Phonon-induced magnetization and its reciprocity:

effects of geometrical phase and nonlinearity

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YRLG Workshop SPICE, Ingelheim

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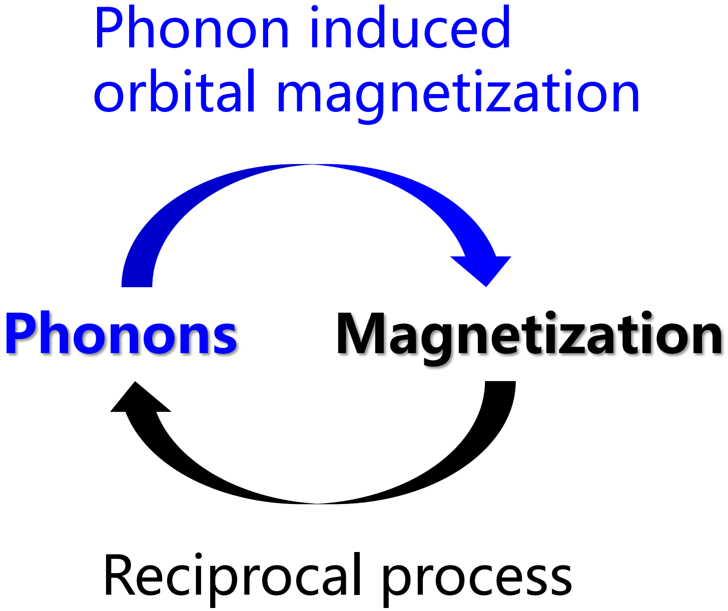
Outline



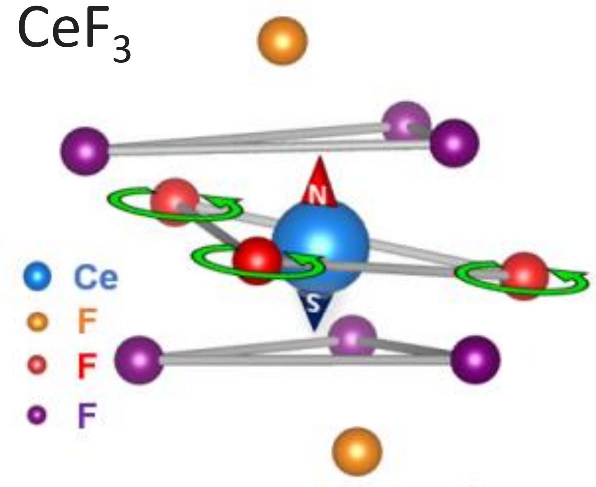
Feedback loop:
Nonlinearity

Phonon Induced Magnetization: Spin vs Orbital

Spin magnetization

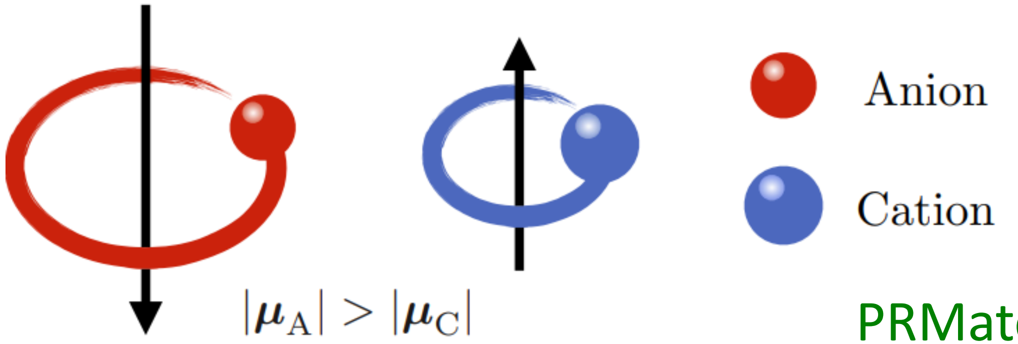
- Paramagnet
- Antiferromagnet

CeF_3



Science **382**, 698 (2023)
Nat. Phys. **13**, 132 (2017)
PRResearch **4**, 013129 (2022)
arXiv:2305.18656 (2023)
arXiv:2306.11630 (2023)

Orbital Magnetization



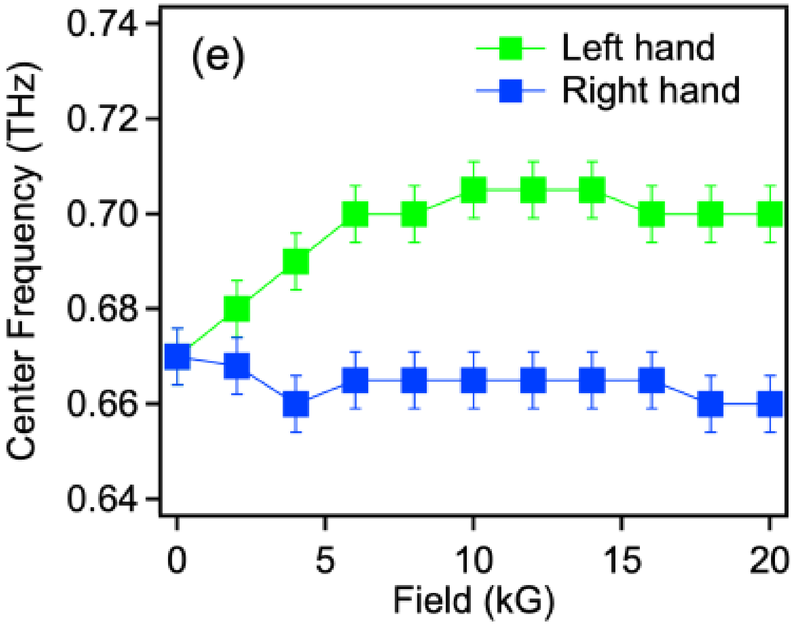
PRMaterials **3**, 064405 (2019)

A classical picture:

- **Linear phonon**: electric dipole $\boldsymbol{P} = \boldsymbol{Z}\boldsymbol{u}$
  - $\boldsymbol{Z}$: Born effective charge tensor
- **Chiral phonon**: $\boldsymbol{M} \sim \boldsymbol{u} \times \boldsymbol{Z}\dot{\boldsymbol{u}}/2$
  - $M\boldsymbol{u} \times \dot{\boldsymbol{u}} \sim \hbar \rightarrow \boldsymbol{M}_{\mathrm{phonon}} \sim \mu_N \sim 10^{-3}\mu_{\mathrm{B}}$

Phonon Magnetic Moment

Phonon Zeeman effect:



Nano Lett. **20**, 5991 (2020)

Cd $4d^{10}5s^2$	As $3d^{10}4s^24p^3$
Pb $5d^{10}6s^26p^2$	Te $4d^{10}5s^25p^4$

Fully filled d orbital

No localized spin

Orbital !

Cd_3As_2 Phonon magnetic moment: $\boldsymbol{M}_{\text{phonon}} \sim 5\ \mu_B$

PbTe Phonon magnetic moment: $\boldsymbol{M}_{\text{phonon}} \sim 0.02\ \mu_B$

PRL **128**, 075901 (2022)

Outline

Quantum description of the phonon induced orbital magnetization?

Phonon induced orbital magnetization

Phonons → **Magnetization**

YR, Xiao, Saparov & Q.N., PRL **127**, 186403 (2021)
Zhang, **YR**, Wang, Cao & Xiao, PRL **130**, 226302 (2023).

A Quantum Picture: Geometrical Phase

Quantum description of electrons:

$$i\hbar \frac{d\Psi}{dt} = H\left(u_x(t), u_y(t)\right)\Psi(t)$$

- $\boldsymbol{u}(t) = (u_x, u_y)$, time periodic
- Adiabatic approximation

➢ Orbital magnetization operator?

$$\boldsymbol{j} = \partial_t \boldsymbol{P} + \nabla \times \boldsymbol{M}$$

➢ $\boldsymbol{P}$: polarization

- $\boldsymbol{P} \sim \int \boldsymbol{A_k} dk, \quad \boldsymbol{A_k} = i\langle \phi_k | \boldsymbol{\partial_k} \phi_k \rangle$
- Apply to phonon induced $\boldsymbol{P}$:

Born effective charge tensor

$$Z_{ij} \sim \frac{\partial P_i}{\partial u_j} \sim \int \Omega_{k_i u_j} dk$$

$$\Omega_{k_i u_j} = -2\mathrm{Im}\langle \partial_{k_i} \phi_k | \partial u_j \phi_k \rangle$$

➢ $\boldsymbol{M}$: magnetization

R. Resta, 1992;
Smith & Vanderbilt, 1993;

Xiao *et al.*, PRL 2009; Dong & Niu, PRB 2018;
YR, Xiao, *et al.*, PRL 2021; Xiao, **YR** & Xiong, PRB 2021

Central Results: Topological Orbital Magnetization

YR, Xiao, Saparov & Niu,
PRL **127**, 186403 (2021)

First Chern form	Second Chern form
$\Omega_{k_x k_y}$ (antisymmetric about two indices)	$\Omega_{k_x u_y}\Omega_{k_y u_x} - \Omega_{k_x u_x}\Omega_{k_y u_y} + \Omega_{k_x k_y}\Omega_{u_x u_y}$ (antisymmetric about four indices)
Dirac monopole (Closed surface in 3D space)	Yang's monopole (Closed surface in 5D space)

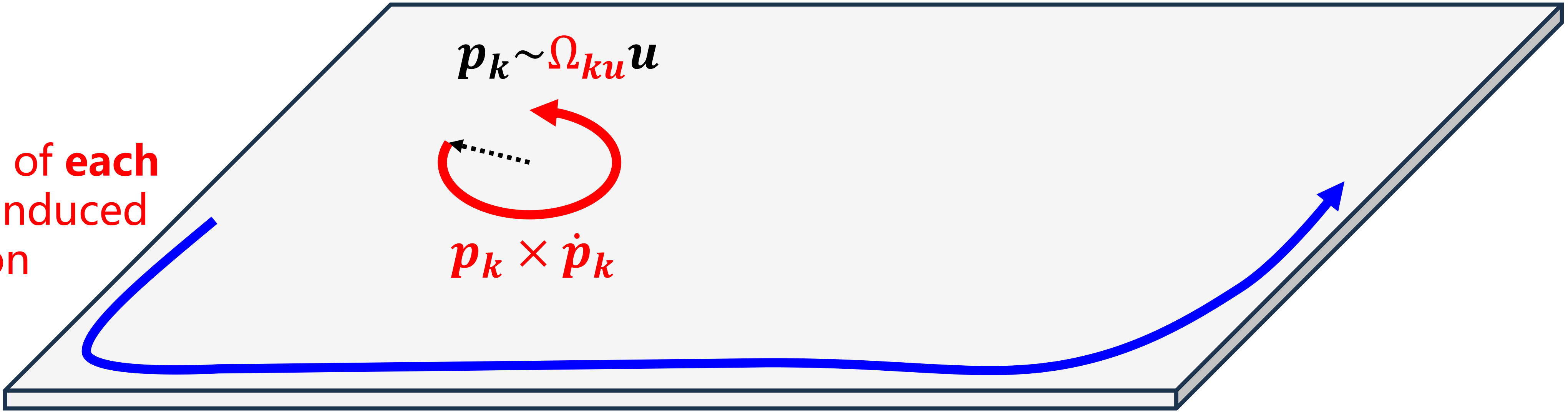
$$\Omega_{ij} = -2\,\mathrm{Im}\langle \partial_i \phi_k | \partial_j \phi_k \rangle$$

$$M_z = \frac{e}{2}(\boldsymbol{u} \times \dot{\boldsymbol{u}}) \int \frac{d\boldsymbol{k}}{(2\pi)^2} \Omega_{k_x u_y}\Omega_{k_y u_x} - \Omega_{k_x u_x}\Omega_{k_y u_y} + \Omega_{k_x k_y}\Omega_{u_x u_y}$$

Central Results: Topological Orbital Magnetization

$\Omega_{\boldsymbol{k}\boldsymbol{u}}$: $\boldsymbol{k}$-resolved Born effective charge

Circular motion of **each** Bloch electron induced by chiral phonon



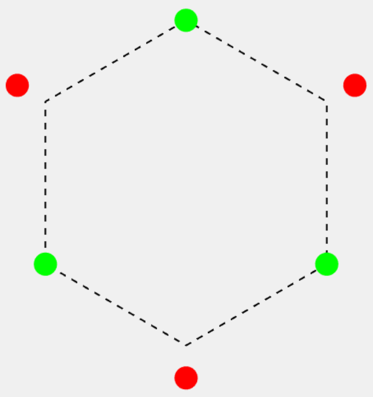
Boundary current induced by chiral phonon

- No classical analogy

$$M_z = \frac{e}{2}(\boldsymbol{u} \times \dot{\boldsymbol{u}}) \int \frac{d\boldsymbol{k}}{(2\pi)^2} \Omega_{k_x u_y}\Omega_{k_y u_x} - \Omega_{k_x u_x}\Omega_{k_y u_y} + \Omega_{k_x k_y}\Omega_{u_x u_y}$$

A Case Study

Gapped graphene + K valley chiral phonon



YR, Xiao, Saparov & Niu, PRL **127**, 186403 (2021)

$$\{|K,B\rangle, |K,A\rangle, |K',A\rangle, |K',B\rangle\}$$

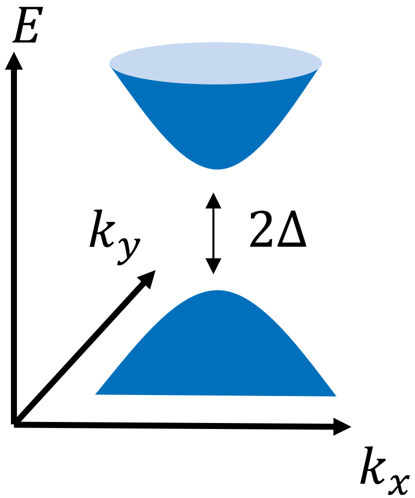
$$\begin{bmatrix} \Delta & v_F\pi^\dagger & \zeta\rho^\dagger & \\ v_F\pi & -\Delta & & \zeta\rho^\dagger \\ \zeta\rho & & -\Delta & -v_F\pi^\dagger \\ & \zeta\rho & -v_F\pi & \Delta \end{bmatrix}$$

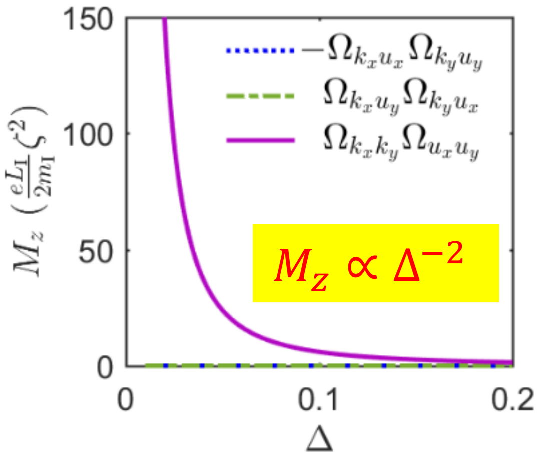
Inter-valley coupling from K-valley phonon

$$\rho = u_y + iu_x$$

$$\pi = k_x + ik_y$$

Energy bands





- Divergently large
- $\Omega_{k_iu_j} = 0 \rightarrow Z_{ij} = 0$
  - Classical estimation $= 0$
  - Quantum estimation $\neq 0$

Different Contributions

- Topological contribution:

$$M_z = \frac{eL_{\mathrm{I}}}{2m_{\mathrm{I}}} \int \frac{d\boldsymbol{k}}{(2\pi)^2} \mathrm{Tr}\left(\Omega_{k_x u_y}\Omega_{k_y u_x} - \Omega_{k_x u_x}\Omega_{k_y u_y} + \Omega_{k_x k_y}\Omega_{u_x u_y} \right)$$

- Orbital, non-topological

Trifunovic, Ono & Watanabe, PRB (2019)

Xiao, **YR** & Xiong, PRB 2021

$$M_z^{nt} = -\frac{e}{2m_{\mathrm{I}}} L_{\mathrm{I}} (\partial_{u_x} F_{u_y} - \partial_{u_y} F_{u_x})$$

$$F_{u_i} = \mathrm{Re} \sum_{n \in \mathrm{occu}} \sum_{n',m' \in \mathrm{unoc}} \int \frac{dk}{(2\pi)^2} \times \frac{\langle n|\partial_{u_i} H|n'\rangle [(\boldsymbol{v}_{n'm'} + \boldsymbol{v}_{nn}\delta_{n'm'}) \times \boldsymbol{v}_{m'n}]_z}{(E_n - E_{n'})^2 (E_n - E_{m'})}$$

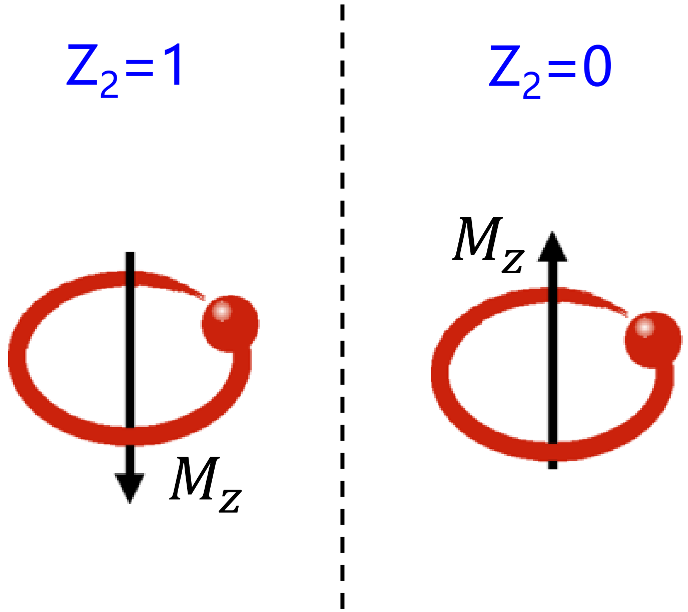
- Spin contribution

Hamada & Murakami,
PRR **2**, 023275 (2020)

Phonon modifies the wavefunction (both spin and orbital parts)

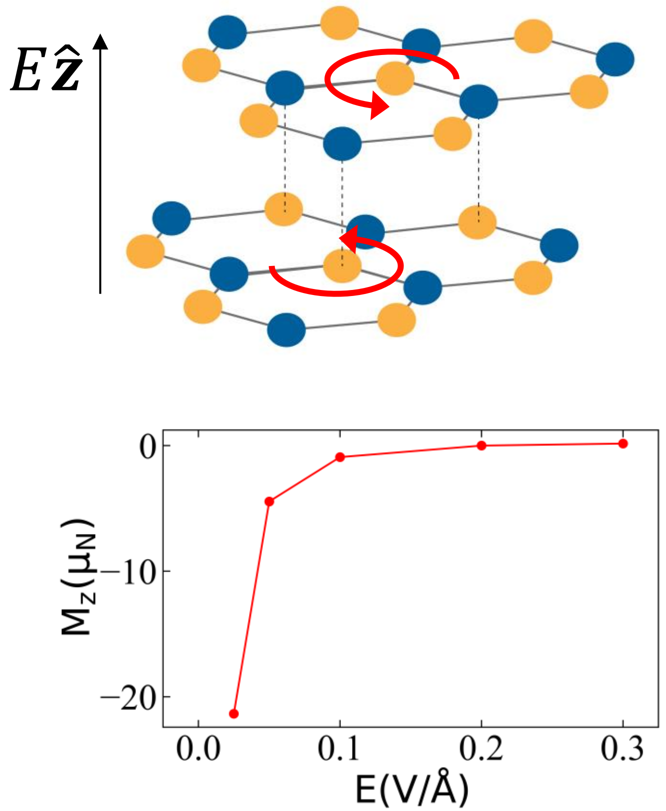
Tunable Phonon Magnetic Moment

By electronic topology (Z_2 TI):



YR, Xiao, Saparov & Niu, PRL (2021)

By electric field (bilayer graphene):

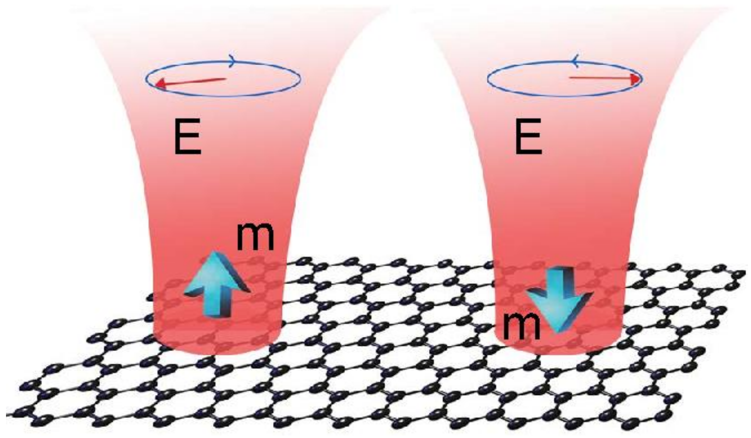


First principles calculation

Zhang, **YR**, Wang, Cao & Xiao
PRL **130**, 226302 (2023)

Topological Contribution: Phonon vs Light

Electron-**light** coupling: Minimal coupling $H(\boldsymbol{k}-\boldsymbol{A})$ with $\boldsymbol{E}=\frac{\partial \boldsymbol{A}}{\partial t}$



PRB **101**, 174429 (2020)

Topological contribution vanishes!

$$\Omega_{k_x A_y}\Omega_{k_y A_x}-\Omega_{k_x A_x}\Omega_{k_y A_y}+\Omega_{k_x k_y}\Omega_{A_x A_y}=0$$

Electron-**phonon** coupling: $H(\boldsymbol{k})+\nabla_{\boldsymbol{u}}H\cdot\boldsymbol{u}$

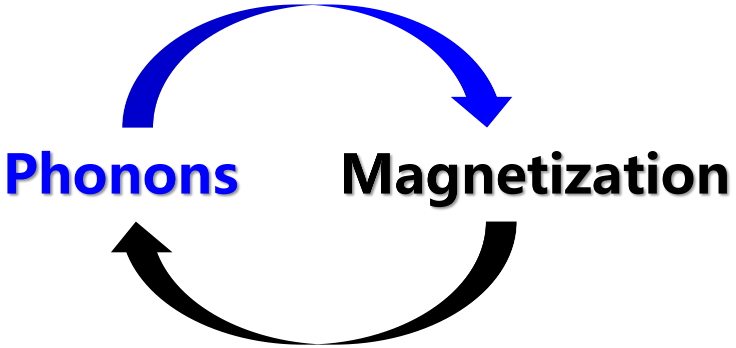
Topological contribution is nonzero!

Novel coupling → Novel physics beyond light driven physics

Outline

Anomalous phonon splitting
without B field?

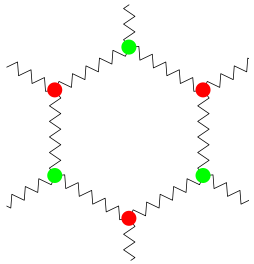
Phonons influence electrons
→ phonon magnetic moment



Magnetization influence phonons
→ Chiral optical phonon

Saparov, Xiong, **YR*** & Niu, PRB **105**, 064303 (2022)

Time Reversal Symmetry Breaking



Electronic **energy** $V(u_i)$:

- Scalar potential, time reversal **even**

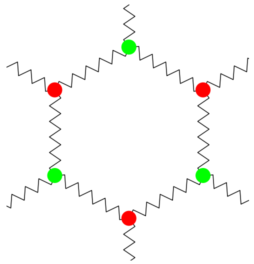
$$V(u_i) \simeq \frac{1}{2}\sum_{ij} u_i D_{ij} u_j$$

1. Angular momentum : $L(-\boldsymbol{k}) = -L(\boldsymbol{k}) \;\rightarrow\; \boldsymbol{L}(\mathbf{0}) = \mathbf{0}$
2. Reciprocal dispersion: $\omega(-\boldsymbol{k}) = \omega(\boldsymbol{k}) \;\rightarrow\; \boldsymbol{v}(\boldsymbol{k}) = -\boldsymbol{v}(\boldsymbol{k})$

How to break time reversal symmetry of phonons?

- Direct Lorentz force
- Lorentz force on electron + electron phonon coupling

Time Reversal Symmetry Breaking



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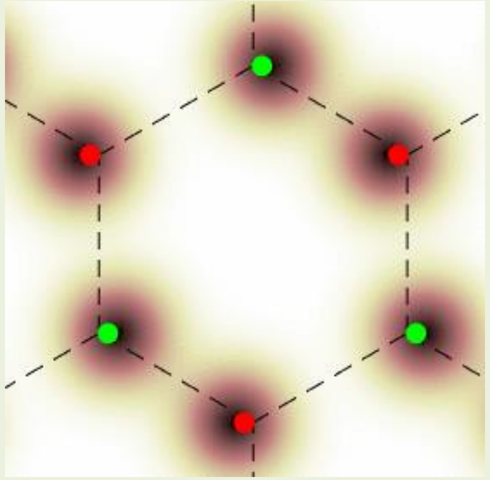


Electronic **wavefunction** $|\Psi_e(u_i)\rangle$

$$\Psi[\boldsymbol{u_i}(\mathrm{T})] = e^{i\gamma} e^{i\int_0^T E(t)dt}\Psi[\boldsymbol{u_i}(0)]$$

Geometric phase $\gamma = \int \boldsymbol{A}_{\boldsymbol{u_i}} \cdot d\boldsymbol{u_i}$

$$\boldsymbol{A}_{\boldsymbol{u_i}} = i\langle\Psi|\partial_{u_i}\Psi\rangle$$

- **Vector** potential, time reversal **odd**

$$T = \sum_i \frac{\left(\boldsymbol{P}_i - \boldsymbol{A}_{\boldsymbol{u_i}}\right)^2}{2M_i}$$

Born & Huang (1954)
Mead & Truhlar, JCP 1979

For stationary states the $\phi_n(x, X)$ can be chosen as real functions; then

$$A_{nn}^{(k)}(X) = -\frac{i\hbar}{2}\frac{\partial}{\partial X_k}\int \phi_n^2(x, X)\, dx = 0, \quad \text{✗} \qquad \text{(VIII.9)}$$

Nonlocal Effective Magnetic Field

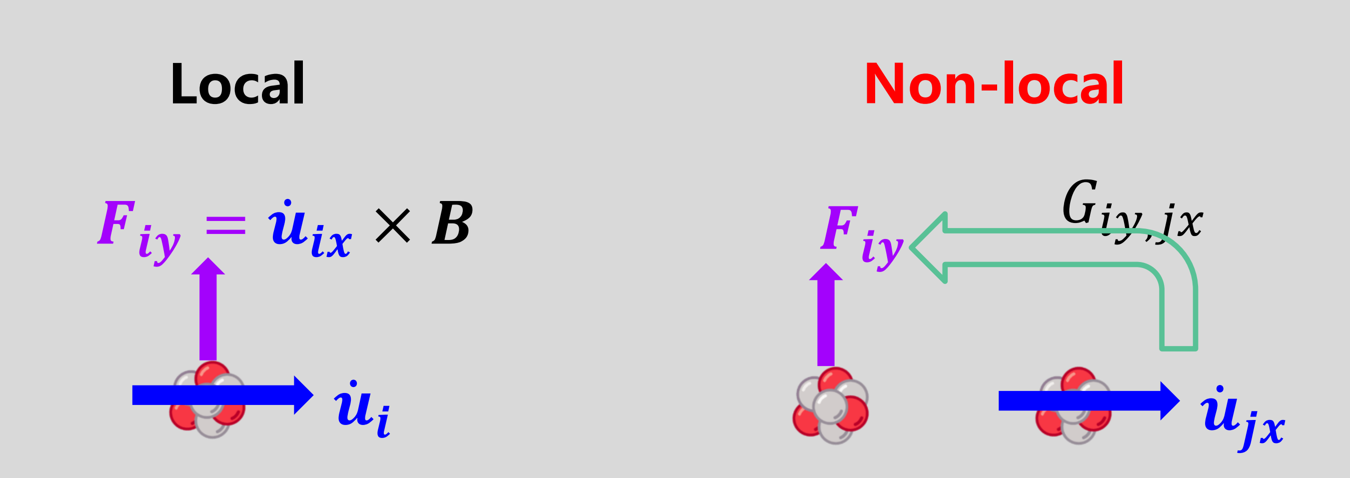
$$M_i \ddot{u}_i = -D_{ij} u_j + G_{ij} \dot{u}_j$$

Spring constant

$D^\dagger = D$

Molecular Berry curvature: $G_{ij} = \partial_{u_i} A_{u_j} - \partial_{u_j} A_{u_i}$

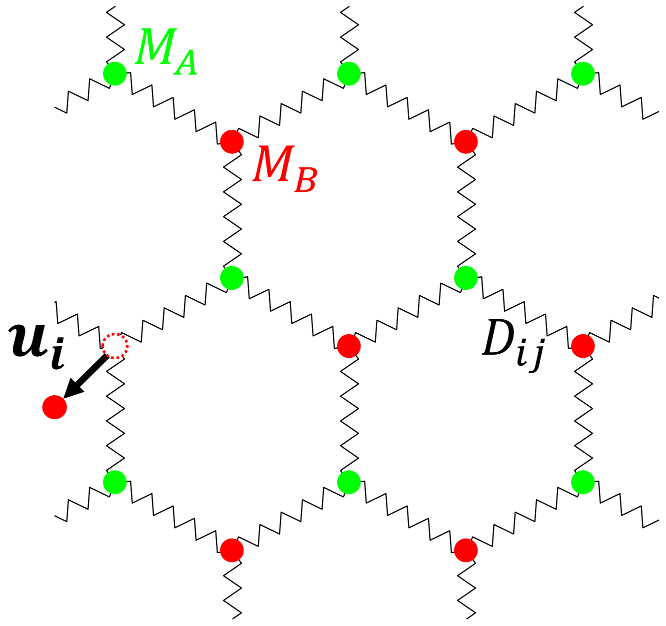
$G^\dagger = -G$ Effective magnetic field



Application to Haldane Model

Lattice part: time reversal invariant

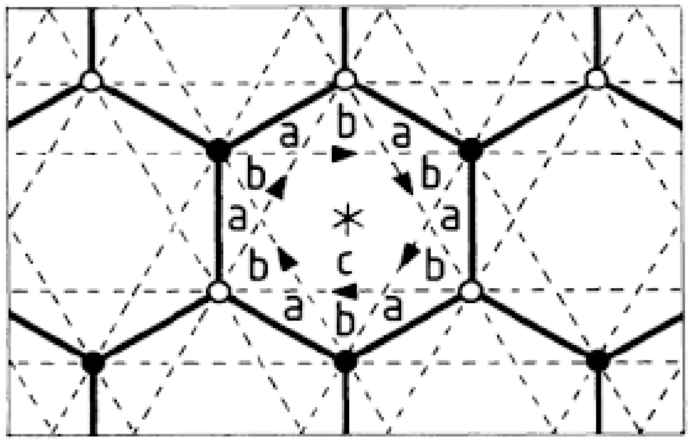
$$H_L = \sum_i \frac{\boldsymbol{P}_i^2}{2M_i} + \sum_{ij} \frac{1}{2} u_i D_{ij} u_j$$



Electronic part: time reversal symmetry breaking

- $H_e(\boldsymbol{u_i})$, e.g., hopping terms
- Electronic ground state $\Psi_e(\{\boldsymbol{u_i}\})$

$$G_{ij} = -2\mathrm{Im}\langle \partial_{u_i}\Psi_e | \partial_{u_j}\Psi_e \rangle$$



Chiral Optical Phonon

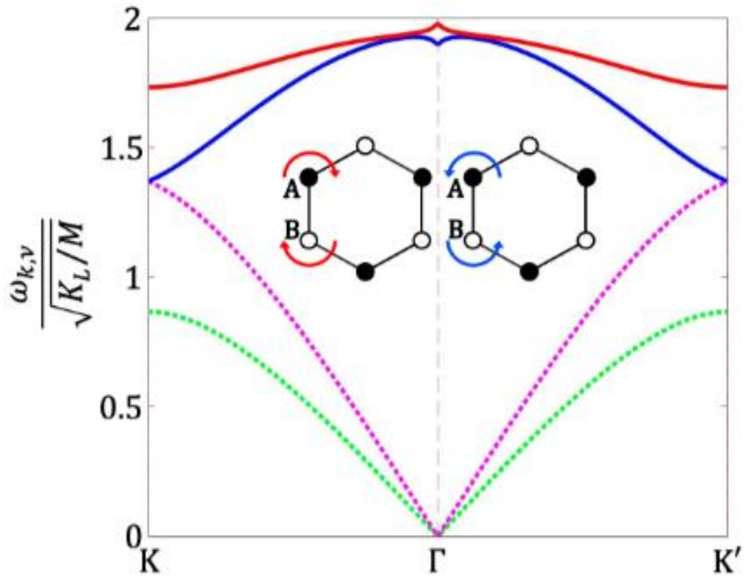
$$u_{\boldsymbol{k}} = \frac{1}{\sqrt{N}} \sum_i u_i e^{i\boldsymbol{k}\cdot \boldsymbol{R}_i^0}$$

$$\ddot{\boldsymbol{u}}_{\boldsymbol{0},A} = -D_{\boldsymbol{0}}^{A,\kappa} u_{\boldsymbol{0}\kappa} + G_0 \hat{z} \times \dot{\boldsymbol{u}}_{\boldsymbol{0},A} - G_0 \hat{z} \times \dot{\boldsymbol{u}}_{\boldsymbol{0},B}$$

- **Acoustic** phonon: $\dot{\boldsymbol{u}}_{\boldsymbol{0},A} = \dot{\boldsymbol{u}}_{\boldsymbol{0},B}$

  → **Zero** Lorentz force
- **Optical** phonon: $\dot{\boldsymbol{u}}_{\boldsymbol{0},A} = -\dot{\boldsymbol{u}}_{\boldsymbol{0},B}$

  → **Nonzero** Lorentz force



Saparov, Xiong, **YR*** & Niu, PRB **105**, 064303 (2022)

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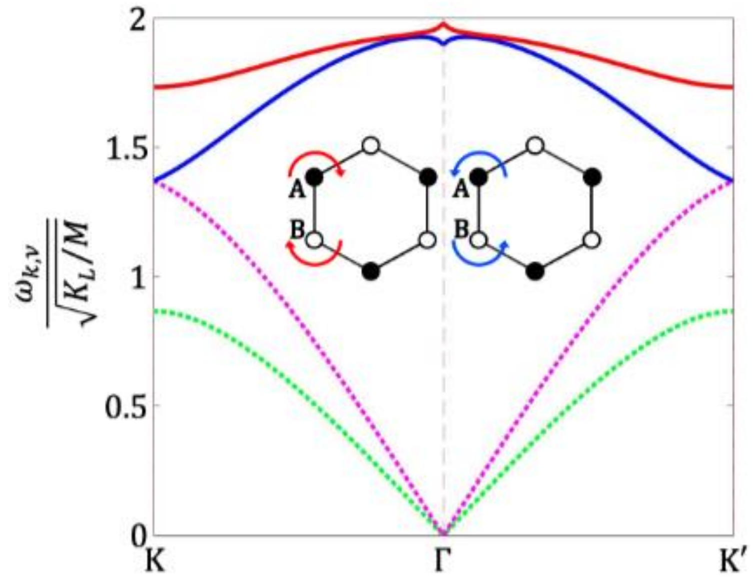
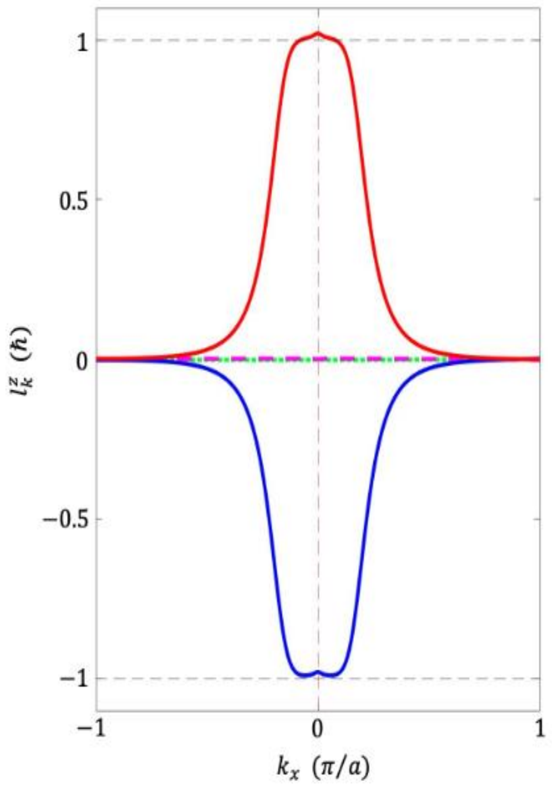
$$\ddot{\boldsymbol{u}}_{\boldsymbol{0},A} = -D_{\boldsymbol{0}}^{A,\kappa} u_{\boldsymbol{0}\kappa} + G_0\hat{z} \times \dot{\boldsymbol{u}}_{\boldsymbol{0},A} - G_0\hat{z} \times \dot{\boldsymbol{u}}_{\boldsymbol{0},B}$$

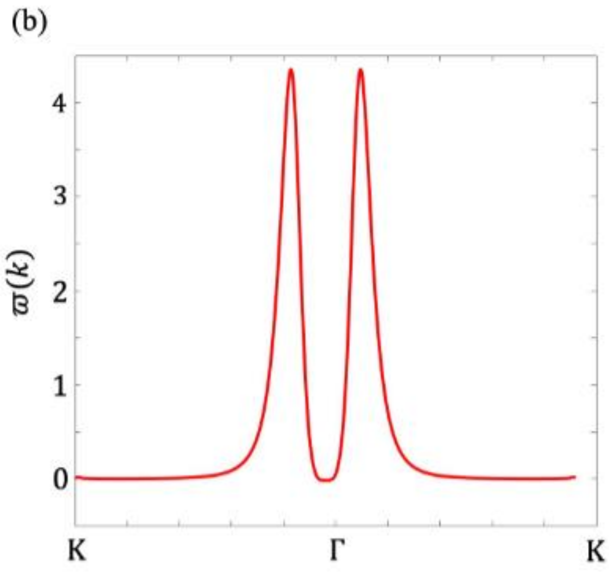


Saparov, Xiong, **YR*** & Niu, PRB **105**, 064303 (2022)

Phonon angular momentum → modify Einstein de Haas effect

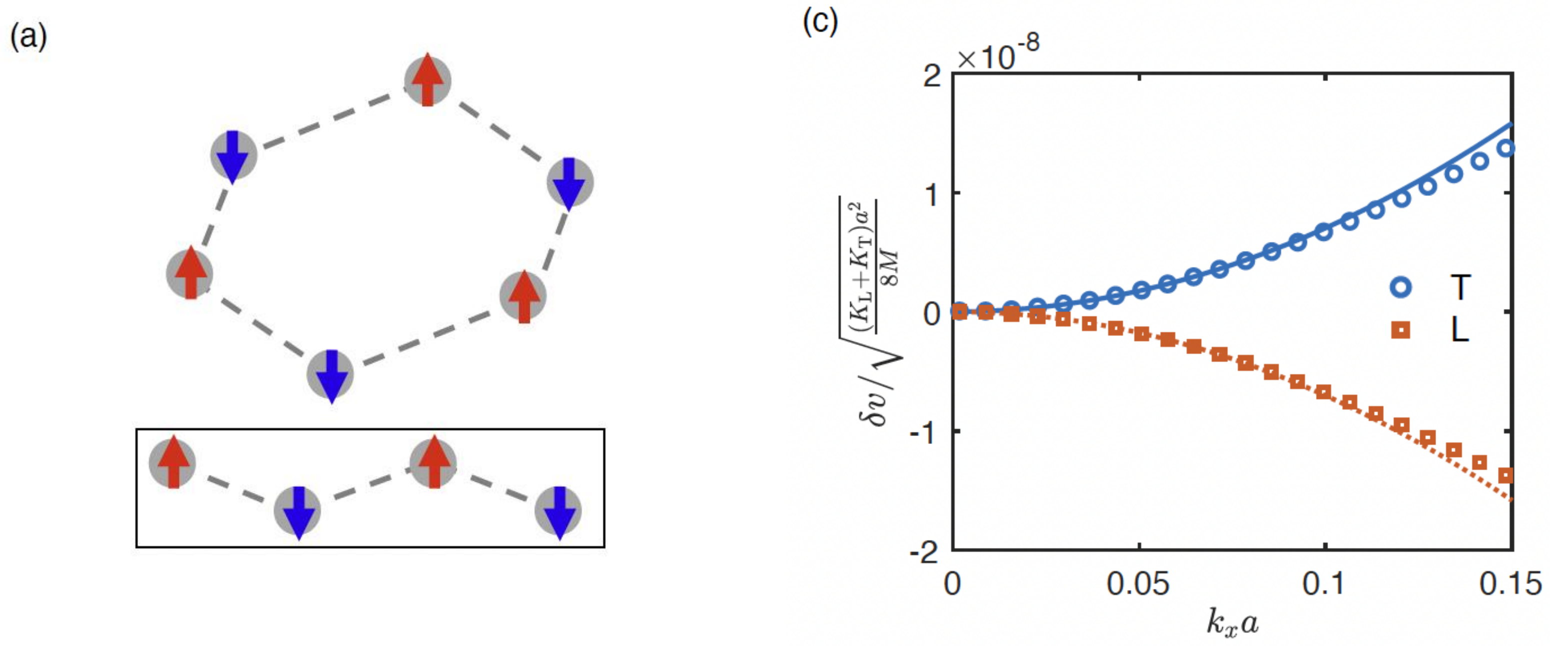


Phonon Berry curvature → phonon Hall effect



Nonreciprocal Acoustic Phonon

Break both time-reversal and inversion symmetries: $\omega(\boldsymbol{k}) \neq \omega(-\boldsymbol{k})$ & $\boldsymbol{v}(\boldsymbol{k}) \neq -\boldsymbol{v}(-\boldsymbol{k})$

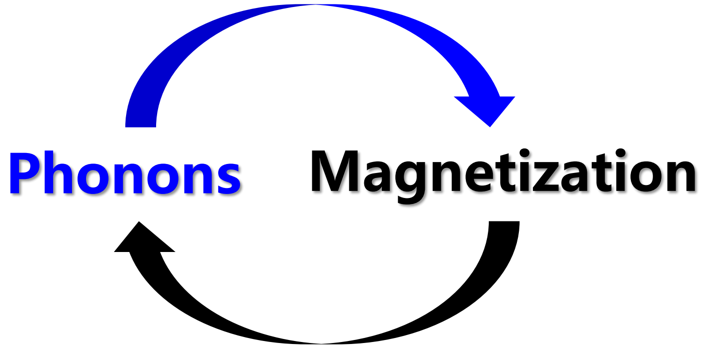


YR, Saparov & Niu, arXiv:2407.09361

Outline

Geometric Phase Effects	
$\Omega_{k_x u_y}$	Born effective charge $Z_{ij} \sim \int \Omega_{k_i u_j} d\boldsymbol{k}$
$\Omega_{k_x k_y}$	Chern number $C \sim \int \Omega_{k_x k_y} d\boldsymbol{k}$
$\Omega_{u_x u_y}$	Molecular Berry curvature $G \sim \int \Omega_{u_x u_y} d\boldsymbol{k}$

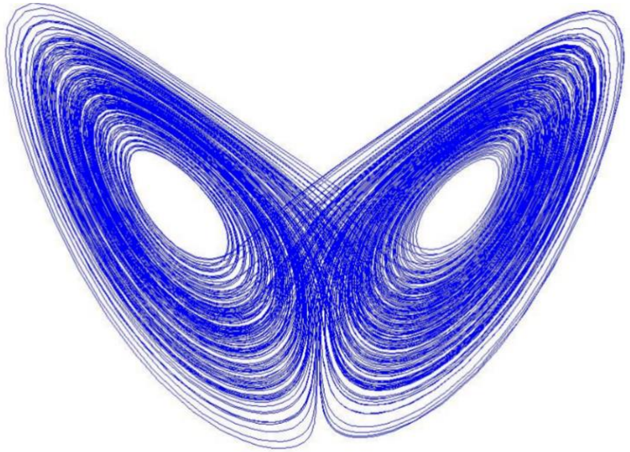
Phonons influence electrons
→ phonon magnetic moment



Electrons influence phonons
→ Chiral optical phonon

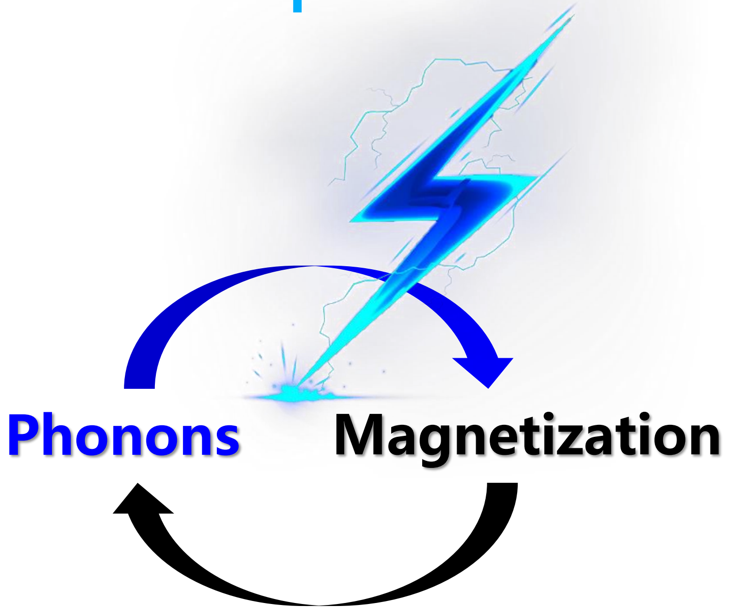
Outline

Nonlinearity:



Chaos, spontaneous symmetry breaking, bifurcation, etc.

Behavior under periodic drive



YR, Rudner & Xiao, PRL **132**, 096702 (2024)

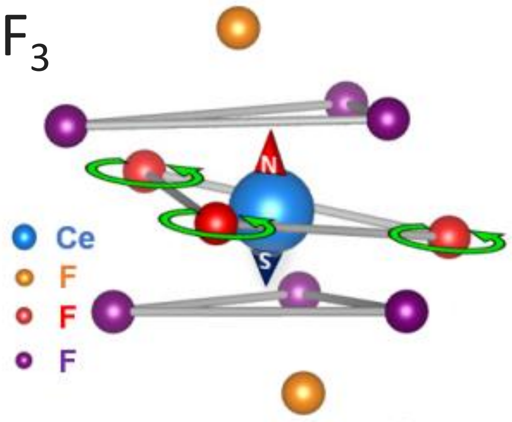
Nonlinearity: Spin-phonon System for Simplicity

Chiral phonon → effective Zeeman field for spin

$$H_{\mathrm{sp}} = g\boldsymbol{S} \cdot \boldsymbol{Q} \times \dot{\boldsymbol{Q}}$$

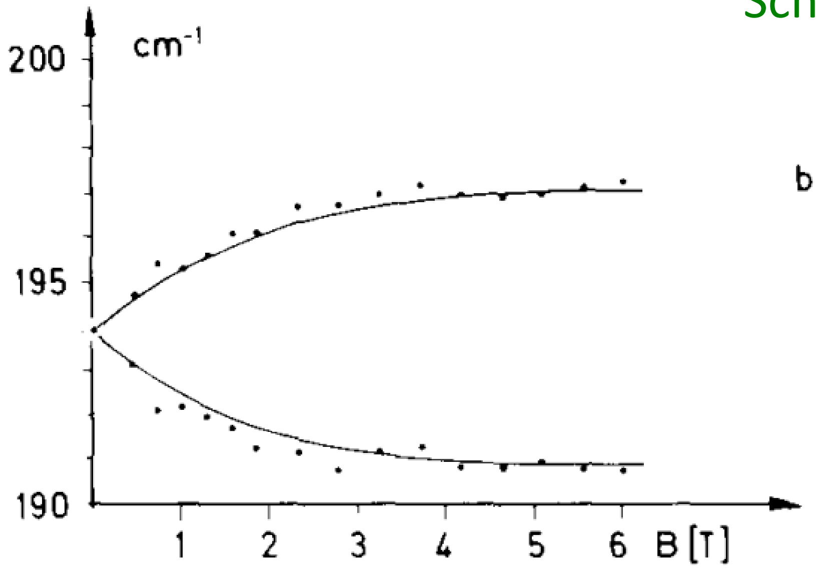
Spin → effective orbital magnetic field for phonon

CeF_3



Science **382**, 698 (2023)

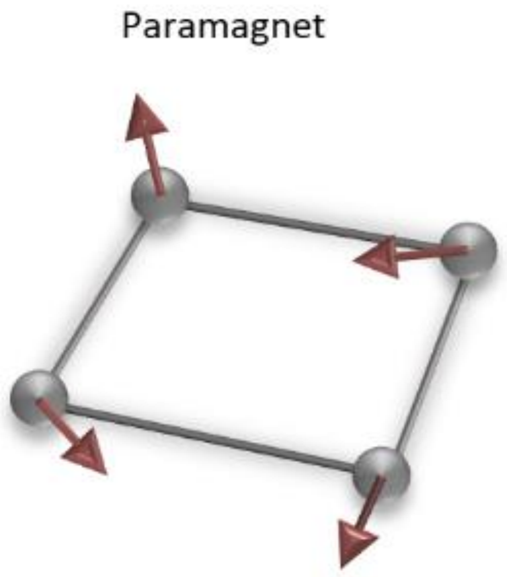
CeF_3, $CeCl_3$, $LaCl_3$ …

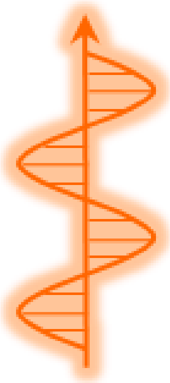


Schaack, 1970s

Light Driven Spin-Phonon Coupled System

Mirror symmetry: $\mathcal{M}_{zx}$ about zx plane





$$\mathcal{L}_{\rm ph} = \frac{1}{2}\dot{\boldsymbol{Q}}^2 - \frac{\omega^2}{2}\boldsymbol{Q}^2 - \frac{gS}{2}\hat{z}\cdot(\boldsymbol{Q}\times\dot{\boldsymbol{Q}}) + \boldsymbol{F}(t)\cdot\boldsymbol{Q}$$

Phonon dynamics

Spin-phonon coupling

Electric dipole $\boldsymbol{P} = \boldsymbol{Z}\boldsymbol{Q}$

$$\boldsymbol{F} = \boldsymbol{E}\boldsymbol{Z} = F(\cos\Omega t, 0, 0)$$

Spontaneous Symmetry Breaking

Equations of motion:

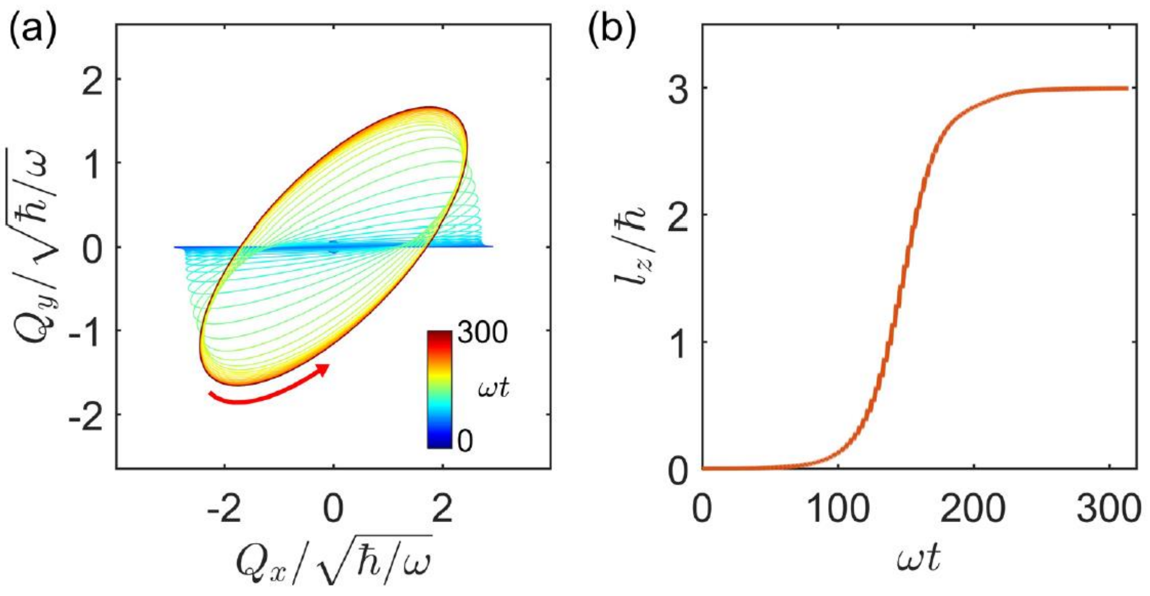
$$\ddot{\boldsymbol{Q}} = -\omega^2 \boldsymbol{Q} + \boldsymbol{F} - g\dot{\boldsymbol{Q}} \times S\hat{z} - \frac{g}{2}\boldsymbol{Q} \times \dot{S}\hat{z} - \dot{\boldsymbol{Q}}/\tau_p$$

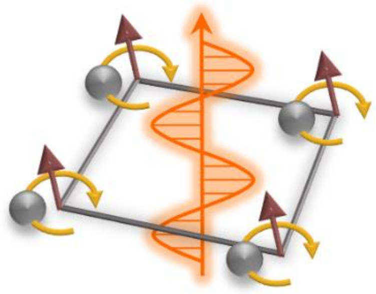
$$\dot{S}(t) = -[S(t) - S_{\rm eq}(l_z(t))]/\tau_s$$

$$S_{\rm eq}(l_z) \approx \chi_p l_z$$

$$\chi_p = g/(2k_B T)$$

Time evolution:



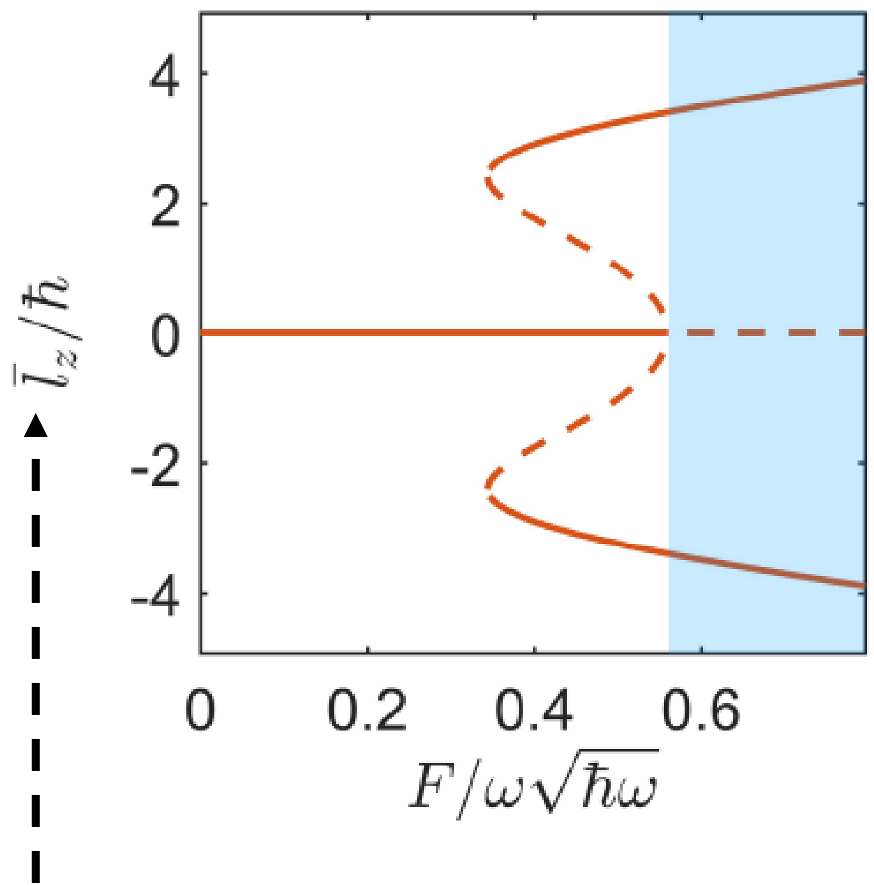


Nonequilibrium steady state

YR, Rudner & Xiao, PRL **132**, 096702 (2024)

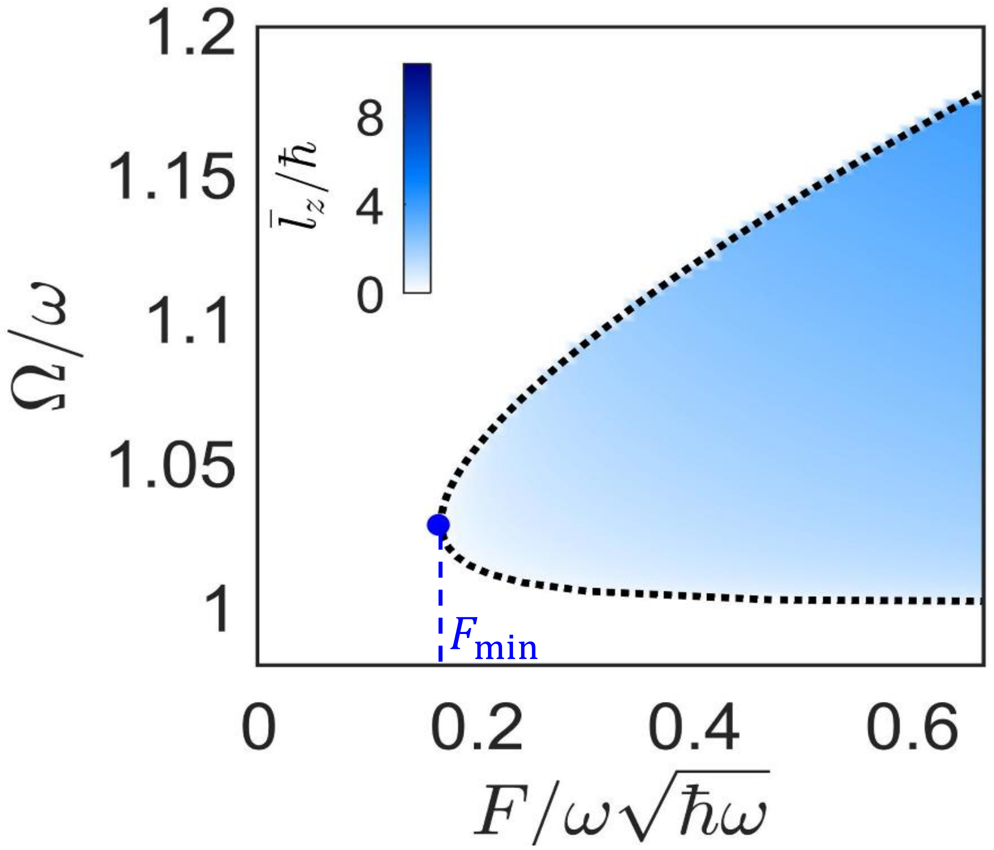
Bifurcation and Phase Diagram

Bifurcation: driving force dependence



Phonon angular momentum at the steady state

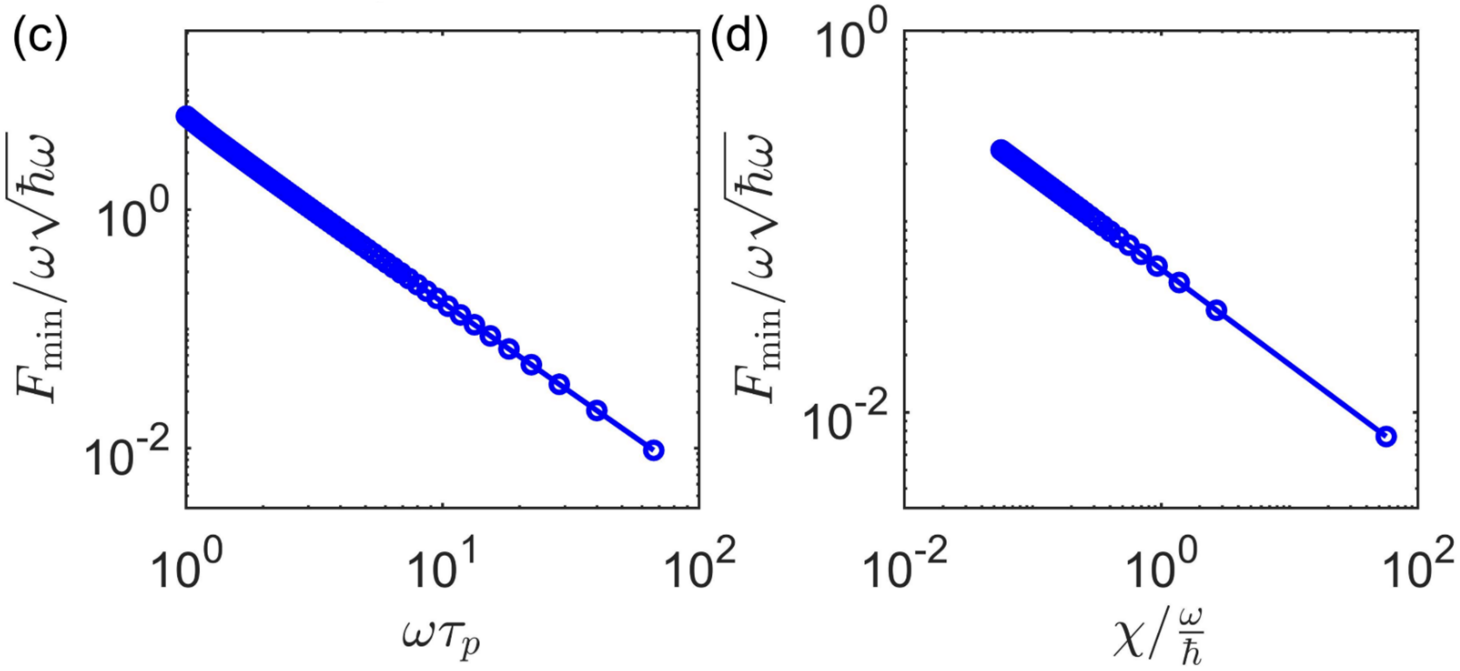
Phase diagram:



YR, Rudner & Xiao, PRL **132**, 096702 (2024)

Threshold Field Strength

$F_{\min} \propto \tau_p^{-1.5}$; $F_{\min} \propto \left(\frac{g^2}{2k_B T}\right)^{-0.5}$



$$\chi \doteq g\chi_p = \frac{g^2}{2k_B T}$$

YR, Rudner & Xiao, PRL **132**, 096702 (2024)

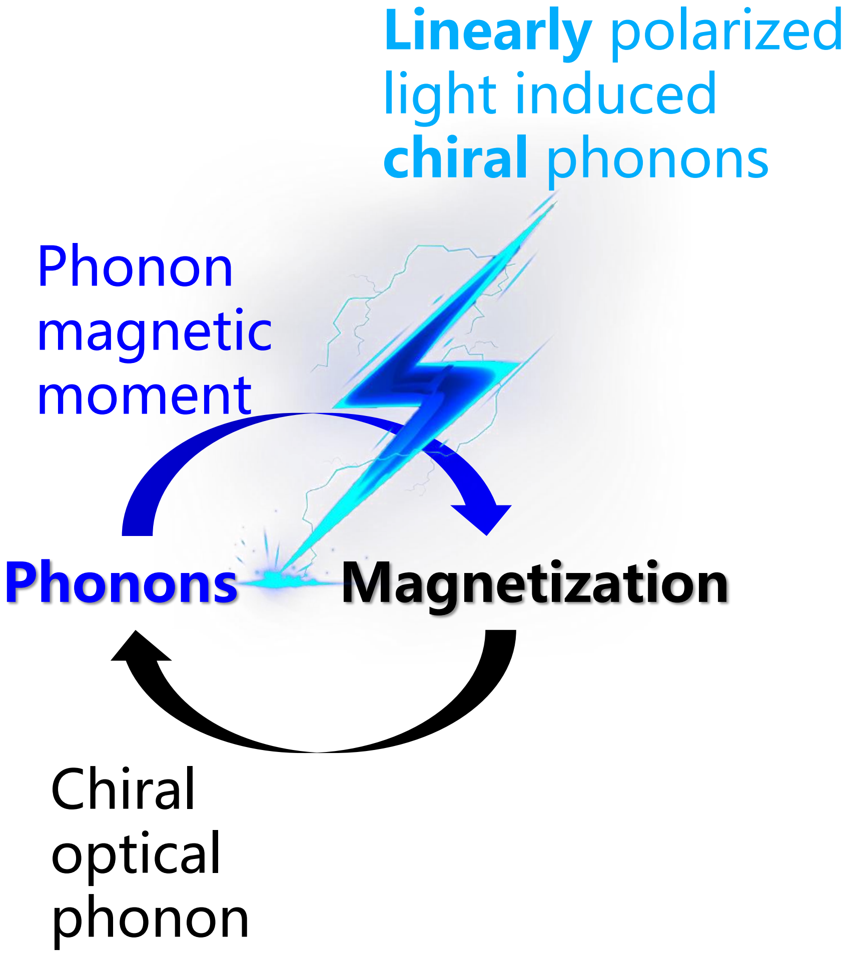
Optimized conditions:

- Longer phonon lifetime
- Stronger coupling
- Large susceptibility
  - For FM/AFM, $T \rightarrow (T - T_c)$

Material candidates:

- *Paramagnetic f-electron spins*: CeF_3, $CeCl_3$
  - $T \sim 1\mathrm{K}, E \sim 1 - 10$ MV/cm
- *d-electron spins:* $CoTiO_3$

Summary



Interplay between phonon and magnetization:

- Nonmagnetic: chiral phonons generate magnetization
- Ferromagnetic: chiral optical phonons
- Antiferromagnet: nonreciprocal acoustic phonons
- Paramagnet: nonlinear feedback loop

YR, Xiao, Saparov & Q.N., PRL **127**, 186403 (2021); Saparov, Xiong, **YR*** & Niu, PRB **105**, 064303 (2022); Zhang, **YR**, Wang, Cao & Xiao, PRL **130**, 226302 (2023); **YR**, Rudner & Xiao, PRL **132**, 096702 (2024); **YR,** Saparov & Niu, arXiv:2407.09361

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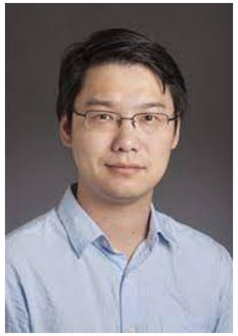

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