CHIRAL PHONONS FROM BROKEN TIME REVERSAL SYMMETRY IN CrI_3

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Motivation

Phonons are typically computed from the force constant matrix ($\mathbf{C}$)

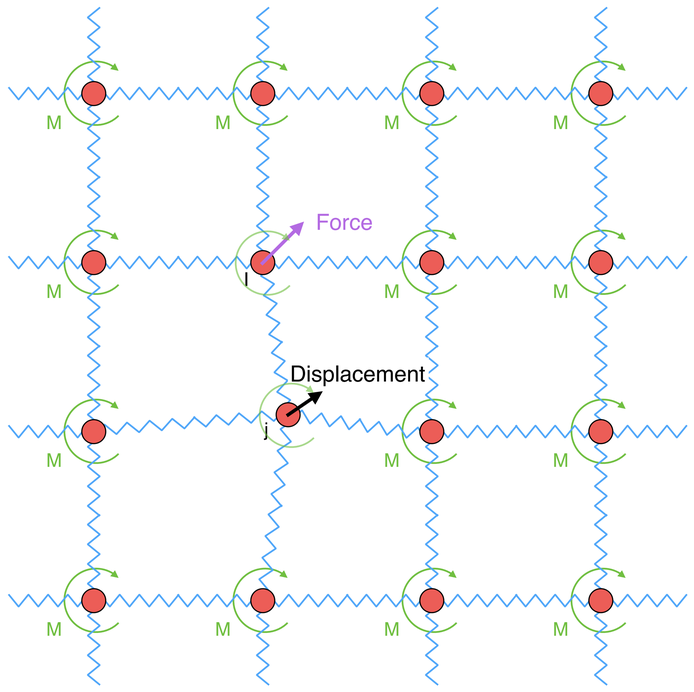
$$\mathbf{F}_i = C_{ij}\Delta\mathbf{u}_j$$

Frequencies and eigenvectors are found by solving

$$\mathbf{C}\eta_n = \mathbf{M}\omega_n^2\eta_n$$

C is invariant under time reversal, even for magnetic systems

$$C_{ij}^{(-M)} = -\frac{\partial^2 E}{\partial\mathbf{u}_i\partial\mathbf{u}_j} = C_{ij}^{(M)}$$



How to obtain broken time reversal ion dynamics from first principles?

Adiabatic theory

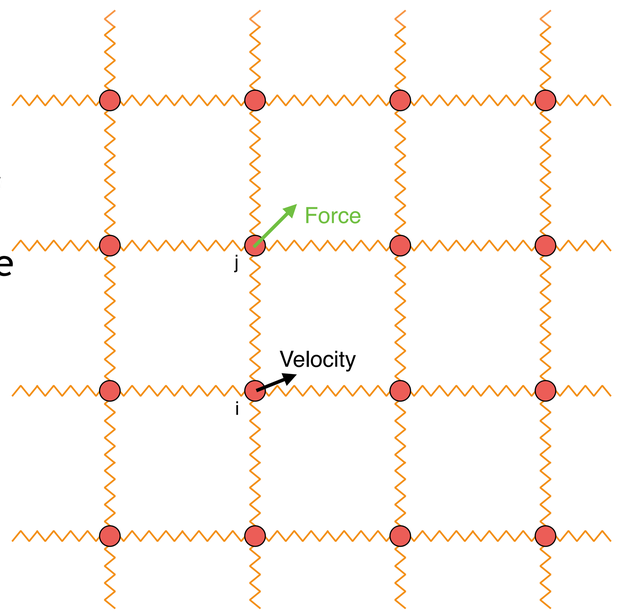
Effective vector potential acting on nuclei

$$\left(\sum_j \frac{\left(-i\hbar\nabla_{u_j} + -i\hbar\langle\psi_0|\nabla_{u_j}|\psi_0\rangle\right)^2}{2m_j} + V_{\text{eff}}\right)\chi_i = E_i\chi_i$$

Results in velocity dependent force

$$-m_I\frac{d^2u_{I\kappa}}{dt^2} = \sum_{J\kappa'} C_{I\kappa,J\kappa'}u_{J\kappa'} + \sum_{J\kappa} G_{I\kappa,J\kappa'}\frac{du_{J\kappa'}}{dt}$$

Nuclei mass; Force constant matrix; Displacement; Force-velocity matrix



Constant is Berry curvature with ionic positions

$$G_{I\kappa,J\kappa'} = 2\hbar\mathrm{Im}\langle\frac{\partial\psi}{\partial u_{I\kappa}}|\frac{\partial\psi}{\partial u_{J\kappa'}}\rangle$$

Now phonons found with:

$$\left(\mathbf{C} - i\omega_n\mathbf{G}\right)\eta_n = \mathbf{M}\omega_n^2\eta_n$$

Mead and Truhlar, Chem. Phys. 70, 2284 (1979)
Qin et al., PRB 86, 104305 (2012)
Saito et al., PRL 123, 255901 (2019)
Saparov et al. PRB 105, 064303 (2022)

Velocity force matrix from first principles

Need $G_{I\kappa,J\kappa'} = 2\hbar\mathrm{Im}\langle\frac{\partial\psi}{\partial u_{I\kappa}}|\frac{\partial\psi}{\partial u_{J\kappa'}}\rangle$

Discrete Berry phase approach:

Assume constant G in small space of nuclear displacements

$$\iint_S \langle\frac{\partial\psi}{\partial u_{I\kappa}}|\frac{\partial\psi}{\partial u_{J\kappa'}}\rangle = \mathrm{Imln}\left[\langle\psi(\underline{R})|\psi(\underline{R}+u_{I\kappa})\rangle\right.$$
$$\langle\psi(\underline{R}+u_{I\kappa})|\psi(\underline{R}+u_{J\kappa'})\rangle$$
$$\left.\langle\psi(\underline{R}+u_{J\kappa'})|\psi(\underline{R})\rangle\right]$$

Divide by area of S to obtain $G_{I\kappa,J\kappa'}$

Example Material CrI_3

FM insulator w/ strong SOC

$R\bar{3}$ space group

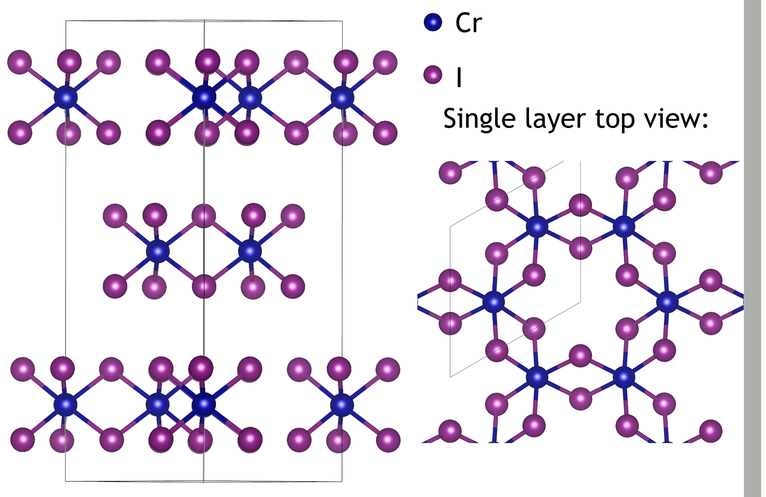
At Γ space group has 2D irreps

→degenerate phonon modes

Time reversal is broken

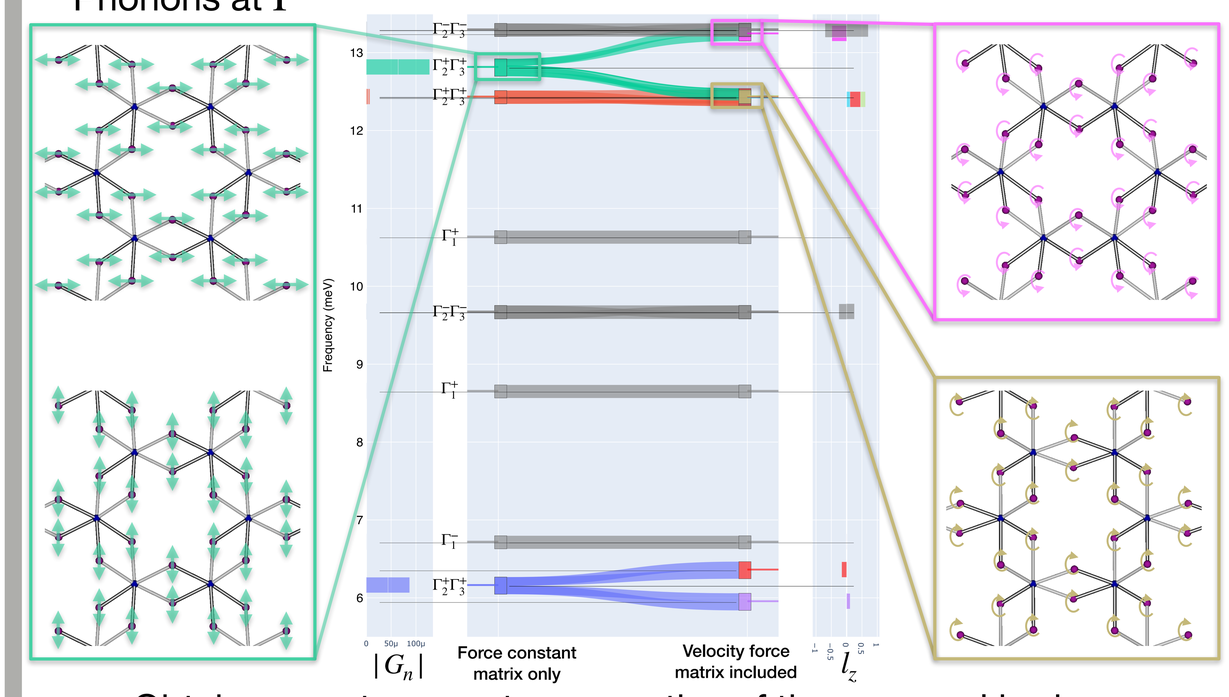
Corresponding magnetic space group has only 1D irreps

→no symmetry enforced degeneracy



First principles adiabatic theory results

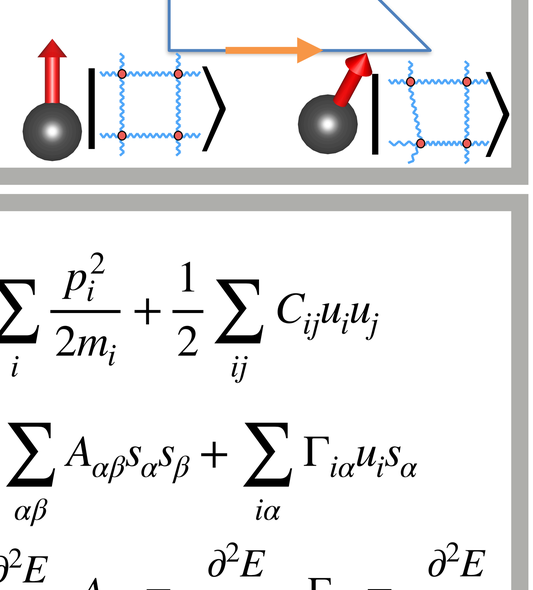
Phonons at Γ



- Obtain correct symmetry properties of time reversal broken system
- Pairs of otherwise degenerate modes now split to left and right chiral states
- Split modes can have angular momentum

Microscopic source of Berry curvature

- Freezing in nuclear displacements can tilt spins on Cr
- On closed path spins can sweep out a solid angle and pick up a phase
- In CrI_3 Berry phase is almost entirely described by Cr spin canting



Beyond adiabatic theory

- Adiabatic theory only appropriate if spin degrees of freedom are "fast" with respect to nuclear dynamics
- Otherwise must consider phonon and spin excitations on the same footing
- Quantitative results change significantly, with largest splitting reduced by over 2 orders of magnitude!

$$H = \sum_i \frac{p_i^2}{2m_i} + \frac{1}{2}\sum_{ij} C_{ij}u_iu_j$$
$$+\frac{1}{2}\sum_{\alpha\beta} A_{\alpha\beta}s_\alpha s_\beta + \sum_{i\alpha}\Gamma_{i\alpha}u_is_\alpha$$

$$C_{ij} = \frac{\partial^2 E}{\partial u_i\partial u_j}\quad A_{\alpha\beta} = \frac{\partial^2 E}{\partial s_\alpha\partial s_\beta}\quad \Gamma_{i\alpha} = \frac{\partial^2 E}{\partial u_i\partial s_\alpha}$$

Mode	Frequency (meV)		
	C only	Adiabatic	Non-adiabatic
12	12.723819	12.36314	12.722631
13	12.723819	13.118631	12.725151

Conclusions

- Magnetic order can split phonon modes, requires a treatment beyond the force constant matrix
- Resulting phonons can have chiral character and angular momentum
- In CrI_3 the adiabatic Berry phase theory vastly overestimates splitting of some modes

Outlook

- Full first principles non-adiabatic magnon-phonon coupling
- DFPT implementation
- Calculations for other materials, including metals
- First principles approaches to Phonon Hall effect and other observables