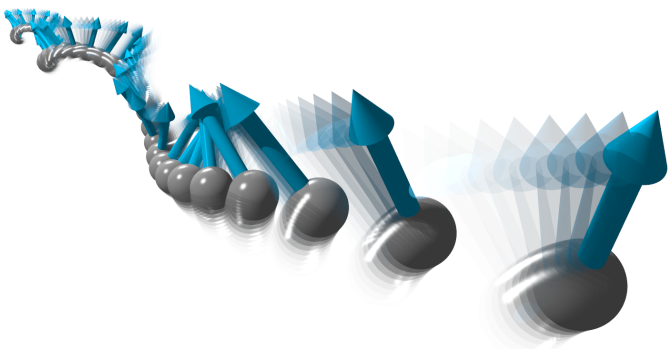
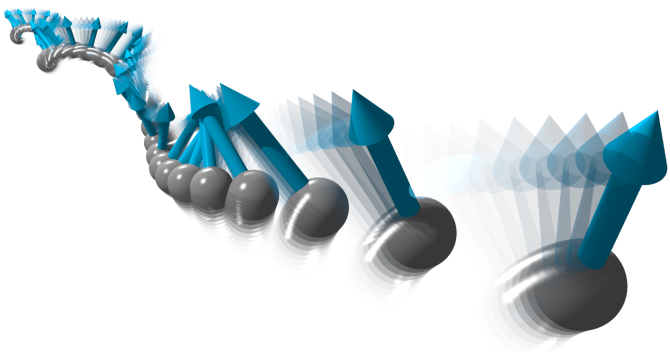
Phonon angular momentum in ultrafast demagnetization and hybrid quasiparticles



Markus Weißenhofer

Department of Physics and Astronomy, Uppsala University

Magnetism gives phonons a twist



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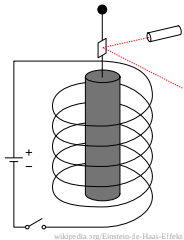
Magnetism and lattice angular momentum

- **Einstein–de Haas effect:** changing magnetic order with an external field leads to rotation

  Einstein and de Haas, Verh. d. DPG 17 (1915)



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- Reciprocal effect: **Barnett effect**

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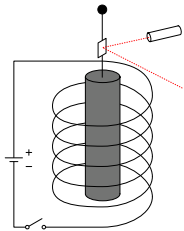

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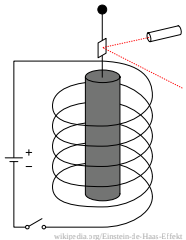

These show that:

1. Magnetization originates from angular momenta
2. Angular momentum can be exchanged between magnetic order and lattice

How fast can we transfer angular momentum?

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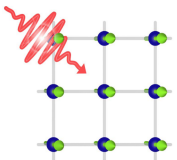
- Femtosecond laser pulse



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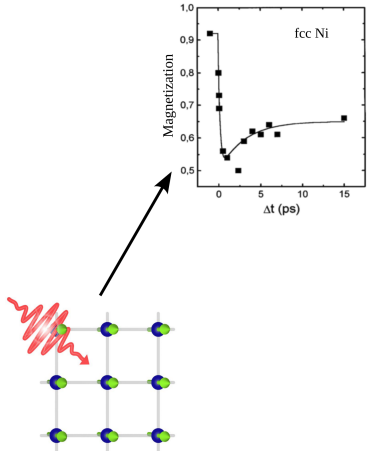
- **Sub-ps** loss of magnetic order



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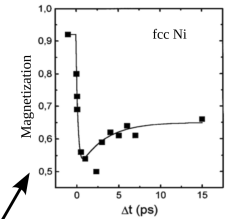

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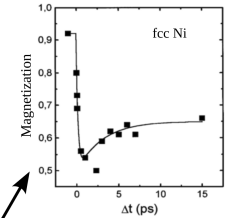
Dornes et al., Nature 565, 209 (2019)

How fast can we transfer angular momentum?

- Femtosecond laser pulse
- **Sub-ps** loss of magnetic order
- Ultrafast Einstein–de Haas effect
- Excitation of **chiral phonons***



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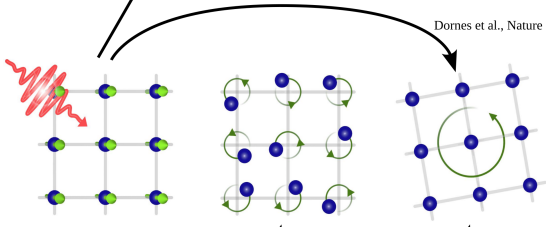

Tauchert et al., Nature 602, 737 (2022)

*Axial phonons *Juraschek et al., Nat. Phys. 21 (2025)*

Ultrafast manipulation of magnetism via chiral phonons

Ultrafast Barnett effect

- Paramagnet CeF_3

  Luo et al. Science 382, 698 (2023)

- Paraelectric diamagnet $SrTiO_3$

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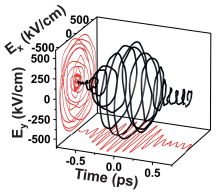

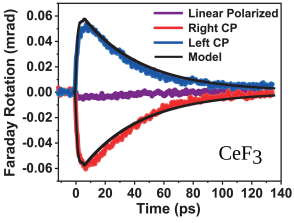

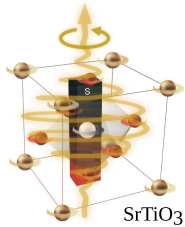

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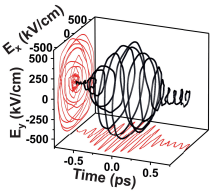
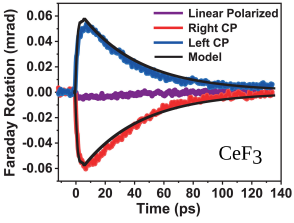
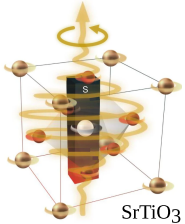


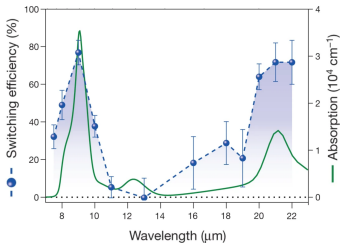




Circular phonon induced switching of magnetization

- Ferrimagnet GdFeCo on Al_2O_3 or SiO_2

  Davies et al., 628, 540 Nature (2024)



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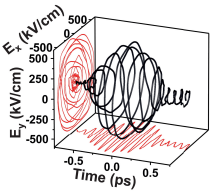

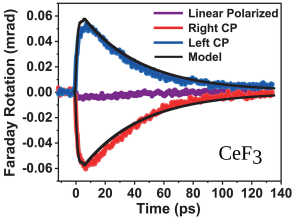

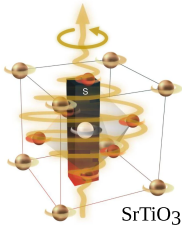

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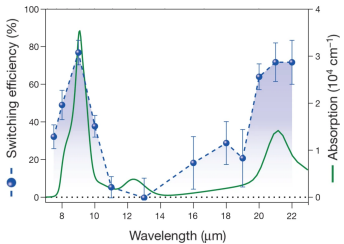

Problem

Mechanisms not understood ↔ lack of microscopic theory

Overview

1. Introduction
2. Phonon angular momentum after ultrafast demagnetization of FePt
3. Atomistic spin-lattice Hamiltonians
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5. Summary

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Time-dependent DFT

Electron dynamics → **time-dependent Kohn-Sham equation**

$$i\hbar\frac{\partial}{\partial t}\psi_{\boldsymbol{k}n}(\boldsymbol{r},t) = \left\{\frac{1}{2m}\left(-i\hbar\boldsymbol{\nabla}+\frac{e}{c}\boldsymbol{A}(t)\right)^2\hat{\sigma}_0+\hat{V}_{\mathrm{KS}}(\boldsymbol{r},t)\right\}\psi_{\boldsymbol{k}n}(\boldsymbol{r},t)$$

- Pauli spinor wavefuction $\psi_{\boldsymbol{k}n}(\boldsymbol{r},t) = (\chi^{\uparrow}_{\boldsymbol{k}n}, \chi^{\downarrow}_{\boldsymbol{k}n})^{\mathrm{T}}$
- Kohn-Sham potential $\hat{V}_{\mathrm{KS}}(\boldsymbol{r},t)$
- Laser excitation is modeled via vector potential $\boldsymbol{A}(t)$

Nuclear dynamics → **Ehrenfest dynamics**

OCTOPUS code *Tancogne-Dejean et al., J. Chem. Phys. 152, 124119 (2020)*

Ultrafast Demagnetization of $L1_0$ FePt

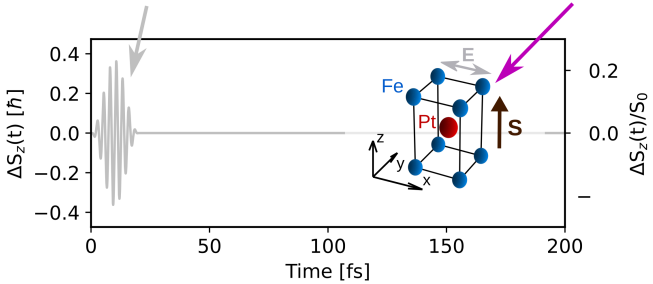
Linear pulse: electric field $\boldsymbol{E} \parallel \boldsymbol{e}_x$

Strong laser field

Central frequency 1.55 eV/ħ
Peak intensity 2 TW/cm^2
Duration 20 fs

Ferromagnet

Magnetization of ≈ 3 μ_B
Strong spin-orbit coupling



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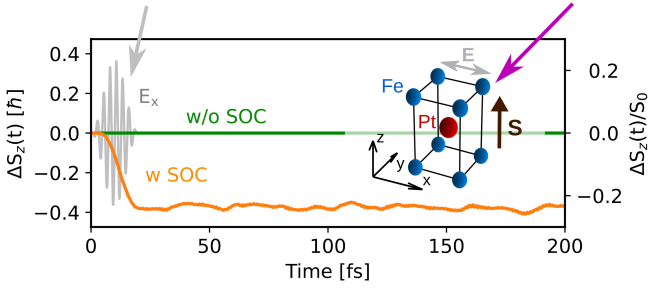

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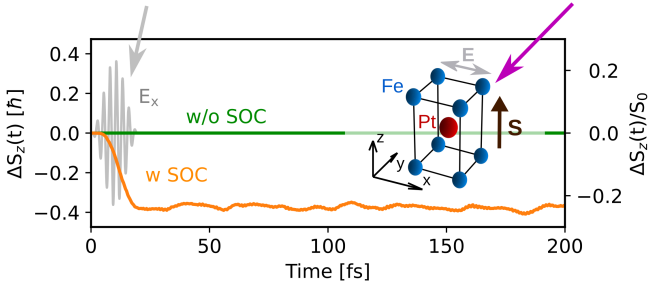
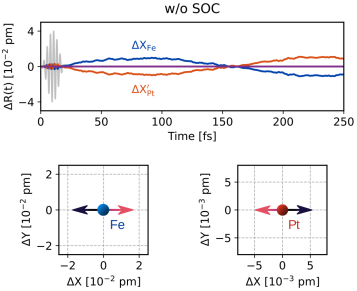


Without SOC: $\partial_t \hat{S} \propto [\hat{\mathcal{H}}, \hat{S}] = 0$

Demagnetization due to optically induced spin-flips *Krieger et al., JCTC 11 (2015); Mrudul and Oppeneer, PRB 109 (2024)*

Laser driven phonon excitation

Linear pulse: electric field $\boldsymbol{E} \parallel \boldsymbol{e}_x$



- Without SOC: linear optical phonon along $\boldsymbol{E}$
- With SOC: **phonon angular momentum**
- Frequency of displacements corresponds to pair of degenerate optical phonon modes
- Consistent with experiment? *Tauchert et al., Nature 602, 737 (2022)*

*Mrudul, **MW** et al., PRB 112, L180407 (2025)*

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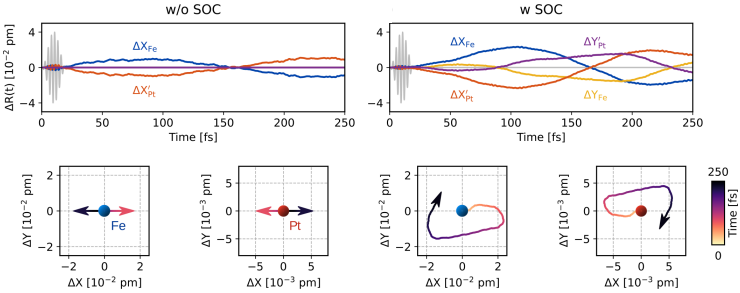

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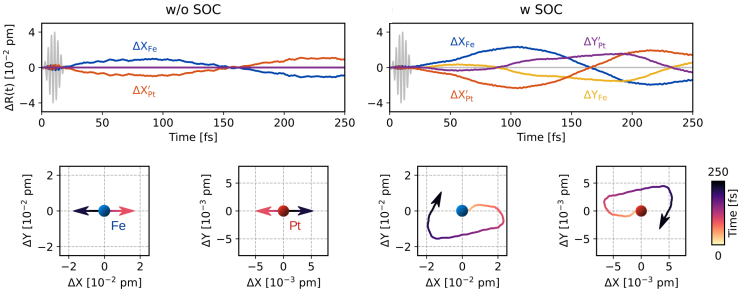

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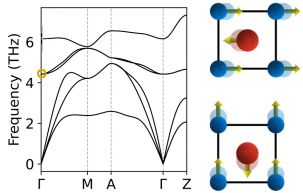

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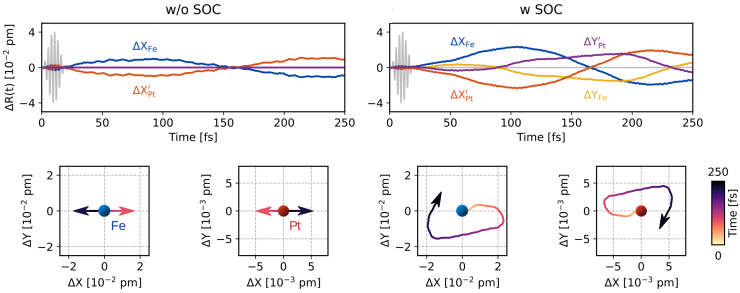

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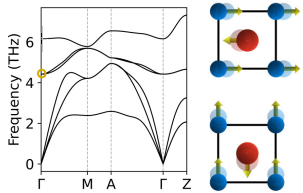

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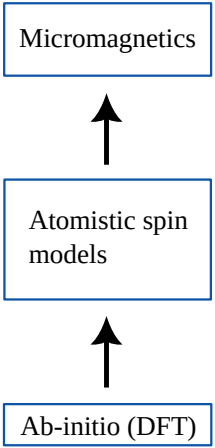
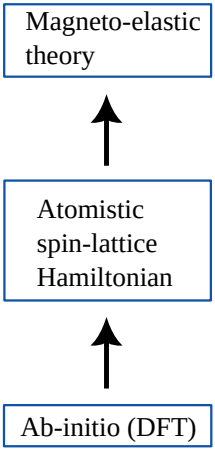
Multiscale approach for spin dynamics



Multiscale approach for spin-lattice dynamics



Multiscale approach for spin-lattice dynamics

Magneto-elastic theory

↑

Atomistic spin-lattice Hamiltonian

↑

Ab-initio (DFT)

Expanding extended Heisenberg Hamiltonian in displacements $\boldsymbol{u}_i = \boldsymbol{r}_i - \boldsymbol{r}_i^{\mathrm{eq}}$:

Hellsvik et al., PRB 99 (2019)

$$\mathcal{H} = \sum_{ij,\alpha\beta} J_{ij}^{\alpha\beta}(\{\boldsymbol{r}_k\}) S_i^\alpha S_j^\beta = \underbrace{\sum_{ij,\alpha\beta} J_{ij}^{\alpha\beta}(\{\boldsymbol{r}^{\mathrm{eq}}\}) S_i^\alpha S_j^\beta}_{\text{spin}-\text{spin interactions}} + \underbrace{\sum_{ijk,\alpha\beta\mu} J_{ij,k}^{\alpha\beta,\mu}(\{\boldsymbol{r}^{\mathrm{eq}}\}) S_i^\alpha S_j^\beta u_k^\mu}_{\text{spin}-\text{lattice interactions}} + \dots$$

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Spin-lattice coupling (SLC) tensors $J_{ij,k}^{\alpha\beta,\mu}(\{\boldsymbol{r}^{\mathrm{eq}}\}) = \partial J_{ij}^{\alpha\beta} / \partial r_k^\mu (\{\boldsymbol{r}^{\mathrm{eq}}\})$ can be calculated from first principles via

- Super cell method
  Hellsvik et al., PRB 99 (2019)
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  Liechtenstein et al., JMMM 67 (1987)
  *Mankovsky, **MW** et al., PRL 129 (2022)*
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Multiscale approach for spin-lattice dynamics

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Magneto-elastic constants B_1, B_2,... can be **calculated from microscopic SLC parameters** *MW et al., PRB 108 (2023); Miranda, MW et al., Phys. Rev. Mat. 9 (2025)*

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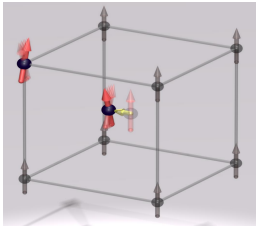
Energy and angular momentum transfer

$$\mathcal{H}^{\mathrm{SLC}} = \sum_{ijk,\alpha\beta\mu} J_{ij,k}^{\alpha\beta,\mu} S_i^{\alpha} S_j^{\beta} u_k^{\mu}$$

SLC contains change in

- isotropic Heisenberg exchange
- Dzyaloshinskii–Moriya interaction (DMI)
- Magnetocrystalline anisotropies

due to displacements



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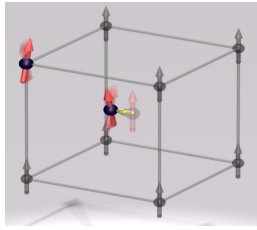
- Magnetocrystalline anisotropies

due to displacements



Example: bcc Fe *Mankovsky, **MW** et al., PRL 129, 067202 (2022)*

1. Correction to isotropic Heisenberg exchange (non-relativistic)

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2. u_k^{μ} break inversion symmetry → **emergent DMI** (relativistic = prop. to spin-orbit interaction)

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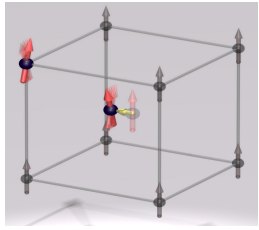

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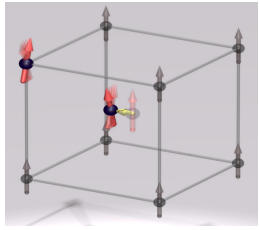

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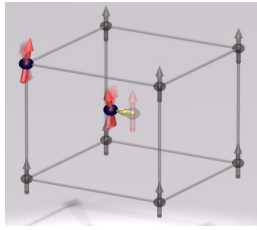

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Overview

1 Introduction

2 Phonon angular momentum after ultrafast demagnetization of FePt

3 Atomistic spin-lattice Hamiltonians

4 **Chiral phonons arising from magnon-phonon interactions**

5 Summary

Phonon angular momentum

Phonon eigenmode $|\psi_{\boldsymbol{k}n}\rangle$ is characterized by

- Wave vector $\boldsymbol{k}$
- Band index n (acoustic/optical)
- Frequency $\omega_{\boldsymbol{k}n}$
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How do phonons "know" about broken $\mathcal{T}$ symmetry in ferromagnets?

Magnon-phonon hybridization in bcc Fe

- Second quantization (Holstein-Primakoff transformation)

$$\mathcal{H}^{\mathrm{SLC}} = \sum_{ijk,\alpha\beta\mu} J_{ij,k}^{\alpha\beta,\mu} S_i^{\alpha} S_j^{\beta} u_k^{\mu} \quad \rightarrow \quad \boxed{\sum_{\boldsymbol{k}\lambda} c_{\boldsymbol{k}\lambda}^{-} \hat{b}_{\boldsymbol{k}} \hat{a}_{\boldsymbol{k}\lambda}^{\dagger} + c_{\boldsymbol{k}\lambda}^{+} \hat{b}_{\boldsymbol{k}}^{\dagger} \hat{a}_{\boldsymbol{k}\lambda}} + \ldots$$

- "Bare" phonon and magnon modes

$$H^{\mathrm{p}} = \sum_{\boldsymbol{k}\lambda} \hbar\Omega_{\boldsymbol{k}} \hat{a}_{\boldsymbol{k}\lambda}^{\dagger} \hat{a}_{\boldsymbol{k}\lambda} \qquad \text{and} \qquad H^{\mathrm{m}} = \sum_{\boldsymbol{k}} \hbar\omega_{\boldsymbol{k}} \hat{b}_{\boldsymbol{k}}^{\dagger} \hat{b}_{\boldsymbol{k}}$$

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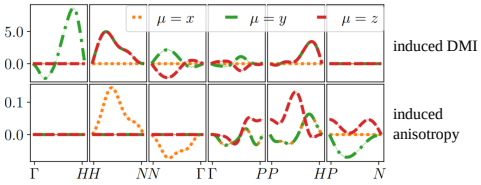
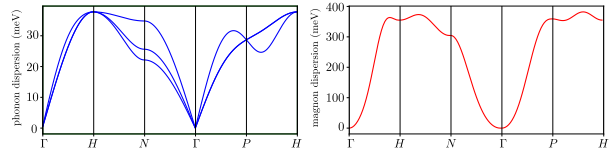
ab initio magnon-phonon coupling



Phonon/magnon energies



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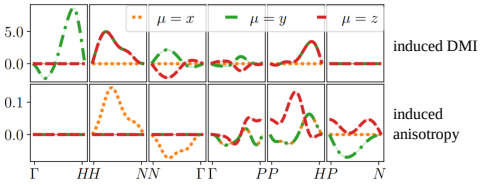

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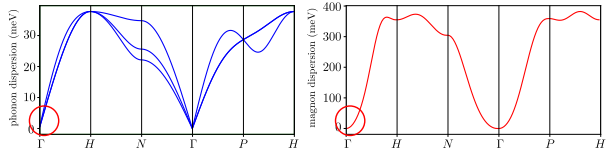

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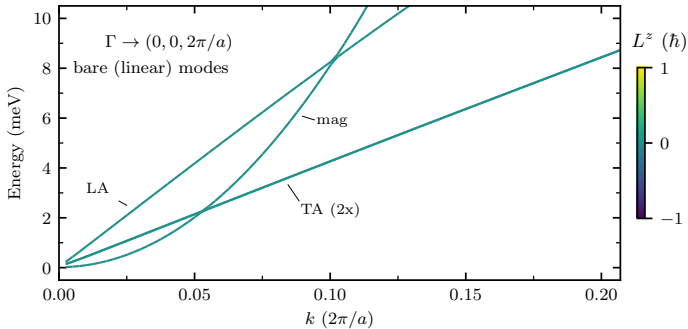
Hybridized magnon-phonon spectrum



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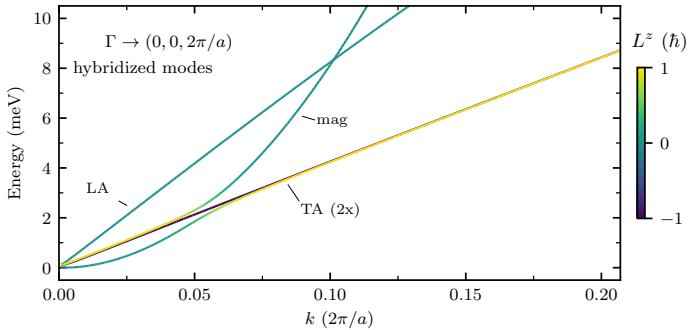

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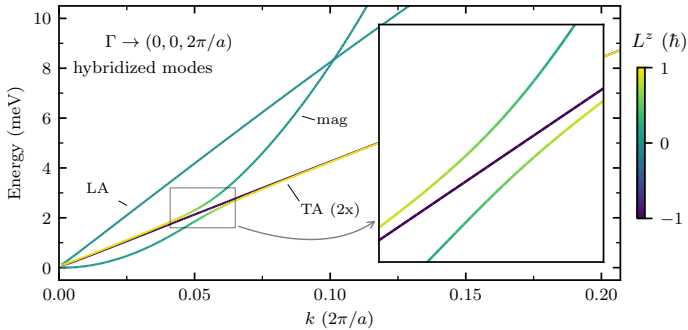

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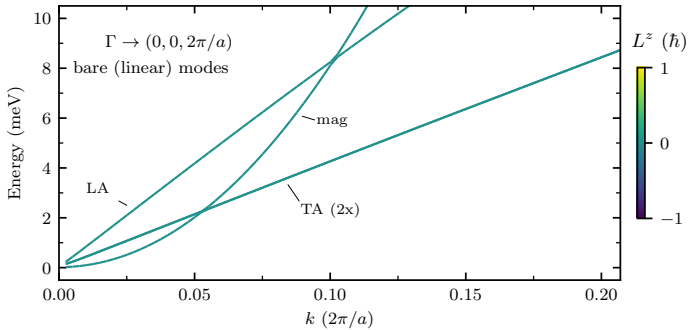

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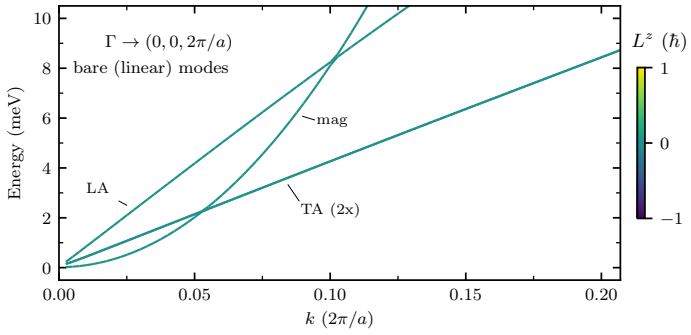

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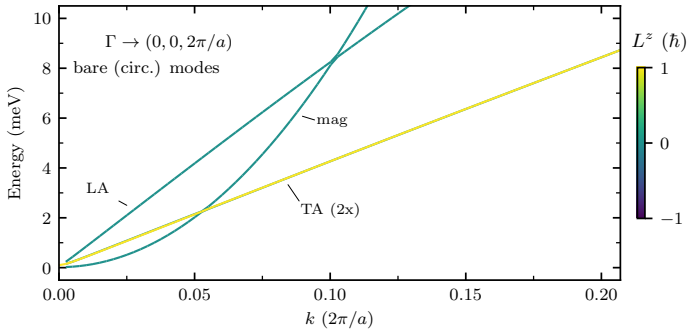

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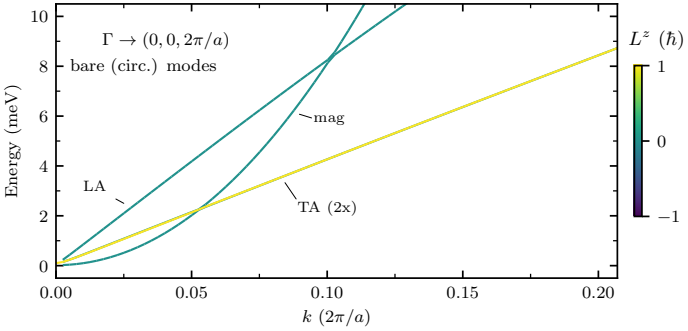

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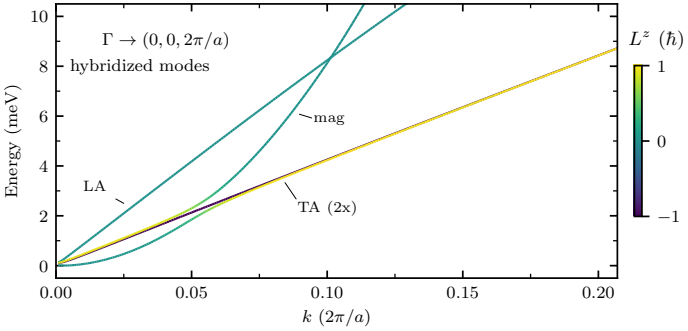

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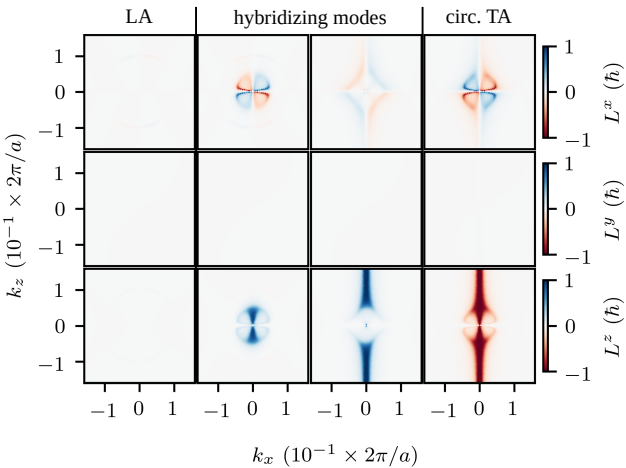
Phonon angular momentum in k_x, k_z-plane



- Magnetization $\parallel \boldsymbol{e}_z$
- $\boldsymbol{L}_{\boldsymbol{k}n} = \langle \psi_{\boldsymbol{k}n} | \sum_i \hat{\boldsymbol{u}}_i \times \hat{\boldsymbol{p}}_i | \psi_{\boldsymbol{k}n} \rangle$
- $k_y = 0$ plane

- Chiral phonons exist close to (but not exclusively at) high-symmetry lines
- Orientation of magnetization can be used to control where in BZ chiral phonons exist

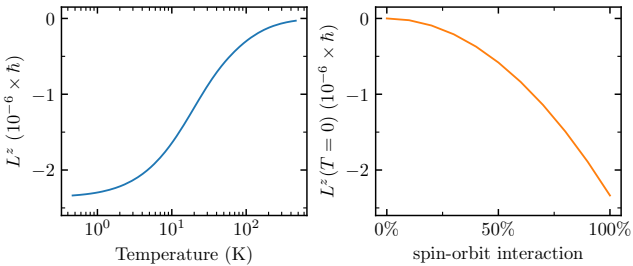
MW et al., PRL 135, 216701 (2025)

Equilibrium angular momentum

$$\boldsymbol{L}(T) = \mathrm{Tr}[\hat{\rho}^{\mathrm{eq}}(T)\hat{\boldsymbol{L}}] = \ldots$$
$$= \sum_{\boldsymbol{k}n} \boldsymbol{L}_{\boldsymbol{k}n} \left(n_{\boldsymbol{k}n}(T) + \tfrac{1}{2} \right)$$

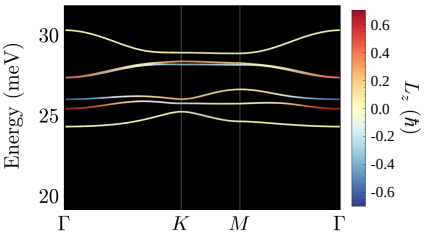
Zhang and Niu, PRL 112, 085503 (2014)

- $\boldsymbol{L}_{\boldsymbol{k}n}$ = ang. momentum of each mode
- $n_{\boldsymbol{k}n}(T)$ = Bose-Einstein distribution
- **Zero-point angular momentum**
- $\boldsymbol{L}(T \to \infty) = \mathrm{Tr}[\hat{\boldsymbol{L}}] = 0$

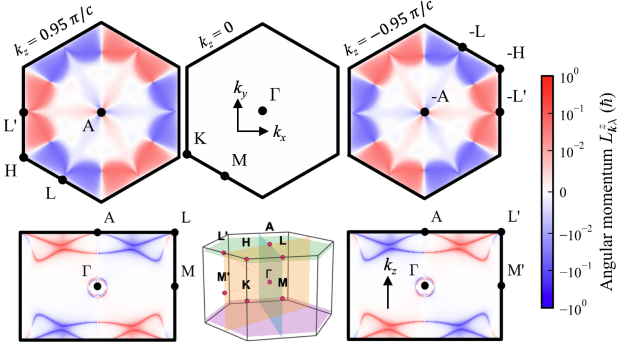


***MW** et al., PRL 135, 216701 (2025)*

Chiral phonons in other materials



2D ferromagnet CrI_3



Bulk g-wave altermagnet CrSb

*Rieger, **MW** et al. (in preparation)*

Floquet engineering of magnons using phonon angular momentum

Non-relativistic spin-lattice Hamiltonian

$$\hat{\mathcal{H}}(t) = \sum_{ij} \left[J_{ij} + \sum_{k} \frac{\partial J_{ij}}{\partial \boldsymbol{r}_{\boldsymbol{k}}} \cdot \boldsymbol{u}_k(t) \right] \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j$$

***MW** et al., arXiv:2605.28425 (2026)*

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- Periodic displacement $\boldsymbol{u}_k(t)$ following linear/circular phonons modes at Γ (E_u, 27.3 meV)

MW *et al., arXiv:2605.28425 (2026)*

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- **Floquet theory** to obtain effective spin Hamiltonian

$$\Delta\hat{\mathcal{H}} \propto \hat{\boldsymbol{S}}_i \cdot (\hat{\boldsymbol{S}}_j \times \hat{\boldsymbol{S}}_k)$$

- Holstein-Primakoff transformation $\rightarrow$ magnon Hamiltonian

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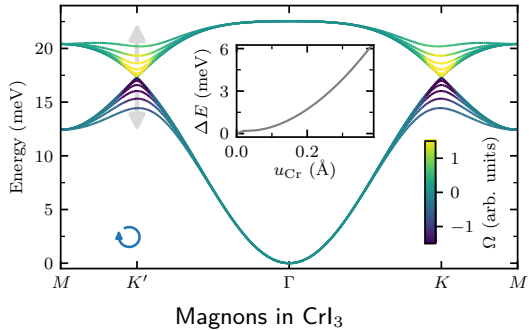

Magnons in CrI_3

MW et al., arXiv:2605.28425 (2026)

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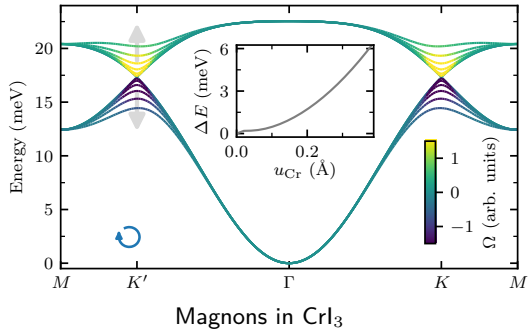
- Holstein-Primakoff transformation → magnon Hamiltonian
- Driving linear phonons does not change magnon energies!



Magnons in CrI_3

$\Delta E \propto |\mathrm{PAM}|$ Chern numbers $\propto \mathrm{sgn}[\mathrm{PAM}]$

PAM = phonon angular momentum

***MW** et al., arXiv:2605.28425 (2026)*

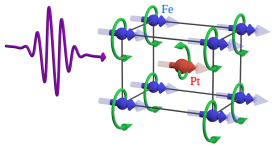
Summary & Acknowledgements

Two *ab initio* approaches to spin-lattice coupling:

1. Time-dependent density functional theory (TDDFT)

Ultrafast demagnetization generates **phonon angular momentum** via spin-orbit coupling

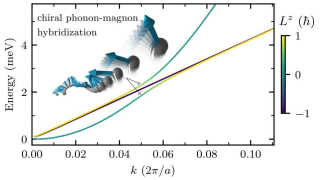
*Mrudul, **MW** and Oppeneer, PRB 112, L180407 (2025)*



2. Atomistic spin-lattice Hamiltonians

$\mathcal{T}$-symmetry breaking creates **chiral phonons** via magnon-phonon coupling

***MW** et al., PRL 135, 216701 (2025)*



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Ultrafast Spin Dynamics

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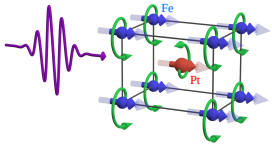

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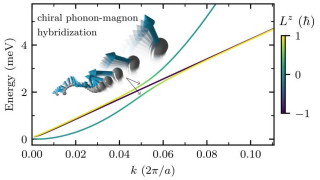

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CRC / TRR 227
Ultrafast Spin Dynamics

Thank you for your attention!

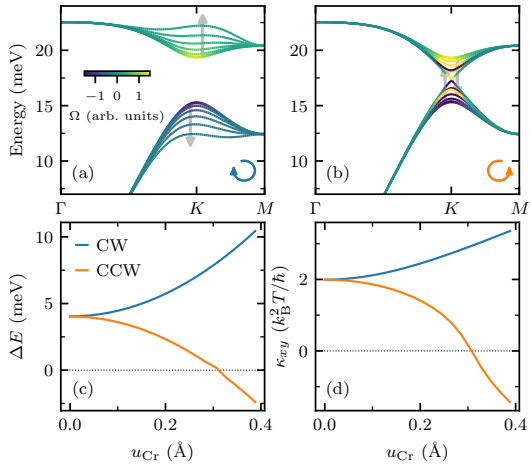
UPPSALA UNIVERSITET

Floquet engineering of magnons using phonon angular momentum

Relativistic spin-lattice Hamiltonian

$$\hat{\mathcal{H}}(t) = \sum_{ij}\left[J_{ij} + \sum_{k}\frac{\partial J_{ij}}{\partial \boldsymbol{r_k}}\cdot \boldsymbol{u}_k(t)\right]\hat{\boldsymbol{S}}_i\cdot\hat{\boldsymbol{S}}_j + \sum_{ij}\boldsymbol{D}_{ij}\times(\hat{\boldsymbol{S}}_i\times\hat{\boldsymbol{S}}_j)$$

- Magnons are already gapped w/o phonons
- Clockwise phonons widen the gap
- Counter-clockwise phonons close and revert the gap



- Anomalous transport $j_x = \kappa_{xy}\partial_y T$ can be controlled by phonon angular momentum

MW et al., arXiv:2605.28425 (2026)

Field-tunable thermal Hall responses of magnon-phonon quasi-particles

Ultrathin Fe(001) film; out-of-plane magnetic field

Anomalous thermal Hall effect
→ transverse heat current

$$J_x = -\kappa_{xy}\partial_y T$$

$$\kappa_{xy} \sim \sum_{\boldsymbol{k}n} \Omega_{xy,\boldsymbol{k}n} c_2(\varepsilon_{\boldsymbol{k}n})$$

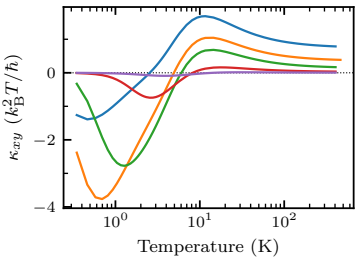
Berry curvature $\Omega_{xy,\boldsymbol{k}n}$

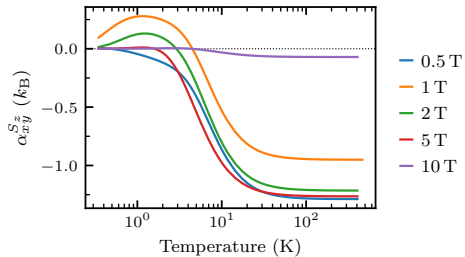
Spin Nernst effect
→ transverse spin current

$$J_x^{S_z} = -\alpha_{xy}^{S_z}\partial_y T$$

$$\alpha_{xy}^{S_z} \sim \sum_{\boldsymbol{k}n} \Omega_{xy,\boldsymbol{k}n}^{S_z} c_1(\varepsilon_{\boldsymbol{k}n})$$

Spin Berry curvature $\Omega_{xy,\boldsymbol{k}n}^{S_z}$





MW et al., arXiv:2411.03879