Inherited TRS Breaking in Chiral Phonons

From Giant Magnetic Moments to Probes of Hidden Order and local Berry curvature

Swati Chaudhary
(Prof. Takashi Oka's group, Institute for Solid State Physics, University of Tokyo)

THE INSTITUTE FOR SOLID STATE PHYSICS
THE UNIVERSITY OF TOKYO

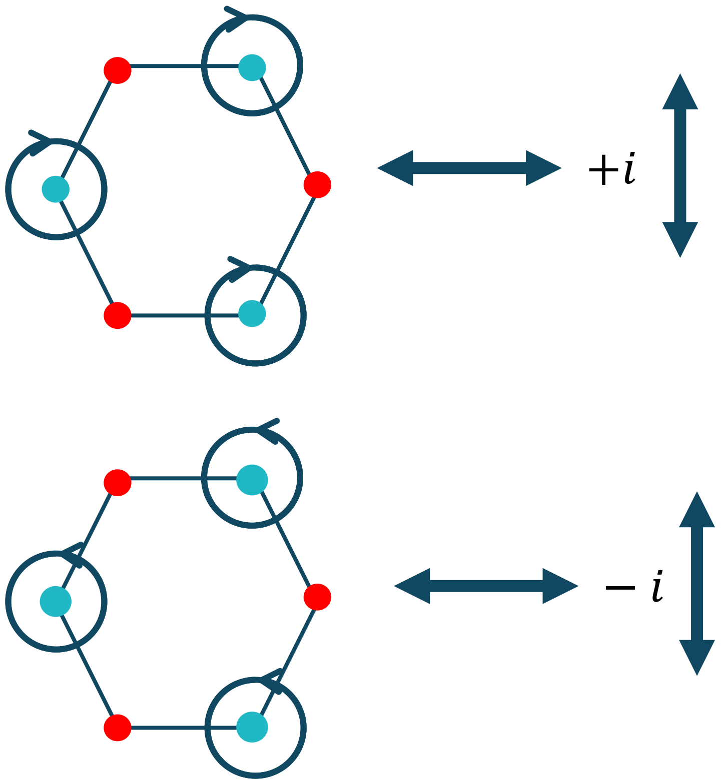
Outline

Chiral phonon, angular momentum, and splitting

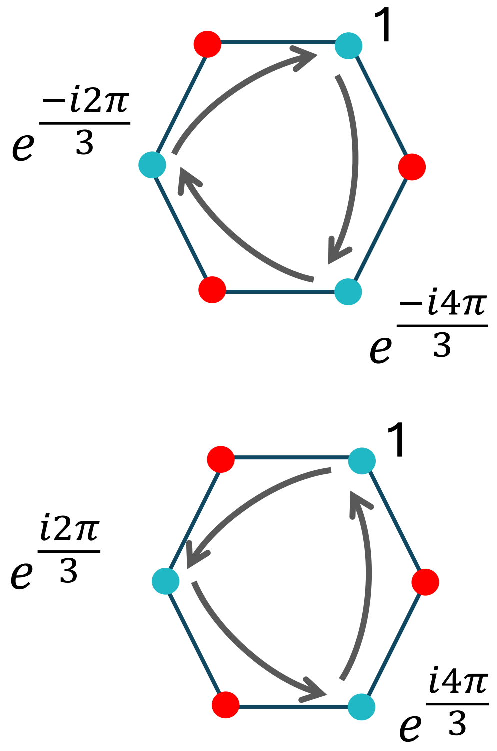
Phonon chirality and TRS breaking

- Giant Magnetic Moments
- Probe of multipolar order
- Imprints of local Berry Curvature

Chiral phonons carry angular momentum



$$C_n \mathbf{u}_{\nu\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_{l\alpha}} = e^{-(2\pi i/n)l_{ph}} \mathbf{u}_{\nu\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_{l\alpha}}$$



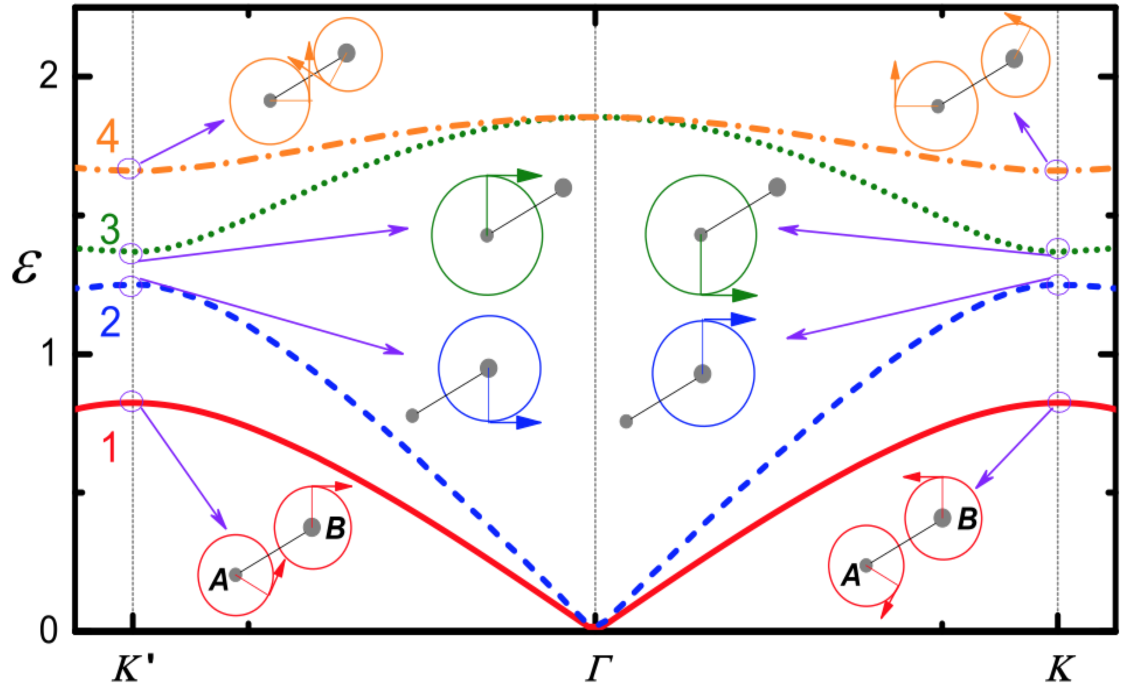
$$\boldsymbol{L} = \boldsymbol{Q} \times \partial_t \boldsymbol{Q}$$

Lifa Zhang, Qian Niu, Phys. Rev. Lett. **112**, 085503 (2014)
Lifa Zhang, Qian Niu, Phys. Rev. Lett. **115**, 115502 (2015)

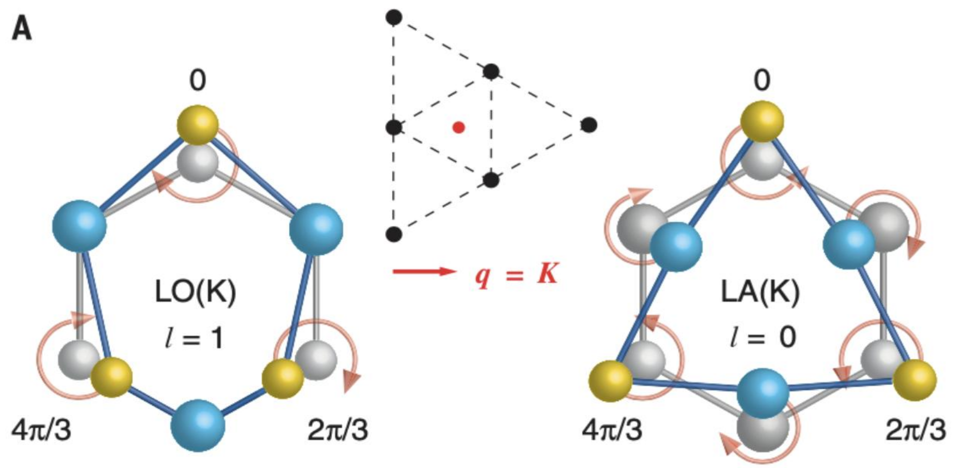
S.C, C.P. Romao, D.Juraschek, arXiv:2504.19121 (2025)

Chiral phonons from Broken inversion

Valley chiral phonons in a hexagonal lattice



Zhang, Lifa, *et.al* PRL **115**, 115502 (2015)

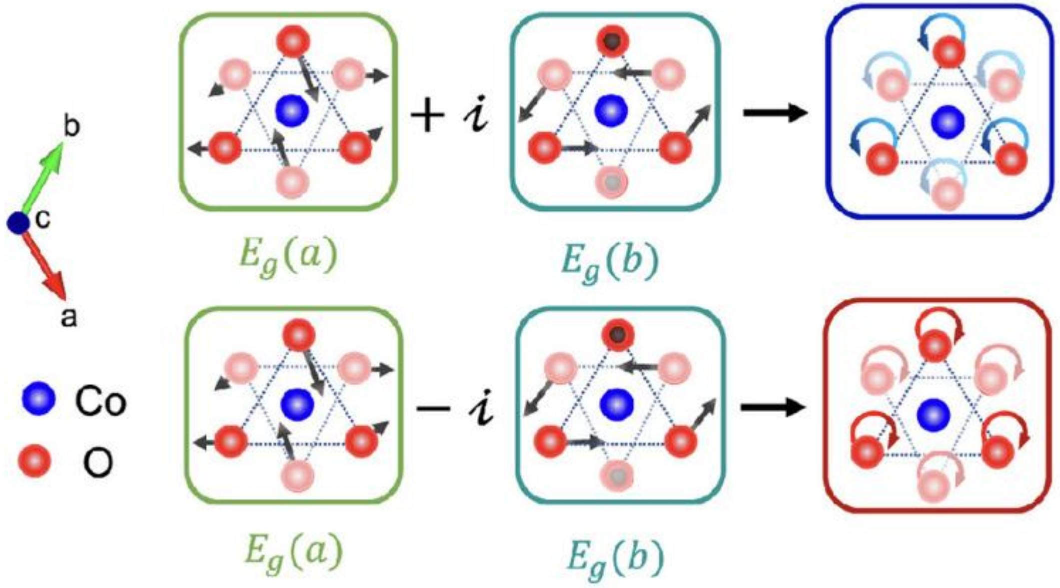


WSe_2

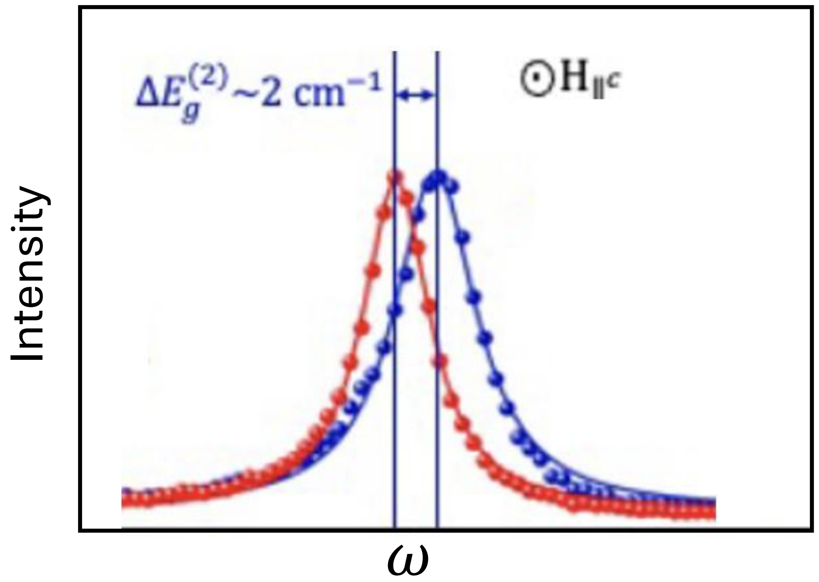
Zhu, Hanyu, et al. *Science* 359.6375 (2018): 579-582.

Zone-centered Chiral phonons require TRS breaking

- ❖ TRS $L \rightarrow -L$



- ❖ TRS breaking lifts degeneracy



D. Lujan*, J. Choe*, **S. Chaudhary***, G. Fiete, & Xiaoqin Li *et al.*, PNAS, 121(11), e2304360121 (2024)

Bonini, John *et.al* PRL **130**, 086701 (2023)

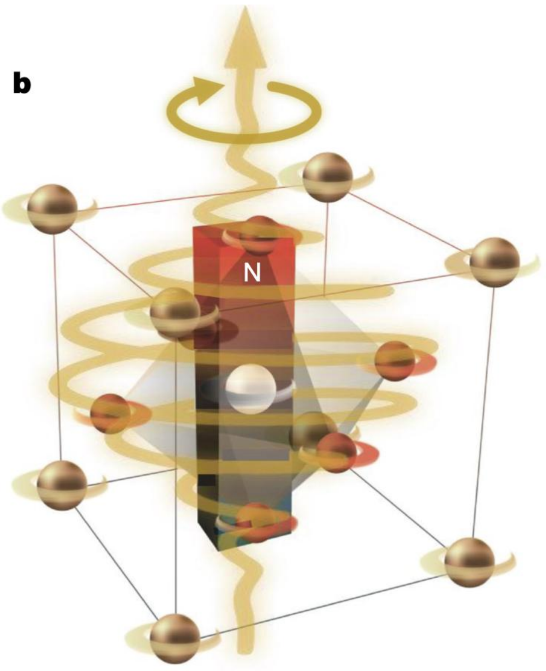
Che, Mengqian, et al. PRL **134**, 196906 (2025)

Zhang *et.al*. PRL **130**, 226302(2023)

Yin *et.al Advanced Materials* 33.36 (2021): 2101618.

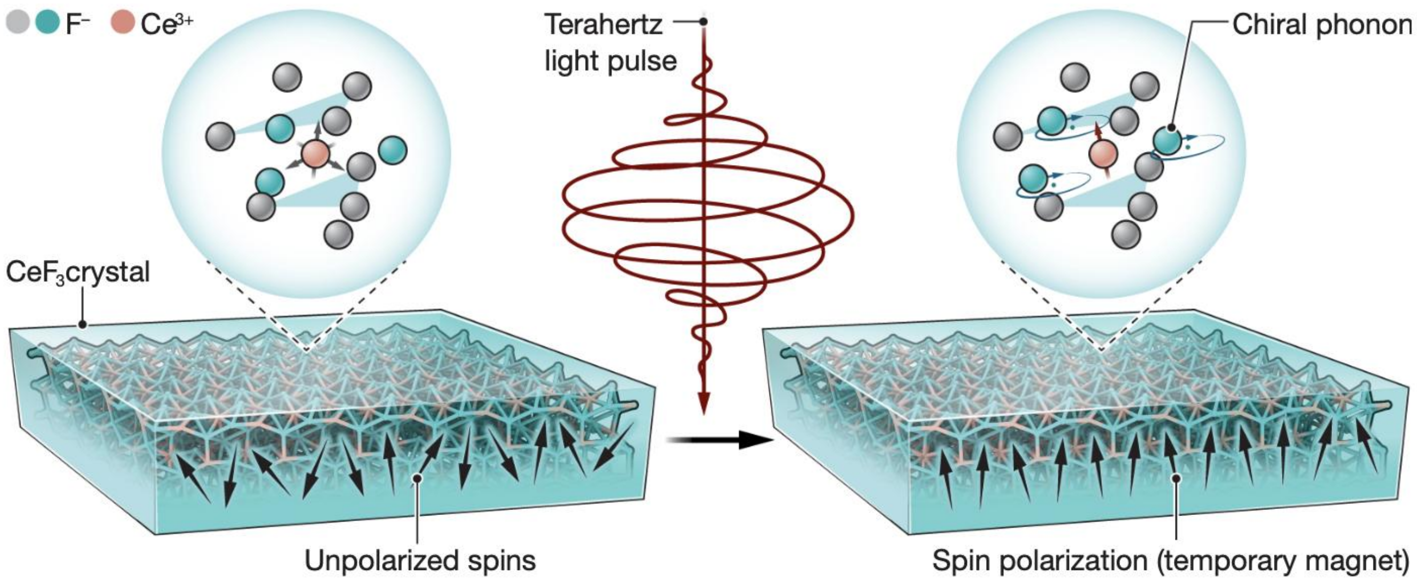
Chiral phonons as mediators between light and magnetism

Phonon Barnett Effect



M. Basini et.al, Nature **628**, 534–539 (2024)
Davies *et.al,* Nature **628**, pages 540–544 (2024)

Giant effective magnetic fields from chiral phonons



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

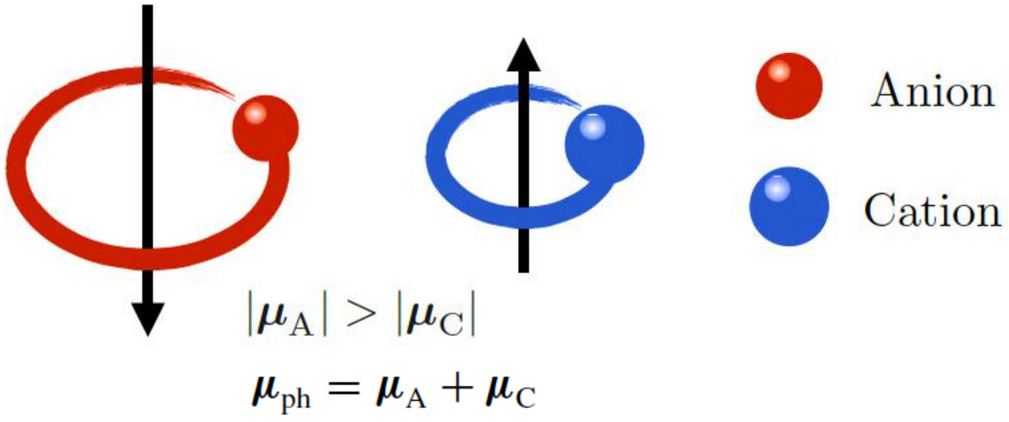
Magnetic response of phonons

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete, Phys. Rev. B **110**, 094401 (2024)

D. Lujan, J. Choe, **S. Chaudhary**, G. Fiete, R. He, & Xiaoqin Li *et al., Spin-Orbit Exciton-Induced Phonon Chirality in a Quantum Magnet* PNAS, 121(11), e2304360121 (2024)

S. Chaudhary*, C.P. Romao*, D. Juraschek, arXiv:2504.19121 (2025)

Magnetic response: classical picture



Material	μ_{ph} (μ_N)	$\Delta\omega_{Zeeman}$ at 100 T	$\Delta\omega_{Zeeman}$ at 1000 T
CsH	1.12	0.06 cm^{-1}	0.6 cm^{-1}
$BaHfO_3$	0.12	0.006 cm^{-1}	0.06 cm^{-1}
$KTaO_3$	0.12	0.006 cm^{-1}	0.06 cm^{-1}
$KNbO_3$	0.18	0.009 cm^{-1}	0.09 cm^{-1}
$SrTiO_3$	0.18	0.009 cm^{-1}	0.09 cm^{-1}

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

Phonon Magnetic moment $\mu_{ph} = \gamma \boldsymbol{L}$

Gyromagnetic ratio $\gamma = \sum_i \gamma_i (\mathbf{q}_{i,x} \times \mathbf{q}_{i,y}),$

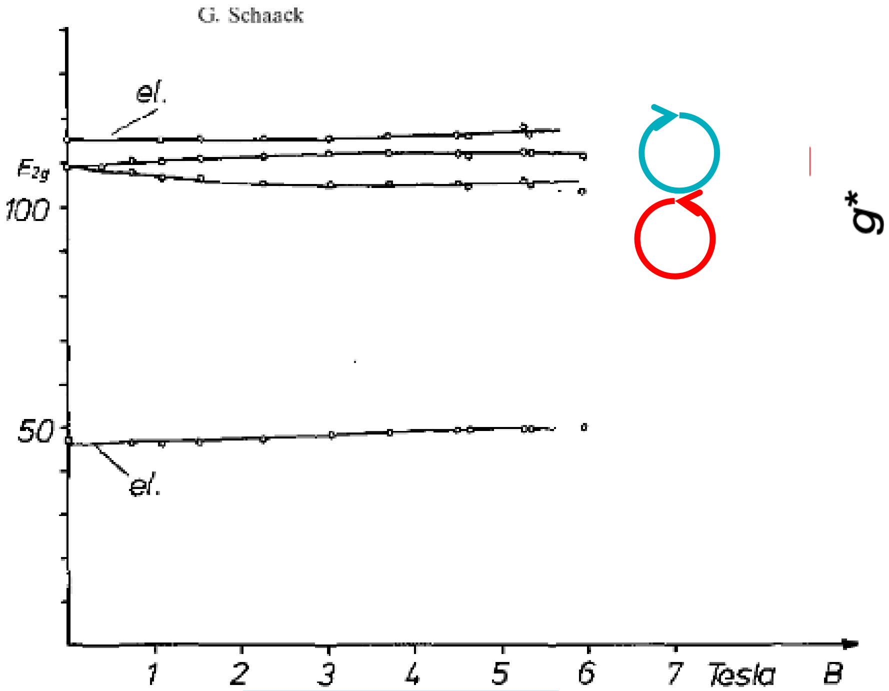
$$\gamma_i = \frac{eZ_i}{2M_i} \longrightarrow \mu_{ph} \approx \mu_N \approx 5 \text{ X } 10^{-4} \mu_B$$

Phonon Zeeman effect $\Delta\omega_{Zeeman} = 2\boldsymbol{\mu}_{ph} B$

Juraschek, Dominik M., and Nicola A. Spaldin. Phys. Rev. Mat. **3** (6) (2019)

Observed Magnetic moment

Rare-Earth Paramagnet $CeCl_3$

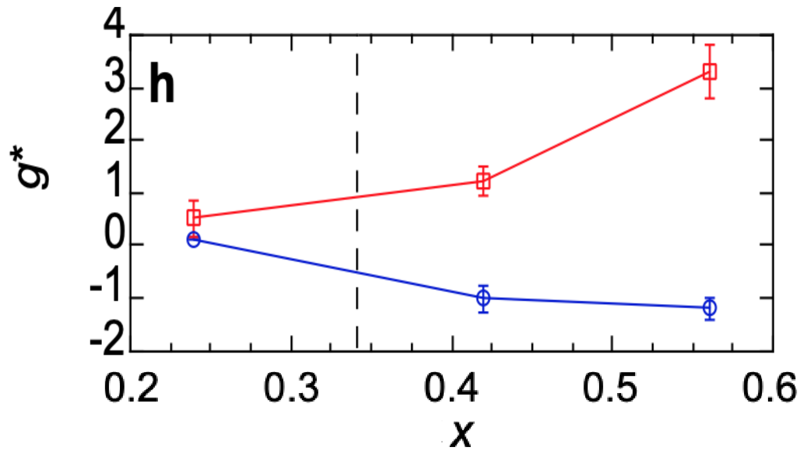


$\mu_{ph} \sim 3\ \mu_B$

G. Schaack *et al.* Z. Physik (1977)

$Pb_{1-x}Sn_xTe$

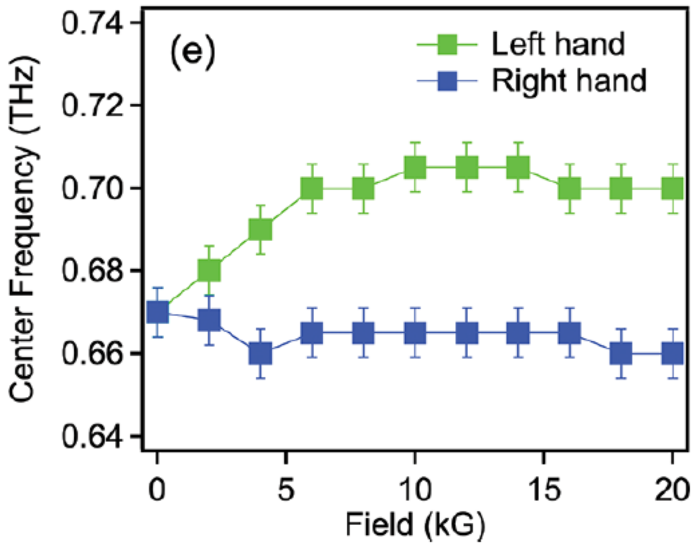
TCI(x>0.32)



$\mu_{ph} \sim \mu_B$

Hernandez, Baydin, Chaudhary, *et al*. Sci. Adv.**9**,eadj4074 (2023).

Dirac SM Cd_3As_2



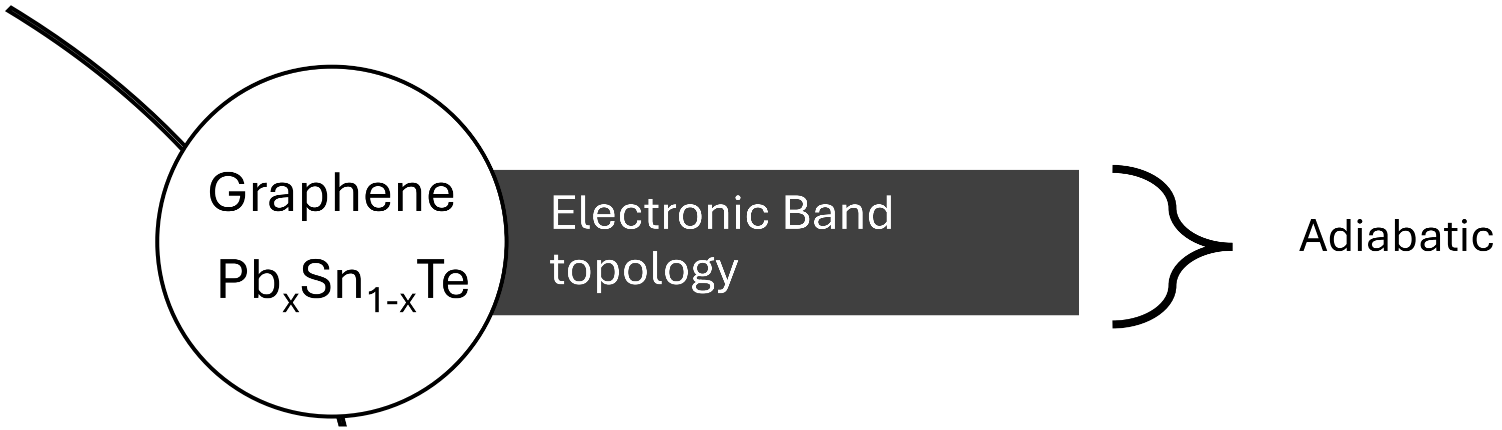
$\mu_{ph} \sim 3\ \mu_B$

B. Cheng. N.P. Armitage *et al.* Nano Lett. 2020, 20, 5991-5996

Possible mechanisms for phonon magnetic moment

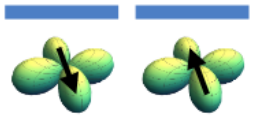
Electronic Contribution

Ren, Y. *et al*. Phys. Rev. Lett. **127**, 186403 (2021)
X. Zhang *et al*. Phys. Rev. Lett. **130**, 226302 (2023)
F. Hernandez, A. Baydin, S. Chaudhary, *et al*. *Sci. Adv.* **9**, eadj4074 (2023).



Phonon magnetic moment from orbital lattice coupling

Degenerate
orbital transition



Degenerate
chiral phonon

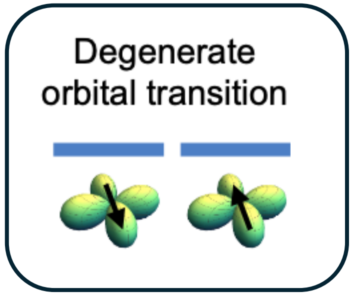
P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

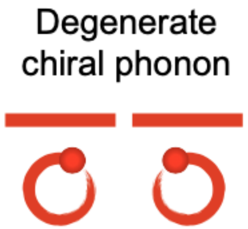
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete, Phys. Rev. B **110**, 094401 (2024)

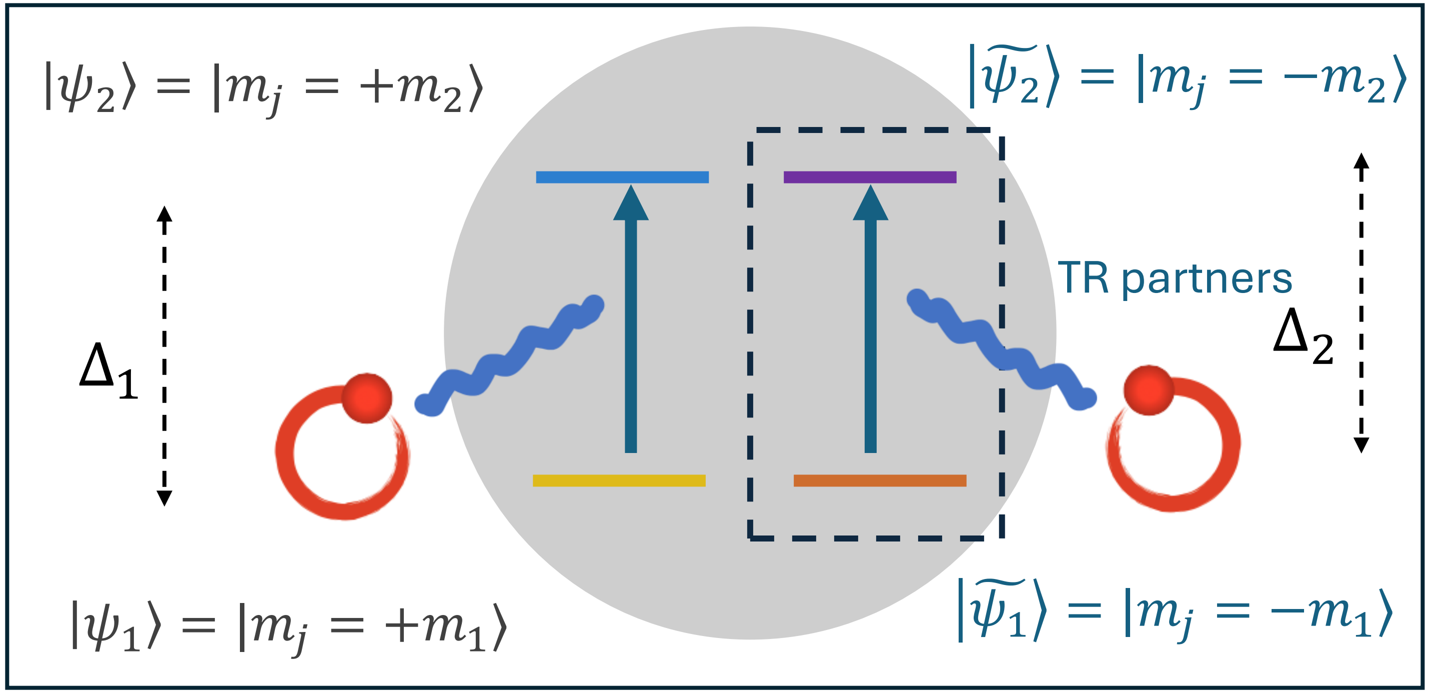
Phonon magnetic moment from orbital lattice coupling

1. Magnetic ion with two Kramers pair states
2. Chiral phonon coupling to excitation between Kramers pairs

$$H_{el-ph} = (u_x + iu_y)|\psi_1\rangle\langle\psi_2| + (u_x - iu_y)|\widetilde{\psi_1}\rangle\langle\widetilde{\psi_2}| + h.c$$



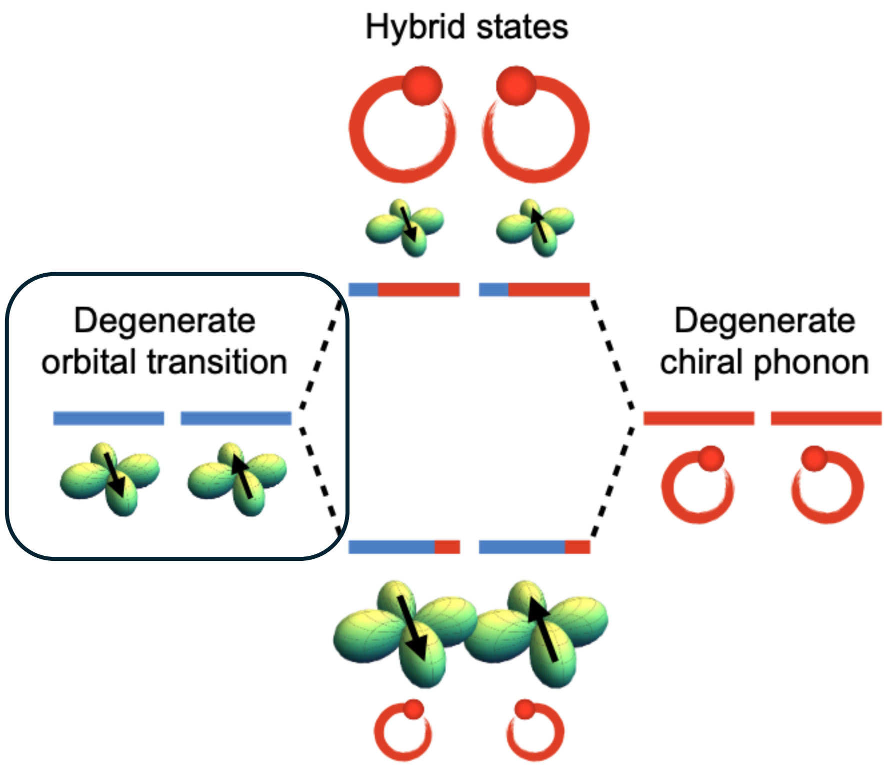




P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete, Phys. Rev. B **110**, 094401 (2024)

Phonon magnetic moment from orbital lattice coupling

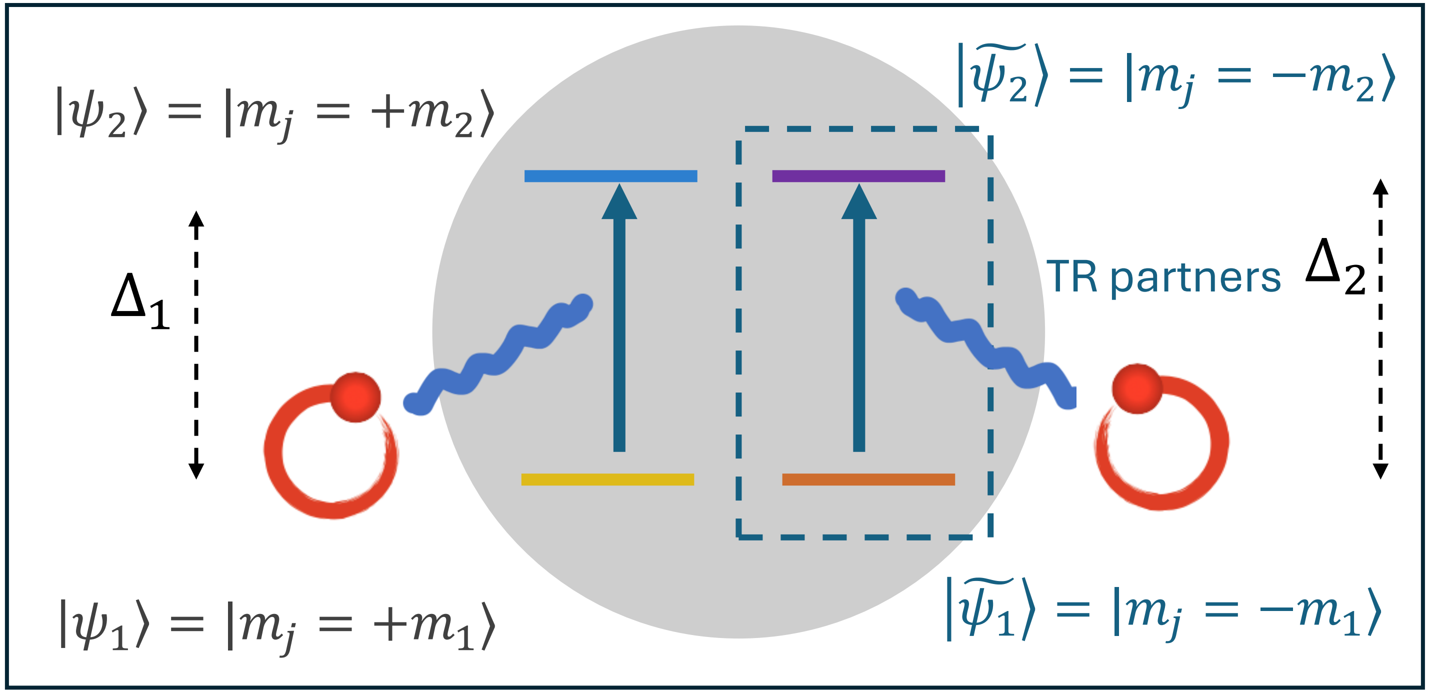


$B = 0$

$\Delta_1 = \Delta_2$

1. Magnetic ion with two Kramers pair states
2. Chiral phonon coupling to excitation between Kramers pairs

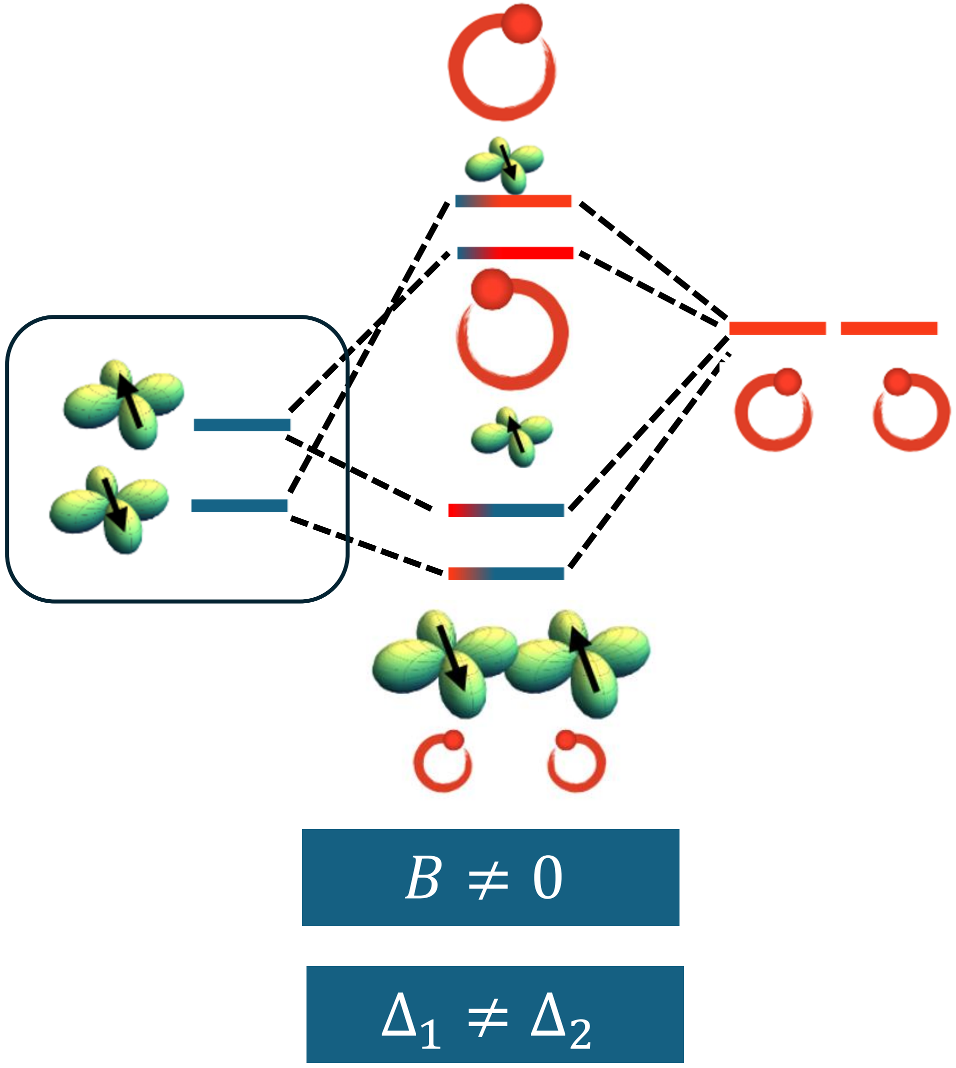
$$H_{el-ph} = (u_x + iu_y)|\psi_1\rangle\langle\psi_2| + (u_x - iu_y)|\widetilde{\psi_1}\rangle\langle\widetilde{\psi_2}| + h.c$$



P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

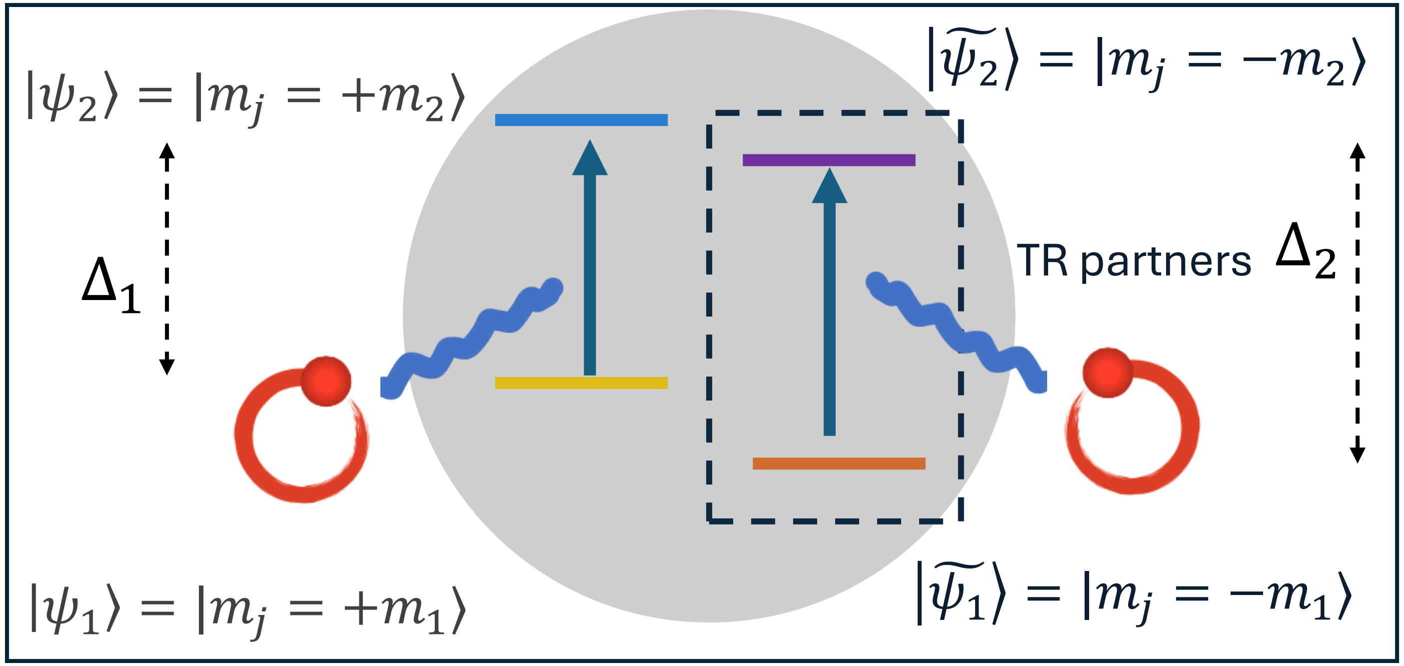
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete, Phys. Rev. B **110**, 094401 (2024)

Phonon magnetic moment from orbital lattice coupling



1. Magnetic ion with two Kramers pair states
2. Chiral phonon coupling to excitation between Kramers pairs

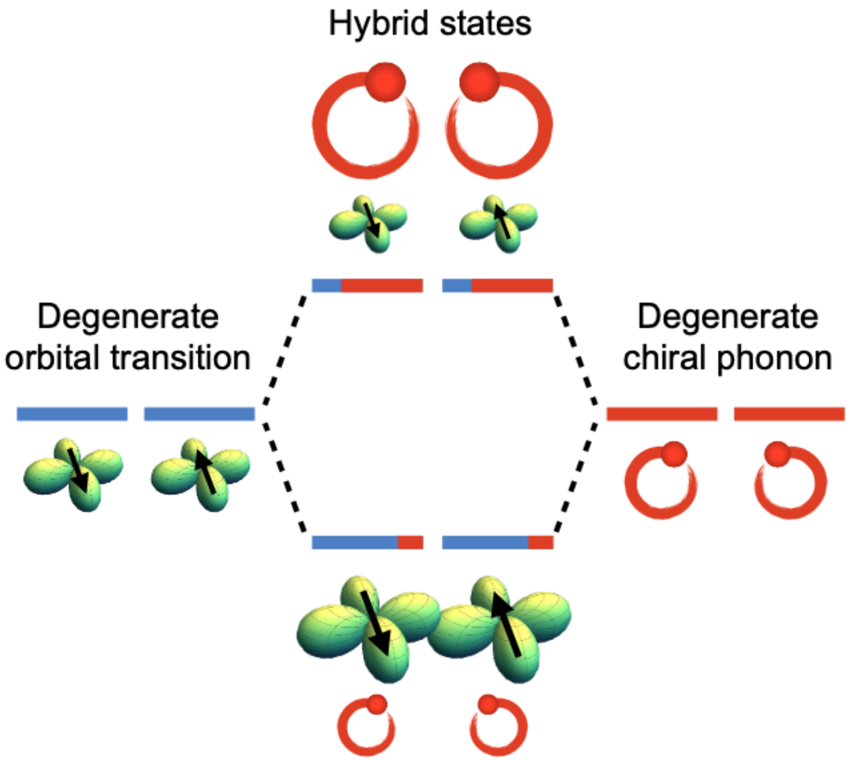
$$H_{el-ph} = (u_x + iu_y)|\psi_1\rangle\langle\psi_2| + (u_x - iu_y)|\widetilde{\psi_1}\rangle\langle\widetilde{\psi_2}| + h.c$$



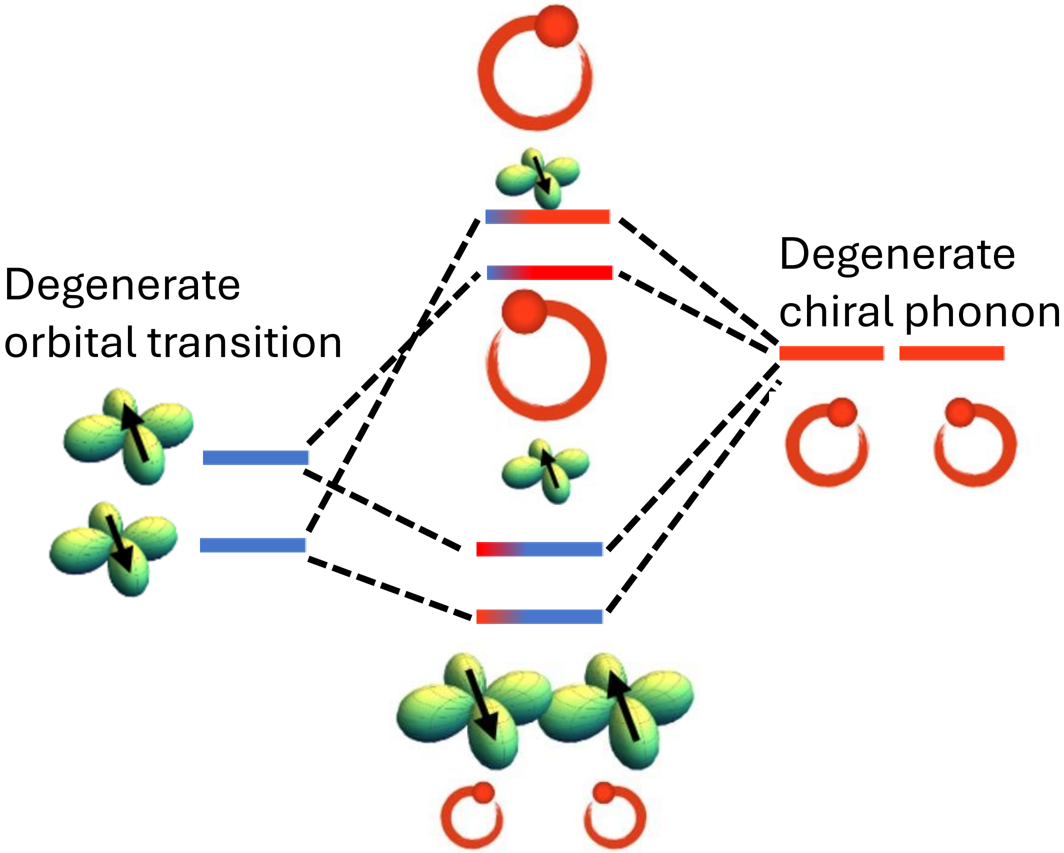
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

Splitting of chiral phonons

$B = 0$



$B \neq 0$



S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

Phonon magnetic moment from orbital lattice coupling

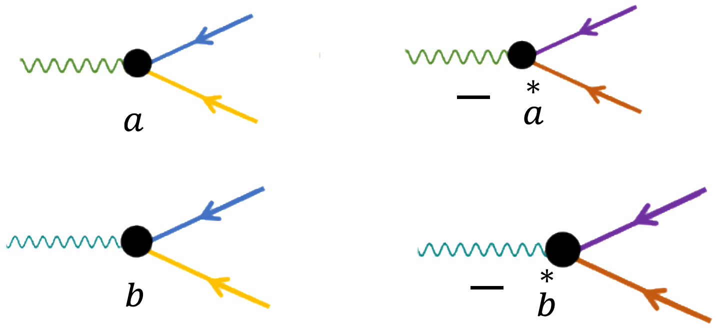
Electron-phonon interaction

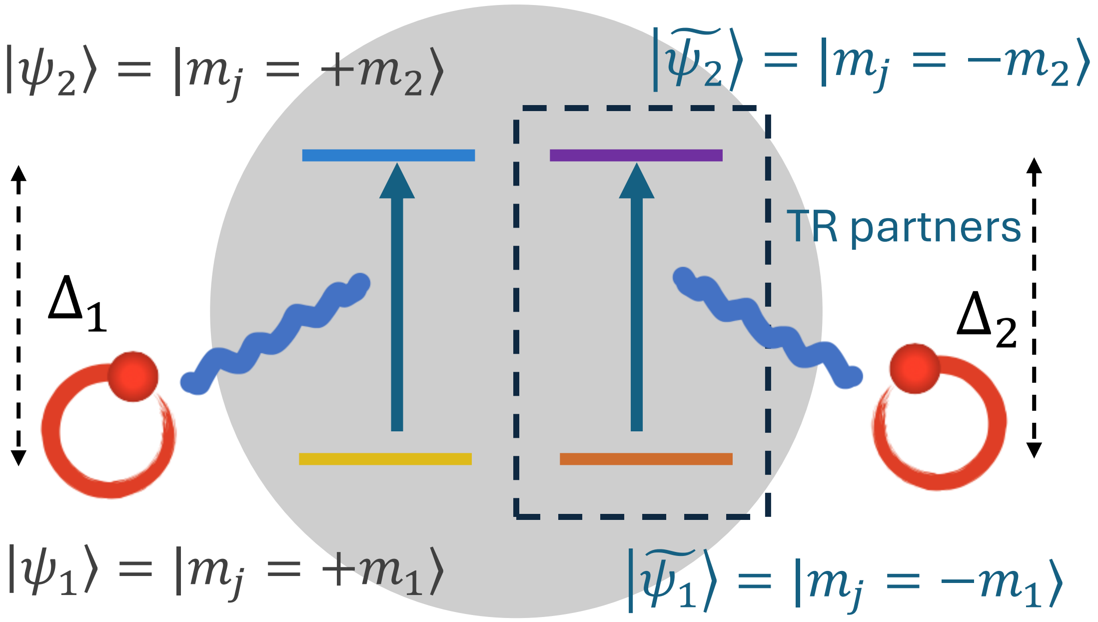
$$H_{ph} = \omega_0 a^\dagger a + \omega_0 b^\dagger b$$

$$H_{el-ph} = (a^\dagger + a)\widehat{\mathbf{O}}_a + (b^\dagger + b)\widehat{\mathbf{O}}_b$$

$$\hat{O}_a = g_a|\psi_1\rangle\langle\psi_2| - g_a^*|\psi_{\tilde{1}}\rangle\langle\psi_{\tilde{2}}| + \text{h.c}$$

$$\hat{O}_b = g_b|\psi_1\rangle\langle\psi_2| - g_b^*|\psi_{\tilde{1}}\rangle\langle\psi_{\tilde{2}}| + \text{h.c}$$





P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

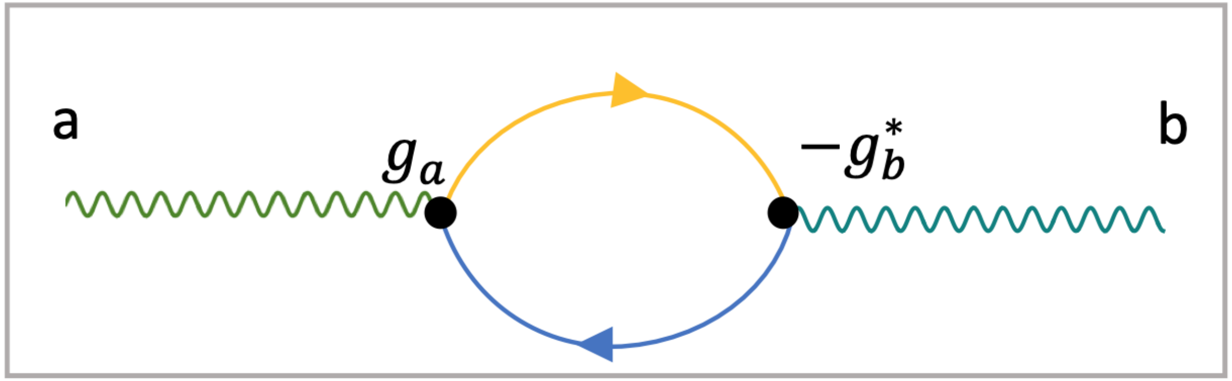
Phonon Green's function

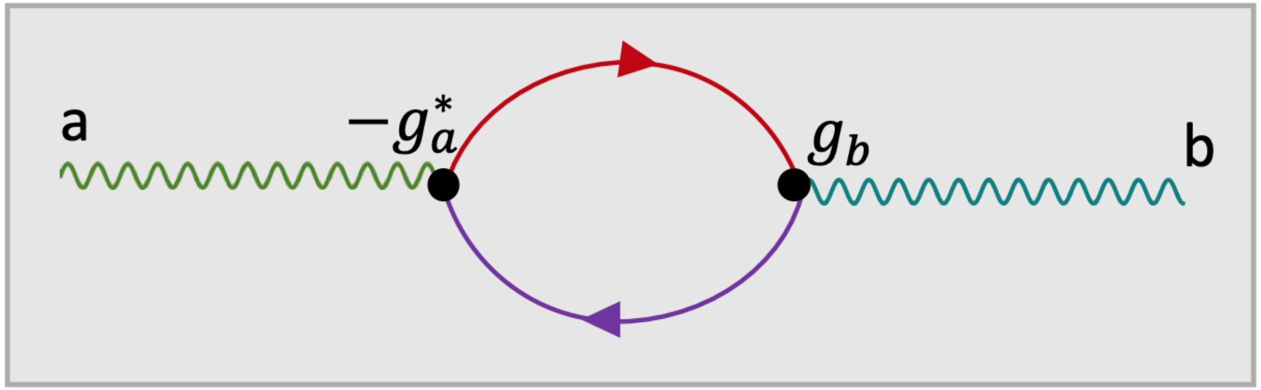
Non-interacting case

$$\widehat{\mathbf{D}}_{\mathbf{0}}(\omega) = \begin{pmatrix} D_0^{aa}(\omega) & 0 \\ 0 & D_0^{bb}(\omega) \end{pmatrix}$$

$$D_0^{aa}(\omega) = D_0^{bb}(\omega) = \frac{2\omega_0}{\omega^2 - \omega_0^2}$$

Corrections





P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

Phonon Green's function

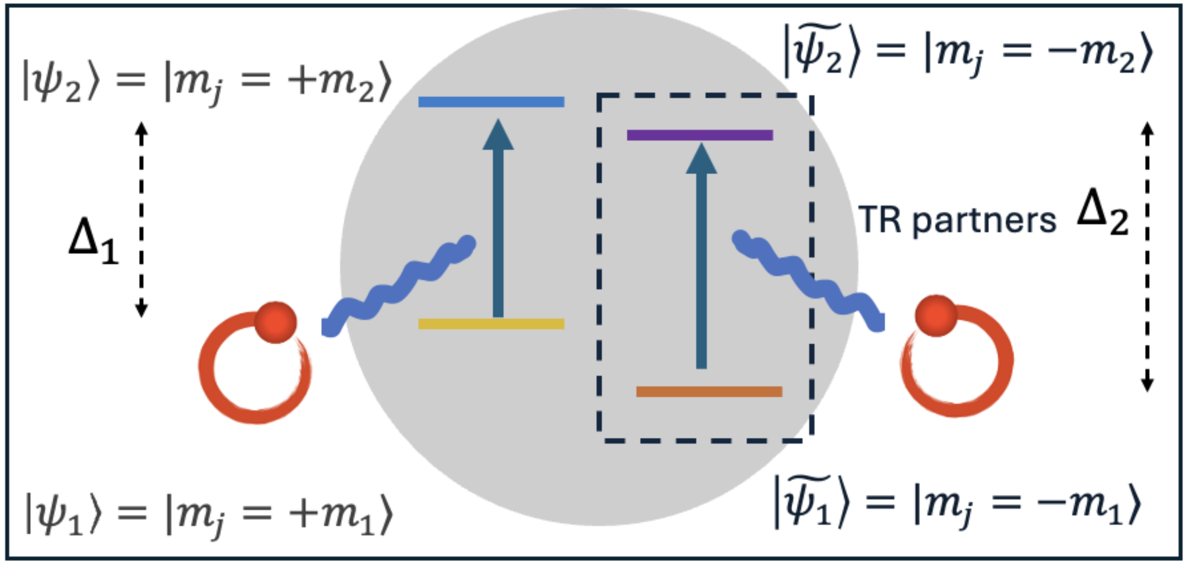
$$\mathbf{D}^{-1} = \begin{pmatrix} \frac{\omega^2-\omega_0^2}{2\omega_0} & -\Pi^{ab} \\ \Pi^{ab} & \frac{\omega^2-\omega_0^2}{2\omega_0} \end{pmatrix}$$

$$\Pi^{ab} \propto \frac{f_1 g_a g_b^*}{\omega - \Delta_1} - \frac{f_1 g_a^* g_b}{\omega + \Delta_1} + \frac{f_{\tilde{1}} g_a^* g_b}{\omega - \Delta_2} - \frac{f_{\tilde{1}} g_a g_b^*}{\omega + \Delta_2}$$

Responsible for splitting of phonon modes

Splitting occurs if

1. $g_a g_b^* = i g^2$
2. $f_1 \neq f_{\tilde{1}}$ or $\Delta_1 \neq \Delta_2$



P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

Phonon energies and eigenmodes

Splitting occurs if

1. $g_a g_b^* = i g^2$
2. $f_1 \neq f_{\tilde{1}}$ or $\Delta_1 \neq \Delta_2$

Det$(D^{-1}(\omega))$=0

$$\frac{\omega_{ph}^{+} - \omega_{ph}^{-}}{\omega_{ph}(B=0)} = 2 \cdot \frac{2g^2\chi + \gamma(\Omega_+^2 - \omega_0^2)/\omega_0}{\sqrt{(\omega_0^2 - \Delta^2)^2 + 16 g^2 f_0 \omega_0 \Delta}} B + \mathcal{O}(B^2)$$

Broken TRS

$$\Delta_1 = \Delta - \gamma B,$$
$$\Delta_2 = \Delta + \gamma B$$
$$f_1 - f_{\tilde{1}} = \chi B$$

New phonon modes

$$\phi_+ = \frac{1}{\sqrt{2}}(\phi_a - i\phi_b)$$
$$\phi_- = \frac{1}{\sqrt{2}}(\phi_a + i\phi_b)$$

Chiral

P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

Factors for chiral phonon splitting

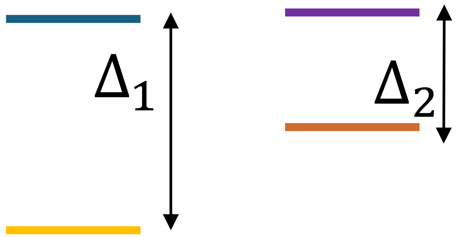
Splitting occurs if

1. $g_a g_b^* = i g^2$
2. $f_1 \neq f_{\tilde{1}}$ or $\Delta_1 \neq \Delta_2$

$$\frac{\omega_{ph}^{+} - \omega_{ph}^{-}}{\omega_{ph}(B=0)} = 2 \cdot \frac{2g^2\chi + \gamma(\Omega_+^2 - \omega_0^2)/\omega_0}{\sqrt{(\omega_0^2 - \Delta^2)^2 + 16 g^2 f_0 \omega_0 \Delta}} B + \mathcal{O}(B^2)$$

Broken TRS

$$\Delta_1 = \Delta - \gamma B,$$
$$\Delta_2 = \Delta + \gamma B$$
$$f_1 - f_{\tilde{1}} = \chi B$$



1. Energy scales: Δ and ω_0
2. Electronic ‘g’ factor
3. Temperature
4. Electron -phonon coupling

P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977)

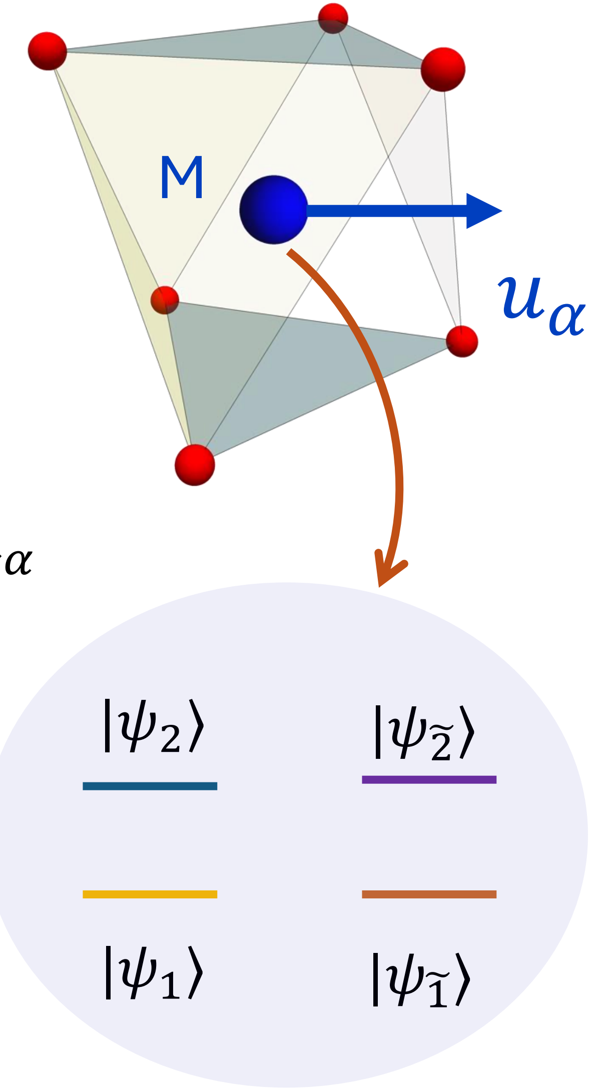
e-ph coupling strength using point charge model

$$H_{el-ph} = (a^\dagger + a)\widehat{\mathbf{O}}_a + (b^\dagger + b)\widehat{\mathbf{O}}_b$$

$$\hat{O}_a = g_a|\psi_1\rangle\langle\psi_2| - g_a^*|\psi_{\tilde{1}}\rangle\langle\psi_{\tilde{2}}|$$

$$\hat{O}_b = g_b|\psi_1\rangle\langle\psi_2| - g_b^*|\psi_{\tilde{1}}\rangle\langle\psi_{\tilde{2}}|$$

$$u_a = \frac{\hbar}{\sqrt{M\hbar\omega_{ph}}}(a + a^\dagger)$$



Modified crystal electric field due to phonon

$$V_M(u_\alpha) = [\alpha_1 xy + \alpha_2 yz + \alpha_3 xz + \alpha_4(x^2 - y^2) + \alpha_5(3z^2 - x^2 - y^2)]u_\alpha$$

$\langle\psi_i|r_\alpha r_\beta|\psi_j\rangle$ → $|\psi_i\rangle = |J_i, m_j^i\rangle$ → $\begin{cases} m_l & \text{Orbital} \\ m_s & \text{Spin} \end{cases}$

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

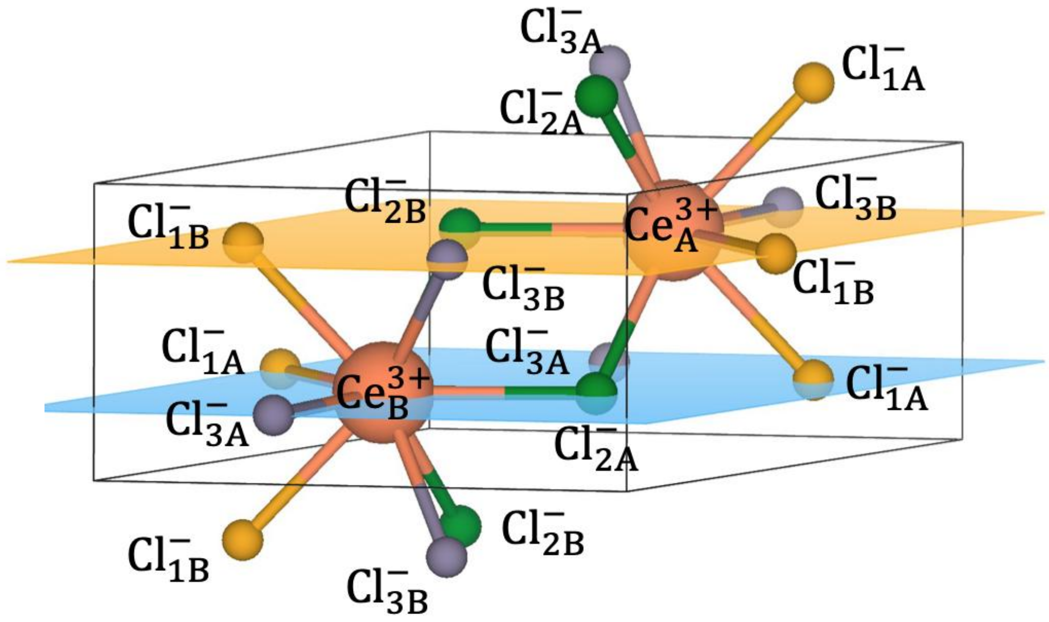
Model applied to Rare earth trihalide $CeCl_3$

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

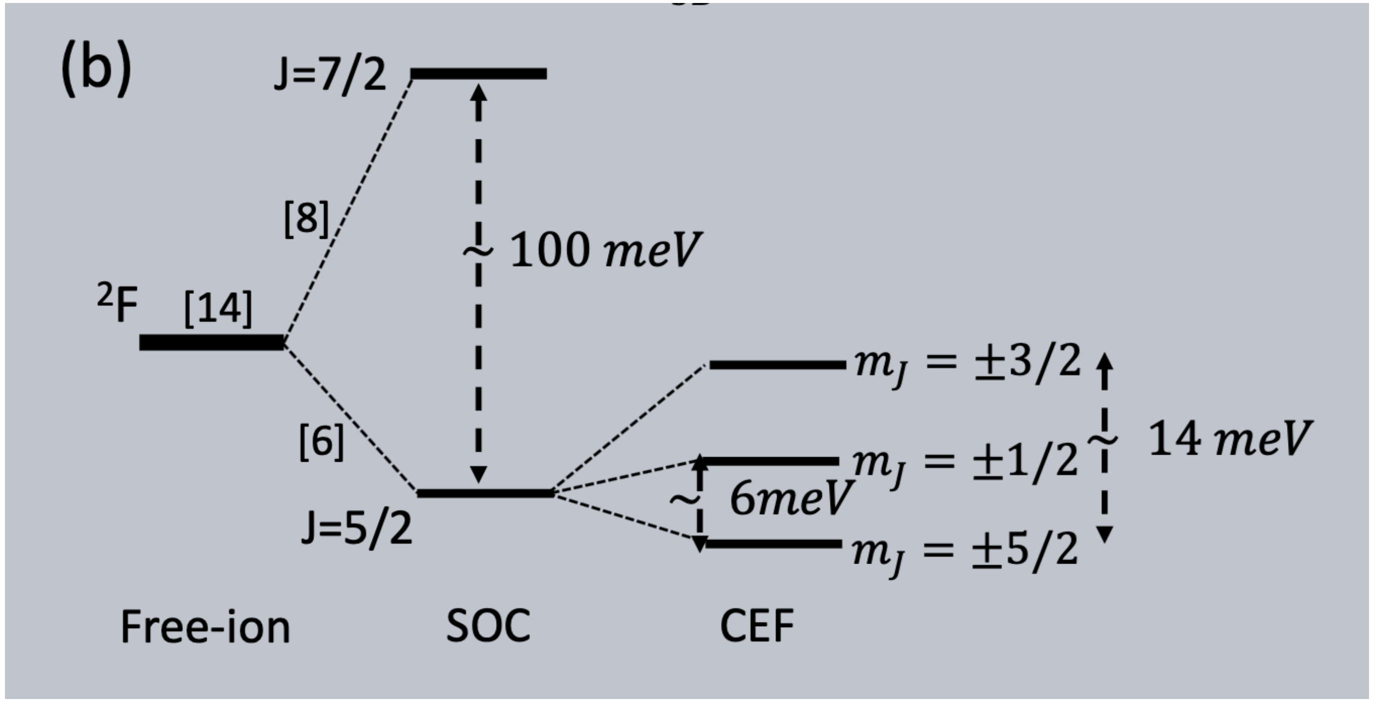
Properties of rare-earth paramagnet $CeCl_3$

$Ce^{3+}: 4f^1$

1. Strong SOC, $\lambda \approx 80\ meV$
2. Weak Crystal-electric fields (CEF)



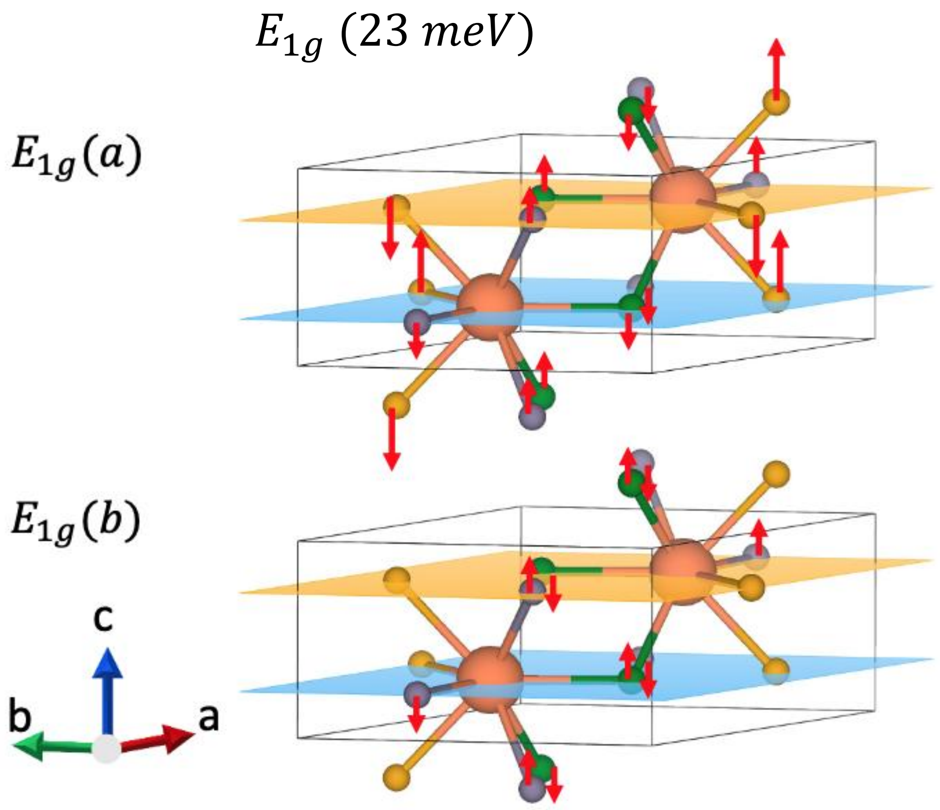
Crystal structure



Electronic states

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

Orbital lattice coupling in $CeCl_3$

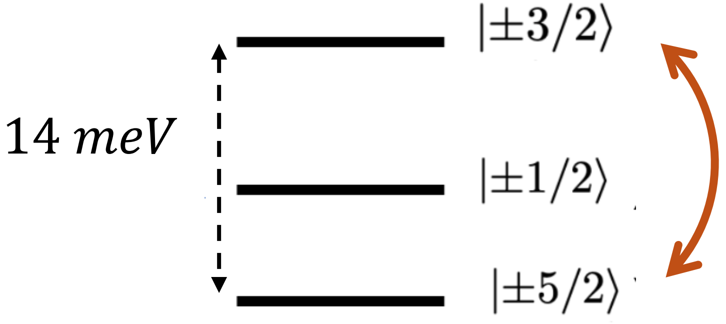


Perturbation to crystal electric field around Ce^{3+} ion

$$V(E_{1g}(a)) = [-0.06\,xz + 0.16\,yz]\;q$$
$$V(E_{1g}(b)) = [0.16\,xz + 0.06\,yz]\;q$$

$$H_1(xz) = -\frac{2}{7\sqrt{5}}\langle r^2\rangle \begin{pmatrix} & \left|\frac{5}{2},\pm\frac{5}{2}\right\rangle & \left|\frac{5}{2},\pm\frac{3}{2}\right\rangle \\ \left|\frac{5}{2}\pm\frac{5}{2}\right\rangle & 0 & \pm 1 \\ \left|\frac{5}{2},\pm\frac{3}{2}\right\rangle & \pm 1 & 0 \end{pmatrix}$$

$$H_1(yz) = \frac{2}{7\sqrt{5}}\langle r^2\rangle \begin{pmatrix} & \left|\frac{5}{2},\pm\frac{5}{2}\right\rangle & \left|\frac{5}{2},\pm\frac{3}{2}\right\rangle \\ \left|\frac{5}{2},\pm\frac{5}{2}\right\rangle & 0 & i \\ \left|\frac{5}{2},\pm\frac{3}{2}\right\rangle & -i & 0 \end{pmatrix},$$



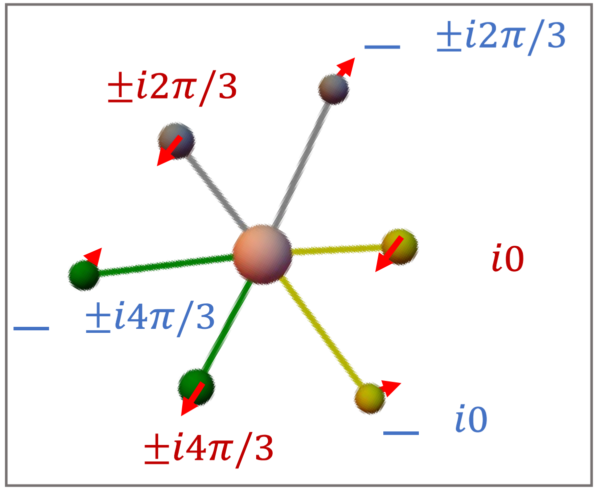
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete
Phys. Rev. B **110**, 094401 (2024)

S. Chaudhary*, C.P. Romao*, D. Juraschek, arXiv:2504.19121 (2025)

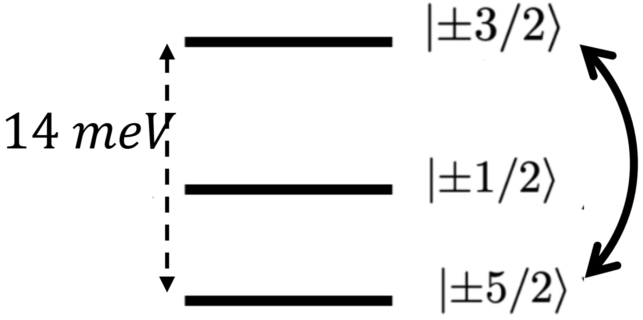
Perturbation to CEFs around Ce^{3+} from phonons in $CeCl_3$

$\square_{1g}$ $\square$23$\square$)

$_{1g}$( ) $\pm$ $\square_{1g}$( )

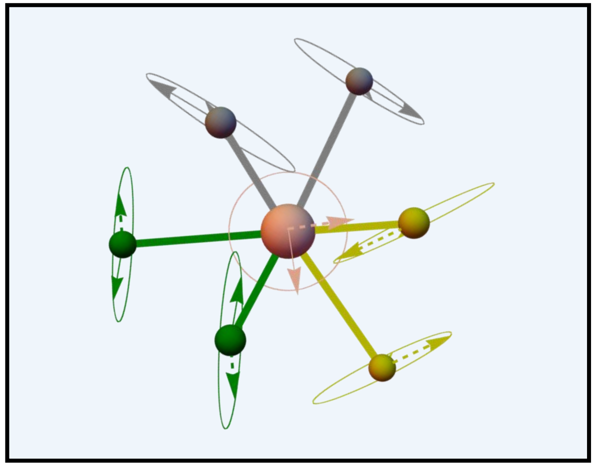


$$V(E_{1g}(a)) = [-0.06\,xz + 0.16\,yz]\;q$$
$$V(E_{1g}(b)) = [0.16\,xz + 0.06\,yz]\;q$$

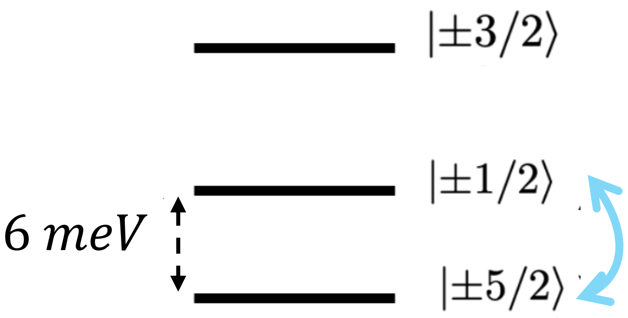


E_{2g}^1 (12 meV)

$E_{2g}^1(a) \pm i\, E_{2g}^1(b)$

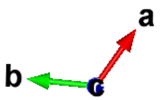


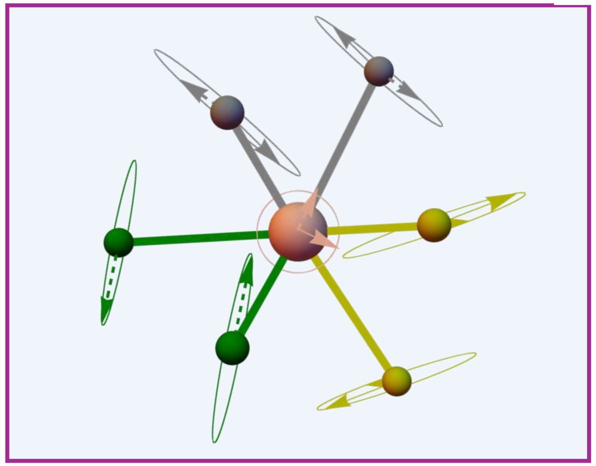
$$V(E_{2g}^1(a)) = [-0.05\,xy - 0.007\,(x^2 - y^2)]\;q$$
$$V(E_{2g}^1(b)) = [0.014\,xy - 0.025\,(x^2 - y^2)]\;q$$



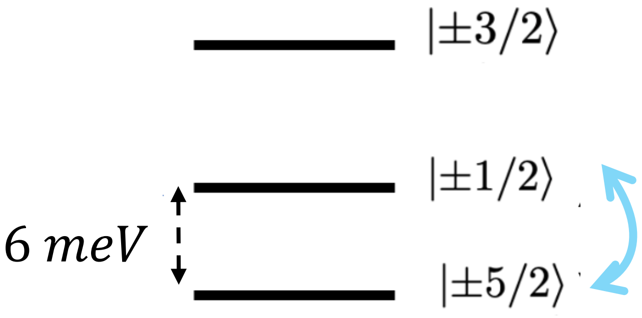
E_{2g}^2 (21.5 meV)

$E_{2g}^2(a) \pm i\, E_{2g}^2(b)$

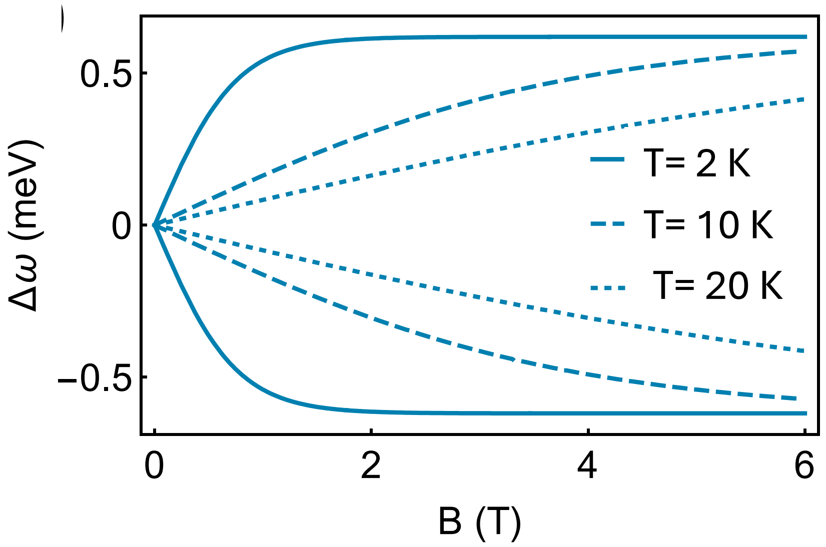




$$V(E_{2g}^2(a)) = [0.08\,xy + 0.01\,(x^2 - y^2)]\;q$$
$$V(E_{2g}^2(b)) = [-0.02\,xy + 0.04\,(x^2 - y^2)]\;q$$



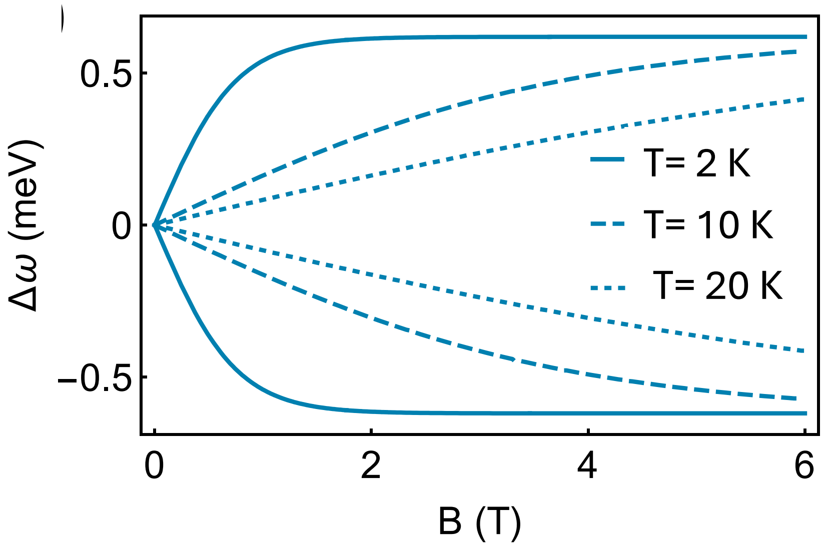
Zeeman splitting and Phonon Magnetic moment in $CeCl_3$

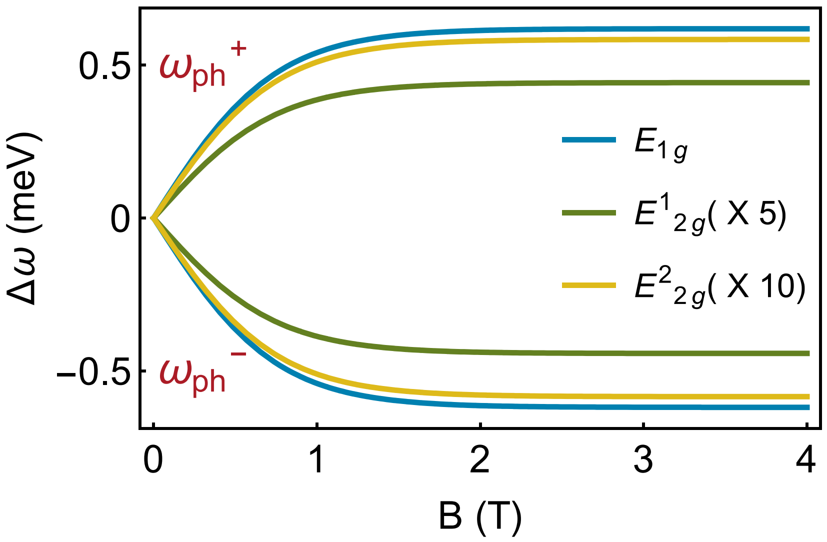


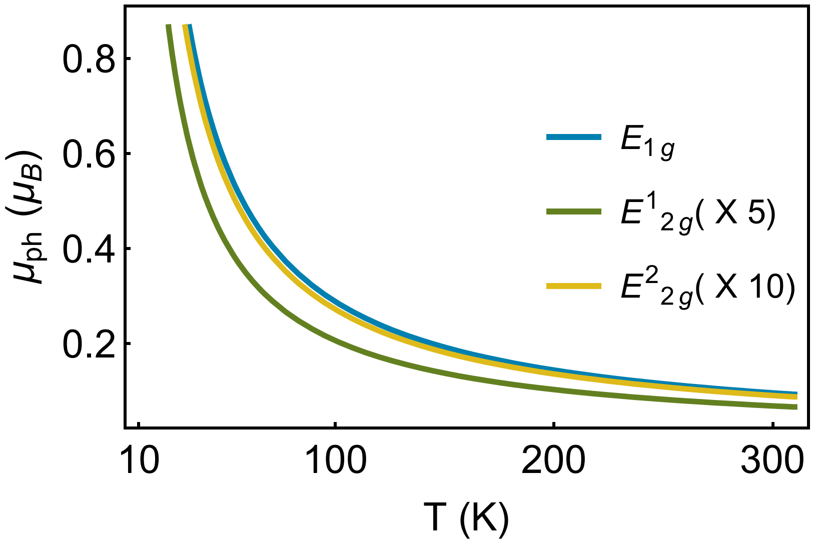
Phonon	μ_{ph} (μ_B) 10 K	μ_{ph}(μ_B) 20 K
E_{1g} (22meV)	2	1
E_{2g}^1(12 meV)	0.5	0.3
E_{2g}^2 (21.5 meV)	0.2	0.1

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

Zeeman splitting and Phonon Magnetic moment in $CeCl_3$







Phonon	μ_{ph} (μ_B) 10 K	$\mu_{ph}(\mu_B)$ 20 K
E_{1g} (22meV)	2	1
E_{2g}^1(12 meV)	0.5	0.3
E_{2g}^2 (21.5 meV)	0.2	0.1

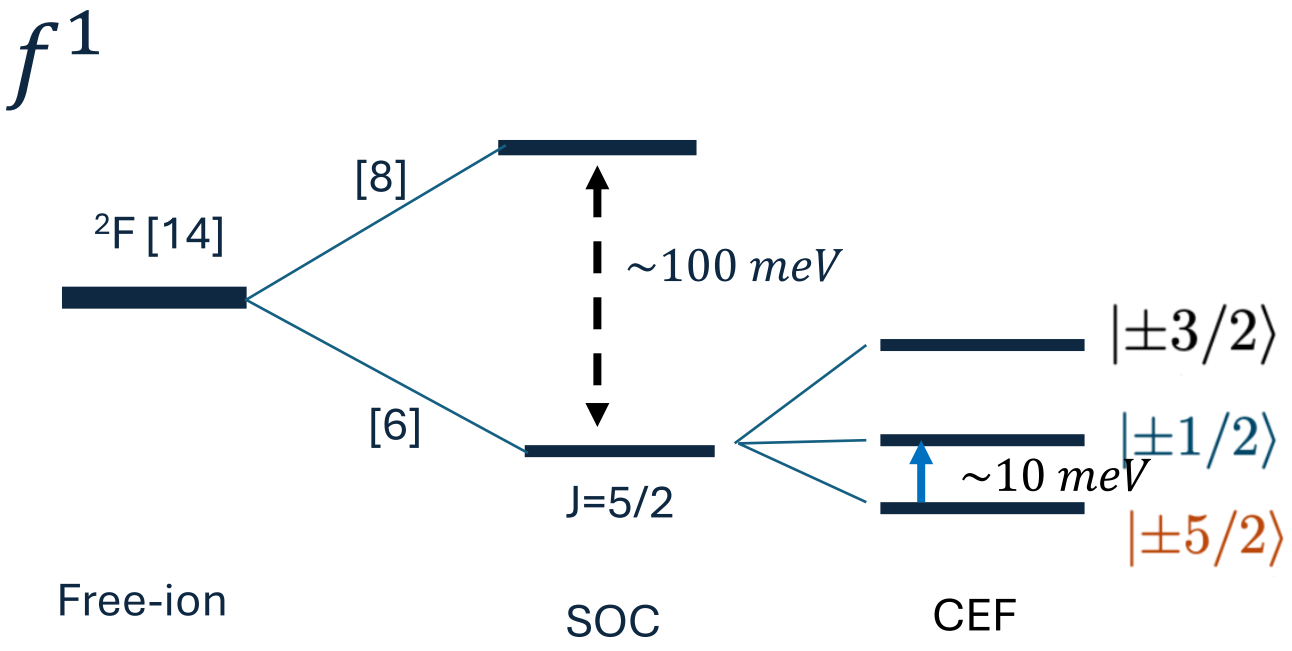
S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete Phys. Rev. B **110**, 094401 (2024)

Model applied to a *d* orbital magnet

Electronic excitations on magnetic ions

f - electrons

1. Strong SOC
2. Weak CEF effects

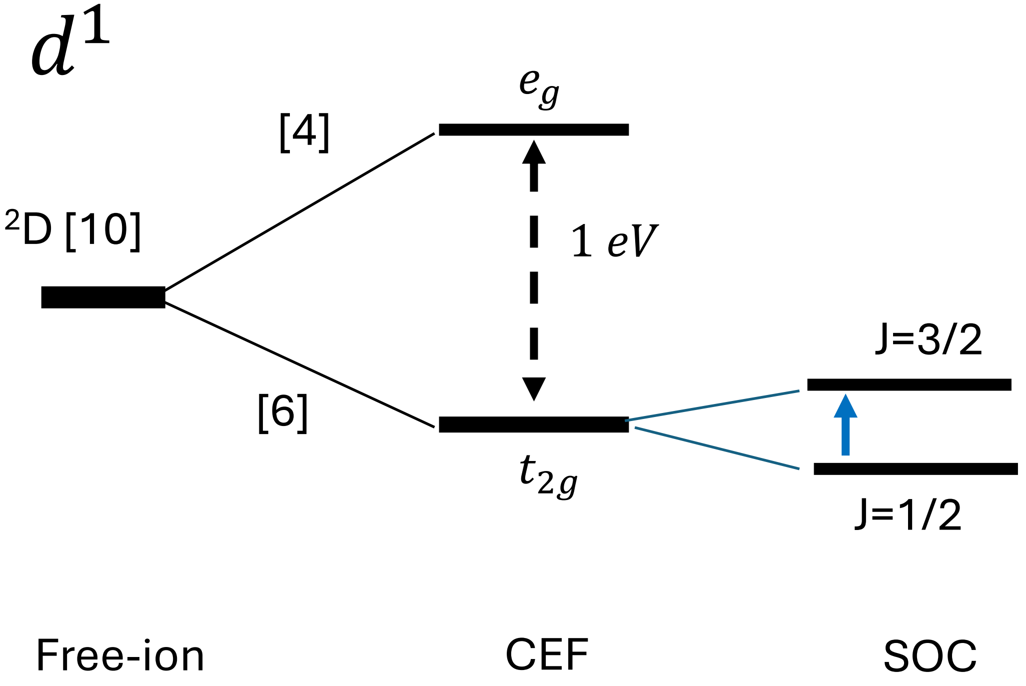


Kramers-doublet ground state

Kramers-doublet excited state

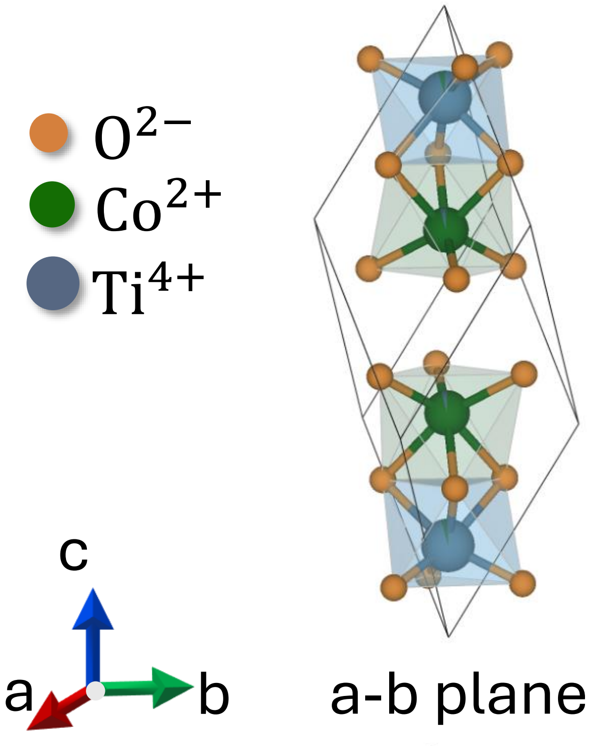
d - electrons

1. Weak SOC
2. Very strong. CEF effects (~1eV)

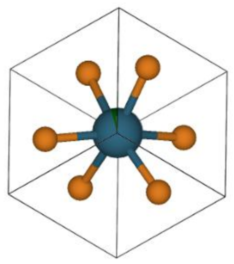


$CoTiO_3$ physical and electronic properties

Rhombohedral setting

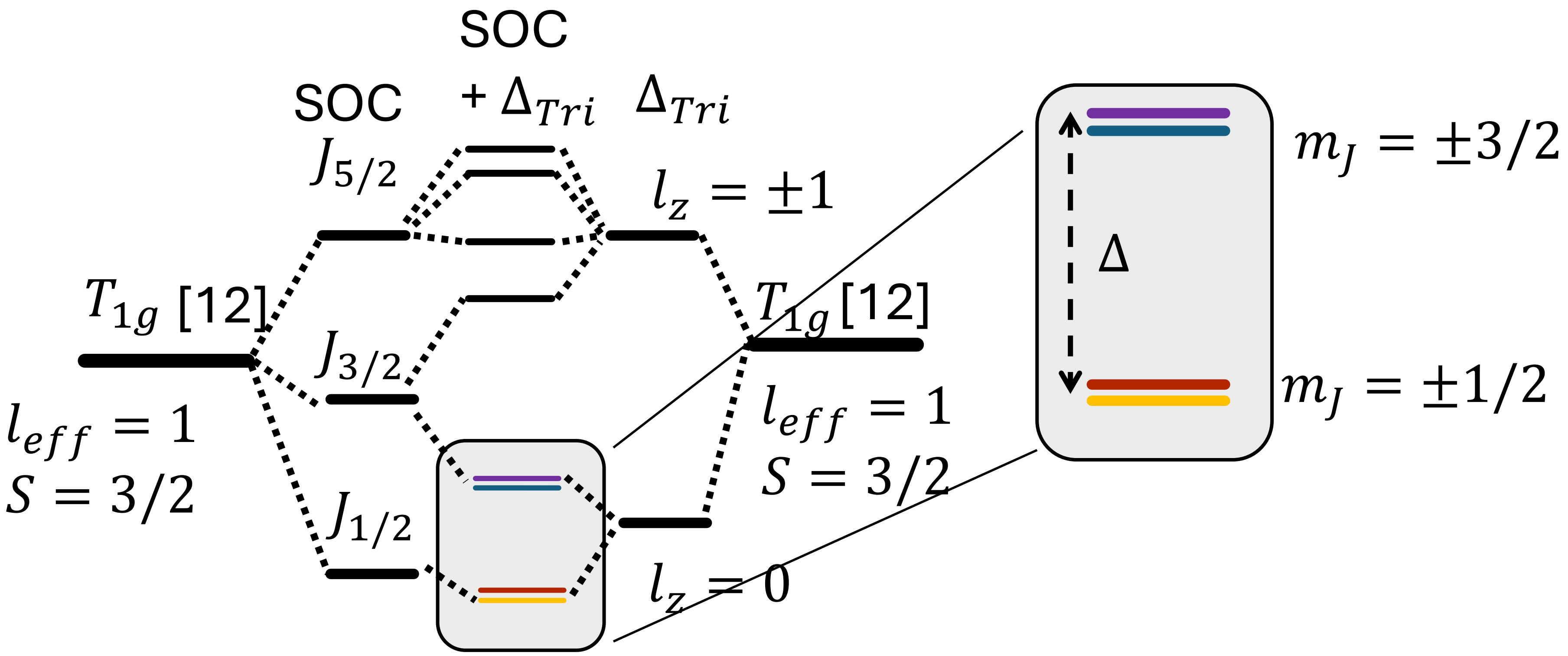


a-b plane



Crystal Structure

SOC +Trigonal distortion

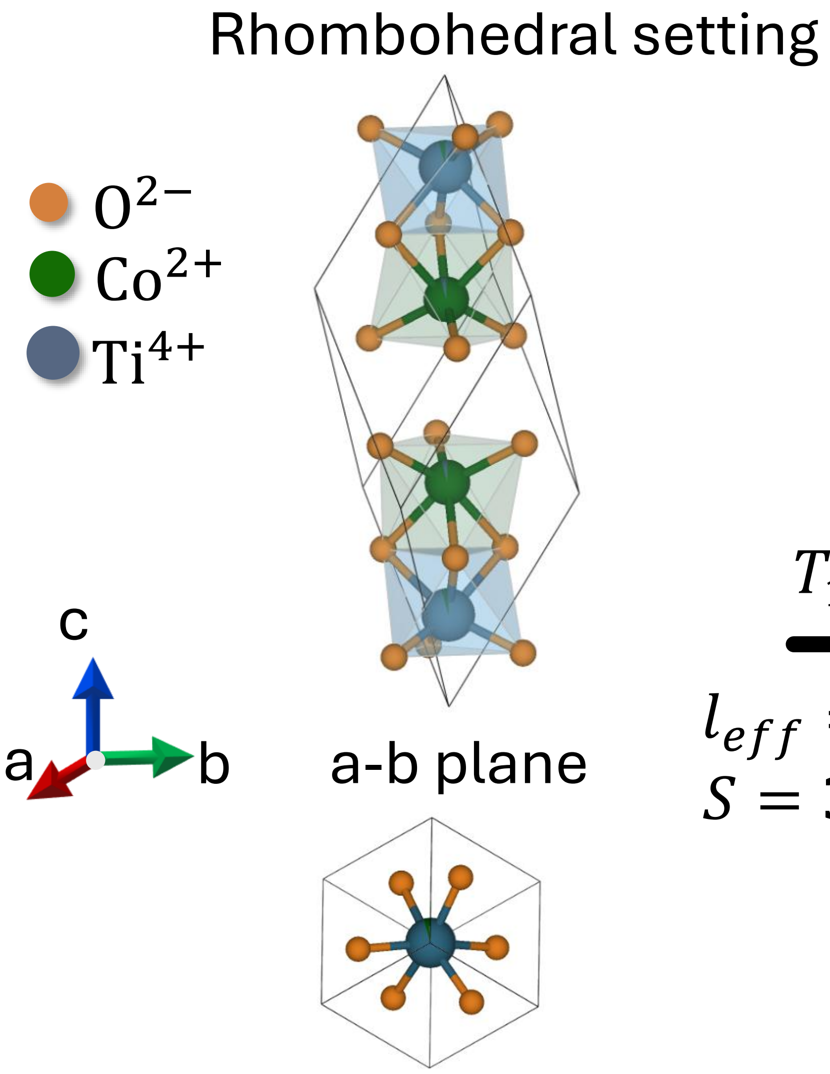


Co^{2+} electronic states (Three holes in 3d orbitals)

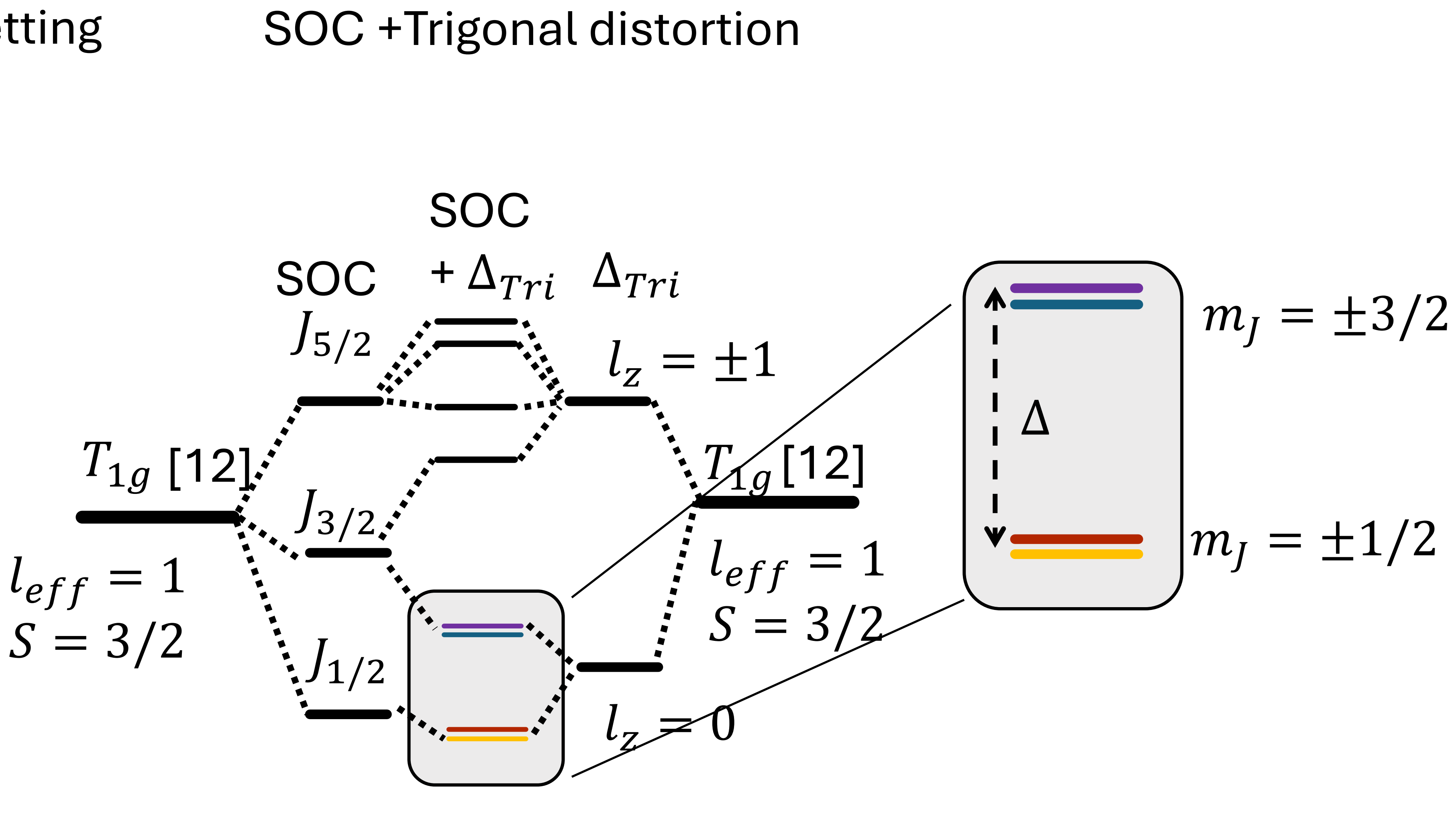
Low-energy Kramers doublets

Nature Communications 12.1 (2021): 3936.

$CoTiO_3$ physical and electronic properties



Crystal Structure

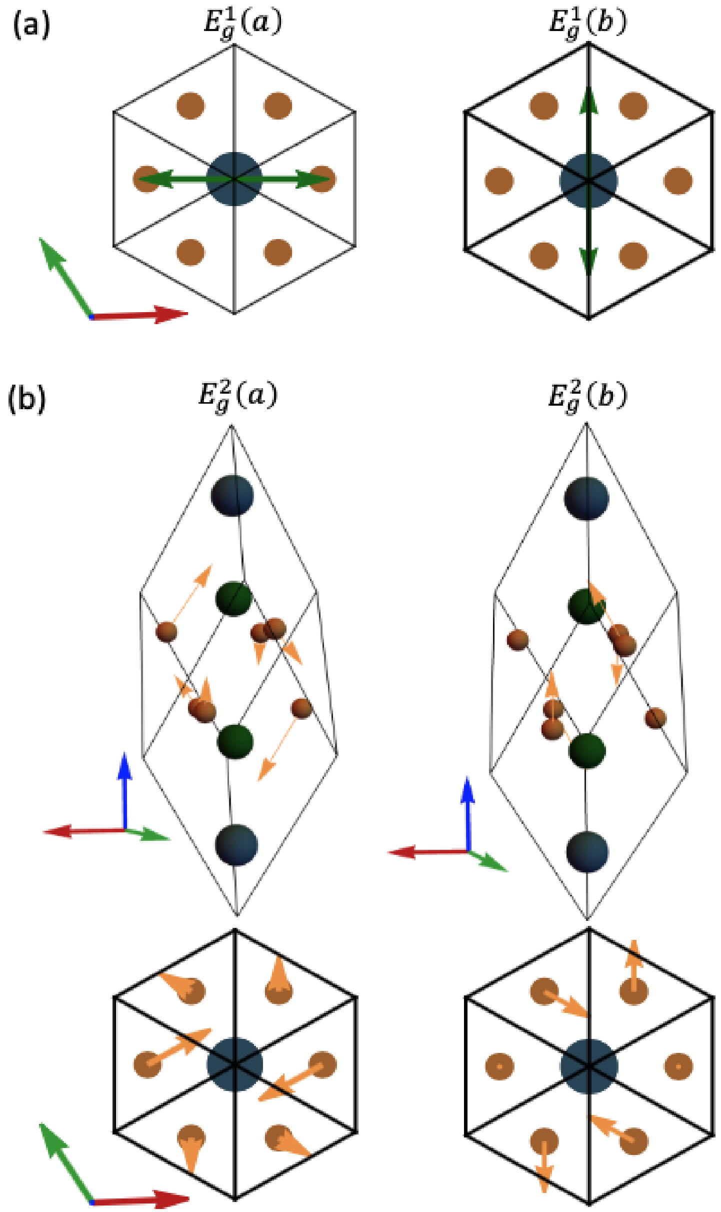


Co^{2+} electronic states (Three holes in 3d orbitals)

Low-energy Kramers doublets

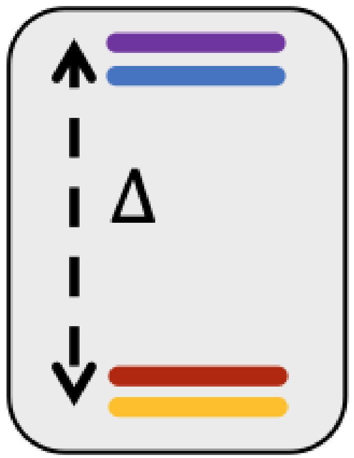
Nature Communications 12.1 (2021): 3936.

$CoTiO_3$ E_g phonon modes



26 meV

33 meV

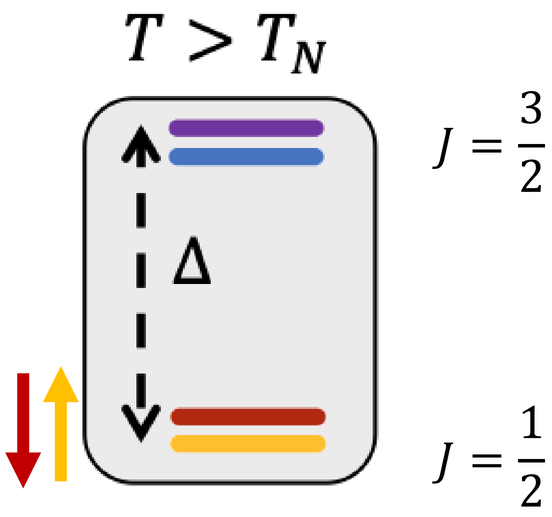


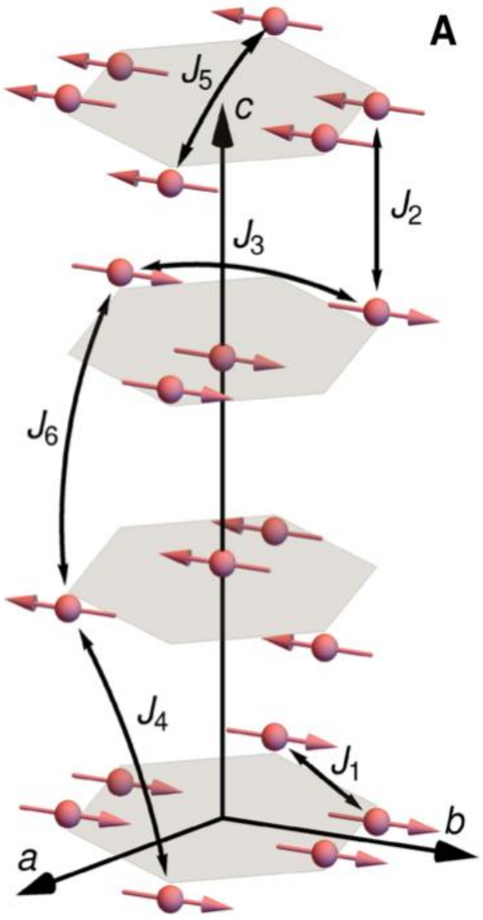
$\Delta = 23\ meV$

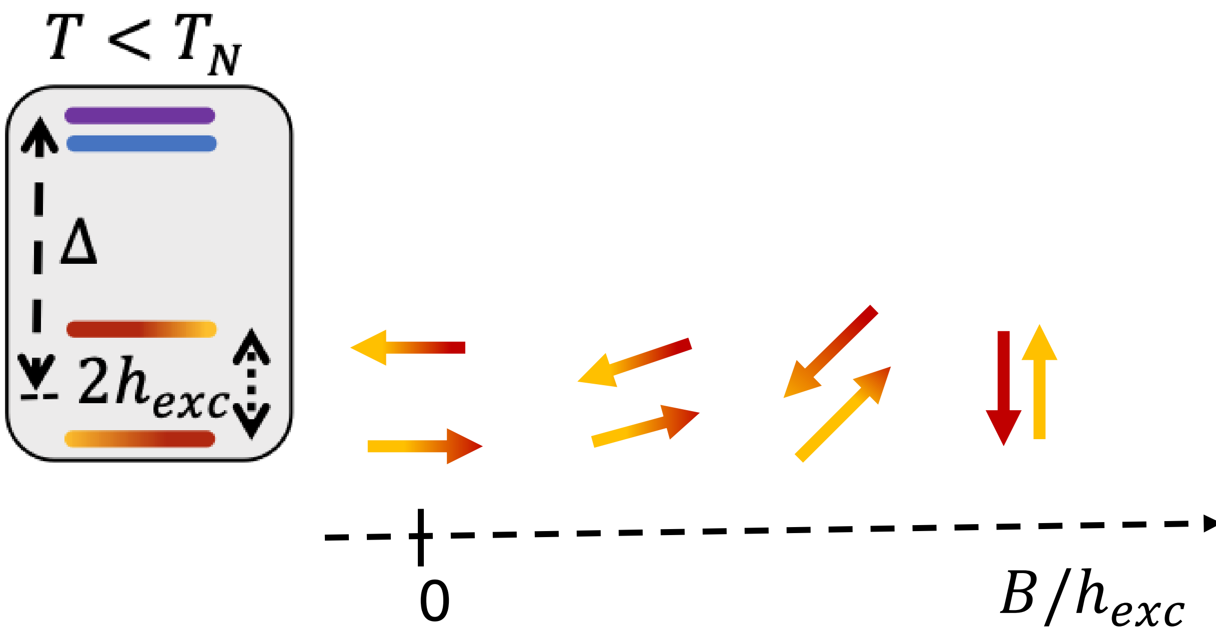
Phys. Rev. B **102**, 134404 (2020)

Nature Communications 12.1 (2021): 3936.

$CoTiO_3$ magnetic properties







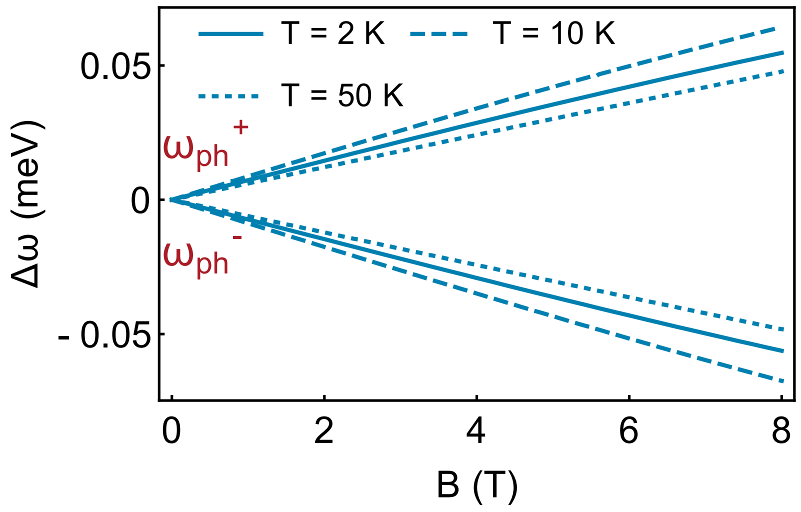
$$\left|J=\frac{1}{2}, m_j=-\frac{1}{2}\right\rangle$$

$$\left|J=\frac{1}{2}, m_j=+\frac{1}{2}\right\rangle$$

$$\frac{1}{\sqrt{2}}\left|J=\frac{1}{2}, m_j=-\frac{1}{2}\right\rangle+\frac{1}{\sqrt{2}}\left|J=\frac{1}{2}, m_j=-\frac{1}{2}\right\rangle$$

$$\frac{1}{\sqrt{2}}\left|J=\frac{1}{2}, m_j=-\frac{1}{2}\right\rangle-\frac{1}{\sqrt{2}}\left|J=\frac{1}{2}, m_j=-\frac{1}{2}\right\rangle$$

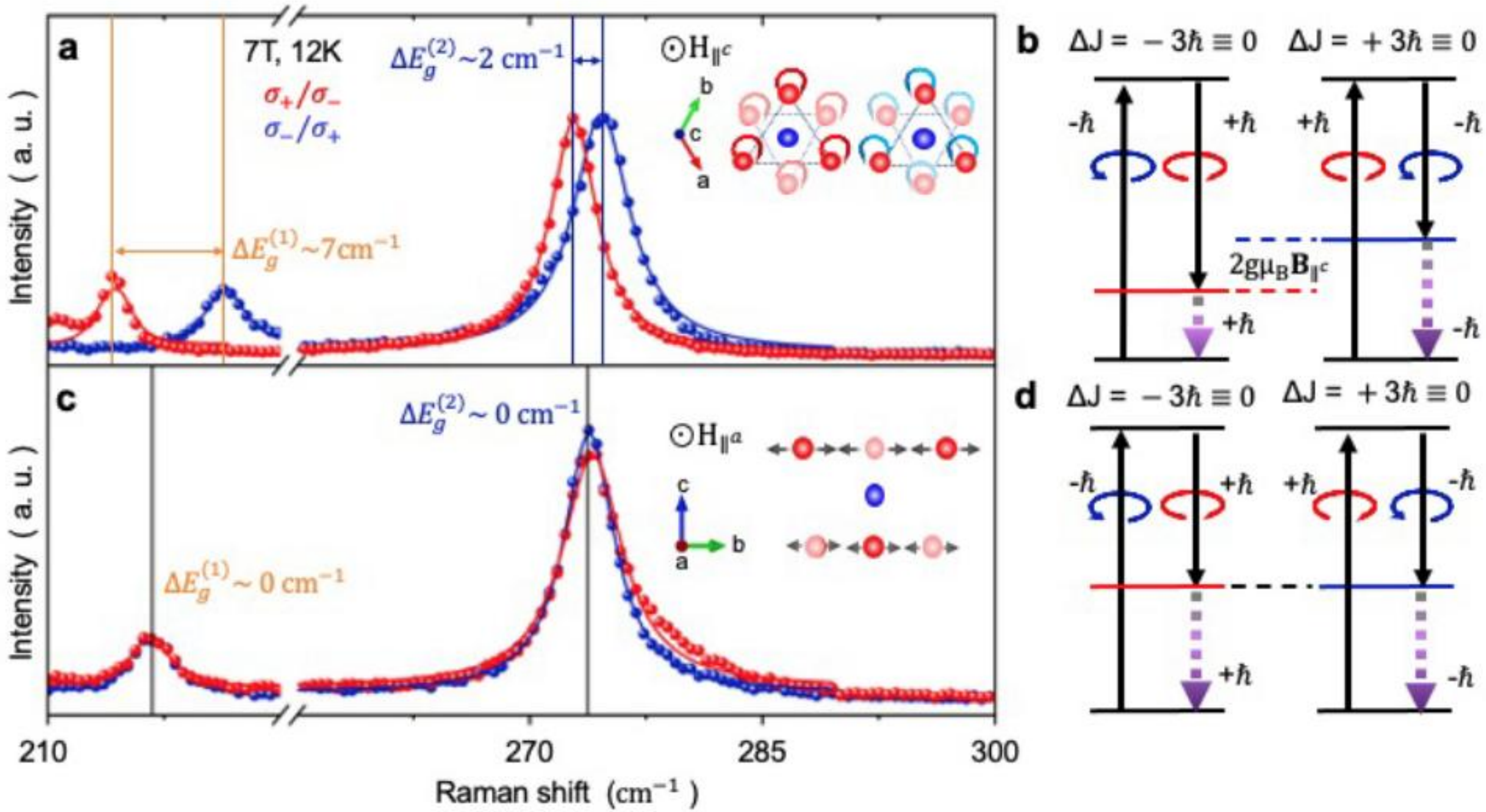
$CoTiO_3$ E_g modes phonon magnetic moment

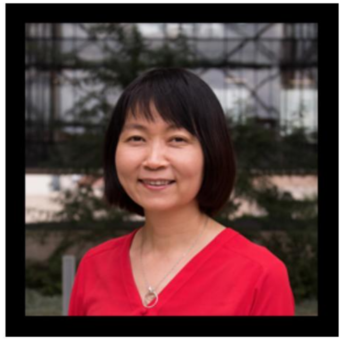


Phonon	μ_{ph} (μ_B) 50 K	$\mu_{ph}(\mu_B)$ 100 K
E_g^1 (22meV)	0.2	0.1
E_g^2 (21.5 meV)	0.12	0.6

- No saturation in the given B field limit
- T trend similar to magnetic susceptibility

Results from helicity-resolved magneto-Raman





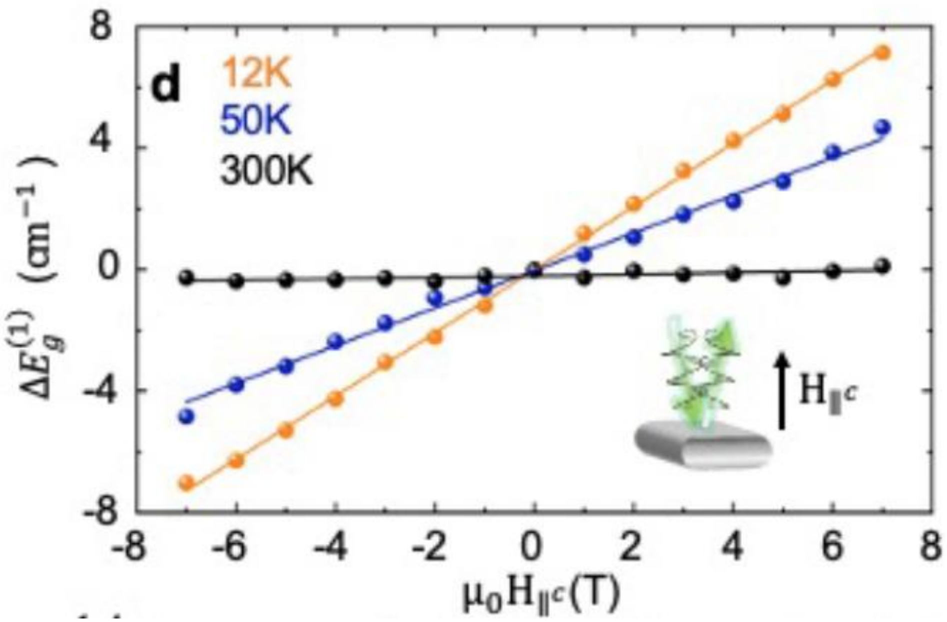
Xiaoqin (Elaine) Li
UT Austin

David Lujan
UT Austin

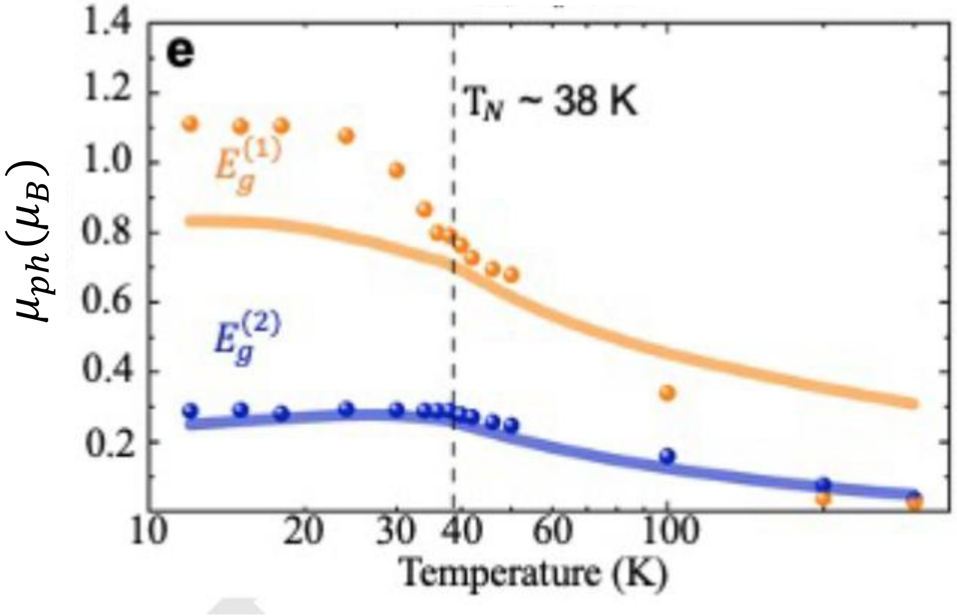
D. Lujan*, J. Choe*, **S. Chaudhary***, G. Fiete, R. He, & Xiaoqin Li *et al., Spin-Orbit Exciton-Induced Phonon Chirality in a Quantum Magnet* PNAS, 121(11), e2304360121 (2024)

Chiral phonons with giant magnetic moment in $CoTiO_3$

Zeeman splitting of E_g^1 phonon



Phonon magnetic moment

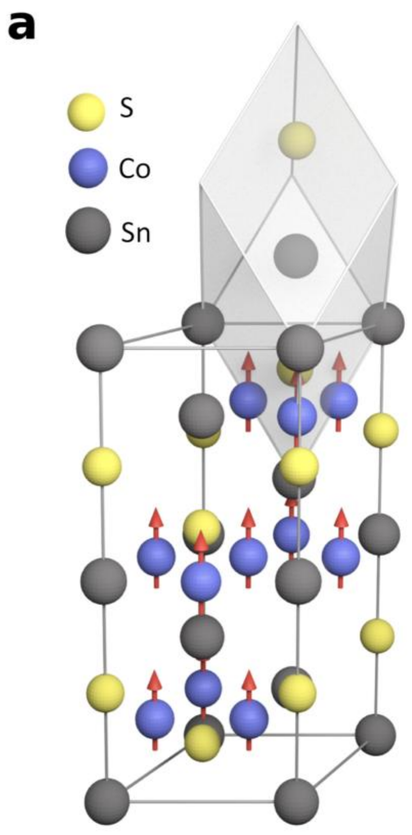


- ❖ Phonon magnetic moment $\boldsymbol{\mu_{ph} \approx \mu_B}$
- ❖ First such example in a ‘**d**’ orbital quantum magnet

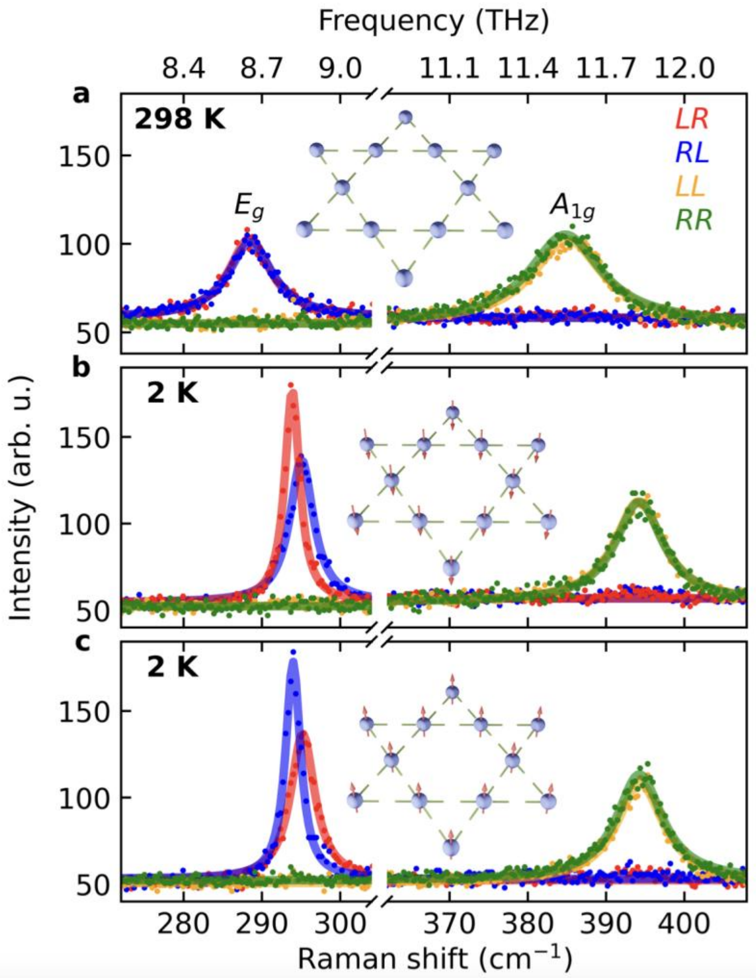
D. Lujan*, J. Choe*, **S. Chaudhary***, G. Fiete, R. He, & Xiaoqin Li *et al.,* PNAS, 121(11), e2304360121 (2024)

Other possibilities: Magnetic order induced phonon chirality

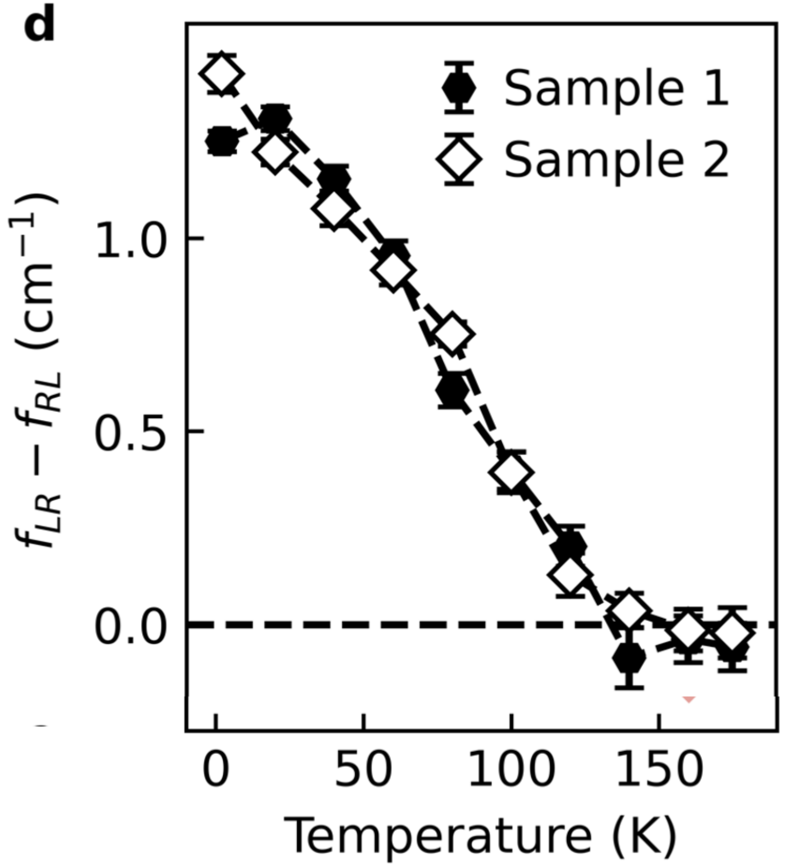
Che, Mengqian, et al. Phys. Rev, Lett. **134**, 196906 (2025)



FM order in $Co_3Sn_2S_2$

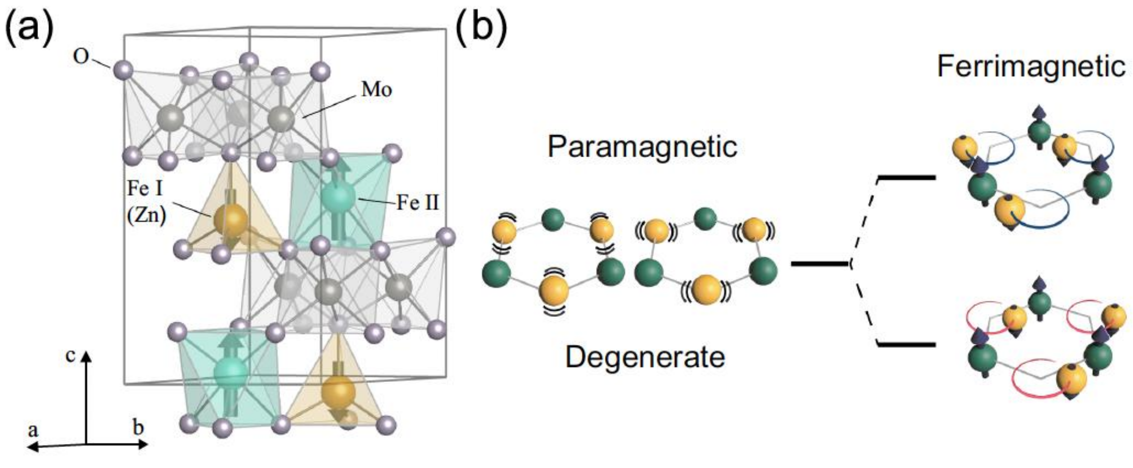


Chiral phonon splitting below T_c

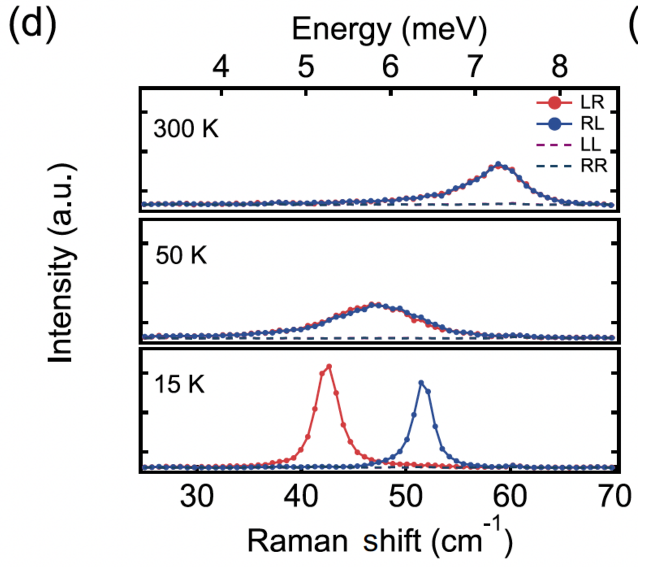


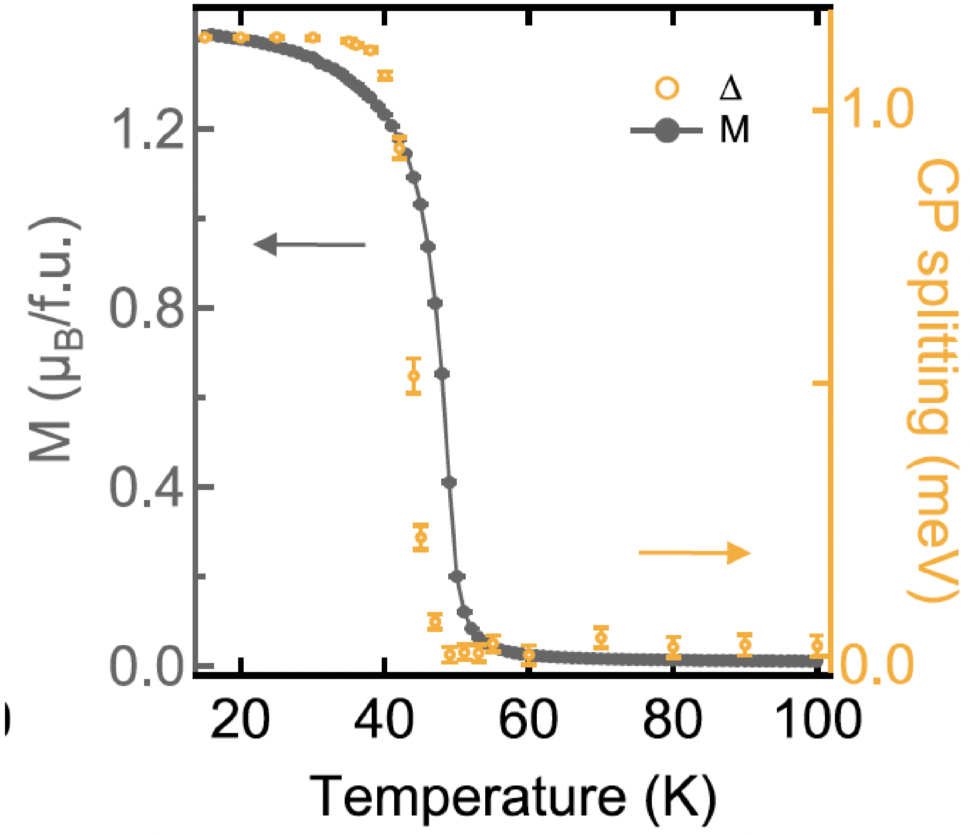
Other possibilities: Magnetic order induced phonon chirality

Wu, Fangliang, et al. Phys. Rev. Lett. **134**, 236701 (2025)

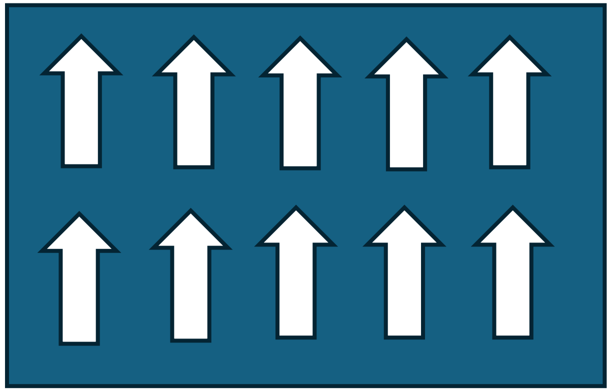


Crystal structure of $Fe_{1.75}Zn_{0.25}Mo_3O_8$ (FZMO)

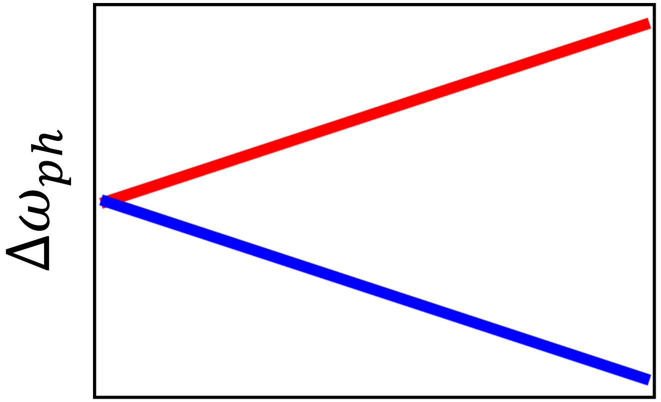




Chiral phonon splitting without magnetization?



Magnetization



Magnetization

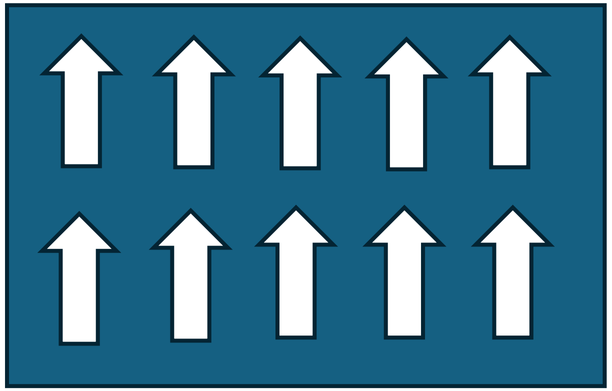
$CeCl_3$, $CoTiO_3$, $Co_3Sn_2S_2$, FZMO

TRS breaking without magnetic dipoles

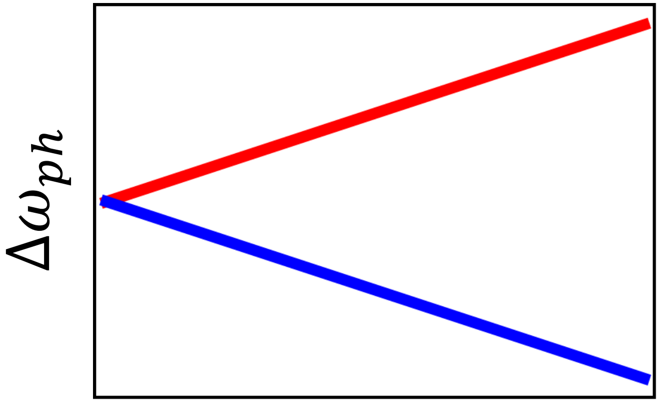
?

Magnetic Multipoles

Chiral phonon splitting without magnetization?



Magnetization



$CeCl_3$, $CoTiO_3$, $Co_3Sn_2S_2$, FZMO

TRS breaking without magnetic dipoles

?

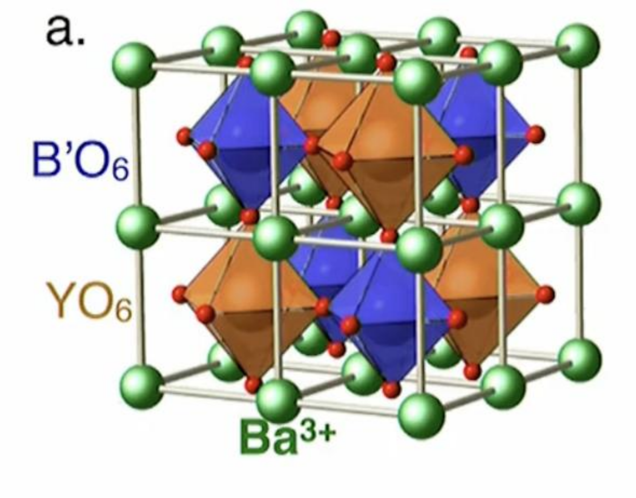
R. Sutcliffe, K. Hart, S. Chaudhary, and A. Paramekanti, *Pseudo-chiral phonon splitting from octupolar magnetic order*, arxiv: 2506.18978 (2025)

Arun Paramekanti (Univ. Of Toronto)

Kathleen Hart

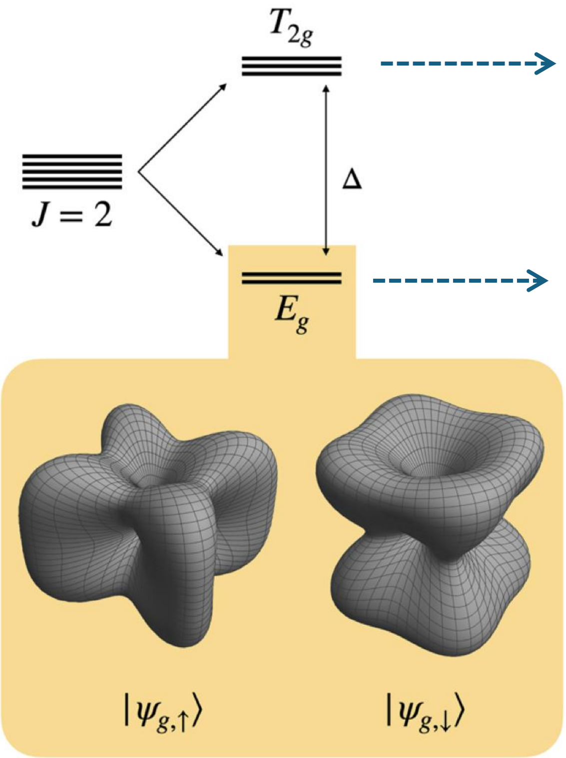
Ruairidh. Sutcliffe

Multipolar ordering in $5d^2$ osmate double perovskites



Double perovskite

Ba_2MOsO_6, M= Ca, Mg



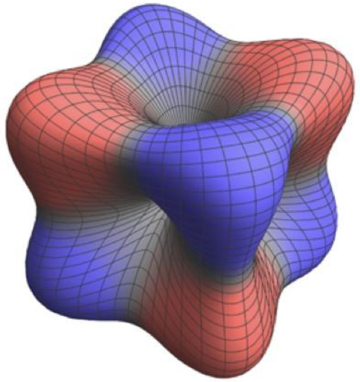
$5d^2$ Os^{6+}

$$\left|\psi_{ex,\pm}\right\rangle = |\pm 1\rangle$$

$$\left|\psi_{ex,0}\right\rangle = \frac{1}{\sqrt{2}}(|2\rangle - |-2\rangle)$$

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

$$\left|\psi_{g,\downarrow}\right\rangle = |0\rangle$$



$|\psi_{g,\uparrow}\rangle + i|\psi_{g,\downarrow}\rangle$

Pseudospin 1/2

$$\tau = (\tau_x, \tau_y, \tau_z)$$

$$\tau_x \propto (J_x^2 - J_y^2)$$

$$\tau_z \propto \left(3J_z^2 - J(J+1)\right)$$

$$\tau_y \propto \overline{J_x J_y J_z}$$ $\mathcal{T}$- odd

Octupolar Order

Voleti, et.al. *npj Quantum materials*, *8*(1), 42 (2023).

Pourovoskii, Phys. Rev. Lett. **127**, 237201 (2021)

Multipolar ordering in $5d^2$ osmate double perovskites

Arun Paramekanti (U Toronto)

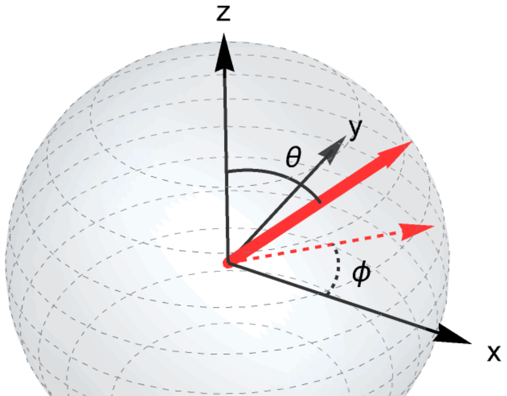
$\tau = (\tau_x, \tau_y, \tau_z)$ $\quad \tau_x \propto (J_x^2 - J_y^2)$ $\quad \tau_z \propto \left(3J_z^2 - J(J+1)\right)$

$\tau_y \propto \overline{J_x J_y J_z}$ $\quad \mathcal{T}$ - odd

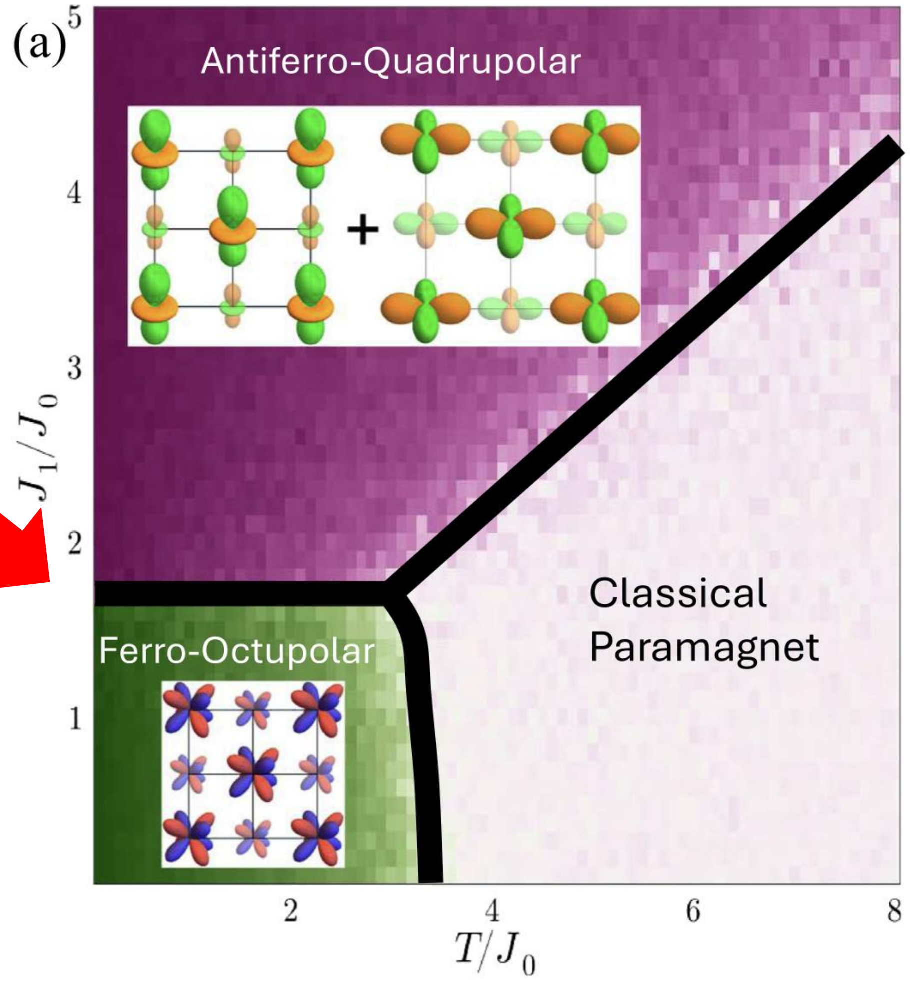
$$H_{\rm sp} = \sum_{\langle \mathbf{r},\mathbf{r}' \rangle} \Big[-J_0 \tau_{\mathbf{r}y} \tau_{\mathbf{r}'y} + J_1 \left(\cos^2\phi_{\mathbf{rr}'} + \gamma \sin^2\phi_{\mathbf{rr}'}\right) \tau_{\mathbf{r}z}\tau_{\mathbf{r}'z} + J_1 (1-\gamma) \sin\phi_{\mathbf{rr}'} \cos\phi_{\mathbf{rr}'} \left(\tau_{\mathbf{r}x}\tau_{\mathbf{r}'z} + \tau_{\mathbf{r}z}\tau_{\mathbf{r}'x}\right) + J_1 \left(\sin^2\phi_{\mathbf{rr}'} + \gamma \cos^2\phi_{\mathbf{rr}'}\right) \tau_{\mathbf{r}x}\tau_{\mathbf{r}'x} \Big],$$

$$\phi_{\mathbf{rr}'} = \{0, 2\pi/3, 4\pi/3\}$$

NN in $\{XY, YZ, ZX\}$ planes



Ground state represented by a pseudo-spin



Pourovoskii, Phys. Rev. Lett. **127**, 237201 (2021)

Hart et.al. PRL **134**, 246701 (2025)

Spin-phonon coupling in double perovskites

Pseudo-spin

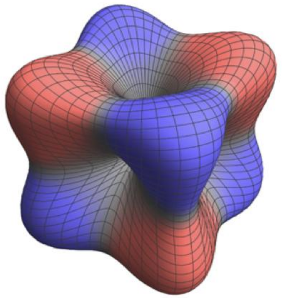
$\mathcal{T}$ - even quadrupolar

$\tau_x \propto (J_x^2 - J_y^2)$

$\tau_z \propto \left(3J_z^2 - J(J+1)\right)$

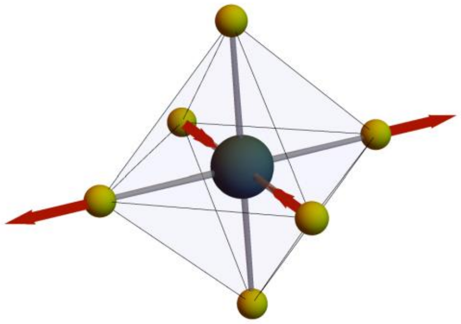
$\mathcal{T}$ - odd octupolar

$\tau_y \propto \overline{J_x J_y J_z}$

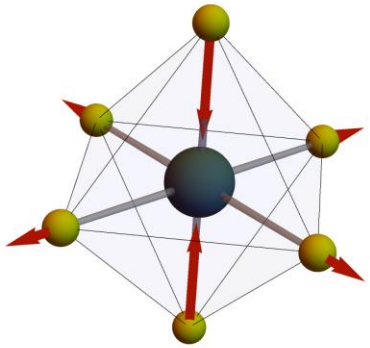


$|\psi_{g,\uparrow}\rangle + i|\psi_{g,\downarrow}\rangle$

E_g Phonons



$Q_x = x^2 - y^2$



$Q_z = 3z^2 - r^2$

Spin-phonon coupling

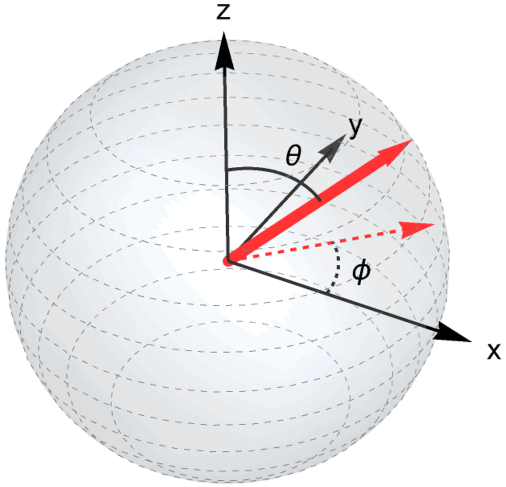
$$H_{sp-ph} = g(Q_x\tau_x + Q_z\tau_y)$$

$$H_{sp-ph} = \lambda\left[(b_x + b_x^\dagger)\tau_x + (b_z + b_z^\dagger)\tau_z\right]$$

Next: Integrate out spins and get effective phonon action.

Voleti, et.al. *npj Quantum materials*, *8*(1), 42 (2023).

Effective phonon action



$$\boldsymbol{\tau} = R(\theta_{\mathbf{r}}, \phi_{\mathbf{r}}) \cdot \tau'$$

$$\begin{aligned} \tau'_{\mathbf{r}y} &= 2(S - \alpha^\dagger_{\mathbf{r}}\alpha_{\mathbf{r}}) \\ \tau'_{\mathbf{r}x} &= \sqrt{2S}(\alpha_{\mathbf{r}} + \alpha^\dagger_{\mathbf{r}}) \\ \tau'_{\mathbf{r}z} &= i\sqrt{2S}(\alpha_{\mathbf{r}} - \alpha^\dagger_{\mathbf{r}}). \end{aligned}$$

$$H_{\mathrm{sp-ph}} = \lambda \sum_{\mathbf{r}} [(b_{\mathbf{r}x} + b^\dagger_{\mathbf{r}x})(\sqrt{2S}\sin\phi_{\mathbf{r}}$$
$$+ \; 2\sin\theta_{\mathbf{r}}\cos\phi_{\mathbf{r}}(S - \alpha^\dagger_{\mathbf{r}}\alpha_{\mathbf{r}}) + i\sqrt{2S}\cos\theta_{\mathbf{r}}$$
$$+ \; (b_{\mathbf{r}z} + b^\dagger_{\mathbf{r}z})(2\cos\theta_{\mathbf{r}}(S - \alpha^\dagger_{\mathbf{r}}\alpha_{\mathbf{r}}) + i\sqrt{2S}\,\mathrm{si}$$

Arun Paramekanti (Univ. Of Toronto)

Ground state calculated using Monte Carlo → Holstein-Primakoff → Spin-phonon interaction in Hol Primakoff boson basis

Kathleen Hart

$$S^{\mathrm{ph}}_{\mathrm{eff}} = S^{\mathrm{ph}}_0 + \langle S_{\mathrm{sp-ph}} \rangle_\alpha - \frac{1}{2}\left[\langle S^2_{\mathrm{sp-ph}} \rangle_\alpha - \langle S_{\mathrm{sp-ph}} \rangle^2_\alpha\right]$$

$$\langle \ldots \rangle_\alpha = \int D[\bar{\alpha}\alpha](\ldots)e^{-S_{\mathrm{sp}}}$$

$$\mathcal{G}^{-1}_{xz}(\mathbf{r}, \omega) = -8S\lambda^2 \left[\frac{i\omega\sin\theta_{\mathbf{r}}\sin\phi_{\mathbf{r}}}{\omega^2 - 4h_r^2}\right]$$

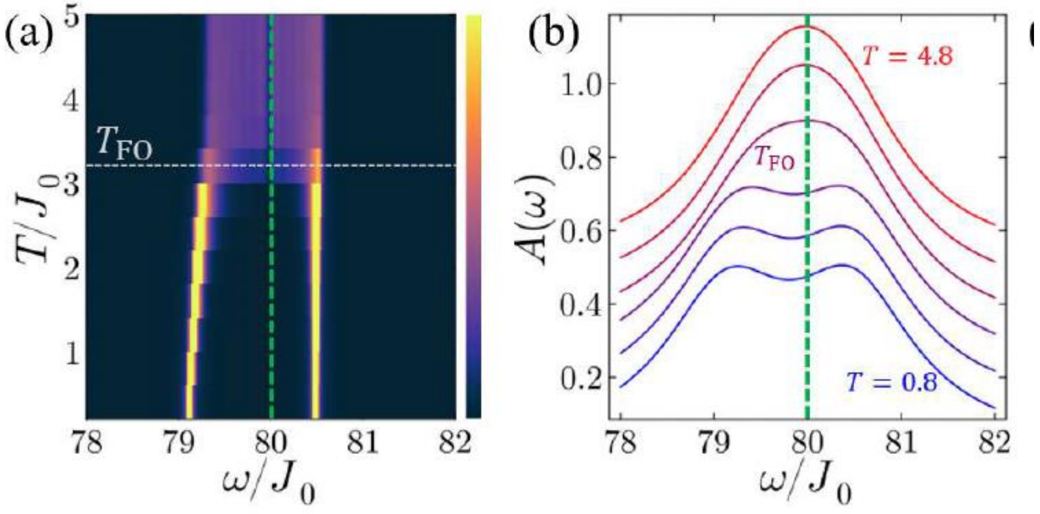
Corrections to Phonon Green's function

Ruairidh. Sutcliffe (U. Toronto)

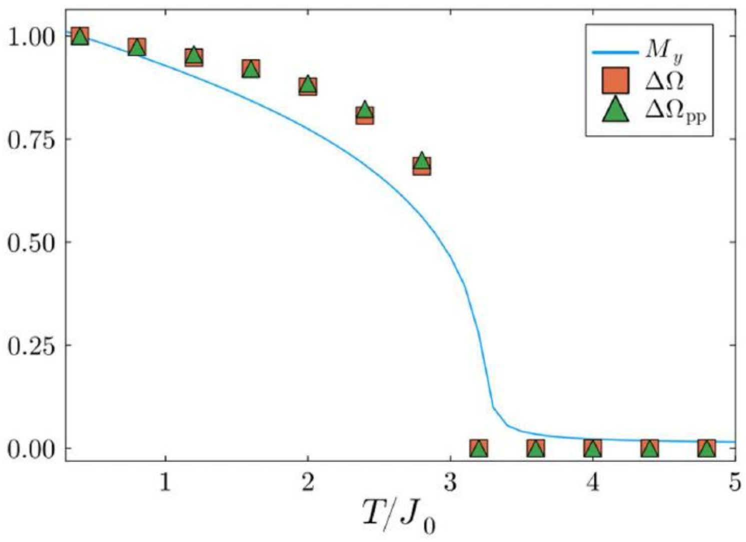
R. Sutcliffe, Kathleen Hart, S. Chaudhary, and Arun Paramekanti, arxiv: 2506.18978 (2025)

Phonon spectra

Phonon spectral function



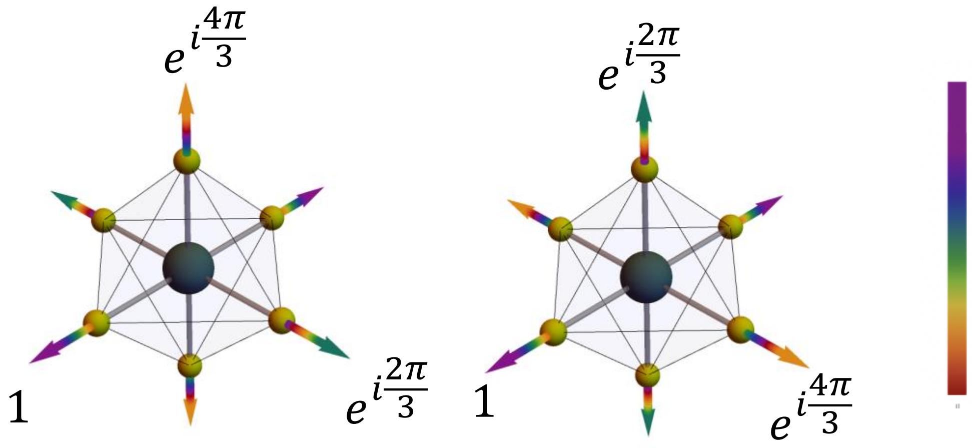
Chiral phonon splitting



Chiral phonons

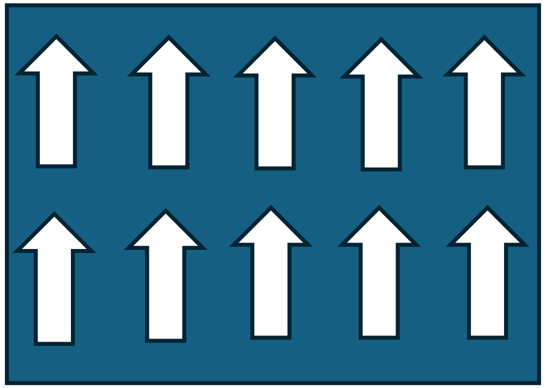
$$Q^{\pm} = Q_x \pm iQ_z$$

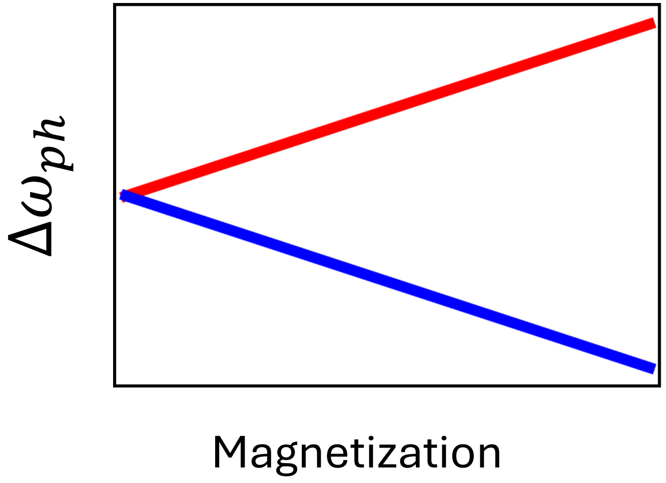
$$C_3^{\hat{n}} Q^{\pm} = e^{\pm i \frac{2\pi}{3} l} Q^{\pm}$$



R. Sutcliffe, Kathleen Hart, S. Chaudhary, and Arun Paramekanti, arxiv: 2506.18978 (2025)

Chiral phonon splitting without spin d.o.f?

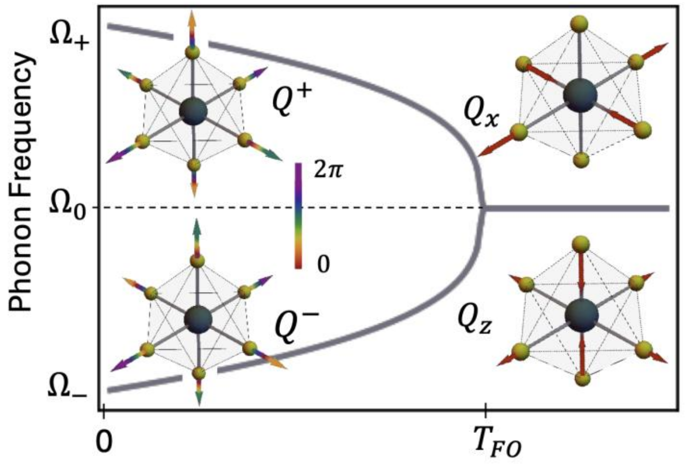




$CeCl_3$, $CoTiO_3$, $Co_3Sn_2S_2$, FZMO

SC, DJ, .., Phys. Rev. B **110**, 094401 (2024)
Che, et al. PRL **134**, 196906 (2025)

TRS breaking from Ferro-octupolar order



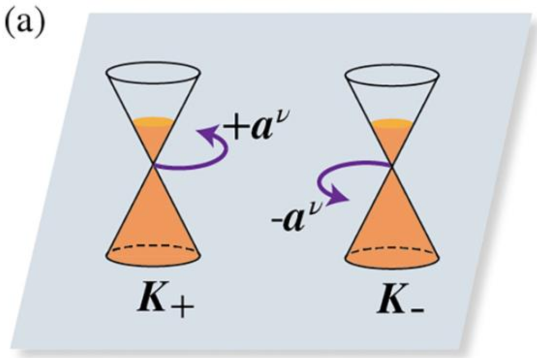
Sutcliffe, Hart, SC, AP, arxiv 2506.18978

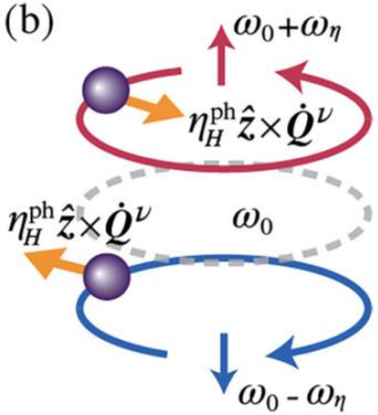
TRS breaking without spin degrees of freedom

?

Chen, Zhang, Su, Cao, Xiao, and Lin, Phys. Rev. B **111**, 035126 (2025)

Chiral phonon splitting and Hall conductivity





A_j : EM fields
a_j : Pseudo gauge fields

$$\mathcal{H}_{\text{eff}} = \sum_j v_j(\hbar k_j - eA_j - e\chi a_j^\nu)\gamma^j$$

Chen, Zhang, Su, Cao, Xiao, and Lin,
PRB **111**, 035126 (2025)

Hu, et.al. PRL **127**, 125901 (2021)

Chern-Simons action for pseudogauge fields

$$S_{\text{CS}}[\boldsymbol{a}^\nu] = \frac{\sigma_{xy}}{2}\int d^3x\epsilon^{ijk}a_i^\nu\partial_j a_k^\nu.$$

Corrections to phonon Lagrangian:

$$\mathcal{L}_\eta = \eta_H^{\text{ph}}(Q_y^\nu\dot{Q}_x^\nu - Q_x^\nu\dot{Q}_y^\nu)$$

$$\eta_H^{\text{ph}} = \sigma_{xy}(g^\nu)^2/(2e^2v_F^2)$$

Hall viscosity

Chiral-phonon splitting

$$\omega_{l/r} = \sqrt{\omega_0^2 + \left(\frac{\eta_H^{\text{ph}}}{\rho_I}\right)^2} \pm \frac{\eta_H^{\text{ph}}}{\rho_I}.$$

Phonon magnetic moment and Hall conductivity

$$\omega_{l/r} = \sqrt{\omega_0^2 + \left(\frac{\eta_H^{\text{ph}}}{\rho_I}\right)^2} \pm \frac{\eta_H^{\text{ph}}}{\rho_I}.$$

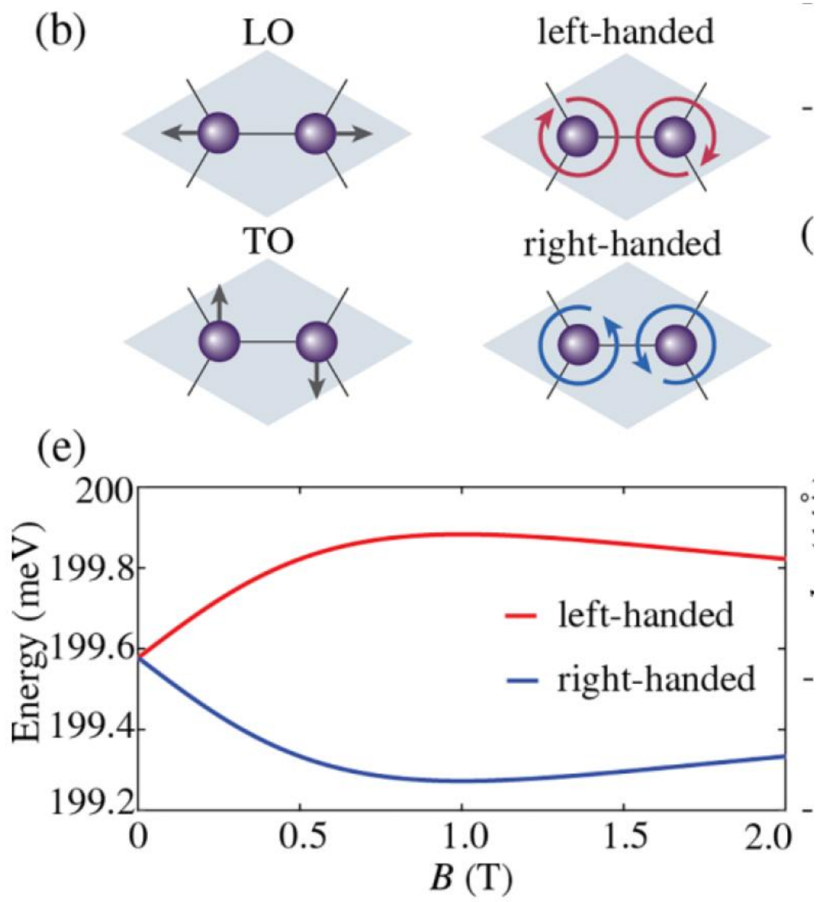
$$\eta_H^{\text{ph}} = \sigma_{xy}(g^\nu)^2/(2e^2 v_F^2)$$

$$\omega_{l/r} = \omega_0\left[\sqrt{1+\left(\xi\frac{\tilde{\mu}_{\text{tr}}B}{1+\tilde{\mu}_{\text{tr}}^2B^2}\right)^2} \pm \xi\frac{\tilde{\mu}_{\text{tr}}B}{1+\tilde{\mu}_{\text{tr}}^2B^2}\right]$$

$$\hbar\omega_{l/r} = \hbar\omega_0 \pm \boldsymbol{\mu}_{\text{ph}} \cdot \boldsymbol{B}$$

$$\mu_{\text{ph}} = \frac{(g^\nu)^2}{v_F^2\rho_I B}\frac{\hbar}{2e^2}\sigma_{xy},$$

$$\mu_{\text{ph}} = 10.6\ \mu_B$$



Chen, Zhang, Su, Cao, Xiao, and Lin,
Phys. Rev. B **111**, 035126 (2025)

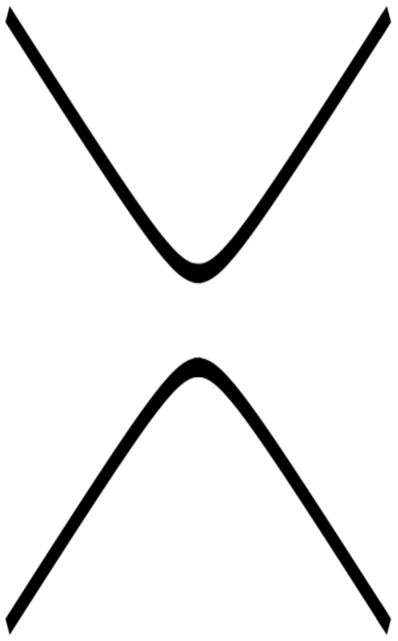
Gapped Dirac system and phonon Hall viscosity

$$\omega_{l/r} = \sqrt{\omega_0^2 + \left(\frac{\eta_H^{\text{ph}}}{\rho_I}\right)^2} \pm \frac{\eta_H^{\text{ph}}}{\rho_I}.$$

$$\eta_H^{\text{ph}} = \sigma_{xy}(g^\nu)^2/(2e^2 v_F^2)$$

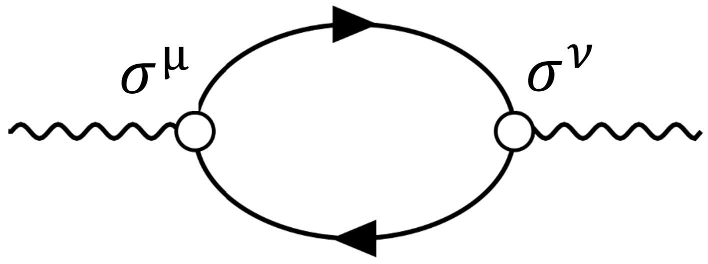
$$\sigma_{xy}(\omega \to 0) = \frac{e^2}{2h}\text{sign}(m)$$

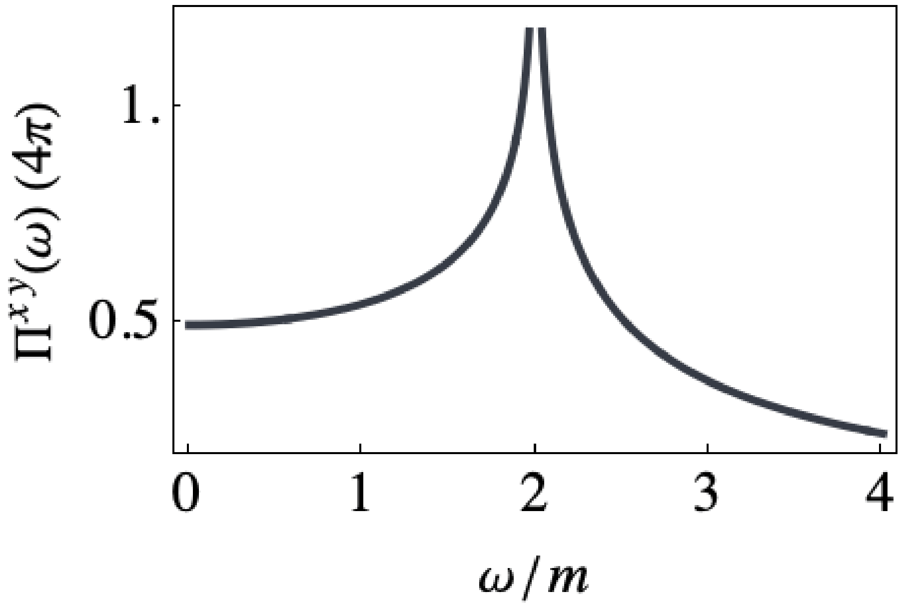
$$\sigma_{xy}(\omega \neq 0) = \frac{e^2}{2h}\text{sign}(m)\Pi^{xy}(\omega)$$



$$H_{el} = vk \cdot \sigma + m\sigma_z$$

$\Pi^{\mu\nu} \equiv$





Chaudhary and Oka, in preparation

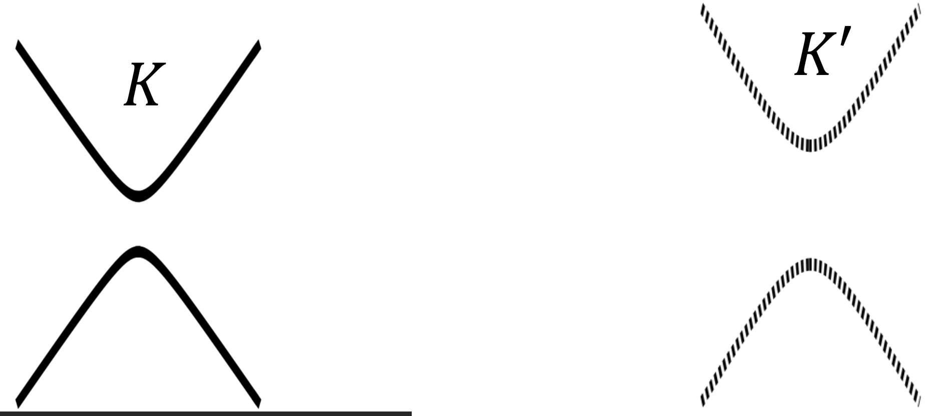
Hu, et.al. PRL **127**, 125901 (2021)

Gapped Dirac system and phonon Hall viscosity

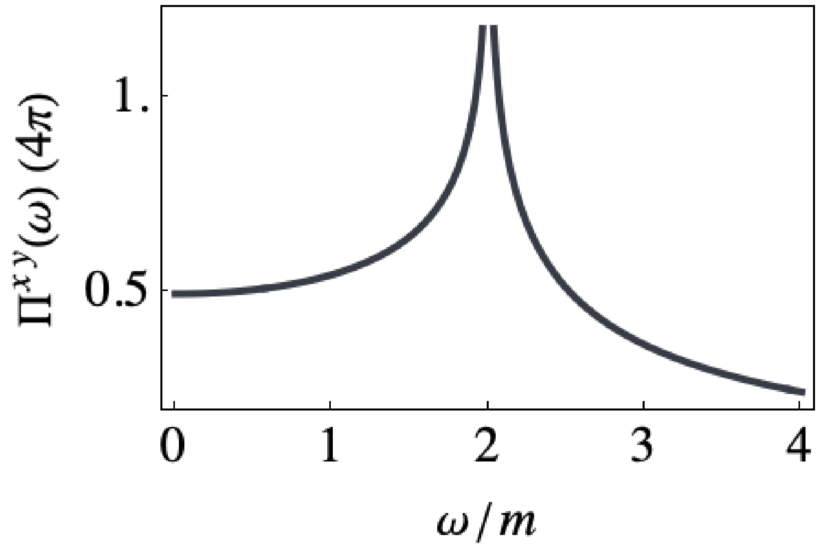
$$\omega_{l/r} = \sqrt{\omega_0^2 + \left(\frac{\eta_H^{\mathrm{ph}}}{\rho_I}\right)^2} \pm \frac{\eta_H^{\mathrm{ph}}}{\rho_I}.$$

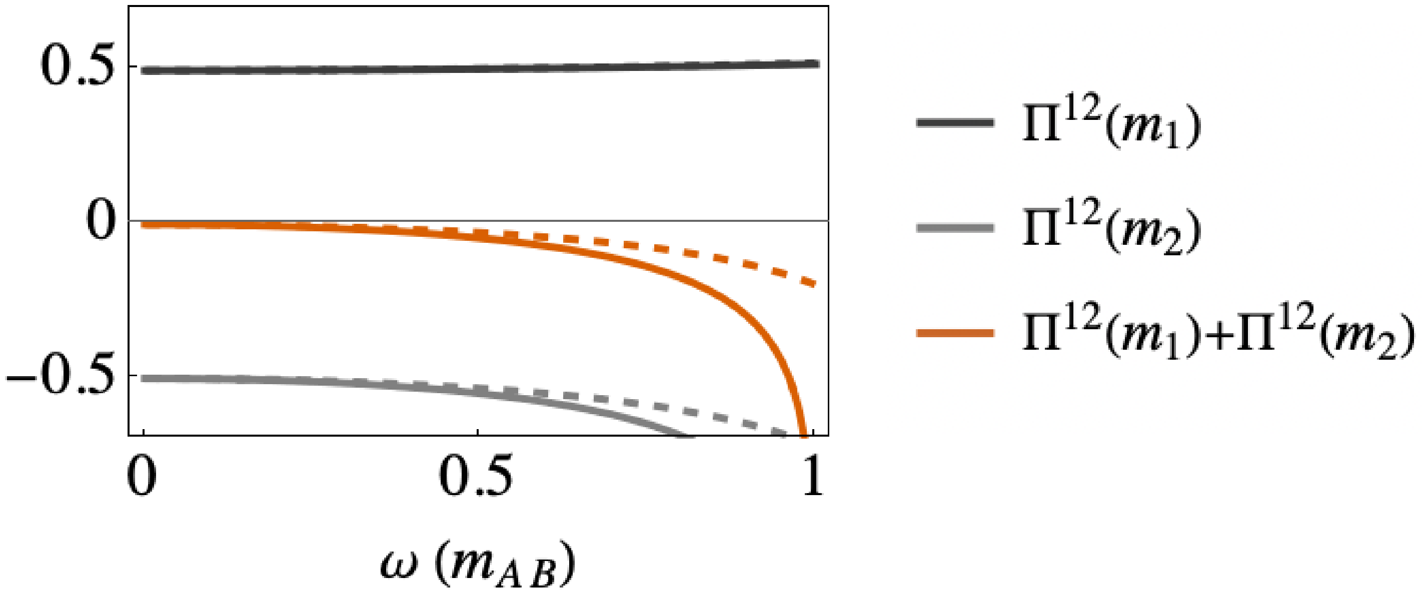
$$\eta_H^{\mathrm{ph}} = \sigma_{xy}(g^\nu)^2/(2e^2 v_F^2)$$

$$\sigma_{xy}(\omega \neq 0) = \frac{e^2}{2h}\mathrm{sign}(m)\Pi^{xy}(\omega)$$

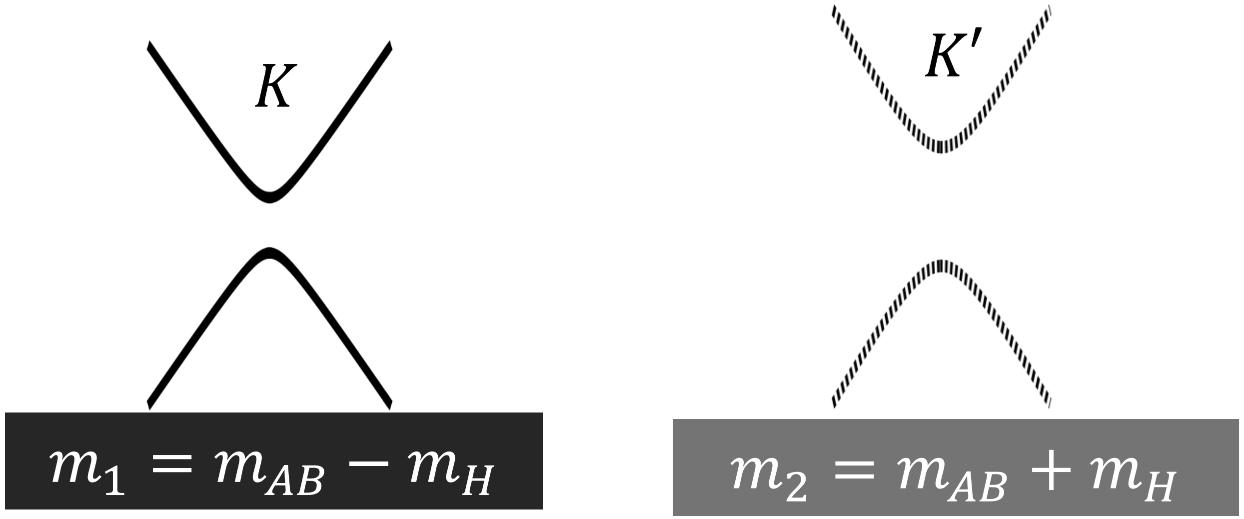


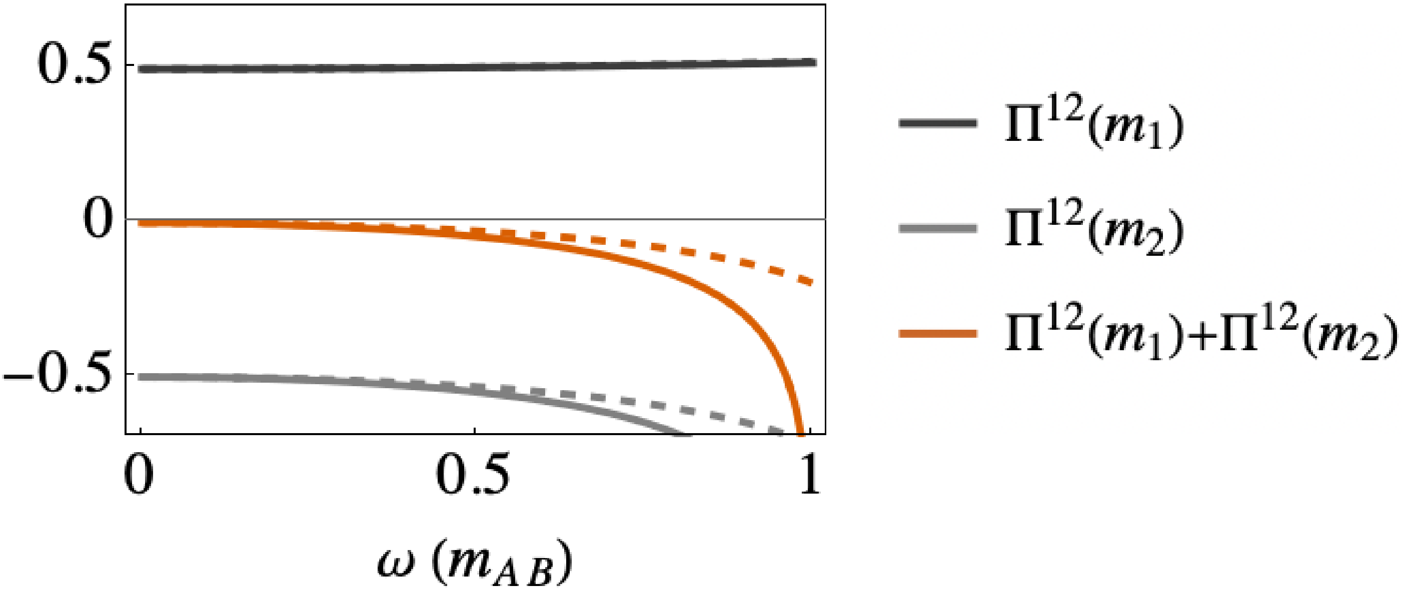
$m_1 = m_{AB} - m_H$ $\quad$ $m_2 = m_{AB} + m_H$

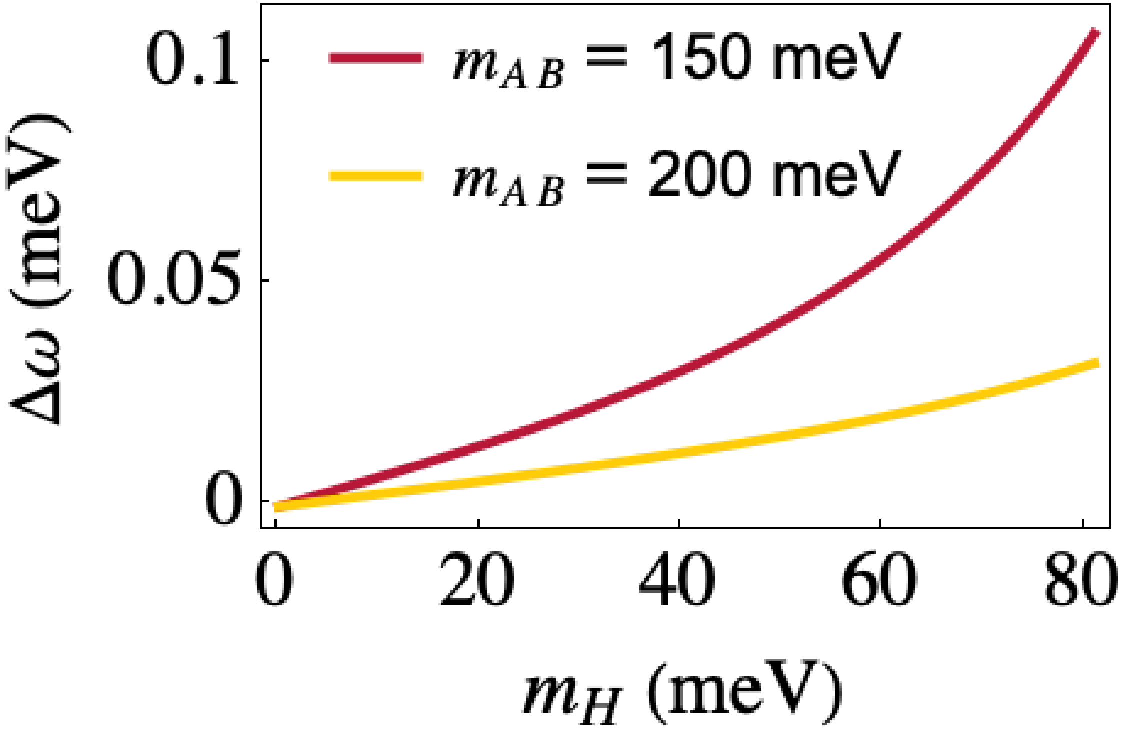




Chiral phonon splitting from local Berry curvature

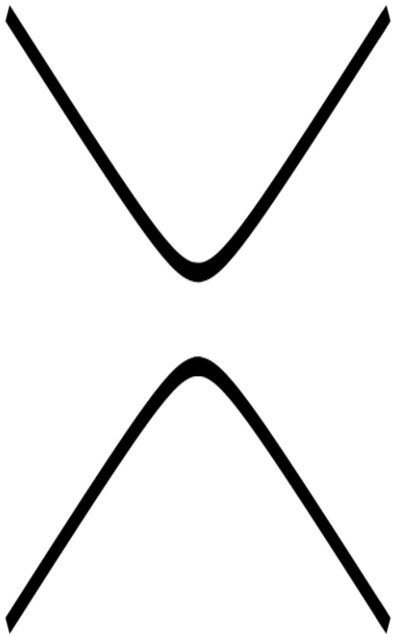


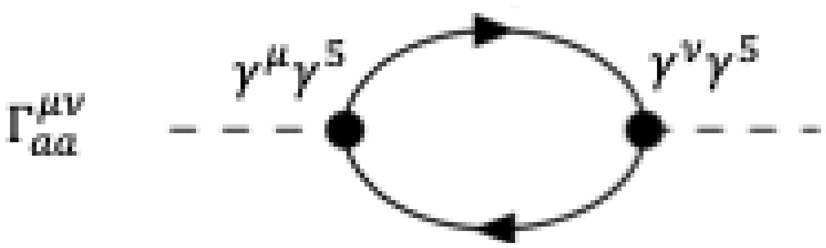




Chaudhary and Oka, in preparation

Dynamics from Chern-Simons like cross-correlations





$$S_{CS}[\boldsymbol{a}^{\nu}] \propto \sigma_{xy} \int d^3x \epsilon^{ijk} a_i^{\nu} \partial_j a_k^{\nu}$$

$$S_{CS}[\boldsymbol{a}^{\nu}, \boldsymbol{A}] \propto \sigma_{xy}^{\text{Valley}} \int d^3x \epsilon^{ijk} a_i^{\nu} \partial_j A_k$$

$$H_{el} = vk \cdot \gamma + m_{AB}\gamma^3 + m_H \gamma^3 \gamma^5$$

$$H_{el-ph} = v(k - a\gamma^5 - A) \cdot \gamma + m_{AB}\gamma^3 + m_H \gamma^3 \gamma^5$$

Chaudhary and Oka, in preparation

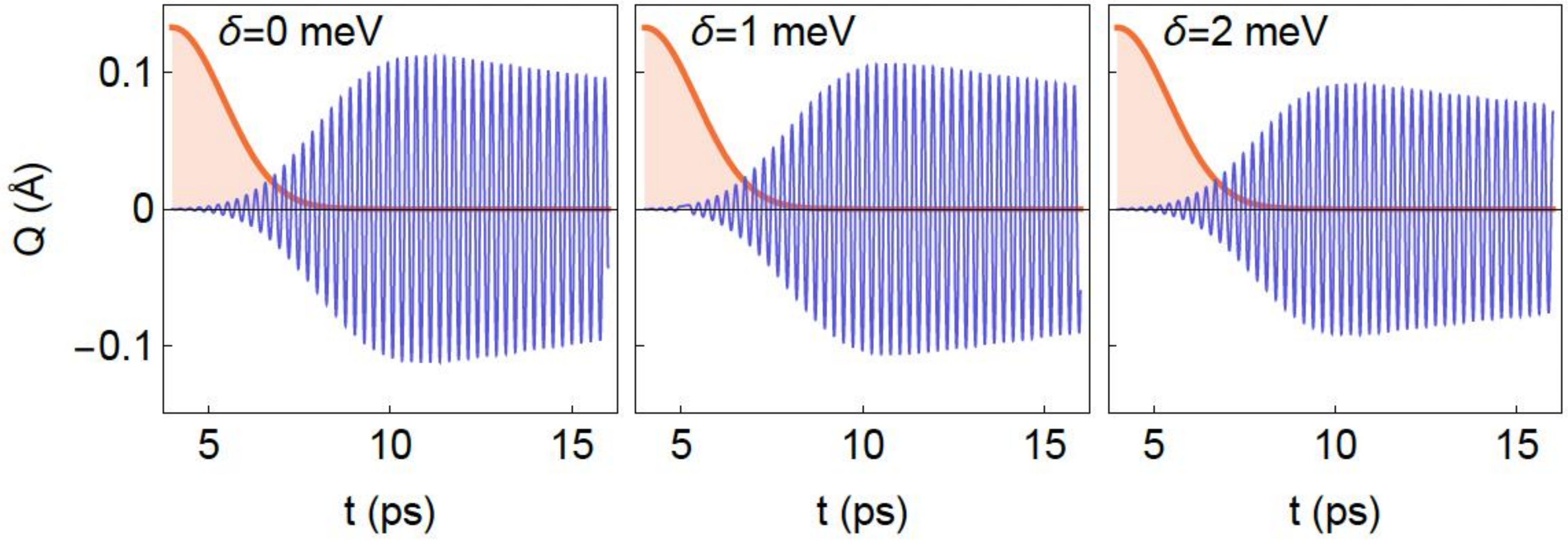
Vaezi, et.al. PRB **88**, 125406 (2013)

Su, Balatsky, Lin, et.al. PRL **134**, 026304 (2025)

Valley Chern number mediates coupling with light

$$\ddot{Q}_{L/R} + \eta \dot{Q}_{L/R} + \omega_0^2 Q_{L/R} = \frac{2\,eg}{2\pi\hbar v_F \rho_I} \tilde{C} \dot{A}_{L/R}$$

Related to geometric
Born effective charge



$E_{peak} = 1\ MV/cm$

Chaudhary and Oka, in preparation

Summary

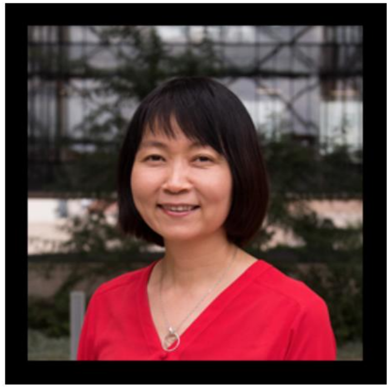
- ❖ Giant magnetic moment from orbit-lattice coupling
- ❖ Chiral phonon splitting from magnetic-octupolar order
- ❖ Raman based possible probe for elusive order
- ❖ Local Berry curvature signatures in phonon spectra

Acknowledgement

Gregory A. Fiete
Northeastern University, Boston

Dominik Juraschek
Eindhoven University of Technology

Martin Rodriguez-Vega
UT Austin

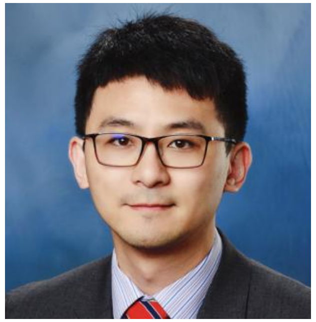


Xiaoqin (Elaine) Li
UT Austin

Jeongheon Choe
UT Austin

David Lujan
UT Austin

Takashi Oka
(ISSP, U Tokyo)



Wencan Jin
Auburn Univ.

Carl P Romao
CTUP, Prague

Arun Paramekanti
(U. Toronto)

Kathleen Hart
(U. Toronto)

Ruairidh. Sutcliffe
(U. Toronto)

Thank you for your attention!