Giant effective magnetic moments of chiral phonons

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Electrons v/s phonons: Magnetic response

Electrons



Phonons



Chiral phonons can carry angular momentum!



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

Chiral phonons: recent examples

Broken inversion symmetry (Valley chiral phonons) Broken time-reversal symmetry (Zone-centered chiral phonons)





Other Works

Bonini, John *et.al* Phys. Rev. Lett. **130**, 086701 (2023) Zhang *et.al* Phys. Rev. Lett. **130**, 226302(2023) Yin *et.al Advanced Materials* 33.36 (2021): 2101618. Liu *et.al* Phys. Rev. Lett. **119**, 255901 (2017)

Magnetic response: classical picture



 $\boldsymbol{L} = \boldsymbol{Q} \times \partial_t \boldsymbol{Q}$ Angular Momentum Phonon Magnetic moment $\mu_{ph} = \gamma L$ $\gamma = \sum_{i} \gamma_i (\mathbf{q}_{i,x} \times \mathbf{q}_{i,y}),$ Gyromagnetic ratio $\gamma_i = \frac{eZ_i}{2M_i}$ $\mu_{ph} \approx \mu_N \approx 5 \,\mathrm{X} \,10^{-4} \mu_B$ $\Delta \omega_{Zeeman} = 2\mu_{ph}B$ Phonon Zeeman effect

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

Observed Magnetic moment



Possible mechanisms for phonon magnetic moment



P. Thalmeier and P. Fulde, Zeitschrift fur Physik B Condensed Matter 26, 323 (1977) **S. Chaudhary**, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete, *Giant effective magnetic moments of chiral phonons from orbit-lattice coupling*, arXiv:2306.11630



2. Chiral phonon coupling to excitation between Kramers pairs



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

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete arXiv:2306.11630



S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete arXiv:2306.11630

- 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 arXiv:2306.11630

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Splitting of chiral phonons



S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete arXiv:2306.11630

Details of model for phonon magnetic moment



Phonon Green's function



Phonon Green's function

e-ph interactions included

$$\begin{aligned} \mathbf{D}^{-1} &= \mathbf{D}_0^{-1} - \mathbf{\Pi}(q, \omega) \\ \Pi^{ab} \propto \frac{f_1 g_a g_b^*}{\omega - \Delta_1} - \frac{f_1 g_a^* g_b}{\omega + \Delta_1} \\ &+ \frac{f_1 g_a^* g_b}{\omega - \Delta_2} - \frac{f_1 g_a g_b^*}{\omega + \Delta_2} \end{aligned}$$

Splitting occurs if

1. $g_a g_b^* = i g^2$

2. $f_1 \neq f_{\widetilde{1}} \text{ or } \Delta_1 \neq \Delta_2$



Phonon energies and eigenmodes

Splitting occurs if

1. $g_a g_b^* = ig^2$

2. $f_1 \neq f_{\widetilde{1}} \text{ or } \Delta_1 \neq \Delta_2$

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

$$\frac{\omega_{ph}^{+} - \omega_{ph}^{-}}{\omega_{ph}(B=0)} = 2\frac{\gamma(\Omega_{+}^{2} - \omega_{0}^{2})/\omega_{0} + \tilde{g}\beta}{\sqrt{(\omega_{0}^{2} - \Delta^{2})^{2} + 8\tilde{g}f_{0}\omega_{0}\Delta}}B + O(B^{2})$$

Broken TRS

$$\begin{split} &\Delta_1 = \Delta - \gamma B, \\ &\Delta_2 = \Delta + \gamma B \\ &f_1 - f_{\widetilde{1}} = \beta B \end{split}$$

New phonon modes

$$\phi_{+} = \frac{1}{\sqrt{2}}(\phi_{a} - i\phi_{b})$$

$$\phi_{-} = \frac{1}{\sqrt{2}}(\phi_{a} + i\phi_{b})$$
Chira

 $\tilde{g} = 4\pi g^2$

Phonon energies and eigenmodes

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- 1. $g_a g_b^* = ig^2$
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- **1.** Energy scales: Δ and ω_0
- 2. Electronic 'g' factor
- 3. Temperature
- 4. Electron -phonon coupling

 $\tilde{g} = 4\pi g^2$

e-ph coupling strength using point charge model

Μ

 u_{α}

$$\begin{split} H_{el-ph} &= (a^{\dagger} + a)\widehat{\mathbf{O}}_{a} + (b^{\dagger} + b)\widehat{\mathbf{O}}_{b} \\ \widehat{O}_{a} &= g_{a}|\psi_{1}\rangle\langle\psi_{2}| - g_{a}^{*}|\psi_{\widetilde{1}}\rangle\langle\psi_{\widetilde{2}}| \\ \widehat{O}_{b} &= g_{b}|\psi_{1}\rangle\langle\psi_{2}| - g_{b}^{*}|\psi_{\widetilde{1}}\rangle\langle\psi_{\widetilde{2}}| \end{split} \qquad u_{a} = \frac{\hbar}{\sqrt{M\hbar\omega_{ph}}} \left(a + a^{\dagger}\right) : \end{split}$$

Modified crystal electric field due to phonon

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

$$\langle \psi_i | r_{\alpha} r_{\beta} | \psi_j \rangle \longrightarrow | \psi_i \rangle = | J_i, \underbrace{m_j^i}_{m_j} \longrightarrow \begin{cases} m_l & \text{Orbital} \\ m_s & \text{Spin} \end{cases} \xrightarrow{|\psi_2\rangle} \frac{|\psi_2\rangle}{|\psi_1\rangle} \xrightarrow{|\psi_1\rangle}$$

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete arXiv:2306.11630

Model applied to Rare earth trihalide $CeCl_3$

Properties of rare-earth paramagnet CeCl₃

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

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



Crystal structure

Electronic states

S. Chaudhary, D. Juraschek, M. Rodriguez-Vega, & G. A. Fiete arXiv:2306.11630

Orbital lattice coupling in CeCl₃



Perturbation to CEFs around Ce³⁺ from phonons in CeCl₃



Zeeman splitting and Phonon Magnetic moment in CeCl₃



Phonon	$\mu_{ph}\left(\mu_B ight)$ 10 K	$\mu_{ph}(\mu_B)$ 20 K
<i>E</i> _{1<i>g</i>} (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 arXiv:2306.11630

Electronic excitations on magnetic ions

Model applied to a *d* orbital magnet

CoTiO₃ physical and electronic properties

Crystal Structure

Co³⁺ electronic states Low-energy Kramers (Three holes in 3d doublets orbitals)_{Nature Communications} 12.1 (2021): 3936.

CoTiO₃ E_g phonon modes

CoTiO₃ magnetic properties

CoTiO₃ E_g modes phonon magnetic moment

Phonon	$\mu_{ph}\left(\mu_B ight)$ 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

Cross-circular channel Raman spectra taken with a magnetic field applied along different crystalline axes Phonon Magnetic moment as a function of temperature

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)

Applications: Mediator between light and magnetism

Phonon Barnett Effect

Giant effective magnetic fields from chiral phonons

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

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

Applications: A new chapter in magnetism

Chiral phonon activated spin Seebeck effect

Chiral phonon-trained octupolar order

K. Kim, et al. Nature Materials 22, 322–328 (2023)

K. Hart, A. Paramkanti, *et al.* arXiv: 2404.17633

Summary and Outlook

- 1. Microscopic model for phonon magnetic moment based on orbital-lattice coupling
- 2. Estimate of phonon magnetic moments in different classes of materials

- 1. Other coupling mechanisms for phonon chirality:
 - > Magnons?
 - Itinerant bands?
- 2. Beyond Gamma phonons consequences for band topology?
- 3. Possible applications Consequences for phonon linewidths?
 - Angular momentum conservation restricts scattering
 - Scattering rates tunable with magnetic field
 - Phonon thermal hall effect

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UT Austin

Thank you for your attention!

Center for Dynamics and Control of Materials: an NSF MRSEC https://mrsec.utexas.edu

Group Theory of chiral phonons

1. E_g , E_u phonons

2. TRS breaking *E* irrep splits into two 1D irreps with complex basis functions

3. Axial vector along C_n axis allowed by symmetry

C ₃	E	C ₃	(C ₃) ²	linear functions, rotations	quadratic functions
A	+1	+1	+1	zRz	x^2+y^2, z^2
Е	+1 +1	+ε +ε [*]	+8 [*] +8	$x+iy; R_x+iR_y$ x-iy; R_x-iR_y	$(x^2-y^2, xy) (yz, xz)$

C_{3h}								
C _{3h}	Е	C ₃ (z)	(C ₃) ²	$\boldsymbol{\sigma}_h$	S ₃	(S ₃) ⁵	linear functions, rotations	quadratic functions
A '	+1	+1	+1	+1	+1	+1	R _z	x^2+y^2, z^2
E'	+1 +1	3+ +8	*3+ +8	+1 +1	3+ +8	*3+ +8	x+iy x-iy	(x^2-y^2, xy)
A"	+1	+1	+1	-1	-1	-1	Z	-
E"	+1 +1	3+ +8*	+8 [*] +8	-1 -1	-8 -8*	- 8 [*] - 8	R _x +iR _y R _x -iR _y	(xz, yz)

C _{3v}	E	2C ₃ (z)	3σ _v	linear functions, rotations	quadratic functions
A ₁	+1	+1	+1	Z	x^2+y^2, z^2
A ₂	+1	+1	-1	R _z	-
E	+2	-1	0	$(\mathbf{x},\mathbf{y}) \left(\mathbf{R}_{\mathbf{x}},\mathbf{R}_{\mathbf{y}}\right)$	$(x^2-y^2, xy) (xz, yz)$

Magnetic point groups for Zone-centered chiral

D	h	0	n	0	n	S

Magnetic point group number	notation
9.1.29	4
10.1.32	$\bar{4}$
11.1.35	4/m
12.4.43	42'2'
14.5.52	$\bar{4}2'm'$
15.6.58	4/mm'm'
16.1.60	3
17.1.62	$\bar{3}$
18.3.67	32'
19.3.70	3m'
20.1.71	$\bar{3}m$

Magnetic point group number	notation
20.5.75	$ar{3}'m'$
21.1.76	6
22.1.79	$\overline{6}$
23.1.82	6/m
24.4.90	62'2'
25.4.94	6m'm'
26.5.99	$ar{6}m'2'$
27.5.104	6'/m'mm'
27.6.105	6/mm'm'
29.1.109	$mar{3}$
32.4.121	$m\bar{3}m'$

CoTiO

$$\begin{pmatrix} \mu_{el}^{gd} B_{z}^{\alpha} & h_{ex}(T) \\ h_{ex}(T) & -\mu_{el}^{gd} B_{z}^{\alpha} \end{pmatrix} |\psi_{\tilde{1}/\tilde{2}}^{\alpha}\rangle = E_{\tilde{1}/\tilde{2}} |\psi_{\tilde{1}/\tilde{2}}^{\alpha}\rangle$$
$$\mathbf{D}^{-1}|_{\alpha\alpha} = \frac{\omega^{2} - \omega_{0}^{2}}{2\omega_{0}} - 2\tilde{g} \left(\frac{f_{\tilde{1}} E_{\tilde{1}3} \left(\cos\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{1}3}^{2}} + \frac{f_{\tilde{1}} E_{\tilde{1}4} \left(\sin\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{1}4}^{2}} \right)$$
$$- 2\tilde{g} \left(\frac{f_{\tilde{2}} E_{\tilde{2}3} \left(\sin\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{2}3}^{2}} + \frac{f_{1} E_{24} \left(\cos\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{2}4}^{2}} \right), \qquad (69)$$

$$\begin{aligned} \mathbf{D}^{-1}|_{ab} &= -\mathbf{D}^{-1}|_{ba} = \\ & 2i\tilde{g}\left(-\frac{f_{\tilde{1}}\omega\left(\cos\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{1}3}^{2}} + \frac{f_{\tilde{1}}\omega\left(\sin\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{1}4}^{2}}\right) \\ & + 2i\tilde{g}\left(-\frac{f_{\tilde{2}}\omega\left(\sin\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{2}3}^{2}} + \frac{f_{\tilde{2}}\omega\left(\cos\frac{\theta}{2}\right)^{2}}{\omega^{2} - E_{\tilde{2}4}^{2}}\right). \end{aligned}$$

Angular momentum of photons

Spin Angular momentum

Orbital Angular momentum

Phys. Rev. A 45, 8185 (1992)

Chiral phonons with giant magnetic moment in CoTiO₃

Prof. Xiaoqin (Elaine) Li

1. Phonon magnetic moment $\mu_{ph} \approx \mu_B$

2. First such example in a 'd' orbital quantum magnet

David Lujan, Jeongheon Choe, **Swati Chaudhary**, *et.al,* Spin-Orbit Exciton-Induced Phonon Chirality in a Quantum Magnet (under review)

Results from helicity resolved magneto Raman spectroscopy

Exp. Data from Li Lab, UT Austin

Cross-circular channel Raman spectra taken with a magnetic field applied along different crystalline axes

Barron criteria: True chirality

Phonon angular momentum estimate: Quantization

Classical Picture

Phonon displacement : $\boldsymbol{u} = (\boldsymbol{u}^1, \boldsymbol{u}^2, ...)$ Normal coordinates :

 $\mathbf{Q} = (\mathbf{u}^1 \sqrt{m^1}, \mathbf{u}^2 \sqrt{m^2}, \dots)$

Circularly polarized phonons $\hat{Q} = Q \sin \omega t \ \hat{x} + Q \cos \omega t \ \hat{y}$

Angular Momentum:

 $\boldsymbol{L} = \boldsymbol{Q} \times \partial_t \boldsymbol{Q} \qquad \boldsymbol{L} = \omega Q^2 \ \hat{z}$

Classical harmonic vibrational energy per unit cell :

$$E_{classical} = \frac{1}{2}\dot{Q}^{2} + \frac{1}{2}\omega^{2}Q^{2} = \omega^{2}Q^{2}$$
$$N_{phonon} = \frac{E_{classical}}{\hbar\omega} = \frac{\omega Q^{2}}{\hbar}$$
$$L = \hbar N_{phonon} \hat{Z}$$

Quantum Picture

$$\begin{aligned} \hat{Q}_{x/y} &= \sqrt{\frac{\hbar}{\omega}} \left(a_{x/y}^{+} + a_{x/y} \right) \\ \hat{Q}_{x/y} &= i\sqrt{\hbar\omega} \left(a_{x/y}^{+} - a_{x/y} \right) \\ \hat{L} &= \hat{Q} \times \dot{\hat{Q}} \equiv i\hbar \left(a_{x}^{+} a_{y} - a_{y}^{+} a_{x} \right) \\ \left| \pm \right\rangle &= \frac{1}{\sqrt{2}} \left(a_{x}^{+} \pm i a_{y}^{+} \right) \left| 0 \right\rangle \\ \hat{L} \left| \pm \right\rangle &= \pm \hbar \left| \pm \right\rangle \end{aligned}$$

$$R^{z}\left(\frac{2\pi}{3}\right)\mathbf{q}_{\mathbf{k}\lambda} = e^{-i(2\pi/3)l_{ph}^{k}}\mathbf{q}_{\mathbf{k}\lambda}$$

D. Juraschek, N. Spaldin, Phys. Rev. Mat. 3, 064405 (2019)

Anharmonic effects

Temperature dependence of Optical phonon frequencies Effects dictated by three phonon scattering processes

$$\omega_p(T) = \omega_p(0) - A\left(1 + \frac{2}{\exp[x] - 1}\right)$$

Temperature dependence of Inverse Lifetime

PHYSICAL REVIEW B 104, L020401 (2021), Martin's paper on Spin-phonon interaction in Yttrium Garnet

Phonon Magnetic moment from band topology in a TCI

F. G. G. Hernandez, A. Baydin, **S. Chaudhary**, F. Tay, & G. A. Fiete, et.al *Chiral Phonons with Giant Magnetic Moments in a Topological Crystalline Insulator*, arXiv:2208.12235 (under review in Science Advances)

$$M_z = \frac{e}{2m_{\rm I}} L_{\rm I} \int \frac{d\mathbf{k}}{(2\pi)^2} \Omega_{k_\alpha k_\beta u_x u_y}$$

Second Chern form

$$\Omega_{k_x k_y u_x u_y} = \Omega_{k_x u_y} \Omega_{k_y u_x} - \Omega_{k_x u_x} \Omega_{k_y u_y} + \Omega_{k_x k_y} \Omega_{u_x u_y}$$

Phy. Rev. Lett. 127, 186403 (2021)

Phonon Magnetic moment from band topology in a TCI

$$H_0 = (m + ck_3^2)\sigma_z + v(k_1s_y - k_2s_x)\sigma_x + v_3k_3\sigma_y,$$
$$H_{ph} = \zeta \left(u_x s_0\sigma_x - u_y s_z\sigma_y\right)$$

