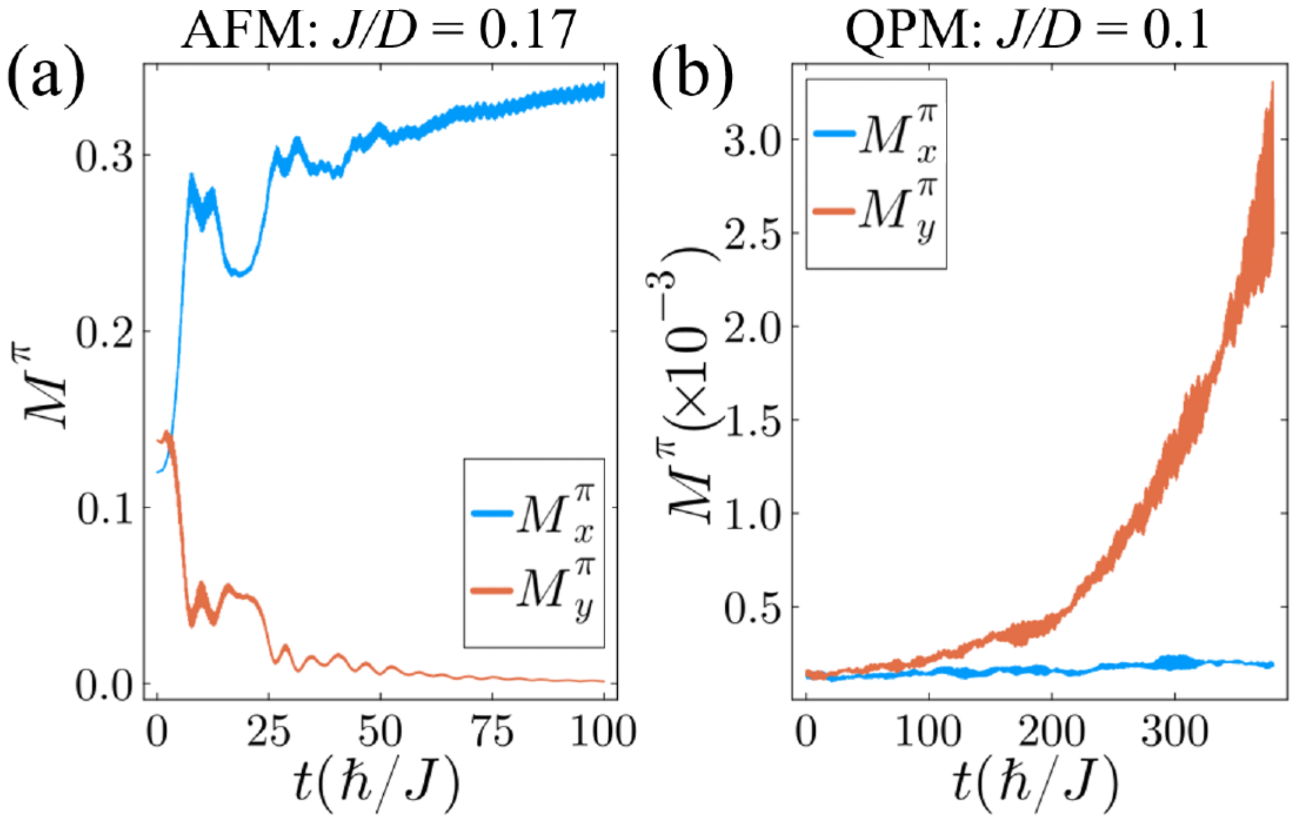
$$H^{\text{sp}}_{\text{drive},0} = 0; \;\; H^{\text{sp}}_{\text{drive},\pm} = -\frac{\lambda Q_0}{2} \sum_{\mathbf{r}} (T_{xz}(\mathbf{r}) \mp i T_{yz}(\mathbf{r}))$$

$$H^{\text{sp,eff}}_{\text{drive}} = \frac{1}{\Omega}[H^{\text{sp}}_{\text{drive},+}, H^{\text{sp}}_{\text{drive},-}] = -\frac{\lambda^2 Q_0^2}{2\Omega} \sum_{\mathbf{r}} S_z(\mathbf{r})$$

- Chiral phonon drive generates an effective Floquet magnetic field
- Lowers Heisenberg symmetry to XY symmetry suppressing AFM fluctuations

Impact of Single Phonon Drive

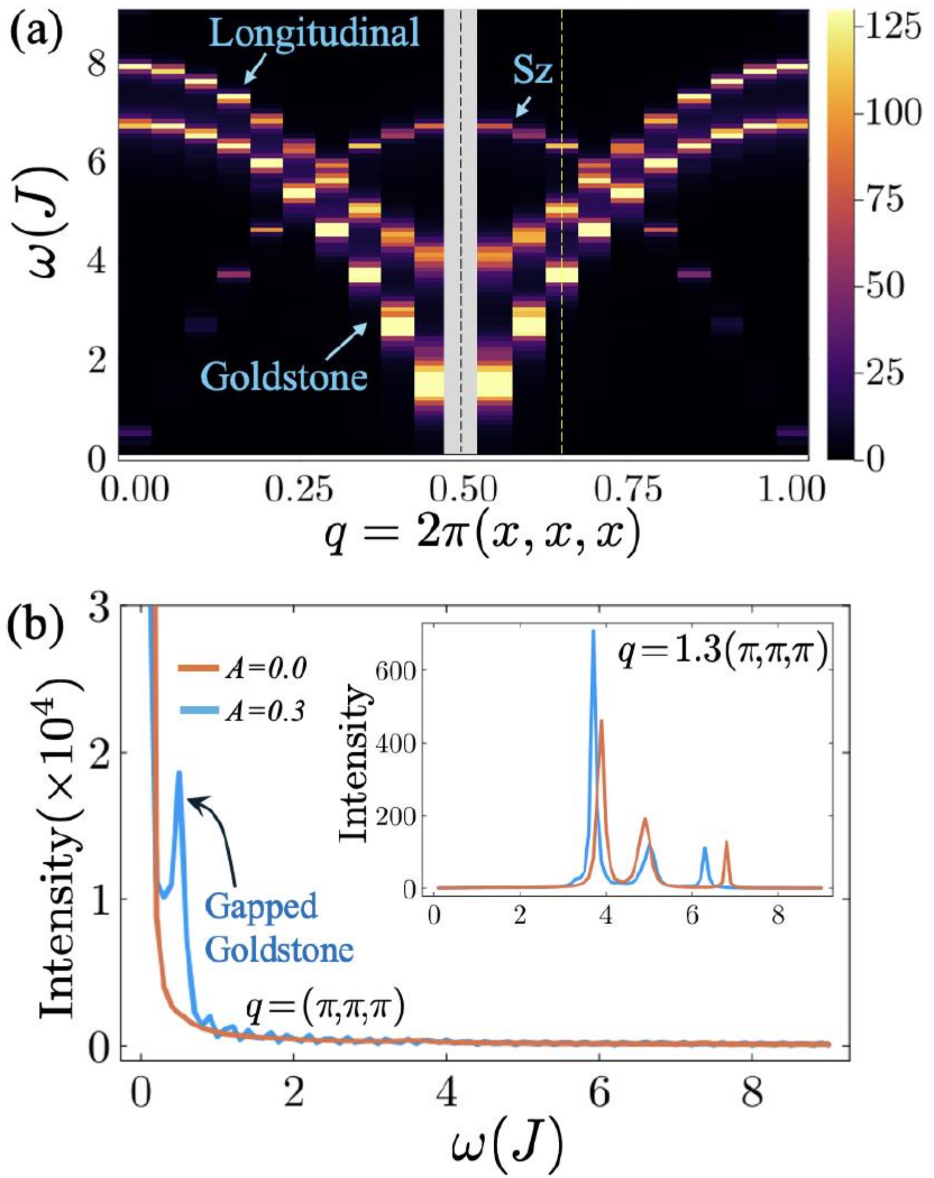
$$H^{\mathrm{ph}}_{\mathrm{drive}} = -A \sum_{\mathbf{r}} \cos(\Omega t) Q_{xz}(\mathbf{r})$$



- Creates uniaxial strengthening of AFM order, breaking XY symmetry
- Creates weak Ising AFM order in the quadrupolar paramagnet

Single Phonon Drive: Gapping the Nambu-Goldstone mode

- Compute S(q,ω) in the full driven system on large lattice sizes
- Phonon drive renormalizes all branches
- Emergence of a gapped NG mode reflects weak drive-induced Ising anisotropy



Collaborators

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ISSP Tokyo

Swati Chaudhary

Bruce Gaulin

Dalini Maharaj

McMaster

Gabriele Sala, Matt Stone,
Edwin Kermarrec, Clemens Ritter,
Francois Fauth, Casey Marjerrison,
John Greedan, Graeme Luke

Caltech

Gil Refael

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