

Non-reciprocal magnons in a two dimensional crystal with off-plane magnetization

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Non reciprocal spin waves have a chiral asymmetry so that their energy is different for two opposite wave vectors. They are found in atomically thin ferromagnetic overlayers with in plane magnetization and are linked to the anti-symmetric Dzyaloshinskii-Moriya surface exchange. We use an itinerant fermion theory based on first principles calculations to predict that non-reciprocal magnons can occur in Fe_3GeTe_2 , the first stand alone metallic two dimensional crystal with off-plane magnetization. We find that both the energy and lifetime of magnons are non-reciprocal and we predict that acoustic magnons can have lifetimes up to hundreds of picoseconds, orders of magnitude larger than in other conducting magnets.

A defining property of elementary excitations in crystals, such as electrons, excitons, phonons, plasmons and magnons is their dispersion curve $E(\vec{q})$. In most cases, the dispersion curves satisfy the reciprocity relation $E(\vec{q}) = E(-\vec{q})$, reflecting the equivalence between the excitation and its mirror image, i.e., their non-chiral nature. In condensed matter systems, non-reciprocal energy dispersions occur under specific circumstances and elicit great attention. Examples are chiral [1] and helical [2] edge states of topological phases of various excitations, including electrons, photons and magnons, as well as Rashba split bands in crystals lacking inversion symmetry and having strong spin orbit coupling [3].

A major driving force for chiral phenomena in magnetism [4–6] is the antisymmetric exchange $\vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$, proposed by Dzyaloshinskii [7] and Moriya [8] (DM). This special type of super-exchange is enabled by the combination of spin orbit coupling [8] and the absence of an inversion center between spins i, j . These conditions are naturally found in overlayers of atomically thin ferromagnets on top of surfaces with high spin orbit coupling. With this background, the existence of non reciprocal spin waves was predicted [9, 10], provided that the DM vector \vec{D} is parallel to the magnetization \vec{M} . Symmetry considerations for this class of systems [11] leads to the conclusion that the interfacial \vec{D} lies in-plane, so that non-reciprocal spin waves in interfaces can only exist for ferromagnets with in plane easy axis, consistent with experimental observations [12, 13]. Logical devices based on non-reciprocal spin-waves have been recently proposed [14].

In this work we show that non-reciprocal spin waves can exist in a newly discovered class of 2D magnets [15], stand alone two dimensional crystals with off-plane magnetization. The survival of magnetism in 2D is definitely linked to a strong spin orbit coupling, that opens up a gap in the magnon spectrum, preventing the infrared catastrophe that destroys long range order in isotropic 2D magnets, as shown by Mermin and Wagner [16], in-

spired [17] by Hohenberg [18].

Here we explore magnons of Fe_3GeTe_2 for several reasons. First, it has a low symmetry magnetic unit cell, without an inversion center. Second, the observation of large Anomalous Hall effect [19], anomalous Nernst effect [20] and skyrmions [21] in thin films strongly suggests that intrinsic DM interaction, as opposed to interfacial, is active in Fe_3GeTe_2 . Third, the system is a conductor, unlike other widely studied 2D crystals such as CrI_3 , and has a large Curie temperature, that can reach room temperature upon gating [22]. Fe_3GeTe_2 was synthesized for the first time, in bulk, in 2006 [23]. Only much more recently, however, high quality few layers samples have been produced [24]. Monolayers have been obtained by exfoliation [25].

Because of its conducting nature and high-temperature ordering, Fe_3GeTe_2 is closer to technological applications. On the theory side, modelling magnons in conducting ferromagnets represents a big challenge due to the non-integer nature of the magnetic moments, the long-range exchange, and the damping of magnons due to their coupling to Stoner excitations. A microscopic description that does not take the itinerant character into account, such as that provide by spin models, will fail to describe most of the relevant physics of these systems.

We compute the magnon spectra of a Fe_3GeTe_2 monolayer using the itinerant fermion picture [10, 26]. With this method we are able to extract magnon energies and lifetimes from a first principles electronic structure calculation, without the need of building an intervening effective spin model. We use Density Functional Theory (DFT) [27, 28] to derive an effective fermionic Hamiltonian to describe the spin dynamics of 2D materials. The unit cell of Fe_3GeTe_2 is shown in Fig. 1. It has three Fe atoms, occupying two nonequivalent positions, A and B . We denote them Fe^A , Fe^{B_1} and Fe^{B_2} . There is no inversion center along the lines joining Fe^A and $\text{Fe}^{B_{1,2}}$.

The DFT calculations were performed using the plane waves code QUANTUM ESPRESSO [29]. The electronic

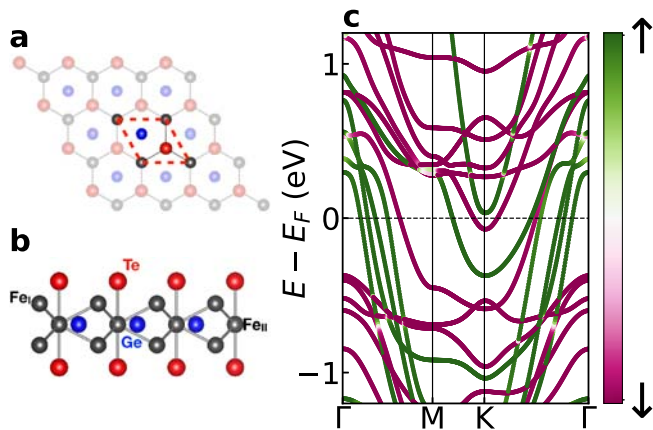


FIG. 1: Top (a) and side (b) views of the lattice structure, showing the unit cell (marked by the dashed red line in a) and the two nonequivalent Fe sites in (b). In (c) we show the band structure along high symmetry points in the 2D Brillouin zone (depicted in the inset of fig. 3a). The color code shows the projection of the electronic eigenvectors on the eigenstates of S_z .

exchange-correlation is described by the generalized gradient approximation (GGA) within the Perdew-Burke-Ernzerhof (PBE) functional [30]. Ionic cores are described using projector augmented wave (PAW) pseudopotentials [31]. The local effective paramagnetic Hamiltonian is obtained using a direct projection of the Kohn-Sham states onto pseudo-atomic orbital (PAO) basis [32], as implemented in the PAOFLow code [33].

The PAO tight-binding Hamiltonian is constructed using a *spd* basis for Fe, Ge and Te atoms. We then add local spin-orbit coupling and intra-atomic Coulomb repulsion [34]. The spin-orbit coupling strengths of Fe, Ge and Te are $\lambda_{\text{Fe}} = 50$ meV, $\lambda_{\text{Ge}} = 200$ meV, $\lambda_{\text{Te}} = 600$ meV [35]. The mean-field self-consistent ground state is obtained [26, 36] by treating every component of the spin moment in each Fe atom as an independent variable. The resulting band structure, shown in figure 1, features several spin polarized bands at the Fermi energy, portraying Fe_3GeTe_2 as a ferromagnetic conductor.

We find that the mean-field spin moments are $s_{B_1} = s_{B_2} = 2.56\mu_B$ and $s_A = 1.52\mu_B$, all of them along the off-plane axis. These values are in excellent agreement with the DFT results, $s_{B_1} = s_{B_2} = 2.54\mu_B$ and $s_A = 1.52\mu_B$. The spin moments of Te and Ge are negligible. We note that, given that the magnetic moments are approximately twice the spin values, the tentative spin values of Fe atoms are clearly not quantized as half integers.

The key quantity in the itinerant fermion theory for spin excitations [10, 26] is the spin-flip spectral density $S(E, \vec{q}) \equiv \text{Im}[\chi^\perp(E, \vec{q})]$, where

$$\chi_{l'l'}^\perp(E, \vec{q}) \equiv \int_{-\infty}^{\infty} dt e^{-i\frac{E}{\hbar}t} \left\{ -i\theta(t) \left\langle [S_{l,\vec{q}}^+(t), S_{l',-\vec{q}}^-(0)] \right\rangle \right\}, \quad (1)$$

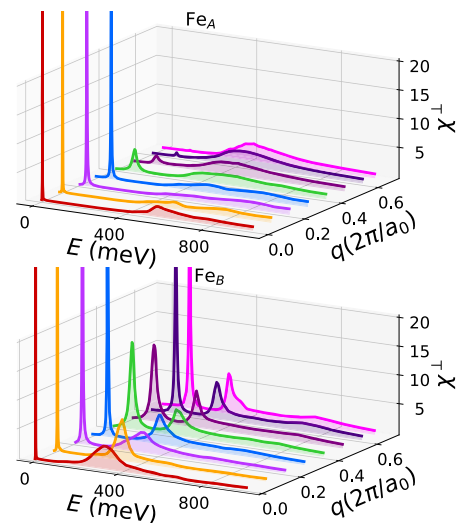


FIG. 2: Magnon spectral density projected at the two nonequivalent Fe sites as a function of energy, for a few selected wave vectors along the $\Gamma-K$ direction. The sharp peak at low energies is associated with the “acoustic” magnon and the broad structure at energies ~ 300 meV is the (strongly damped) “non-bonding” magnon.

l, l' are atomic site indices, E is the excitation energy, \vec{q} is the magnon wave vector, $\theta(t)$ is the Heaviside unit step function and $\langle \cdot \rangle$ denotes thermal average. The four fermion correlator in eq. (1) is computed in the Random Phase Approximation [10, 26, 37].

The diagonal entries, $\chi_{AA}^\perp(E, \vec{q})$ and $\chi_{B_1B_1}^\perp(E, \vec{q}) = \chi_{B_2B_2}^\perp(E, \vec{q})$ of the spin-flip spectral density are shown in figure 2 for a few selected wave vectors. For a given value of \vec{q} the spin flip spectral density has, in general, two types of features. First, symmetric peaks, with a width ΔE much smaller than peak energy E . These peaks are not present in the spectral density of the non-interacting susceptibility. These are magnons modes, featured by all ferromagnets. Second, broad asymmetric features, that correspond to the so called Stoner excitations and are only present in conducting ferromagnets.

Two well defined magnon branches are identified in Fig. 2. For reasons that will become apparent later, we refer to the lower energy, narrow peaks as the acoustic branch and to the higher energy, broader peaks as the non-bonding branch. When SOC is included, the acoustic branch has a gap at the Γ point is $\Delta_\Gamma = 2.9$ meV, that accounts for the magnetic anisotropy. Its magnitude is compatible with existing measurements [38] and DFT calculations [39]. The acoustic branch has weight distributed between A and B sublattices, although most of it lies on the B sites. In contrast, the non-bonding branch is missing entirely from the A site. A broad feature appears at higher ($\gtrsim 400$ meV), energies, localized in the A site, whose nature is discussed below.

The magnon dispersion relation along high-symmetry

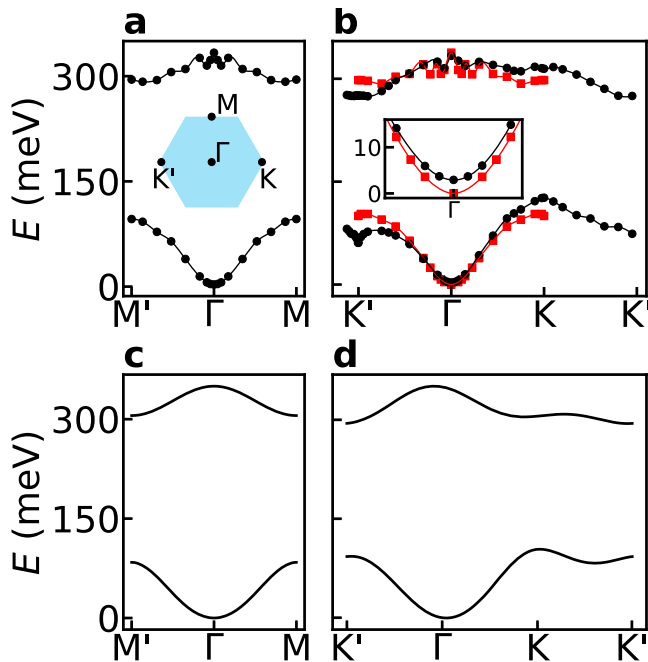


FIG. 3: Fe_3GeTe_2 magnon dispersion relation along high symmetry lines in the 2D Brillouin zone. The dispersion relation along Γ -M (a) is reciprocal, whereas along Γ -K (b, black circles) it shows strong non-reciprocity. In the inset we show a zoom of the dispersion relation for the acoustic magnon close to the Γ point, where the magnetocrystalline anisotropy gap $\Delta_\Gamma \equiv E(\Gamma) = 2.93$ meV can be clearly seen. For comparison, we also show the dispersion relation calculated without spin-orbit coupling (red squares), which is perfectly reciprocal and shows no anisotropy gap, as expected. In (c) and (d) we show the dispersion relations obtained with the localized spins model, including the second neighbor Dzyaloshinskii-Moriya coupling. The Brillouin zone is shown in the inset of panel a.

lines in the Brillouin zone is shown in figure 3, calculated both with and without spin orbit coupling. The bandwidth of the acoustic magnon (~ 120 meV) is much larger than that obtained in other 2D magnets, such as CrI_3 [26], and reflects a large exchange coupling between the magnetic moments in neighbouring Fe atoms, in line with the larger Curie temperature of Fe_3GeTe_2 .

Importantly, when the SOC is included in the calculation, both the acoustic and the non-bonding bands become non-reciprocal in the $K-\Gamma-K'$ direction, but not on the $\Gamma-M$ direction. It is noteworthy that the dispersion relation of the acoustic mode around the Γ -point fits almost perfectly to a function of wave vector q of the form $\Delta_\Gamma + Dq^2$, with negligible linear component. This is in contrast with the behavior of magnons in ultrathin transition metal films on heavy substrates [10], where a sizeable linear term is induced by the DM coupling, and has also been observed in relation to the calculation of static spin spirals in Fe_3GeTe_2 [40].

At this point we introduce a model Hamiltonian for the magnons, in order to gain physical insight on the

origin of the most salient features of the results obtained with the itinerant model. The departure point is a spin Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{Heis}} + \mathcal{H}_{\text{DM}} + \mathcal{H}_{\text{anis}}, \quad (2)$$

composed of an isotropic Heisenberg term $\mathcal{H}_{\text{Heis}}$, a Dzyaloshinskii-Moriya interaction \mathcal{H}_{DM} and a single-ion anisotropy term $\mathcal{H}_{\text{anis}}$. Explicit expressions and further detail can be found in the supplemental material (SM). We build the magnon model using the conventional Holstein-Primakoff linear spin wave theory for a quantized spin model. The spins live in a *decorated honeycomb* lattice with three sites per unit cell, A, B_1, B_2 (see Fig. 1 of the SM), with spins S_A and S_B .

Given that sites B_1 and B_2 are equivalent, we can introduce two new modes, symmetric and anti-symmetric combinations of B_1 and B_2 , so that one of them becomes effectively decoupled from A . The decoupling naturally leads to three bands. One is associated with the anti-symmetric B mode. The other two describe a honeycomb ferromagnet with broken inversion symmetry, on account of the different nature of A and B , and are separated by a gap. The projections of the magnon wave functions over the different sites (see Fig. 3 in the SM) show that the spin model naturally accounts for the fact that the acoustic branch is predominantly located in the symmetric B mode, the \vec{k} dependence of the weight on the A site, and the complete localization of non-dispersive band on the B mode. This behavior is qualitatively identical to that of the magnon wave functions extracted directly from the fermionic model (see SM for details).

We are now in position to address the origin of the non-reciprocal dispersion, obtained with the itinerant model, using the spin model. The fact that it only arises when spin orbit coupling is included is a clear indication that the its origin has to come from the non-Heisenberg terms in the Hamiltonian. We have considered both first and second neighbour DM couplings, $D_{a,a'}^{(1)}$ and $D_{a,a'}^{(2)}$, where a, a' label the sites in the unit cell that do not possess an inversion center. We only consider the DM vector \vec{D} parallel to the magnetization, *i.e.*, in the off-plane direction.

We find that first and second neighbour DM coupling yield non-reciprocal dispersions. However, only a finite $D^{(2)}$ coupling for the B sublattice gives a non-reciprocal dispersion in the $K-\Gamma-K'$ line, and reciprocal dispersion along the $\Gamma-M$ direction. Therefore, the non-reciprocal dispersion is consistent with a second neighbour DM interaction in the B sublattice, for which the super-exchange pathways occurs via Tellurium atoms, the ones with the largest SOC in the crystal.

We now shift our attention to one of the hallmarks of itinerant magnetism: the fact that magnons have finite lifetimes, because of their coupling with the continuum of uncorrelated electron-hole excitations known as the

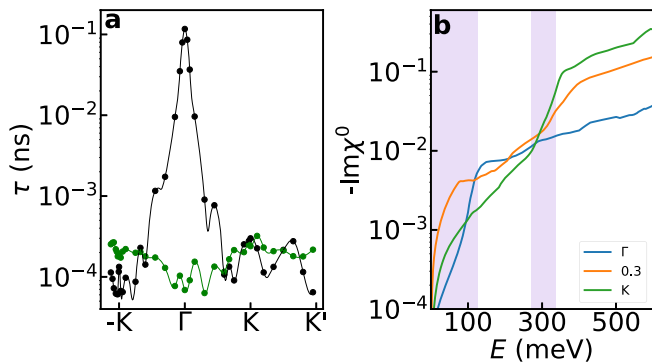


FIG. 4: **a**: Magnon lifetimes as a function of wave vector. **b**: Spectral density of Stoner modes as a function of energy for three different wave vectors along the $\Gamma - K$ line. The shaded regions mark the bandwidths of the acoustic and non-bonding magnons.

Stoner continuum. In figure 4a we show the magnon lifetimes as a function of wave vector. The lifetime is related to the linewidth of the spectral density via $\tau \equiv \frac{2\hbar}{\Delta E}$. Remarkably, the acoustic magnons close to the Γ point have very long lifetimes (~ 100 ps), given that the longest lifetimes measured in ultrathin conducting magnets [41] are ~ 0.4 ps. A long magnon lifetime is a very important figure of merit for potential applications of magnons as carriers of information, for example.

The lifetime of a magnon with energy E and wave vector \vec{q} scales inversely with the weight of the Stoner spectral density at the same energy and wave vector. Due to the spin polarization of the d bands, the density of Stoner modes is very small for energies much smaller than the exchange splitting (roughly proportional to magnetization and the intra-atomic Coulomb repulsion strength). It grows abruptly as the excitation energy approaches the exchange splitting, as seen in fig. 4b. For Fe_3GeTe_2 , the energies of acoustic magnons lie in the region of small density of Stoner modes, whereas the non-bonding magnons live in the energy range where the Stoner spectral density is considerable. This is the origin of the large difference between acoustic and non-bonding magnons lifetimes.

In this context, we can understand why the itinerant picture leads to only two magnon modes, whereas the localized spins model has three. Basically, the third magnon band, still higher in energy than the second, is degenerate with the continuum of Stoner spin flip excitations. As a result, the spectral weight of the high energy optical magnon mode is transferred to the incoherent features predominantly localized in the A site, shown in Fig. 2. The difference between the two theories highlights the limitations of the spin Hamiltonian, most notably in the case of itinerant magnets.

The acoustic magnon lifetimes are also non-reciprocal. This effect is *not* exclusively related to the non-

reciprocity of the energy dispersion: lifetimes are shorter in general for higher energy states. We find that, although magnons around the K point have both energies and lifetimes larger than those at K' . The ultimate reason of this non-reciprocal lifetimes stems from the fact that the density of Stoner modes in Fe_3GeTe_2 is also non-reciprocal.

Besides endowing magnons with finite lifetimes, the Stoner continuum renormalizes the magnon energies, much like friction changes the natural frequency of an harmonic oscillator. This is the origin of the oscillations in the dispersion relation of the non-bonding magnons, seen in figure 3c. The dispersion relation of the acoustic magnons close to the K point also display some oscillations of the same origin.

In conclusion, we have calculated magnons in monolayer Fe_3GeTe_2 using an itinerant fermion description derived from first principles calculations, and we have compared those results with the simple magnon theory for a spin model Hamiltonian for a decorated honeycomb lattice with three spins per unit cell. Due to broken mirror symmetry and spin-orbit coupling, magnons' energies and lifetimes show non-reciprocal behavior along the $\Gamma - K$ direction. Our findings are consistent with a second neighbour DM coupling in the B sublattice, but this deserves further attention. The coupling of magnons to Stoner excitations results in an intrinsic broadening of the two lowest energy magnon branches, and the melting of the optical mode, expected in the spin model, into a broad spectral feature at high energies. From our results we infer a value for the exchange stiffness that is compatible with the large magnetic transition temperatures observed experimentally. Furthermore, we find that the acoustic magnons are extremely long-lived for a conducting two-dimensional ferromagnet ($\tau \sim 100$ ps at the Γ point), which make this material potentially very useful for magnonics and spintronics applications. Our work shows that non-reciprocal magnons can exist in 2D crystals with off-plane magnetization due to their intrinsic DM interaction and suggest that Fe_3GeTe_2 is a very interesting material to explore non-trivial magnon effects.

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Supplemental Material for Non-reciprocal magnons in a two dimensional crystal with off-plane magnetization

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THE SPIN MODEL

We derive here the magnon Hamiltonian for a quantum spin model defined on a decorated honeycomb lattice. We consider a unit cell with 3 atoms, A , B_1 and B_2 , depicted in fig. 1. Atoms B_1 and B_2 are stacked vertically on top of each other. When seen from the top, the decorated honeycomb lattice looks like a regular honeycomb lattice, as atoms B_1 and B_2 are aligned.

We shall consider a Hamiltonian with three types of coupling: Heisenberg, Dzyaloshinskii-Moriya (DM) and single ion anisotropy,

$$\mathcal{H} = \mathcal{H}_{\text{Heis}} + \mathcal{H}_{\text{DM}} + \mathcal{H}_{\text{anis}} \quad (1)$$

In order to expedite the derivation of the spin wave theory taking advantage of the crystal translational invariance, we write the spin Hamiltonian making explicit the unit cell index \vec{R} and the intra cell label $a = A, B_1, B_2$. The Heisenberg coupling reads,

$$\mathcal{H}_{\text{Heis}} = \sum_{\vec{R}, \vec{R}', a, a'} J_{a, a'} (\vec{R} - \vec{R}') \vec{S}(\vec{R}, a) \cdot \vec{S}(\vec{R}', a') \quad (2)$$

Below we consider up to second neighbor exchange.

Taking into account only the z component of the Dzyaloshinskii-Moriya vector, the DM term reads,

$$\mathcal{H}_{\text{DM}} = - \sum_{\vec{R}, \vec{R}', a, a'} D_{a, a'} (\vec{R} - \vec{R}') \left[S_x(\vec{R}, a) S_y(\vec{R}', a') - S_y(\vec{R}, a) S_x(\vec{R}', a') \right]. \quad (3)$$

The single ion anisotropy reads,

$$\mathcal{H}_{\text{anis}} = \sum_{a, R} -|K_a| S_z^2(\vec{R}, a). \quad (4)$$

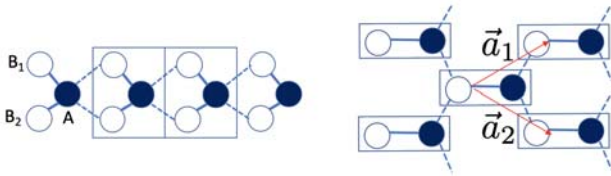


FIG. 1: Decorated Honeycomb lattice. Left panel: side view, showing 3 atoms per unit cell. Right panel: Top view, showing the honeycomb arrangement, and the Bragg vectors \vec{a}_1 and \vec{a}_2 .

LINEAR SPIN WAVE THEORY

We now introduce the standard bosonic Holstein-Primakoff (HP) representation of the spin operators [1]. We assume that the magnetization of the classical ground state lies along the \hat{z} axis and we keep only quadratic terms in the boson operator, which amounts to ignoring magnon-magnon interactions. This is the so-called linear spin wave theory. This is accomplished by expressing the spin operators in terms of the HP bosons c as

$$\begin{aligned} \vec{S}(\vec{r}) \cdot \vec{\Omega}(\vec{r}) &= S - c^\dagger(\vec{r})c(\vec{r}) \\ S^{(+)}(\vec{r}) &\simeq \sqrt{2S}c(\vec{r}) \\ S^{(-)}(\vec{r}) &\simeq \sqrt{2S}c^\dagger(\vec{r}), \end{aligned} \quad (5)$$

where \vec{r} labels every spin in the crystal. Thus, \vec{r} is completely specified by \vec{R}, a . The representation of the S_z^2 terms we only keep terms quadratic in the c operators, dropping the for boson operators.

Momentum representation of the magnon Hamiltonian

After a lengthy but straightforward calculation, we derive the a quadratic Hamiltonian for the HP bosons in the reciprocal space. For that matter, we represent the 1 using the 5, keeping only terms bilinear in the HP bosons, and we define

$$c_{\vec{k}, a} = \frac{1}{\sqrt{N}} \sum_{\vec{R}} c_{\vec{R}, a} e^{-i\vec{k} \cdot \vec{R}}. \quad (6)$$

We consider Heisenberg and DM coupling between first and second neighbors. The resulting magnon Hamiltonian reads,

$$\mathcal{H} = \sum_{\vec{k}, a, a'} c_{\vec{k}, a}^\dagger \mathcal{H}_{a, a'}(\vec{k}) c_{\vec{k}, a'}, \quad (7)$$

with

$$\mathcal{H}_{a, a'}(\vec{k}) \equiv \mathcal{H}_{a, a'}^{\text{Heis}} + \mathcal{H}_{a, a'}^{\text{DM}} + \mathcal{H}_{a, a'}^{\text{anis}}. \quad (8)$$

The wave vector \vec{k} spans the Brillouin zone of the triangular lattice.

Decoupling transformation and analytical expressions

Explicit expressions are given below. Importantly, the Hamiltonian has the form

$$\mathcal{H}_{a,a'}(\vec{k}) \equiv \begin{pmatrix} h_{AA}(\vec{k}) & h_{AB}(\vec{k}) & h_{AB}(\vec{k}) \\ h_{AB}(\vec{k})^* & h_{BB}(\vec{k}) & h_{12}(\vec{k}) \\ h_{AB}(\vec{k})^* & h_{12}(\vec{k}) & h_{BB}(\vec{k}) \end{pmatrix}, \quad (9)$$

reflecting the equivalence of sites B_1 and B_2 . By introducing a change of basis for the B_1 and B_2 sites, that defines the symmetric and antisymmetric modes, $c_{\vec{k},\pm} = \frac{1}{\sqrt{2}}(c_{\vec{k},B_1} \pm c_{\vec{k},B_2})$, we can write the magnon Hamiltonian matrix, in the $A, +, -$ basis as,

$$\mathcal{H}_{a,a'}(\vec{k}) \equiv \begin{pmatrix} h_{AA} & \sqrt{2}h_{AB} & 0 \\ \sqrt{2}h_{AB}^* & h_{BB} + h_{12} & 0 \\ 0 & 0 & h_{BB} - h_{12} \end{pmatrix} \quad (10)$$

where we have used that h_{12} is real and we have removed the explicit dependence on \vec{k} to simplify the notation. This equation shows that the antisymmetric ($-$) mode is decoupled from the other two.

The remaining coupled modes, A and B_+ , are isomorphic to quasiparticles in a honeycomb lattice with broken inversion symmetry, on account of the different couplings and spins of A and B sites. The decoupled expression (10) permits to obtain analytical expressions for the magnon bands,

$$\begin{aligned} E_{\text{ac}}(\vec{k}) &= \epsilon_0(\vec{k}) - \sqrt{\epsilon_1(\vec{k})^2 + 2|h_{AB}(\vec{k})|^2}, \\ E_{\text{op}}(\vec{k}) &= \epsilon_0(\vec{k}) + \sqrt{\epsilon_1(\vec{k})^2 + 2|h_{AB}(\vec{k})|^2}, \\ E_{\text{non-bonding}}(\vec{k}) &= h_{BB}(\vec{k}) - h_{12}(\vec{k}), \end{aligned} \quad (11)$$

where we have defined

$$\begin{aligned} \epsilon_0(\vec{k}) &= \frac{h_{AA}(\vec{k}) + h_{BB}(\vec{k}) + h_{12}(\vec{k})}{2}, \\ \epsilon_1(\vec{k}) &= \frac{h_{AA}(\vec{k}) - h_{BB}(\vec{k}) - h_{12}(\vec{k})}{2}. \end{aligned} \quad (12)$$

The acoustic and optical bands $E_{\text{ac}}(\vec{k})$, $E_{\text{op}}(\vec{k})$ are associated to magnons that live both in the A and B sublattice, with the B modes in phase, whereas $E_{\text{non-bonding}}(\vec{k})$ is *sublattice polarized*.

In the following we derive explicit expressions of the magnon bands in terms of the exchange, DM and anisotropy terms, and derive their values so that the simple model agrees with the magnon bands obtained with the itinerant fermion picture.

Magnon bands for the isotropic case

We consider first the case only symmetric exchange interactions are included. This permits to compare with the calculations without SOC.

We introduce the first neighbor exchange coupling constants, $J_{AB}^{(1)}$ and $J_{BB}^{(1)}$ as well as the second neighbour exchange for the A sublattice, $J_{AA}^{(2)}$, and the intralayer (or direct) and interlayer (or crossed) for the B sublattices $J_{BB,2d}^{(2)}$, $J_{BB,2c}^{(2)}$.

First neighbour approximation

We now keep only the first neighbour interaction. Introducing the notation $\phi_n = \vec{k} \cdot \vec{a}_n$, with $n = 1, 2$, we can write

$$\begin{aligned} h_{AB}(\vec{k}) &= -\sqrt{S_A S_B} J_{AB}^{(1)} (1 + e^{i\phi_1} + e^{i\phi_2}) \\ h_{AA} &= 6J_{AB}^{(1)} S_B \\ h_{BB} &= 3J_{AB}^{(1)} S_A + J_{BB}^{(1)} S_B \\ h_{12} &= -J_{BB}^{(1)} S_B \end{aligned} \quad (13)$$

Here the factors 6 and 3 reflect the number of first neighbours of the A and B sublattice that are connected via the $J_{AB}^{(1)}$. We obtain:

$$\epsilon_0(\vec{k}) = \frac{3J_{AB}^{(1)}}{2} (2S_B + S_A)$$

and

$$\epsilon_1(\vec{k}) = \frac{3J_{AB}^{(1)}}{2} (2S_B - S_A).$$

We now obtain the magnon energies at the high symmetry points. At Γ and K we have $f(\Gamma) = 3$ and $f(K) = f(K') = 0$. We thus have:

$$\begin{aligned} E_{\text{ac}}(\Gamma) &= 0 & E_{\text{ac}}(K) &= 3J_{AB}^{(1)} S_A \\ E_{\text{op}}(\Gamma) &= 3J_{AB}^{(1)} (2S_B + S_A) & E_{\text{op}}(K) &= 6J_{AB}^{(1)} S_B \end{aligned}$$

whereas the non-bonding state is dispersion less:

$$E_{\text{non-bonding}} = E_{\text{ac}}(K) + 2J_{BB}^{(1)} S_B.$$

From these equations we see that the gap between the optical and acoustic branch is due both to the different inter-sublattice coordination of A (6) and B (3) sites and their different spin $S_A \neq S_B$. We also see that the non-bonding state is inside the gap, as long as $2J_{BB}^{(1)} S_B < 3J_{AB}^{(1)} (2S_B - S_A)$.

We now use these results to obtain a rough estimate of the first neighbour exchange constants. At this level of the theory, the values of S_A and S_B do not need to be integer or half-integer, and we take we take $S_A = M_A/2 = 0.76$ and $S_B = M_B/2 = 1.26$. The value of the acoustic band at the K point permits to estimate $J_{AB}^{(1)}$, using $3J_{AB}^{(1)} S_A \simeq 100$ meV. We obtain $J_{AB}^{(1)} \simeq 44$ meV

Given that the band at around 300 meV is exclusively localized in the B sublattice, we infer that this is the non-bonding band. We thus have the equation $2J_{BB}^{(1)} S_B \simeq 200$ meV, from which we obtain $J_{BB}^{(1)} \simeq 80$ meV.

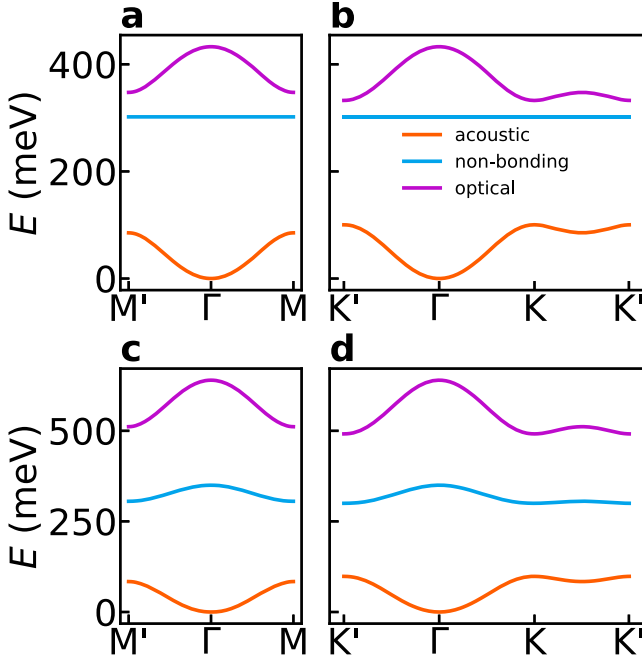


FIG. 2: Top panels: Magnon dispersion relations along high-symmetry lines in the triangular lattice's Brillouin zone, computed within the nn approximation. We take $J_{AB}^{(1)} \simeq 44$ meV, $J_{BB}^{(1)} \simeq 80$ meV. Bottom panels: same as above, but including nnn exchange. We take $J_{AB}^{(1)} \simeq 65$ meV, $J_{BB}^{(1)} \simeq 80$, $J_{BBs}^{(2)} = -4.4$ meV, $J_{BBd}^{(2)} = J_{AAd}^{(2)} = 0$.

Next nearest neighbour contribution

In order to obtain a dispersive non-bonding band, still in the case without SOC, it is necessary to include next nearest neighbour (nnn) exchange. This includes the AA nnn exchange $J_{AA}^{(2)}$ that will contribute to the acoustic and optical bands, as well as the BB exchange couplings, including the *same layer* $J_{B_1B_1}^{(2)} = J_{B_2B_2}^{(2)} \equiv J_{BBs}^{(2)}$ and the *different layer* $J_{B_1B_2}^{(2)} \equiv J_{BBd}^{(2)}$. Unlike the nn, the nnn gives diagonal contributions both for the Ising and flip-flop exchange in the AA and BBs channels. We thus have the following contributions to the Hamiltonian matrix:

$$\begin{aligned} h_{AA}^{(2)}(\vec{k}) &= J_{AA}^{(2)} S_A \left[6 - f_2(\vec{k}) \right] \\ h_{BB}^{(2)}(\vec{k}) &= \left\{ J_{BBs}^{(2)} \left[6 - f_2(\vec{k}) \right] + 6J_{BBd}^{(2)} \right\} S_B \\ h_{12}^{(2)}(\vec{k}) &= -S_B J_{BBd}^{(2)} f_2(\vec{k}) \end{aligned} \quad (14)$$

where

$$f_2(\vec{k}) = 2[(\cos(\phi_1) + \cos(\phi_2) + \cos(\phi_1 - \phi_2))]. \quad (15)$$

We thus see that the non-bonding band dispersion, given by eq. (11), now reads,

$$E_{\text{non-bonding}} = 3J_{AB}^{(1)} S_A + J_{BB}^{(1)} S_B +$$

$$\begin{aligned} &+ \left\{ J_{BBs}^{(2)} \left[6 - f_2(\vec{k}) \right] + 6J_{BBd}^{(2)} \right\} S_B + S_B J_{BBd}^{(2)} f_2(\vec{k}) = \\ &3J_{AB}^{(1)} S_A + J_{BB}^{(1)} S_B + \\ &+ \left[6(J_{BBd}^{(2)} + J_{BBs}^{(2)}) + (J_{BBd}^{(2)} - J_{BBs}^{(2)}) f_2(\vec{k}) \right] S_B. \end{aligned} \quad (16)$$

We note that the opposing sign in the amplitude of the dispersive part of the of the non-bonding part arises from the antisymmetric nature of this mode.

Given that $f_2(\Gamma) = 6$ and $f_2(K) = -3$, the fact that the maximum of the non-bonding band is at the Γ point indicates that $J_{BBd}^{(2)} - J_{BBs}^{(2)}$ is a positive number. Since we expect that, in absolute value $J_{BBd}^{(2)}$ is smaller than $J_{BBs}^{(2)}$, on account of the larger distance, this probably entails that second neighbour interactions are AF. For the sake of simplicity we assume $J_{BBd}^{(2)} = 0$ and we infer $J_{BBs}^{(2)} = -\frac{W}{9S_B}$, where W is the bandwidth of the non-bonding band in the $\Gamma - K$ direction and $9 = f_2(\Gamma) - f_2(K)$. Since $W \simeq 50$ meV, we estimate $J_{BBs}^{(2)} = -4.4$ meV.

The second neighbour exchange also affects the bandwidth of the acoustic magnon band as well as the center of mass position of the non-bonding band. As a result, we need to readjust the values of the first neighbour exchange interactions in the model to obtain magnon bands in agreement with those computed with the itinerant fermion picture.

Magnon wave functions

In figure 3 we plot the projection of the magnon wave functions for the three bands over the two modes that are mixed by the magnon Hamiltonian (10), namely, the A site and the symmetric B mode. The results of the figure are computed including the nnn exchange, with the same parameters than the lower panels of Fig. 2. We see that the acoustic band is predominantly located on the B sublattice. The non-bonding band has 100% of the weight on the anti-symmetric bond. The $A - B$ mixing on the acoustic and optical magnons is minimal at the Dirac points, as expected in a honeycomb lattice.

Magnons with DM and anisotropy

We now consider the two terms in the Hamiltonian, anti-symmetric DM exchange and anisotropy, that can only be present in the spin Hamiltonian when spin-orbit interactions are present in the parent fermion Hamiltonian.

For the DM term we make the following assumptions. The first neighbor DM occurs only for the AB dimers, and the AB_1 and AB_2 couplings are the same. We assume the second neighbor coupling is active only in the B sublattice, is diagonal in the B subspace, and identical for

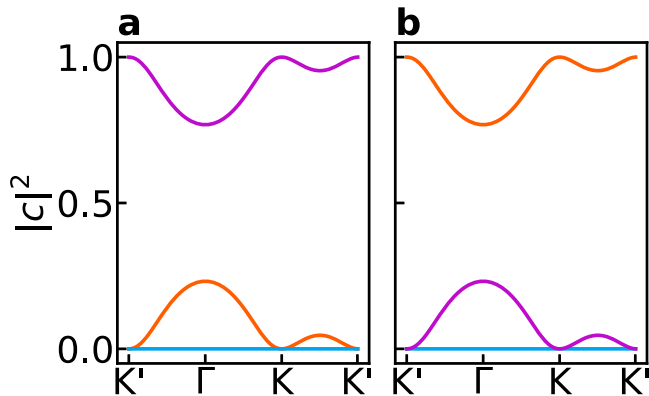


FIG. 3: Projection of the magnon wave functions over the A site (left) and the symmetric mode of the B sublattice (right).

B_1 and B_2 .

$$\mathcal{H}_{a,a'}^{DM}(\vec{k}) \equiv \begin{pmatrix} 0 & d_1(\vec{k}) & d_1(\vec{k}) \\ d_1^*(\vec{k}) & d_2(\vec{k}) & 0 \\ d_1^*(\vec{k}) & 0 & d_2(\vec{k}) \end{pmatrix}, \quad (17)$$

where $d_1(\vec{k}) = i\sqrt{S_A S_B}[f(\vec{k}) - 1]$ and $d_2(\vec{k}) = 2D_2 S_B [\sin(\vec{k} \cdot \vec{a}_1) + \sin(\vec{k} \cdot \vec{a}_2) + \sin(\vec{k} \cdot (\vec{a}_1 - \vec{a}_2))]$

Both d_1 and d_2 are odd functions of the momentum, $d_1(\vec{k}) = -(d_1(-\vec{k}))^*$, and $d_2(\vec{k}) = -d_2(-\vec{k})$. However, they enter in the energy dispersion in different manners. The nn DM only gives a non-reciprocal dispersion along the $\Gamma - M$ direction for the acoustic band, leaving the non-bonding band unaffected. In contrast, the nnn DM gives a non-reciprocal dispersion along the $K - \Gamma, K'$ direction, and affects both the acoustic and the non-bonding bands. The nnn DM coupling enters linearly in the dispersion of the non-bonding band. As a result, we can use the valley splitting of the non-bonding band $\delta = E_{\text{non-bonding}}(K) - E_{\text{non-bonding}}(K') = -4\sqrt{3}D_2 S_B$. Since the valley splitting is $\delta = 45$ meV, we infer $D_2 = \frac{\delta}{4\sqrt{3}S_B} \simeq -5$ meV.

This model accounts for the non-reciprocal dispersion along the K, K' direction and the reciprocal dispersion along the $\Gamma - M$ direction. However, it also shifts laterally the low energy magnon dispersion, in disagreement with the itinerant fermion results. This matter deserves further exploration. We also note that the linear contribution makes the energy of the acoustic band negative in a small range of momenta. This could be easily fixed by adding the single ion anisotropy terms.

MAGNON WAVE FUNCTIONS FROM THE FERMIONIC METHOD

Some physical insight about the character of the magnon modes can be obtained directly from the results of the fermionic approach. The magnon wave func-

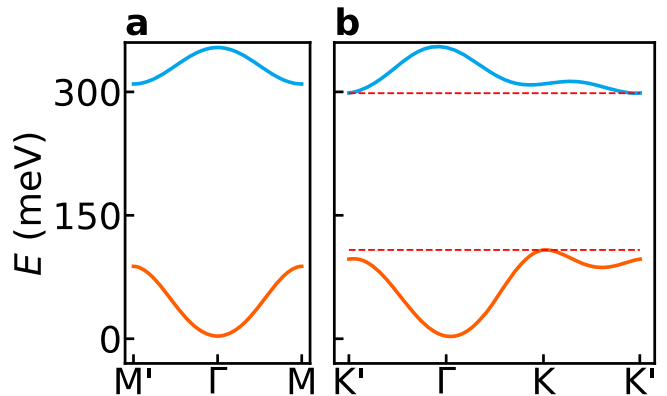


FIG. 4: Complete energy dispersion, including 1st and 2nd neighbour exchange and second neighbour DM interaction. We take $J_{AB}^{(1)} \simeq 65$ meV, $J_{BB}^{(1)} \simeq 80$, $J_{BBs}^{(2)} = -4.4$ meV, $J_{BBd}^{(2)} = J_{AAd}^{(2)} = 0$ and $D_2 = -5$ meV. In panel b we draw red horizontal lines to emphasize the non-reciprocity.

tions can be obtained from the transverse spin susceptibility matrix $\chi_{aa'}^\perp(\omega, \vec{k})$. This object can be interpreted as the single-particle Green function matrix associated with the magnons. As such, it shares eigenvectors with the magnon Hamiltonian (to which we do not have direct access within the fermionic model). Thus, by diagonalizing $\chi_{aa'}^\perp(\omega, \vec{k})$ at an (ω, \vec{k}) pair where its imaginary part has a peak, we obtain the eigenvectors associated with the magnon modes [2, 3]. Of course, in metallic magnets are not true eigenstates of the system's hamiltonian, since they hybridize with the Stoner excitations. Nevertheless, the eigenvectors of $\chi_{aa'}^\perp(\omega, \vec{k})$ still provide useful information, as long as the magnon feature in the spectral density can be clearly distinguished from the background of Stoner modes.

As we have shown in the paper, Fe_3GeTe_2 has only two well-defined magnon modes. By looking at the dispersion of the lowest energy mode (fig. 3b of the main paper), it is clear that it can be identified with the acoustic mode of the spin model of the previous section. Analysis of its wave function coefficients, shown in fig. 5 confirms that identification.

The character of the higher-energy mode is less evident from the dispersion relations alone, since the spin model predicts two modes with energies higher than that of the acoustic magnons. Comparison of the wave function coefficients obtained in the fermionic method with those obtained from the spin model indicate that this mode corresponds to the second mode predicted by the spin model. This conclusion is supported not only by the fact that $|c_A|^2 = 0$ along the whole $\Gamma - K$ line, as seen in figs. 3 and 5, but also by the relative phase of the c_{B_1} and c_{B_2} coefficients, that both methods indicate to be π , independent of the magnon wave vector.

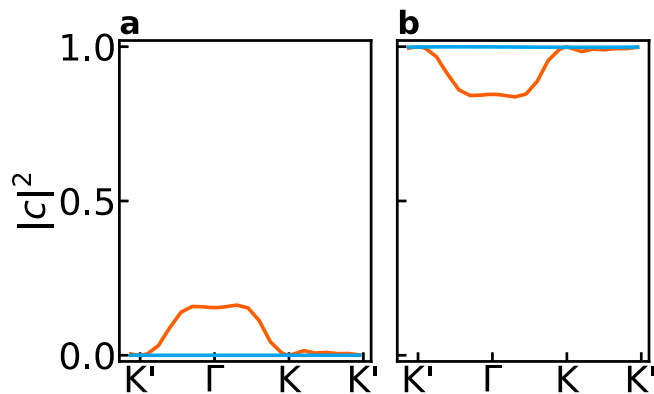


FIG. 5: Projections of the magnon wave functions on the A site (a) and on the symmetric and antisymmetric modes of the B sublattice (b) along the $\Gamma - K$ line, extracted directly from the fermionic method. The orange lines correspond to the projections of the acoustic mode on A and on the *symmetric* B mode. The blue lines are the projections of the non-bonding mode on A and on the *antisymmetric* B mode. As discussed in the text, the third mode, predicted by the spin model, does not have a clearly identifiable signature in the magnon spectral density derived from the fermionic approach, due to strong hybridization with the Stoner continuum.

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