Nonlinear Hall effect from hidden magnetic octupoles

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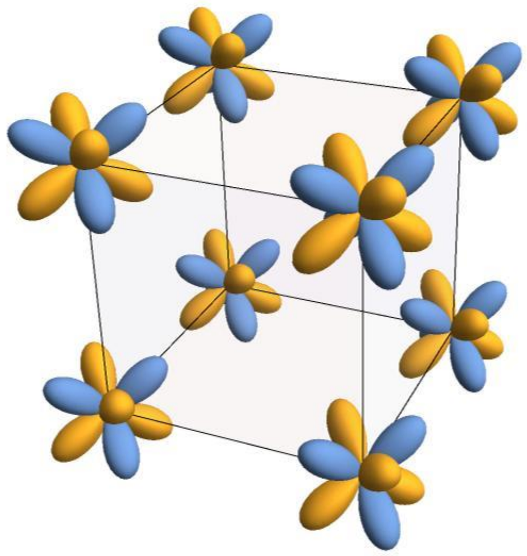
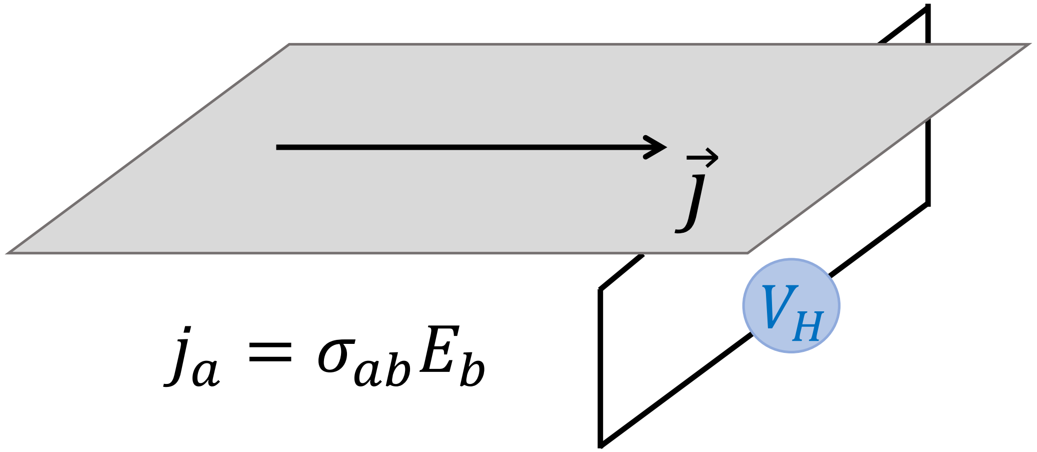


Collaborator

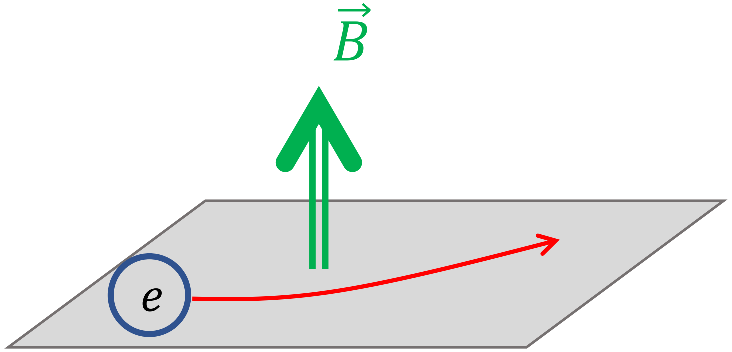
Dr. Adarsh S. Patri
Massachusetts Institute of Technology

S.S., A. S. Patri, arXiv: 2311.03435

Anomalous Hall effect



$$j_a = \sigma_{ab} E_b$$



$$\sigma_{xy} = \sigma_{xy}^{B} + \sigma_{xy}^{int} + \sigma_{xy}^{ext}$$

Equations of motion for electron wave packets

(R. Karplus, J. M. Luttinger, Phys. Rev. 95, 1154, 1954)

$$\frac{d\vec{r}_n}{dt} = \frac{1}{\hbar}\vec{\nabla}_{\vec{k}}\varepsilon_{\vec{k}n} + \frac{e}{\hbar}\vec{E} \times \vec{\boldsymbol{\mathcal{B}}}_{\vec{k}n}$$

$$\frac{d\vec{k}}{dt} = -\frac{e}{\hbar}\vec{E}$$

$$\sigma_{ab}^{H,int} = \frac{e^2}{h}\;\frac{(2\pi)^3}{V}\sum_{\boldsymbol{kn}\,\in occ.} \varepsilon_{abc}\,\boldsymbol{\mathcal{B}}_{\vec{k}n,c}$$

Berry curvature

$$\vec{\boldsymbol{\mathcal{B}}}_{\vec{k}n} = \vec{\nabla}_{\vec{k}} \times i\left\langle \vec{k}n \middle| \vec{\nabla}_{\vec{k}} \middle| \vec{k}n \right\rangle$$

vector potential

Berry connection

For a review of AHE: see e.g. NA Sinitsyn, J. Phys. Condens. Matter 20, 023201, 2007; N. Nagaosa, et al, RMP 82, 1539, 2010

Properties of anomalous Hall effect

$$\sigma_{ab}^{H,int} = \frac{e^2}{h} \frac{(2\pi)^3}{V} \sum_{\boldsymbol{k}n \in occ.} \varepsilon_{abc} \boldsymbol{\mathcal{B}}_{\vec{k}n,c}$$

Local Berry curvature

$$\vec{\boldsymbol{\mathcal{B}}}_{\vec{k}n} = \vec{\nabla}_{\vec{k}} \times \vec{\boldsymbol{\mathcal{A}}}_{\vec{k}n}$$

Relation with topology

$$\sum_{n=occupied} \int_{T^2} d^2k \; \boldsymbol{\mathcal{B}}_{\vec{k}n} = Chern\ number$$

$\vec{\boldsymbol{\mathcal{A}}}_{\vec{k}n}$ = U(1) gauge field
& Brillouin zone is a torus

Relation with symmetry

$$\sigma_{ab}^{H,int} \sim \varepsilon_{abc} \boldsymbol{\mathcal{M}}_c$$

Hall vector

Order parameter $\boldsymbol{\mathcal{M}}_c$
- Break time reversal
- Axial vector

$\boldsymbol{\mathcal{M}}_c$ activates a net Berry curvature; vice versa

Activation of net Berry curvature in magnets

$$\sigma_{ab}^{H,int} \sim \varepsilon_{abc}\mathcal{M}_c$$

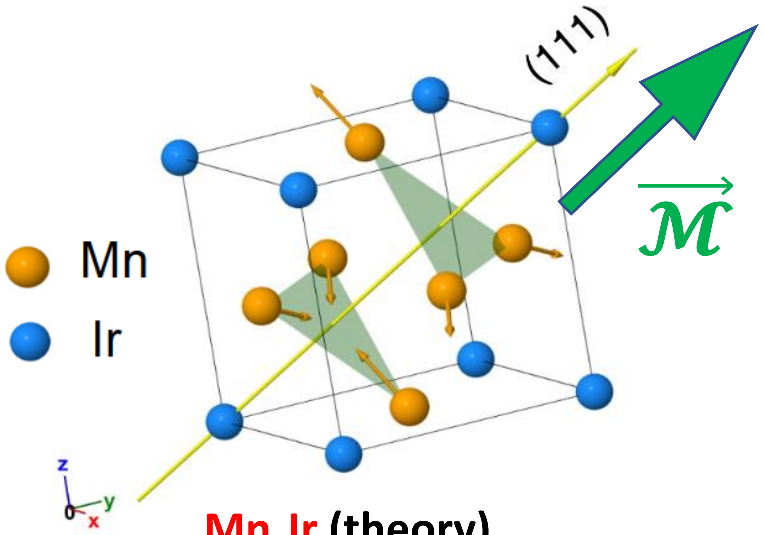
In antiferromagnets, **combination of crystal and magnetic order** can sufficiently lower the symmetry

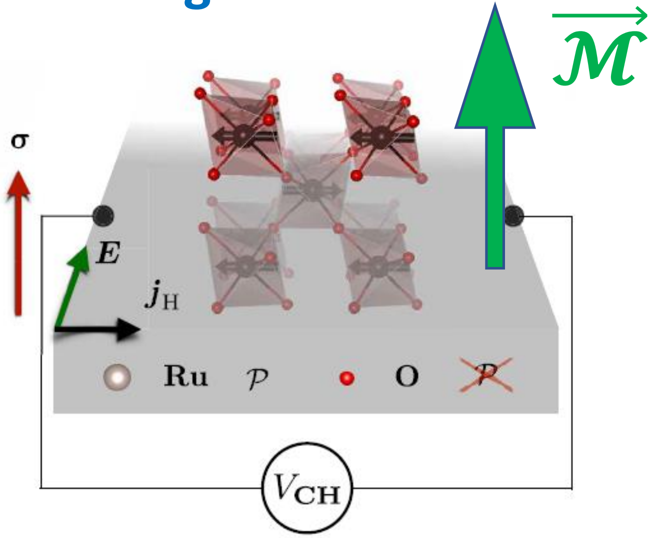
In ferromagnets, $\vec{\mathcal{M}} \leftrightarrow$ magnetization

$$\rho_{xy}^{AHE} \propto M$$

(N. Nagaosa, et al, RMP 82, 1539, 2010)

Large AHE but miniscule magnetization





Mn_3Ir (theory)
H. Chen, et al, PRL 112, 017205 (2014)

Mn_3Sn (exp.)
S. Nakatsuji et al, Nature 527, 212, (2015)

RuO_2 (theory)
L. Šmejkal, et al, Sci. Adv. 6, eaaz8809 (2020)

MnTe (theory + exp.)
R.D. Gonzalez Betancourt, et al, PRL, 036702 (2023)

For a review of AHE in antiferromagnets: see e.g. L. Šmejkal, et al, Nat. Rev. Mater. 7, 482, (2022)

What if such
magnetic-dipole-like order parameters
are absent?

What happens to Hall effect?

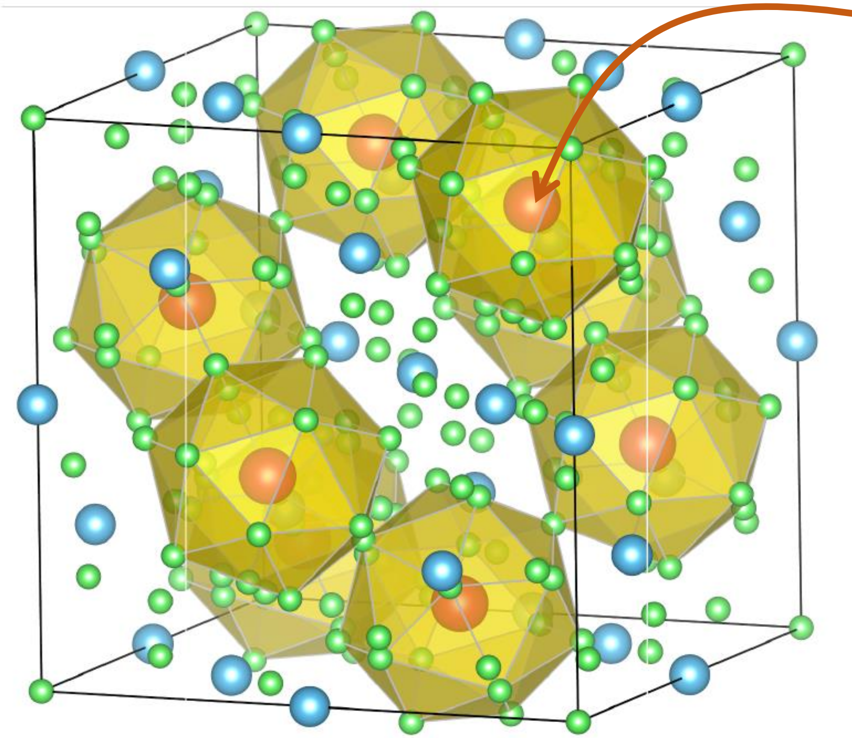
Nonlinear Hall response becomes relevant!

Outline

- Multipolar ordering in heavy-fermion compounds
- From octupolar order to Berry curvature quadrupole & 3rd order Hall effect
- Aspects of the 3rd order Hall effect
- 3rd order Hall effect in other systems

Multipolar ordering in heavy-fermion systems

- Heavy-fermion systems supporting multipolar orders



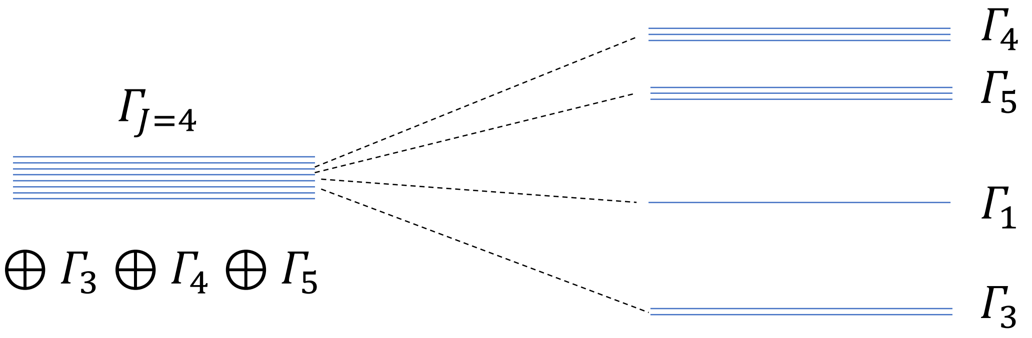
Unit cell of Pr-1-2-20 family compounds
Ex: $Pr(Ti,V)_2Al_{20}$, $PrIr_2Zn_{20}$

Pr^{3+} ion surrounded by a cage with octahedral crystal field environment

$4f^2$ electronic configuration

Applying Hund's rule

$$S_{tot} = 1, \qquad L_{tot} = 5, \qquad J_{tot} = 4$$



$$\Gamma_{J=4} = \Gamma_1 \oplus \Gamma_3 \oplus \Gamma_4 \oplus \Gamma_5$$

Evidence from inelastic neutron exp that Γ_3 is the ground state

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

Multipolar ordering in heavy-fermion systems

- Quenching of dipole moment in Γ_3 subspace

$$(J_i)^{\Gamma_3} = \begin{pmatrix} \langle \Gamma_3^{(1)} | \hat{J}_i | \Gamma_3^{(1)} \rangle & \langle \Gamma_3^{(1)} | \hat{J}_i | \Gamma_3^{(2)} \rangle \\ \langle \Gamma_3^{(2)} | \hat{J}_i | \Gamma_3^{(1)} \rangle & \langle \Gamma_3^{(2)} | \hat{J}_i | \Gamma_3^{(2)} \rangle \end{pmatrix} = 0$$

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

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

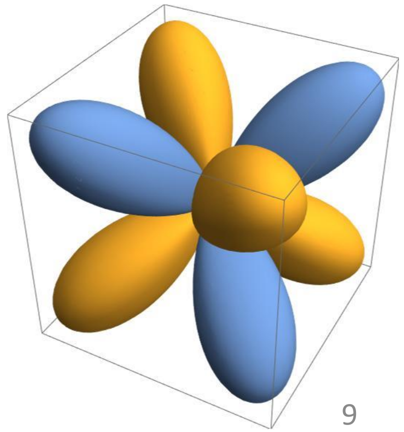
- Using representation theory, one can show Γ_3 supports higher moments

$$\hat{J}_x^2 - \hat{J}_y^2 \qquad 2\hat{J}_z^2 - \hat{J}_x^2 - \hat{J}_y^2$$

Charge quadrupole moments

$$\overline{\hat{J}_x\hat{J}_y\hat{J}_z} = \frac{1}{3!}(\hat{J}_x\hat{J}_y\hat{J}_z + permute\ x, y, z)$$

Magnetic octupole moments



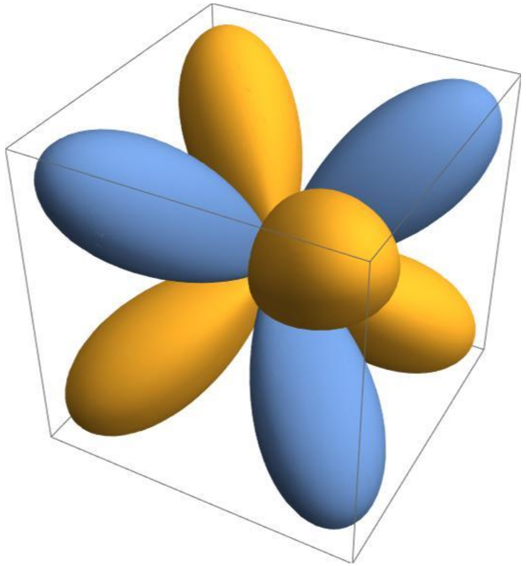
Multipolar ordering in heavy-fermion systems

- Γ_3 doublet supports an octupolar moment

$$\phi \propto \langle \overline{J_x J_y J_z} \rangle$$

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

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



- Odd under time reversal
- Even under inversion
- Possible to develop a long-ranged ferroic ordering
- Hard to detect using conventional local probes, e.g. neutron scattering or magnetic resonance
- Proposal for probably most direct probe of ferro-octupolar order is angular dependence of magnetostriction (A.S.Patri, et al, Nat. Commun. 10, 4092, 2019.)

$$F \sim \phi_{abc}\varepsilon_{ij}h_k$$

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- Aspects of the 3rd order Hall effect
- 3rd order Hall effect in other systems

Cubic lattice model

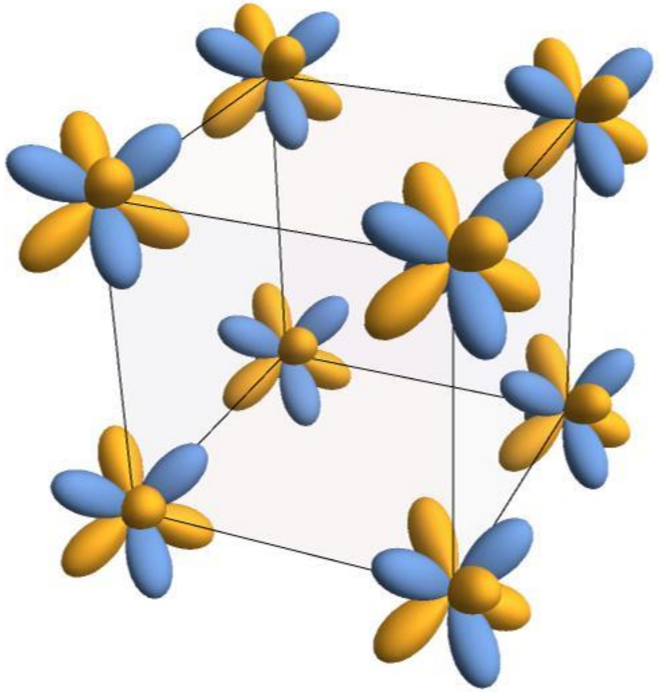
- Slater-Koster tight-binding e_g orbital $\{d_{x^2-y^2}, d_{3z^2-r^2}\}$
- Coupling to order parameter ϕ (A. Patri et al, PRX 2020; A.Patri et al, PRR 2020)

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_\phi$$

$$\mathcal{H}_0(\vec{k}) = h_0(\vec{k})\,\tau^0 + h_3(\vec{k})\,\tau^z + h_1(\vec{k})\,\tau^x$$

$$\mathcal{H}_\phi(\vec{k}) = \phi\,\tau^y$$

Octupolar order parameter
breaking time reversal symmetry

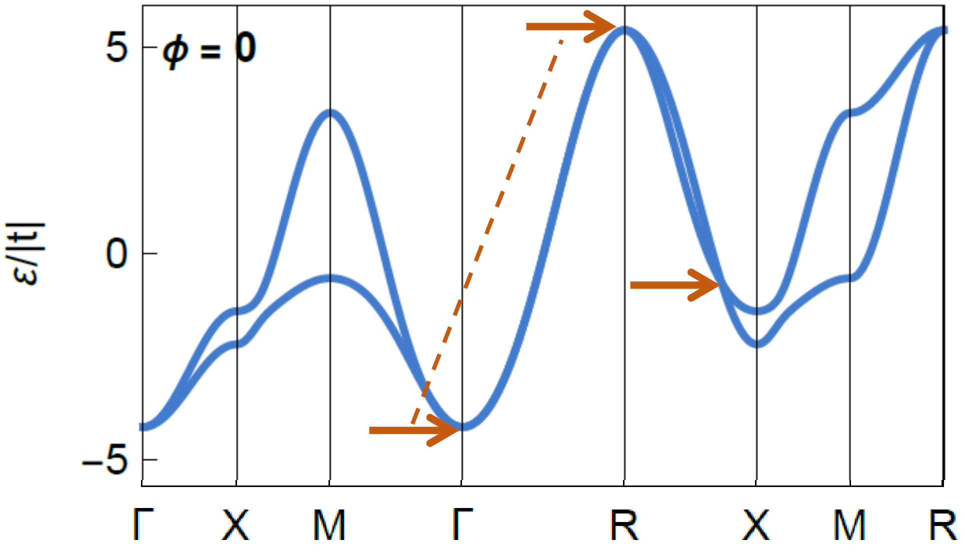


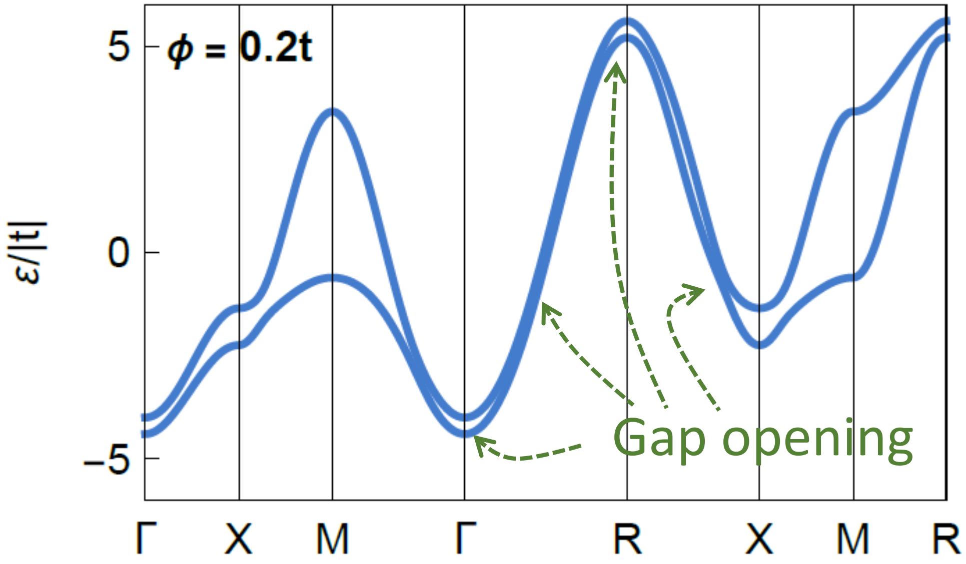
Ferro-octupole ordering on cubic lattice

Ordered state has $m\bar{3}m'$ magnetic point group

Band structure and gap opening

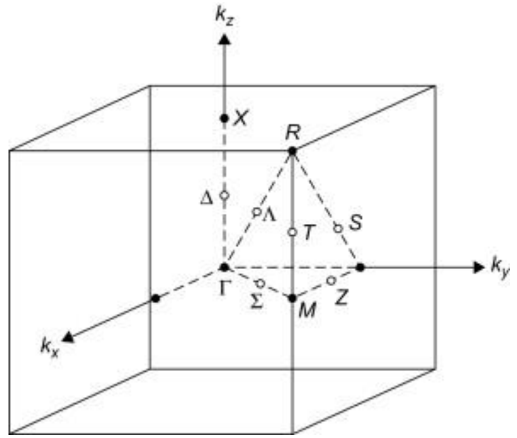
Mirror + C_3 symmetry protection

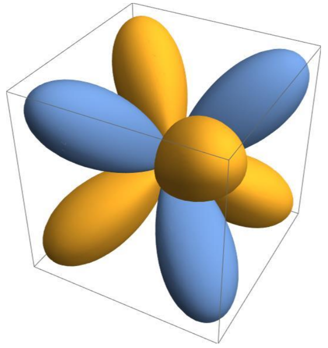




Symmetry protection: $C_3^{[111]}$ & m_1

m_1: mirror plane formed by [111] and [100]



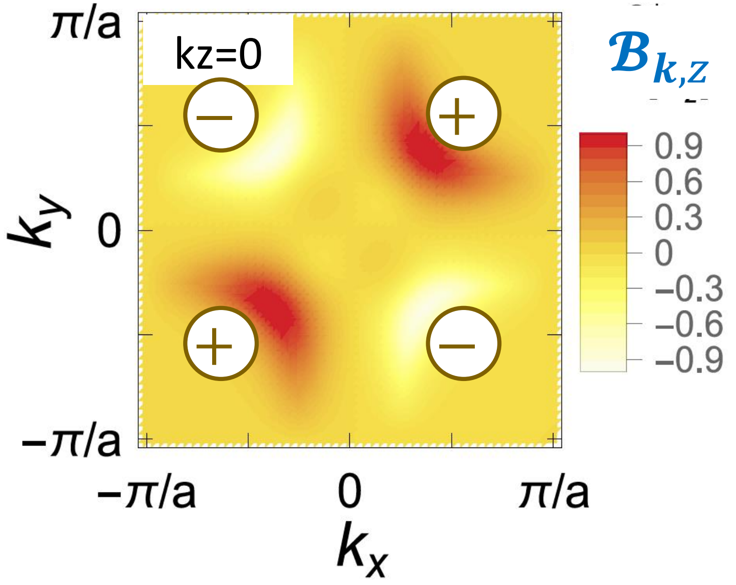


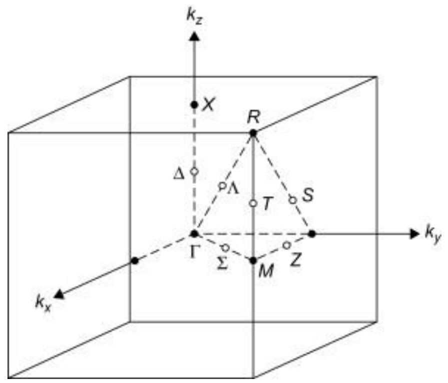
$\langle \overline{J_x J_y J_z} \rangle$

Berry curvature quadrupole

For ϕ $\propto \langle\overline{J_x J_y J_z}\rangle \neq 0$

Berry curvature quadrupole structure





$$\sigma^H_{ab} \propto \varepsilon_{abh} \sum_{kn\,\in occ.} \mathcal{B}_{kn,h} = 0$$

Consistent with the symmetry of the order parameter: $C_{4z}T$

Berry curvature quadrupole

$$Q_{abc} = \int f_0\, \partial_{k_a} \partial_{k_b} \boldsymbol{\mathcal{B}}_c$$

The only independent component of the Berry curvature quadrupole is Q_{xyz}

Hall conductivity tensor

$$\sigma^H_{abcd} \sim \epsilon_{abh} Q_{cdh} + \epsilon_{ach} Q_{dbh} + \epsilon_{adh} Q_{bch}$$

Schematically: $j_H = \sigma^H EEE$

Boltzmann formalism for nonlinear response

- Boltzmann equation for nonequilibrium distribution function $f(\vec{k}, t)$

I. Sodemann, L. Fu, PRL 115, 216806 (2015), T. Morimoto, et al, PRB, 94, 245121, (2016), C.-P. Zhang, et al, PRB 107, 115142 (2023)

$$\frac{d\vec{k}}{dt}.\nabla_{\vec{k}} f(\vec{k}, t) + \partial_t f(\vec{k}, t) = \frac{f_0(\vec{k}) - f(\vec{k}, t)}{\tau}$$

Relaxation time approximation

- Solved together with the semi-classical equations of motion for electron

$$\frac{d\vec{k}}{dt} = -\frac{e}{\hbar}\vec{E}$$

$$\frac{d\vec{r}}{dt} = \frac{1}{\hbar}\nabla_{\vec{k}}\varepsilon_{\vec{k}} + \frac{e}{\hbar}\vec{E} \times \vec{\boldsymbol{\mathcal{B}}}$$

Berry curvature

$$\vec{\boldsymbol{\mathcal{B}}} = \vec{\nabla}_{\vec{k}} \times i \left\langle \vec{k}n \middle| \vec{\nabla}_{\vec{k}} \middle| \vec{k}n \right\rangle$$

- Expansion of the current $\vec{j}(t) = -e \int d\vec{k}\; f(\vec{k}, t) \frac{d\vec{r}}{dt}$

Second-order conductivity

- Second-order current (I. Sodemann, L. Fu, Phys. Rev. Lett. 115, 216806 (2015))

$$E_a(t) = Re[\, E_a(\omega)\, e^{i\omega t}\,]$$

$$j_a^{(2)}(t) = Re\left[j_a^{(2)}(\Omega) e^{i\Omega t} \right]$$

$$j_a^{(2)}(\omega \pm \omega) = \sigma_{abc}^{(\omega \pm \omega)} E_b(\omega)\, E_c(\pm\omega)$$

- Second-har
- Conductivit

Must be noncentrosymmetric systems!

$$\sigma_{abc}^{D} \sim \int f_0 \partial_{k_a} \partial_{k_b} \partial_{k_c} \varepsilon_{\vec{k}n}$$

Joule heating $\sim j_a^{(2)D} E_a \neq 0$

$$\sigma_{abc}^{H} \sim \epsilon_{abh} D_{ch} + \epsilon_{ach} D_{bh}$$

No Joule heating $\sim j_a^{(2)H} E_a = 0$

Berry curvature dipole moment

$$D_{ch} = \int f_0\, \partial_{k_c} \boldsymbol{\mathcal{B}}_h$$

Observation of Berry curvature dipole effect

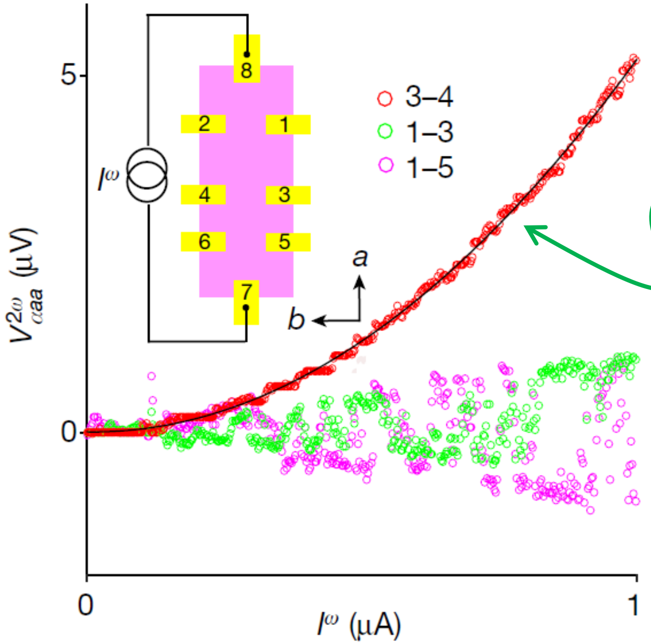
LETTER

https://doi.org/10.1038/s41586-018-0807-6

Observation of the nonlinear Hall effect under time-reversal-symmetric conditions

Qiong Ma[1,13], Su-Yang Xu[1,13], Huitao Shen[1,13], David MacNeill[1], Valla Fatemi[1], Tay-Rong Chang[2], Andrés M. Mier Valdivia[1], Sanfeng Wu[1], Zongzheng Du[3,4,5], Chuang-Han Hsu[6,7], Shiang Fang[8], Quinn D. Gibson[9], Kenji Watanabe[10], Takashi Taniguchi[10], Robert J. Cava[9], Efthimios Kaxiras[8,11], Hai-Zhou Lu[3,4], Hsin Lin[12], Liang Fu[1], Nuh Gedik[1]* & Pablo Jarillo-Herrero[1]*

Apply: sinusoidal longitudinal current
Measure: transverse voltage at second harmonics



Quadratic I-V relation for second harmonic generation in transverse voltage in WTe_2

See more data on WTe_2 in K. Kang et al, Nat. Mat. 18, 324, (2019)
Also experiments on WSe_2 (M. Huang, et al, Nat. Sci. Rev. nwac232(2022) & J.-X. Hu et al, Commun. Phys. 5, 255 (2022))

However, not directly relevant since our system is centrosymmetric.

Third-order Hall effect

- Third-order current (C.-P. Zhang, et al, Phys. Rev. B 107, 115142 (2023))

$$j_a^{(3)}(t) = Re\left[j_a^{(3)}(\Omega) e^{i\Omega t}\right]$$

$$E_a(t) = Re[\, E_a(\omega)\, e^{i\omega t}\,]$$

$$j_a^{(3)}(2\omega \pm \omega) = \sigma_{abcd}^{(2\omega\pm\omega)} E_b(\omega)\, E_c(\omega)\, E_d(\pm\omega)$$

- Third-harmonic generation: $j_a^{(3)}(3\omega) = \sigma_{abcd}^{(3\omega)} E_b(\omega)\, E_c(\omega)\, E_d(\omega)$
- Conductivity tensor

$$\sigma_{abcd}^{(3\omega)} = \sigma_{abcd}^{D} + \sigma_{abcd}^{H}$$

Drude term

$$\sigma_{abcd}^{D} \sim \int f_0 \partial_{k_a}\partial_{k_b}\partial_{k_c}\partial_{k_d}\varepsilon_{\vec{k}n}$$

Joule heating $\sim j_a^{(3)D} E_a \neq 0$

Hall term

$$\sigma_{abcd}^{H} \sim \epsilon_{abh} Q_{cdh} + \epsilon_{ach} Q_{dbh} + \epsilon_{adh} Q_{bch}$$

No Joule heating $\sim j_a^{(3)H} E_a = 0$

Berry curvature quadrupole moment

$$Q_{cdh} = \int f_0\, \partial_{k_c}\partial_{k_d}\, \boldsymbol{\mathcal{B}}_h$$

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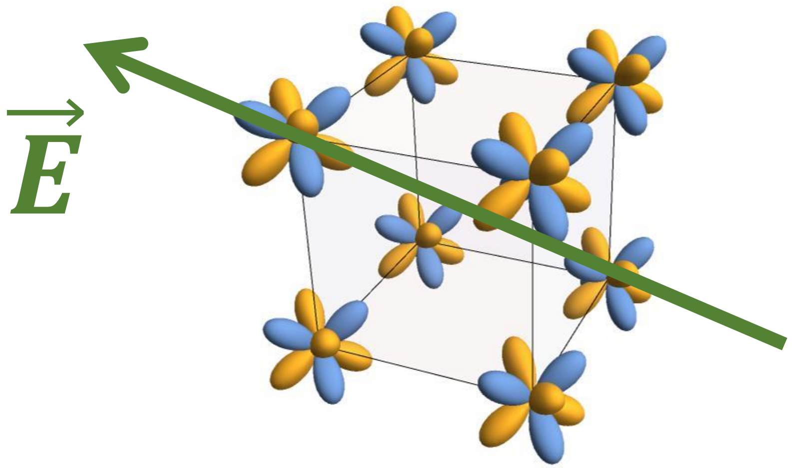
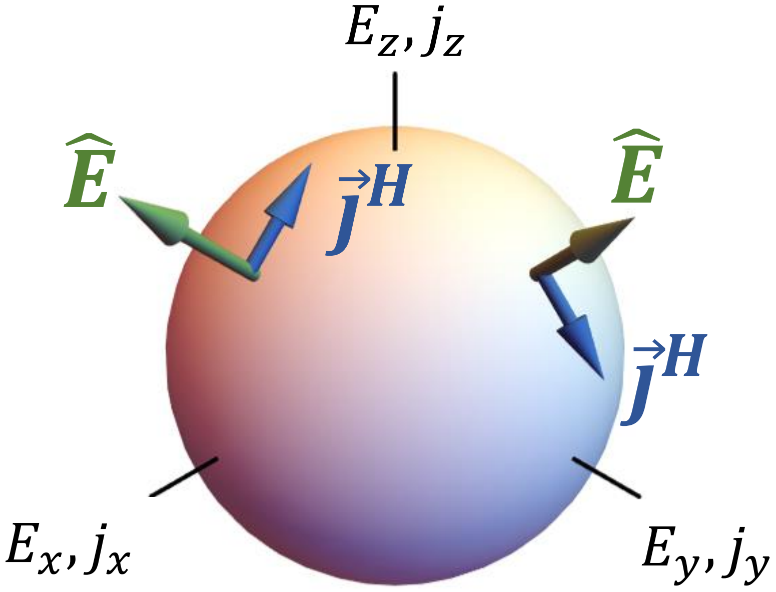
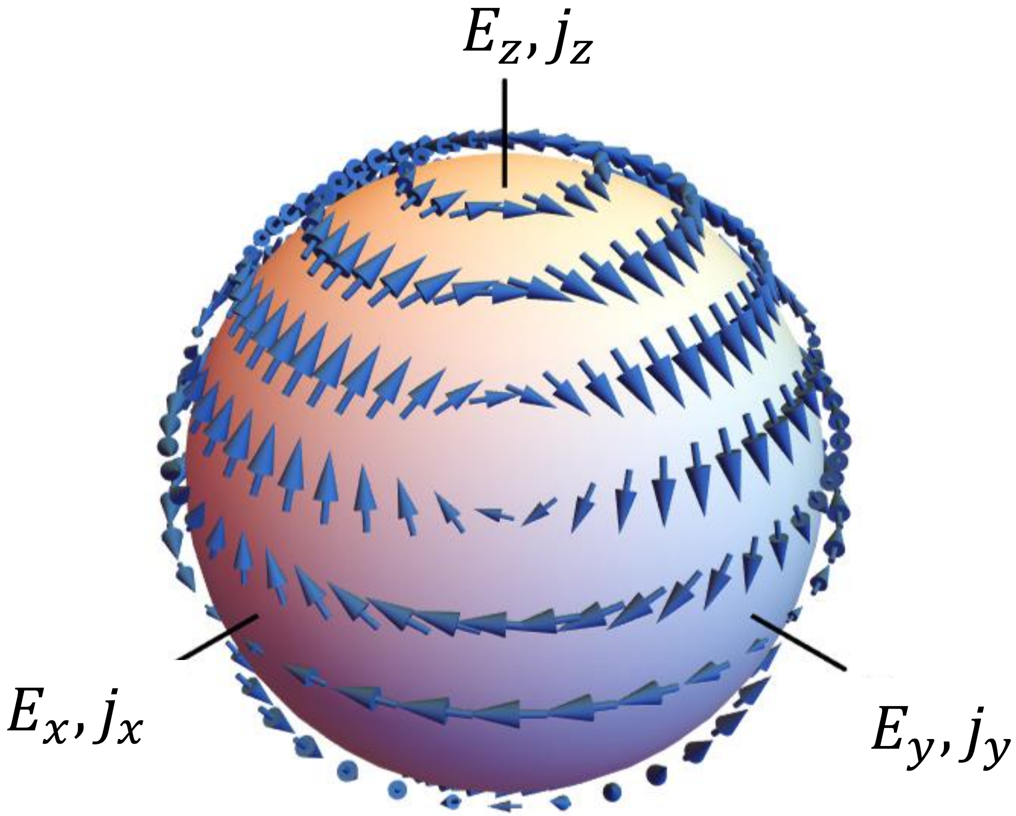
Dependence on orientation of Hall plane



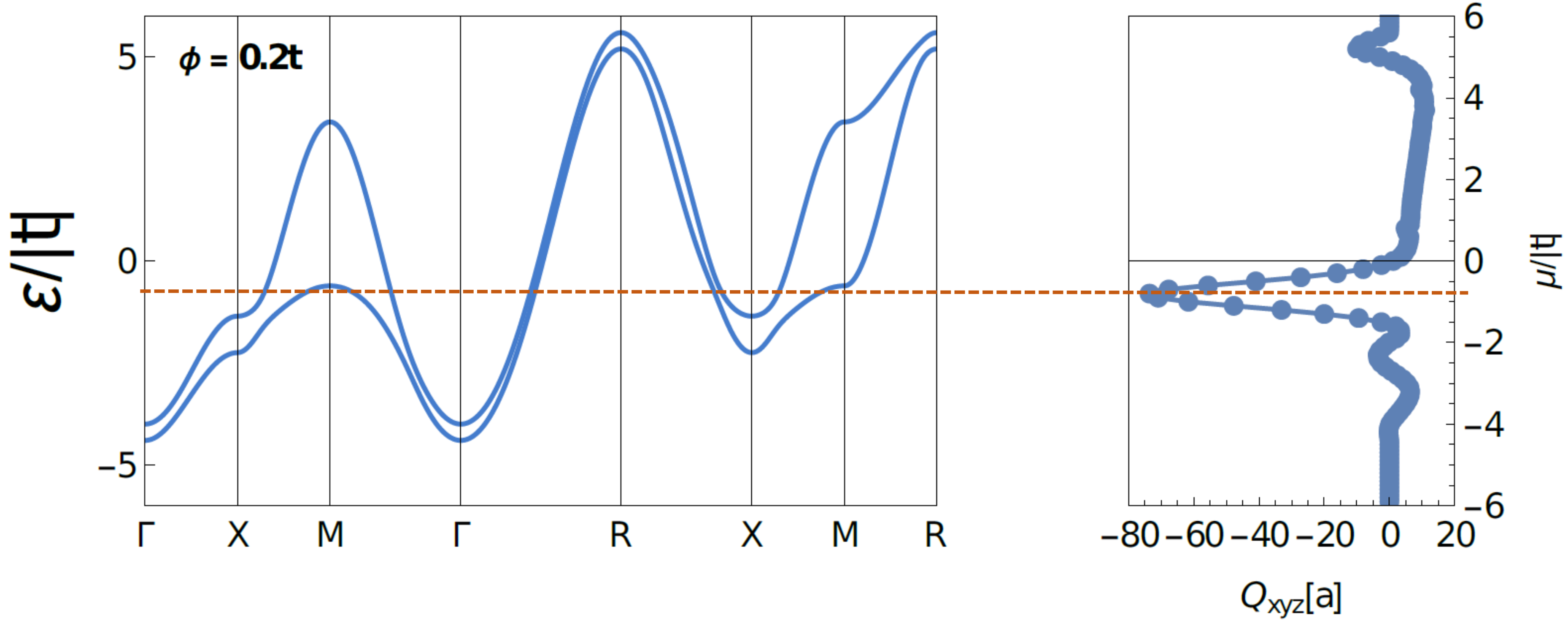
$$j_a^H(3\omega) = \sigma_{abcd}^H \, E_b(\omega)\, E_c(\omega)\, E_d(\omega)$$



$$\vec{j}^H \perp \hat{E}$$

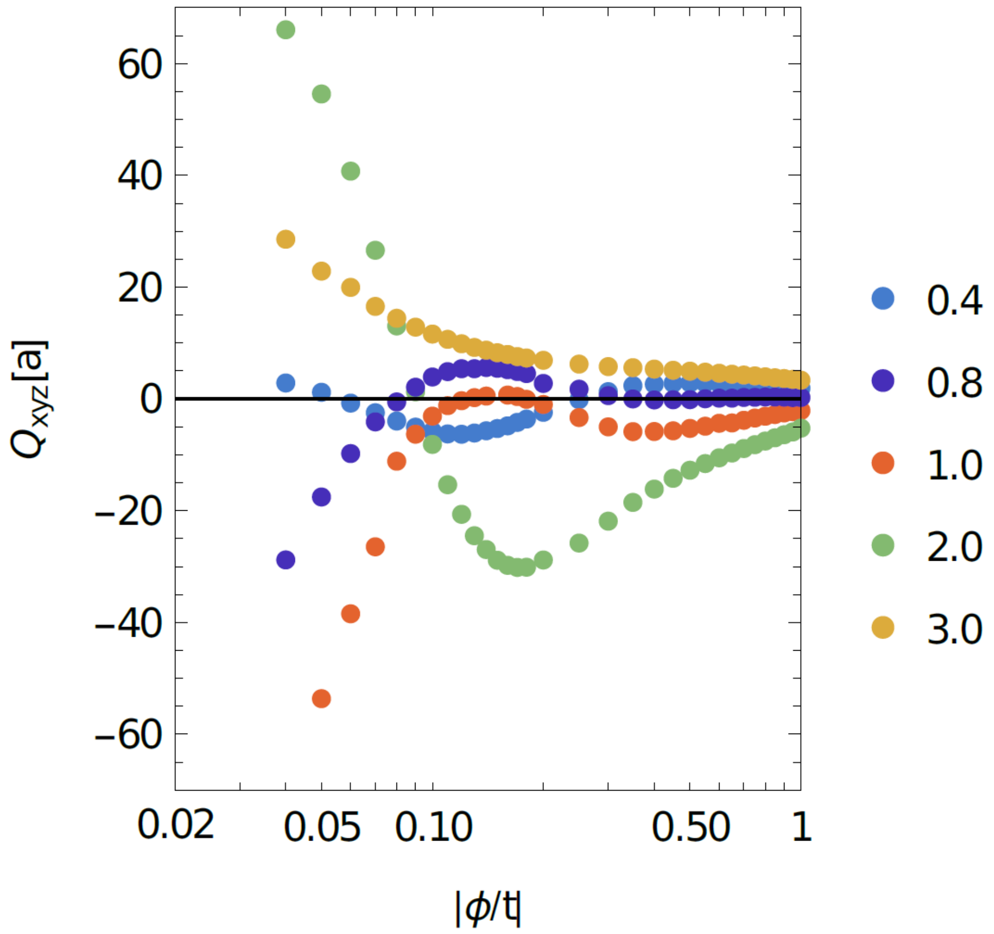


Chemical potential dependence



Nonmonotonic dependence on order parameter

At a fixed electronic density

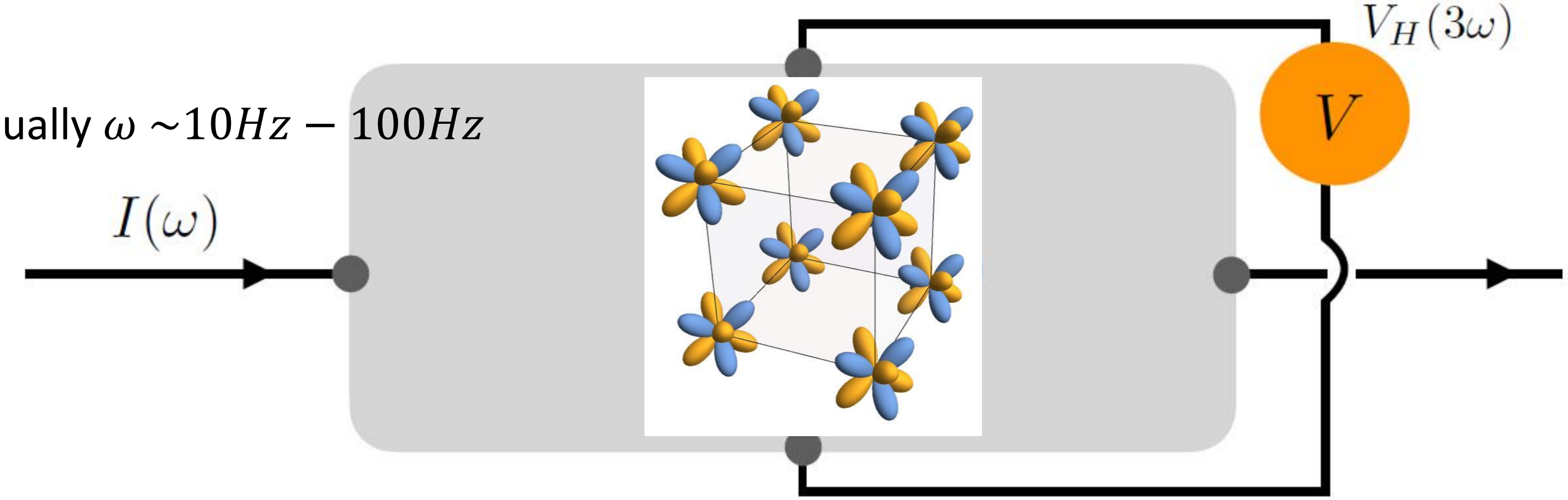


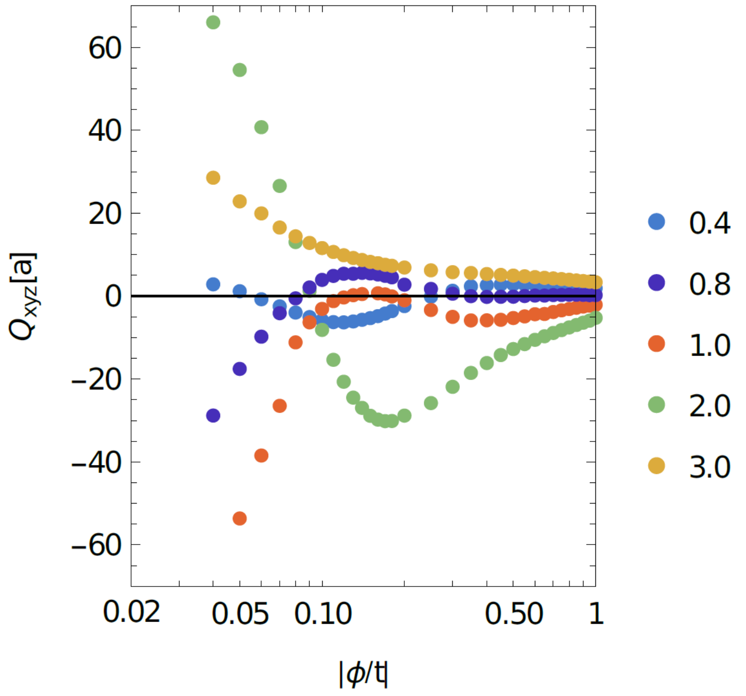
$$Q_{xyz} = \int f_0 \, \partial_{k_x} \partial_{k_y} \, \boldsymbol{\mathcal{B}}_z$$

Hall experiment: third harmonic generation

$$j_a^H(3\omega) = \boldsymbol{\sigma}_{\boldsymbol{a;bcd}}^{\boldsymbol{H,3\omega}} E_b(\omega)E_c(\omega)E_d(\omega)$$

Usually $\omega \sim 10Hz - 100Hz$





a.c. Hall measurement: Applied $\boldsymbol{I(\omega)}$ and measure $\boldsymbol{V_H(3\omega)}$

Estimate of Q_{xyz} assuming lattice spacing of Pr-1-2-20 compounds $\sim 1nm$: $Q_{xyz} \sim 10\text{Å} - 100\ \text{Å}$

Comparable Q_{abc} has been estimated in FeSn where the 3rd Hall effect was measured.
(S. Sankar, et al, arXiv: 2303.03274 (2023))

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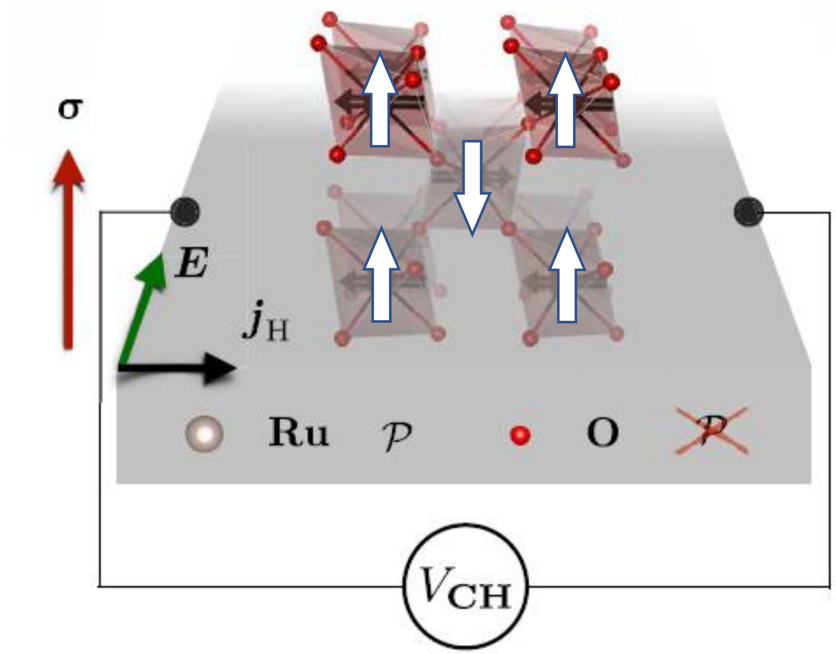
Third order Hall in altermagnetic candidates

PHYSICAL REVIEW X **14**, 011019 (2024)

Ferroically Ordered Magnetic Octupoles in *d*-Wave Altermagnets

Sayantika Bhowal and Nicola A. Spaldin
Materials Theory, ETH Zurich, Wolfgang-Pauli-Strasse 27, 8093 Zurich, Switzerland



AHE no!

System	**n**	st-MPG	mod-MPG
Fe	(001)	$4/mm'm'$ (16)	$m\bar{3}m$ (48)
Co	(0001)	$6/mm'm'$ (24)	$6/mmm$ (24)
Ni	(111)	$\bar{3}m'$ (12)	$m\bar{3}m$ (48)
FeO	(111)	$\bar{3}m1'$ (24)	$\bar{3}m1'$ (24)
Mn_2Au	(100), (110)	$m'mm$ (8)	$4/m'mm$ (16)
RuO_2	(001)	$4'/mm'm$ (16)	$4'/mm'm$ (16)
RuO_2	(100), (110)	$m'm'm$ (8)	$4'/mm'm$ (16)
MnTe	$(11\bar{2}0)$	mmm (8)	$6'/m'm'm$ (24)
MnTe	$(1\bar{1}00)$	$m'm'm$ (8)	$6'/m'm'm$ (24)

(I. Turek, Phys. Rev. B 106, 094432 (2022))

RuO_2 (theory)
L. Šmejkal, et al, Sci. Adv. 6, eaaz8809 (2020)

Theoretical proposal for 3[rd] order Hall for altermagnets
Y. Fang, et al, arXiv: 2310.11489 (2023)

Summary

- First proposal to use Hall measurement to directly detect presence of hidden octupolar orders in heavy-fermion compounds.
- 3rd order Hall effect becomes the leading effect
- Measure 3rd harmonic generation of transverse voltage.
- 3rd order Hall is also important other systems, e.g. d-wave altermagnetic candidates
- Outlook:
  - Experimental realization of our proposal
  - Quantitative estimate taking into account inter-band and extrinsic effects
  - Quantitative understanding the intrinsic effects beyond semi-classical formalism

Reference: <u>S.S.</u>, A. S. Patri, arXiv: 2311.03435