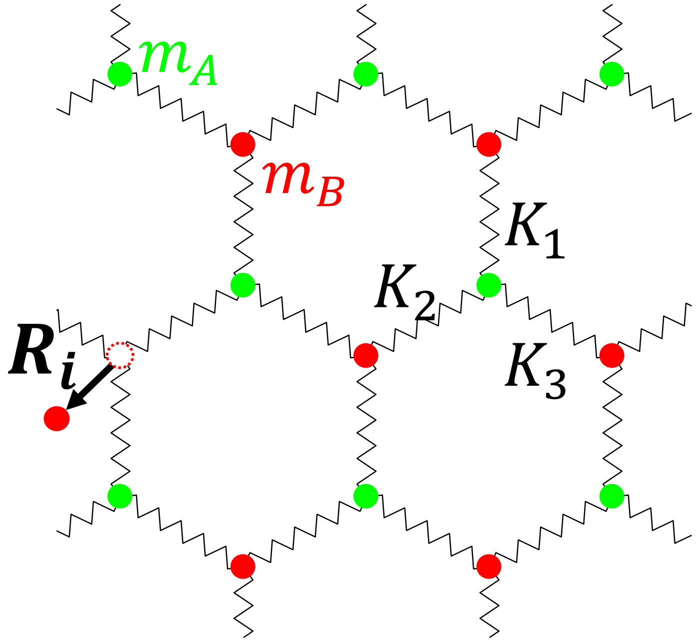
LECTURE 8
Molecular Berry curvature

- Lattice dynamics: dispersion & polarization
- Phonon chirality: angular momentum
- Molecular Berry curvature: zero B field
- Molecular Berry curvature: with B field
- Summary

Lattice Dynamics



Potential energy $V(\{\boldsymbol{R}\})$

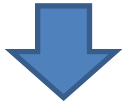
Coupled equations of motion

$$M_\kappa \ddot{\boldsymbol{R}}_{l,\kappa} = -\nabla_{\boldsymbol{R}_{l,\kappa}} V(\{\boldsymbol{R}\})$$

Harmonic approximation:

$$\boldsymbol{R}_{l,\kappa} = \boldsymbol{R}^0_{l,\kappa} + \boldsymbol{u}_{l,\kappa}$$

$$V(\{\boldsymbol{R}\}) \simeq V(\{\boldsymbol{R}^0\}) + \frac{1}{2}\sum D^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l, \boldsymbol{R}^0_{l'})\, u_{l,\kappa,\alpha} u_{l',\kappa',\alpha'}$$



$$\partial_{l,\kappa,\alpha}\partial_{l',\kappa',\alpha'} V$$

Translational symmetry and Fourier transform

Translational symmetry:

$$D^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l,\boldsymbol{R}^0_{l'}) = D^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l - \boldsymbol{R}^0_{l'})$$

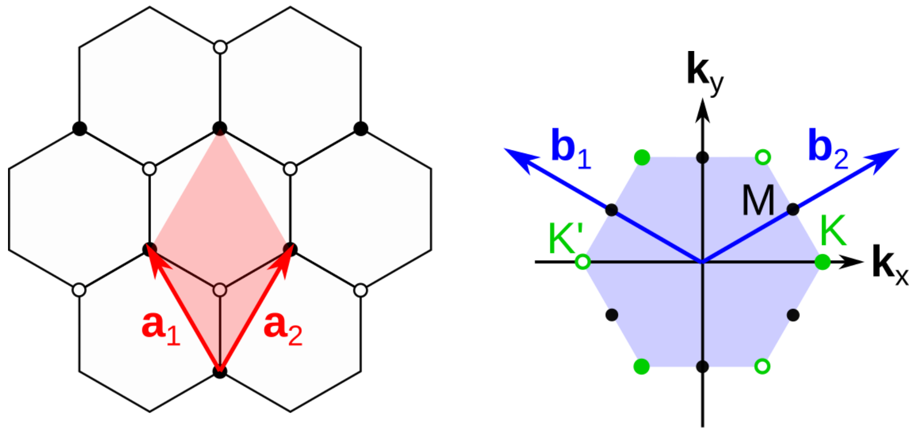
$$D^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{k}) = \sum_l D^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l - \boldsymbol{R}^0_0)e^{i\boldsymbol{k}\cdot(\boldsymbol{R}^0_l - \boldsymbol{R}^0_0)}$$

Lattice wave:

$$\boldsymbol{u}_{\boldsymbol{k},\kappa} = \frac{1}{\sqrt{\mathrm{N}}}\sum_i e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{i,0}}\,\boldsymbol{u}_{i,\kappa}$$

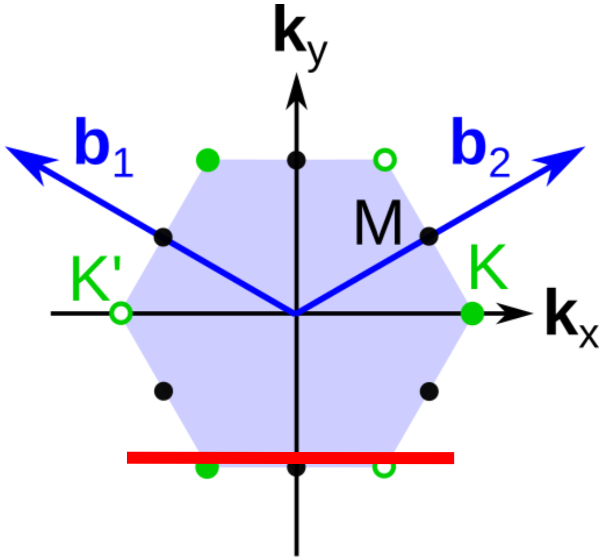
Equation of motion:

$$M_\kappa \ddot{\mathbf{u}}_{\boldsymbol{k},\kappa} = -\boldsymbol{D}^{\kappa}_{\kappa'}(\boldsymbol{k})\boldsymbol{u}_{\boldsymbol{k},\kappa'}$$



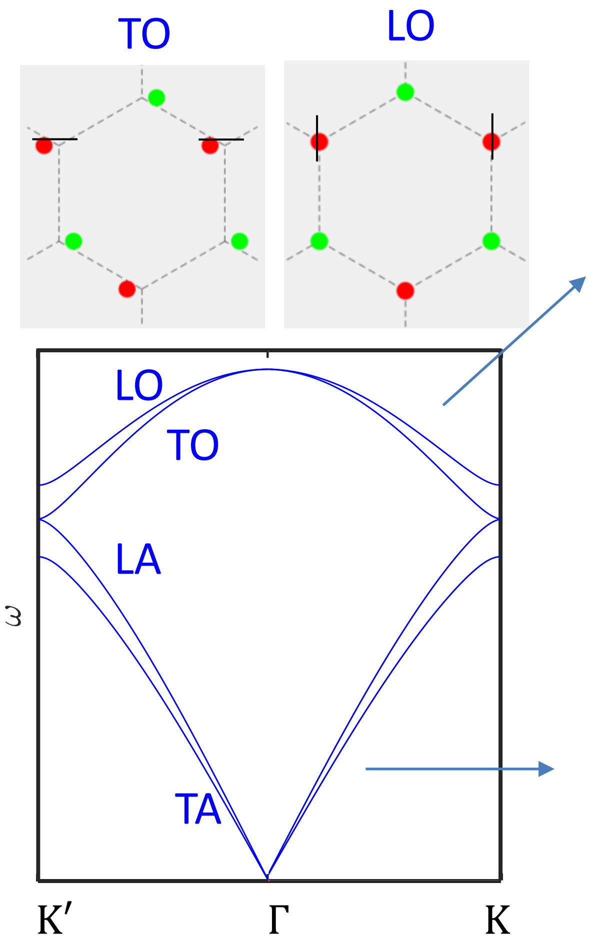
Lattice and momentum space

Dispersion and Polarization



$$\omega_\alpha(\boldsymbol{k}) = \omega_\alpha(-\boldsymbol{k})$$

Reciprocity: time reversal symmetry



Optical branch:

electric dipoles

Acoustic branch:
ions move together

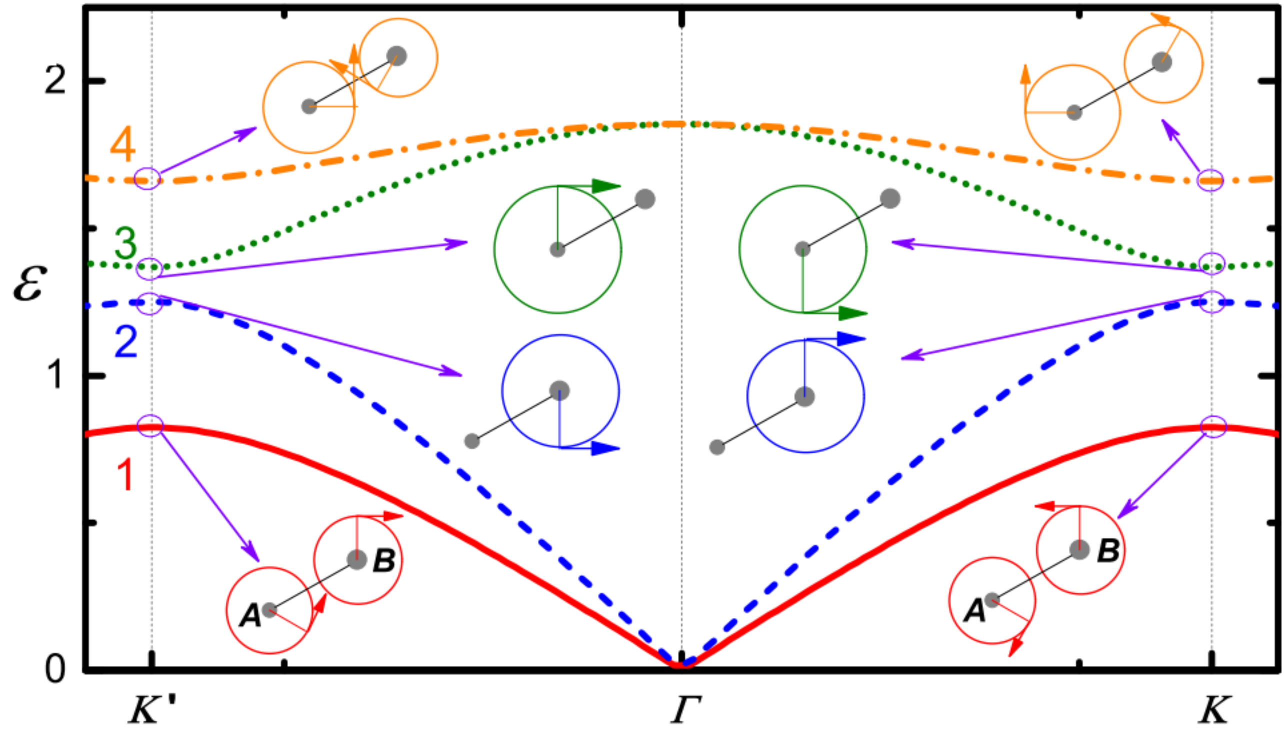
Quantization and phonons

- Quantization:
  - n phonon: $E_n = (n + \frac{1}{2})\hbar\omega$
  - $\boldsymbol{p} = \hbar\boldsymbol{k}$
- Bose-Einstein distribution:
  - Averaged phonon number

    $n(\omega) = \frac{1}{e^{\hbar\omega/k_B T} - 1}$

- Internal energy:
  - $U = \sum_{\boldsymbol{k},\alpha}[n(\omega_\alpha) + \frac{1}{2}]\,\hbar\omega_\alpha$
- Heat capacity: $C_V = \left(\frac{\partial U}{\partial T}\right)_V$
  - Quantum: $C_V \propto T^3$ at low T
  - Classical: C_V constant

Inversion symmetry breaking: phonon chirality



Zhang and Niu, PRL 2015
MoS2: inversion symmetry is broken

Phonon angular momentum:

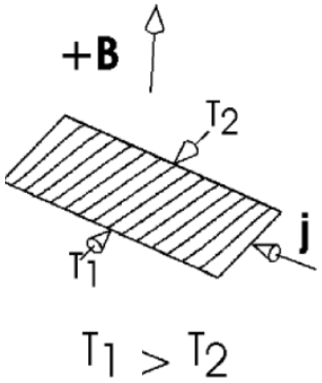
$$\boldsymbol{L}(\boldsymbol{k}) = \sum_{\kappa} M_{\kappa} \boldsymbol{u}_{\kappa} \times \dot{\boldsymbol{u}}_{\kappa}$$

odd under time reversal or spatial inversion

- Breaking inversion symmetry
  - $\boldsymbol{L}(\boldsymbol{k}) \neq \boldsymbol{0}$ at nonzero $\boldsymbol{k}$
  - Chiral phonon at K/K'
- Time-reversal symmetry still exists
  - Γ point: no chirality
  - $\omega(\boldsymbol{k}) = \omega(-\boldsymbol{k})$

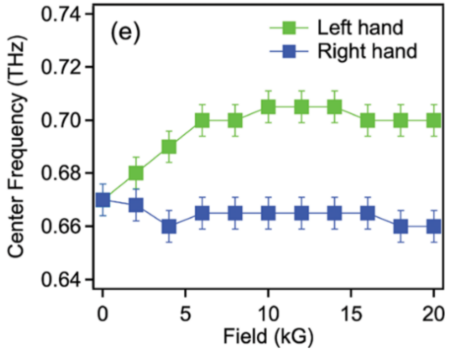
Effects of Breaking Time Reversal Symmetry

Thermal Hall effect



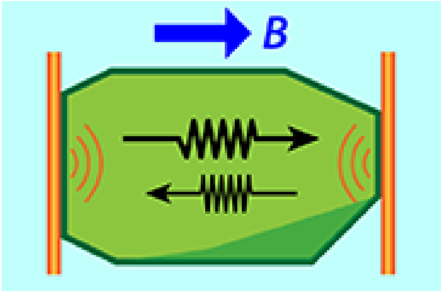
PRL **95**, 155901 (2005)

Chiral phonon at Γ:
Phonon Zeeman splitting



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

Phonon nonreciprocity:
$\omega(\boldsymbol{k}) \neq \omega(-\boldsymbol{k})$

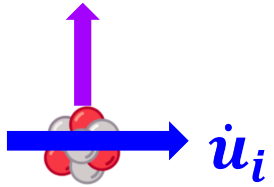


PRL **122**, 145901 (2019)

Old Ways to Break Time Reversal Symmetry

Lorentz force:

$$\boldsymbol{F}_i = Z\dot{\boldsymbol{u}}_i \times \boldsymbol{B}$$

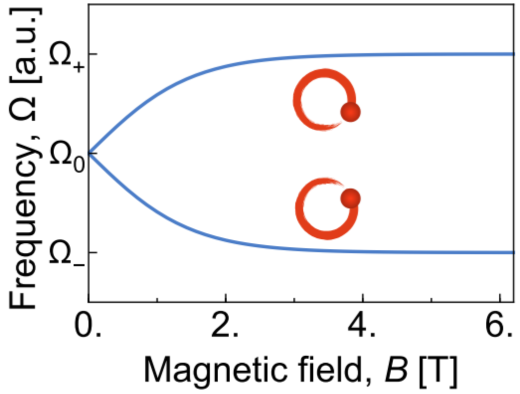


Z: ionic charge

Spin-Lattice coupling:

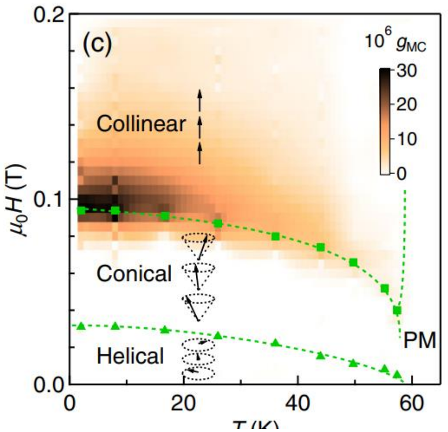
$$H^{SL} = \lambda\langle\boldsymbol{S}\rangle \cdot \boldsymbol{u} \times \dot{\boldsymbol{u}}$$

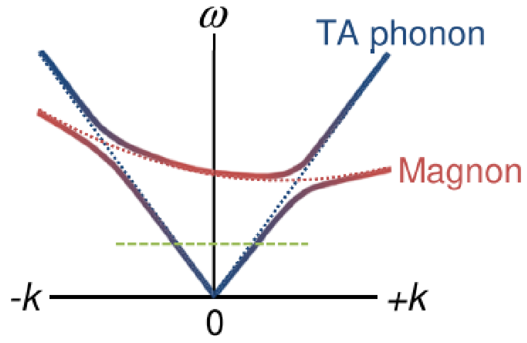
Effective local magnetic field, saturate as B field increases



PRR **4**, 013129 (2022)

Phonon-magnon coupling:





PRL **122**, 145901 (2019)

New Way: Molecular Berry Curvature

Time-dependent variational principle:

$$\mathcal{L} = \sum_{l,\kappa} \frac{M_\kappa}{2} \dot{\boldsymbol{R}}_{l,\kappa}^2 + \langle \Phi_0 | i\hbar d_t - H_e | \Phi_0 \rangle$$

$$= \sum_{l,\kappa} \frac{M_\kappa}{2} \dot{\boldsymbol{R}}_{l,\kappa}^2 + \langle \Phi_0 | i\hbar d_t | \Phi_0 \rangle - V_{\text{eff}}(\{\boldsymbol{R}\})$$

$$= \sum_{l,\kappa} \frac{M_\kappa}{2} \dot{\boldsymbol{R}}_{l,\kappa}^2 + \boxed{\hbar \boldsymbol{A}_{l,\kappa} \cdot \dot{\boldsymbol{R}}_{l,\kappa}} - V_{\text{eff}}(\{\boldsymbol{R}\})$$

Vector potential

Scalar potential

$$\boldsymbol{A}_{l,\kappa} = \langle \Phi_0 | i \nabla_{\boldsymbol{R}_{l,\kappa}} | \Phi_0 \rangle$$

Born & Huang (1954)
Mead & Truhlar, JCP 1979, Mead, RMP 1992
Ceresoli & Tosatti, PRL 2002
Qin, Zhou & Shi, PRB 86, 104305 (2012)

Saito, Misaki, Ishizuka & Nagaosa, PRL 2019
Bistoni Mauri & Calandra, PRL 2021
Saparov, Xiong, Ren & Q.N., PRB 105, 064303 (2022)
Bonini, et al., PRL 130, 086701 (2023)

Molecular Berry curvature $G_{\kappa',\alpha'}^{\kappa,\alpha}(\boldsymbol{R}_l^0, \boldsymbol{R}_{l'}^0) = \partial_{l,\kappa,\alpha} A_{l',\kappa',\alpha'} - \partial_{l',\kappa',\alpha'} A_{l,\kappa,\alpha}$

- act as an effective magnetic field

Nonlocal Lorentz force

$$d_t \frac{\partial \mathcal{L}}{\partial \dot{\boldsymbol{R}}_{l,\kappa}} - \frac{\partial \mathcal{L}}{\partial \boldsymbol{R}_{l,\kappa}} = 0 \quad \Rightarrow \quad M_\kappa \ddot{\boldsymbol{R}}_{l,\kappa} = -\partial_{\boldsymbol{R}_{l,\kappa}} V + \sum_{l',\kappa'} \boldsymbol{G}^{\kappa}_{\kappa'}(\boldsymbol{R}^0_l, \boldsymbol{R}^0_{l'}) \dot{\boldsymbol{R}}_{l',\kappa'}$$

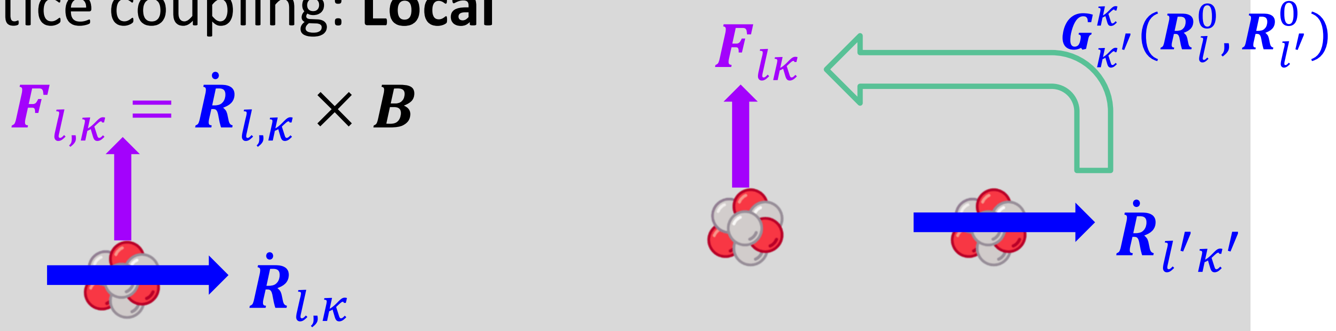
$G^\dagger = -G$: real **antisymmetric**

- **Non-local** effective B field

$\boldsymbol{B}$ field/Raman spin-lattice coupling: **Local**

$\boldsymbol{F}_{l,\kappa} = \dot{\boldsymbol{R}}_{l,\kappa} \times \boldsymbol{B}$

Non-local



Molecular Berry curvature at zero u

- Linearization: $\boldsymbol{R}_{l,\kappa} = \boldsymbol{R}^0_{l,\kappa} + \boldsymbol{u}_{l,\kappa}$
- Translation symmetry: $G^{\kappa,\alpha}_{\kappa',\alpha'}\left(\boldsymbol{R}^0_l, \boldsymbol{R}^0_{l'}\right) = G^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l - \boldsymbol{R}^0_{l'})$

$$G^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{k}) = \sum_l G^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{R}^0_l - \boldsymbol{R}^0_0)e^{i\boldsymbol{k}\cdot(\boldsymbol{R}^0_l - \boldsymbol{R}^0_0)}$$

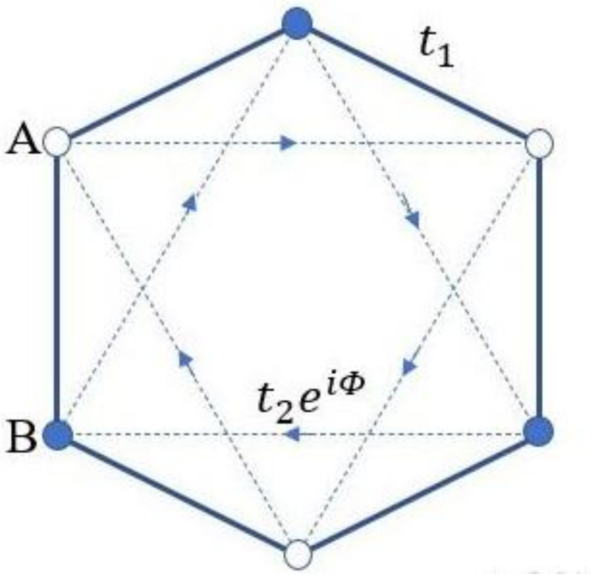
- Equation of motion for the Fourier amplitudes:

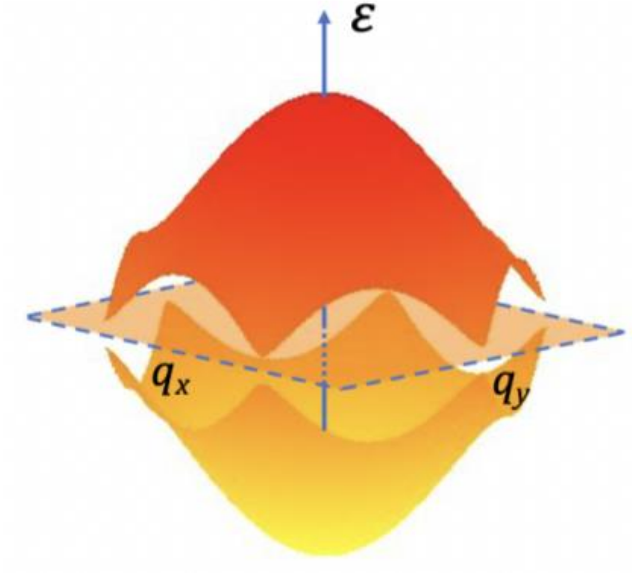
$$M_\kappa \ddot{\boldsymbol{u}}_{\boldsymbol{k},\kappa} = -\boldsymbol{D}^{\kappa}_{\kappa'}(\boldsymbol{k})\boldsymbol{u}_{\boldsymbol{k},\kappa'} + \boldsymbol{G}^{\kappa}_{\kappa'}(\boldsymbol{k})\dot{\boldsymbol{u}}_{\boldsymbol{k},\kappa'}$$

- Symmetry properties:

$D^\dagger = D$: Hermitian $\qquad$ $G^\dagger = -G$: anti-Hermitian

Application to Haldane Model





$$H_e = -\sum_{\langle i,j\rangle} t_{ij} a_i^\dagger a_j - \sum_{\langle\langle ij\rangle\rangle} t'_{ij}\, a_i^\dagger a_j + \mathrm{h.c.}$$

Depends on $\{\boldsymbol{u_i}\}$

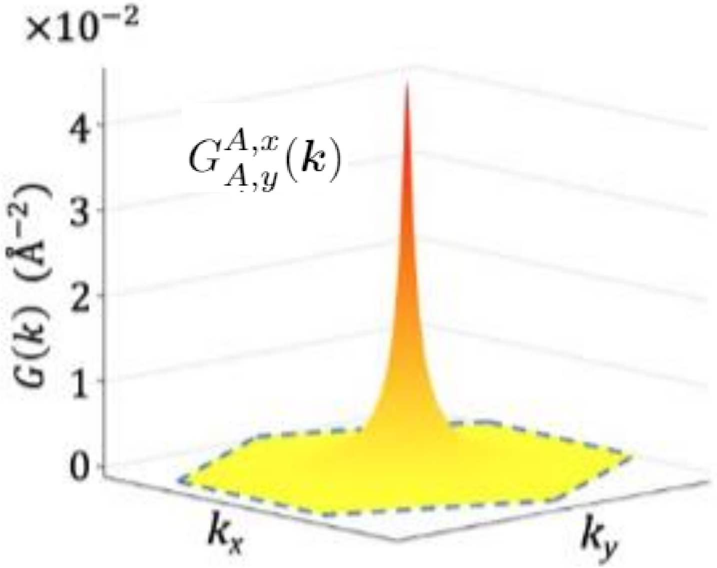
Assume lower band is filled:
a topological insulator

Electronic ground state $\Psi_e(\{\boldsymbol{u_i}\})$ can be calculated perturbatively for small u.

Calculation of molecular Berry curvature

$$G^{\kappa,\alpha}_{\kappa',\alpha'}(\boldsymbol{k}) = \frac{i}{N}\sum_{\boldsymbol{q}}\sum_{\substack{\varepsilon_m<\mu \\ \varepsilon_{m'}>\mu}} \frac{\phi^{\dagger}_{m,\boldsymbol{q}}\mathcal{M}_{\boldsymbol{k},\kappa\alpha}\phi_{m',\boldsymbol{q}+\boldsymbol{k}}\phi^{\dagger}_{m',\boldsymbol{q}+\boldsymbol{k}}\mathcal{M}_{-\boldsymbol{k},\kappa'\alpha'}\phi_{m,\boldsymbol{q}}}{\left(\varepsilon_{m,\boldsymbol{q}} - \varepsilon_{m',\boldsymbol{q}+\boldsymbol{k}}\right)^2} - (\boldsymbol{k} \Leftrightarrow -\boldsymbol{k}, \kappa\alpha \Leftrightarrow \kappa'\alpha')$$

$$G_{\boldsymbol{k}} = \begin{pmatrix} G^{A,x}_{A,x}(\boldsymbol{k}) & G^{A,x}_{A,y}(\boldsymbol{k}) & G^{A,x}_{B,x}(\boldsymbol{k}) & G^{A,x}_{B,y}(\boldsymbol{k}) \\ G^{A,y}_{A,x}(\boldsymbol{k}) & G^{A,y}_{A,y}(\boldsymbol{k}) & G^{A,y}_{B,x}(\boldsymbol{k}) & G^{A,y}_{B,y}(\boldsymbol{k}) \\ G^{B,x}_{A,x}(\boldsymbol{k}) & G^{B,x}_{A,y}(\boldsymbol{k}) & G^{B,x}_{B,x}(\boldsymbol{k}) & G^{B,x}_{B,y}(\boldsymbol{k}) \\ G^{B,y}_{A,x}(\boldsymbol{k}) & G^{B,y}_{A,y}(\boldsymbol{k}) & G^{B,y}_{B,x}(\boldsymbol{k}) & G^{B,y}_{B,y}(\boldsymbol{k}) \end{pmatrix}$$



Phonon spectrum

Non-hermitian eigenvalue problem:

$$\omega_{\boldsymbol{k}} \begin{pmatrix} u_{\boldsymbol{k}} \\ p_{\boldsymbol{k}} \end{pmatrix} = i \begin{pmatrix} \tilde{G}_{\boldsymbol{k}} & \mathbb{1} \\ -D_{\boldsymbol{k}} & \tilde{G}_{\boldsymbol{k}} \end{pmatrix} \begin{pmatrix} u_{\boldsymbol{k}} \\ p_{\boldsymbol{k}} \end{pmatrix}$$

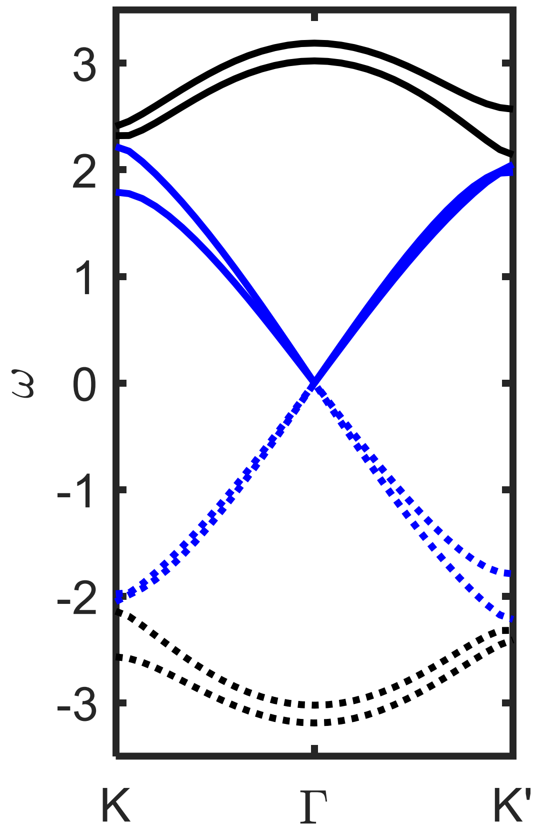
Can be made Hermitian if D is positive definite

Particle-hole symmetry:

$$\omega_{\boldsymbol{k},\nu} = -\omega_{-\boldsymbol{k},-\nu}$$

Quantization:

phonon energy corresponds to positive frequencies



Non-reciprocity if inversion symmetry is also broken

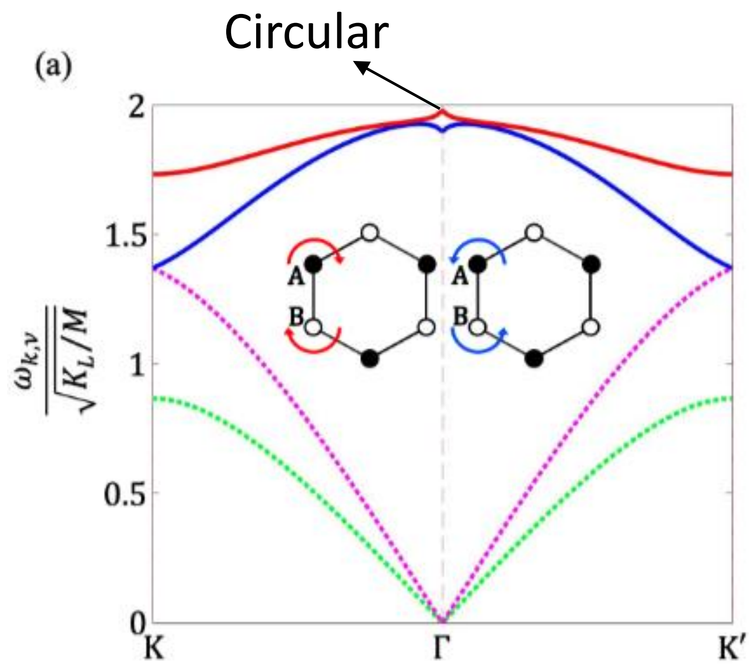
Chiral Optical Phonon

Berry curvature at Γ point:

$$G_\Gamma = \begin{pmatrix} 0 & B & 0 & -B \\ -B & 0 & B & 0 \\ 0 & -B & 0 & B \\ B & 0 & -B & 0 \end{pmatrix}$$

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

Berry curvature affects relative motion only



Inversion symmetry:
$\omega(\boldsymbol{k}) = \omega(-\boldsymbol{k})$

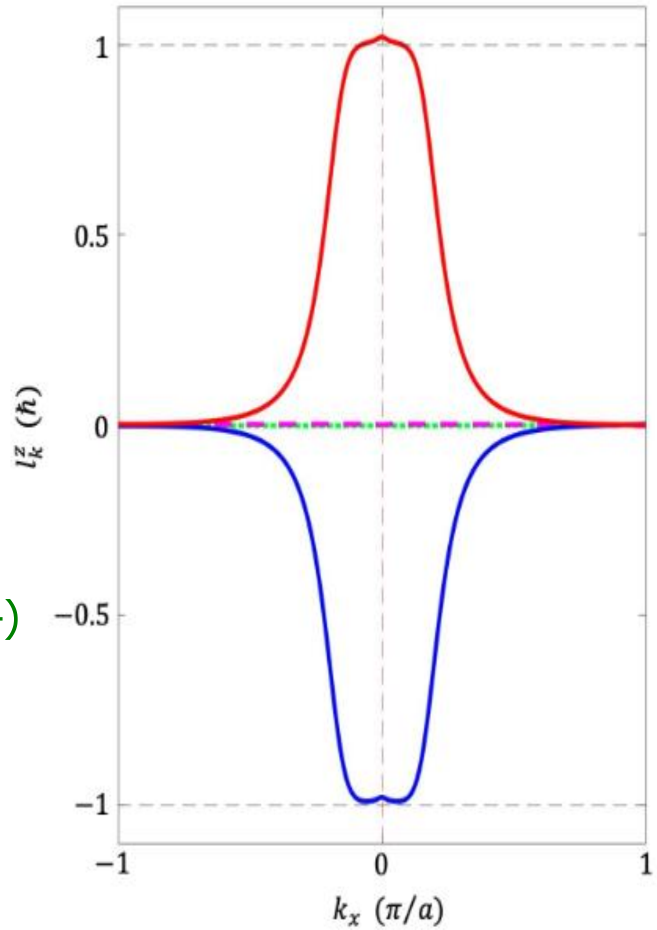
Phonon Angular Momentum

$$\boldsymbol{J}^{\mathrm{ph}} = \sum_{l\alpha} \boldsymbol{u}_{l\alpha} \times \dot{\boldsymbol{u}}_{l\alpha}$$

Nonzero angular momentum

Einstein-de Haas effect

Zhang & Niu PRL (2014)



Saparov, Xiong, Ren & Niu, PRB (2022)

Daniyar Saparov

Yafei Ren

Bangguo Xiong

Under Magnetic Field

Effective Lagrangian:

$$\mathcal{L} = \frac{1}{2} M_\kappa \left(\dot{u}^l_{\kappa\alpha}\right)^2 + Z_\kappa \dot{u}^l_{\kappa\alpha} A^l_{\kappa\alpha} + \left\langle \Phi_e \middle| i d_t - H_{\text{el}}(\boldsymbol{A}, \{\boldsymbol{u}^l_\kappa\}) \middle| \Phi_e \right\rangle$$

Zabalo, Dreyer & Stengel, PRB 2022

$$\left\langle \Phi_e \middle| i d_t - H_{el}(\{\boldsymbol{u}^l_\kappa\}) \middle| \Phi_e \right\rangle$$

$$\langle \Phi_e | \boldsymbol{A} \cdot \boldsymbol{j}_{\text{el}} | \Phi_e \rangle$$

$$\boldsymbol{A} = \frac{1}{2} \boldsymbol{r} \times \boldsymbol{B}$$

$$\boldsymbol{B} \cdot \langle \Phi_e | \boldsymbol{r} \times \boldsymbol{j}_{\text{el}} | \Phi_e \rangle$$

$-\frac{\partial \mathcal{L}}{\partial \boldsymbol{B}} = \langle \boldsymbol{m}_0 \rangle$: Magnetic moment (zero order in ionic velocities)

Phonon magnetic moment

Expanding to first order in ionic velocity:

$$-\frac{\partial \mathcal{L}}{\partial \boldsymbol{B}} = \langle \boldsymbol{m}_0 \rangle + \left\langle \frac{\partial \boldsymbol{m}}{\partial \dot{u}^l_{\kappa\alpha}} \dot{u}^l_{\kappa\alpha} \right\rangle + \frac{1}{2} \left\langle \frac{\partial^2 \boldsymbol{m}}{\partial u^l_{\kappa\alpha} \partial \dot{u}^{l'}_{\kappa'\alpha'}} u^l_{\kappa\alpha} \dot{u}^{l'}_{\kappa'\alpha'} \right\rangle$$

Lattice motion induced magnetic moment

Orbital magnetization: topological

Ren, Xiao, Saparov & Q.N., PRL (2021)

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

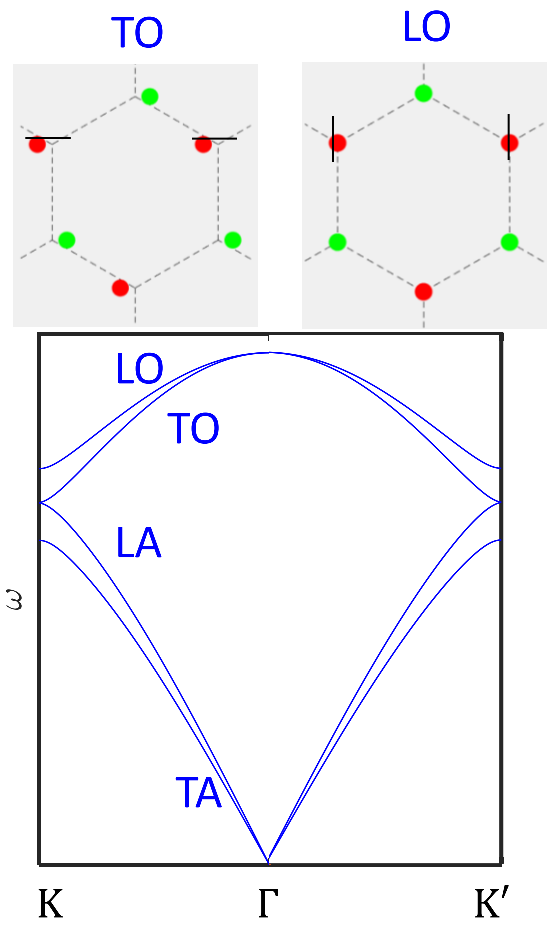

Yafei Ren


Daniyar Saparov

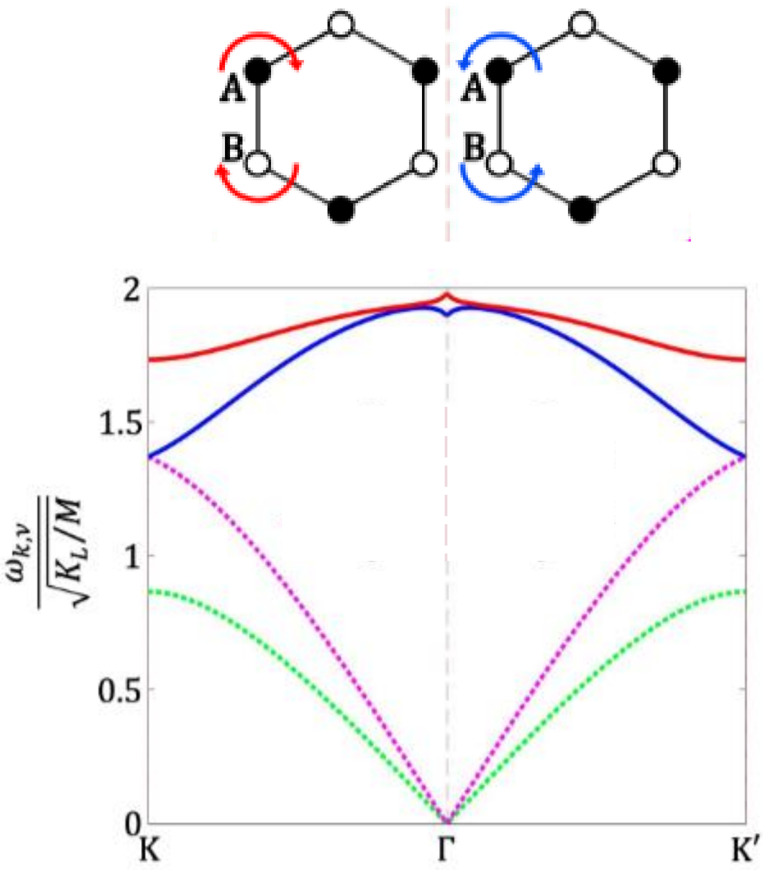

Cong Xiao

Summary

Only scalar potential for ions



Both **scalar** and **vector** potentials for ions:
time reversal symmetry breaking



$\boldsymbol{L}(\boldsymbol{k}) \neq \boldsymbol{0}$

$\omega(\boldsymbol{k}) \neq \omega(-\boldsymbol{k})$

Phonon Hall effect

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