Damped Topological Magnons in the Kagome-Lattice Ferromagnets

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Damped Topological Magnons in the Kagomé-Lattice Ferromagnets

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We demonstrate that interactions can substantially undermine the free-particle description of magnons in ferromagnets on geometrically frustrated lattices. The anharmonic coupling, facilitated by the Dzyaloshinsky-Moria interaction, and a highly-degenerate two-magnon continuum yield a strong, non-perturbative damping of the high-energy magnon modes. We provide a detailed account of the effect for the $S=1/2$ ferromagnet on the kagomé lattice and propose further experiments.

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Theoretical proposals and experimental discoveries of electronic topological materials having bulk bands with nonzero topological invariants and protected edge states [1, 2] have lead to an active search for similar effects in systems with different quasiparticles [3–5]. Among the latter are magnon excitations in ferromagnets on frustrated lattices, with several materials identified, synthesized, and studied since the original proposal [5–9].

Simple Heisenberg ferromagnets have a classical, fully polarized ground state and their excitation spectra are affected by quantum effects only at a finite temperature [10], regardless of the underlying lattice. However, the lower symmetries of the geometrically-frustrated lattices, such as kagomé and pyrochlore, allow for a rather significant Dzyaloshinskii-Moriya (DM) interaction [8, 9]. While in their simplest form, the DM terms are frustrated, leaving the fully saturated ferromagnetic ground state intact, such a protection does not hold for the excited states. Instead, the DM interaction generates complex hopping amplitudes for the spin flips that translate into fluxes of fictitious fields, see Fig. 1(a), leading to Berry curvature of magnon bands. Among the consequences of this band transformation are unusual transport phenomena such as magnon Hall and spin Nernst effects [5–7, 11–14].

On closer inspection, the sought-after nontrivial topological character of magnon bands is intimately tied to several aspects of the underlying structures. In particular, their non-Bravias lattices necessarily host optical magnon branches, while the geometrically-frustrating lattice topology favors underconstrained couplings that result in the "flat" excitation branches featuring degeneracy points with the dispersive magnon bands, see Fig. 1(b). This degeneracy is lifted by the DM interaction, giving rise to the Berry curvature of the bands, which is responsible for nontrivial transport properties.

It has also been suggested that, in a minimal model, the topology of the bands can be "tuned" by manipulating the direction of magnetization [9, 14]. Using a small field to change the mutual orientation of magnetization $\mathbf{M}$ and DM vector $\mathbf{D}$ from $\mathbf{M} \parallel \mathbf{D}$ to $\mathbf{M} \perp \mathbf{D}$, one formally turns the DM-induced complex hoppings and the concomitant topological effects from "on" to "off" [9].

We point out that in all these constructions, an idealized, non-interacting free-boson description of magnons is simply taken for granted [11, 12, 15]. Below we demonstrate that such a free-quasiparticle picture of magnons in ferromagnets on the geometrically-frustrated lattices is missing a crucial physical effect, which, in turn, challenges conclusions reached within the idealized picture.

The key idea is that, for $\mathbf{M} \nparallel \mathbf{D}$, the DM interaction is also a source of the anharmonic, particle-non-conserving coupling of magnons. The coupling is hidden for the ground state, but not for excitations, similarly to the complex hopping effect. Its most important outcome is a significant, non-perturbative damping of the flat and dispersive optical modes in the proximity of their degeneracy point, the effect precipitated by the divergent density of states in the two-magnon continuum. The resultant broadening at $\mathbf{k} \to 0$ is proportional to the first power of the DM term, $\Gamma \propto |\mathbf{D}|$, same as the band-splitting effect for $\mathbf{M} \parallel \mathbf{D}$. Interestingly, a sizable broadening has been noted as an unexpected result in a recent study of the kagomé-lattice ferromagnet, Cu(1-3,bdc), see Ref. [9].

Model and magnon interaction.—The nearest-neighbor model of a ferromagnet with the DM term is

$$\hat{\mathcal{H}} = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j) , \qquad (1)$$

where $J>0$, $\langle ij \rangle$ runs over bonds of the kagomé lattice, and Fig. 1(a) shows the order of i and j in the DM term, see [16]. While the DM interaction in the kagomé lat-

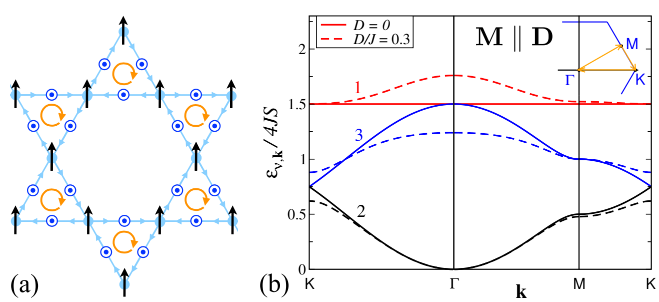

FIG. 1: (a) A ground state of (1) with $\mathbf{D}_{ij} = D\hat{\mathbf{z}}$; arrows on bonds show ordering of $\mathbf{S}_i$ and $\mathbf{S}_j$ in the DM term with fictitious fluxes indicated. (b) Magnon bands along the KΓMK path for $D=0$ (solid) and for $D/J=0.3$ with $\mathbf{M} \parallel \mathbf{D}$ (dashed).

tice can have both in- and out-of-plane components, the latter is dominant [17, 18]. In the following, we consider Hamiltonian (1) with $\mathbf{D}_{ij}=D\hat{\mathbf{z}}$ as a minimal model that illustrates a dramatic effect of magnon interactions.

Usually, the out-of-plane DM coupling would favor a canted in-plane order of spins with reduced magnetic moment due to quantum fluctuations in the ground state [19]. However, for ferromagnets on the geometrically-frustrated lattices it is the DM term that is frustrated. Thus, counterintuitively, magnetization remains fully saturated, $|\mathbf{M}|=SN$, regardless of its orientation with respect to $\mathbf{D}$. This is because the mean-field tug of the DM interactions on a given spin from its neighbors vanishes identically due to its cancellation from different bonds, see Fig. 1(a). For the same reason, the DM term cannot generate fluctuations in the saturated ground state. One can immediately see that the same is not true for magnon excitations, because spin flips violate cancellation of the DM contributions from different bonds. Therefore, while the ground state is insensitive to the DM interaction, the spectrum is not.

For the uniform out-of-plane $\mathbf{D}$, there are two principal directions for magnetization: $\mathbf{M}\parallel\mathbf{D}$ and $\mathbf{M}\perp\mathbf{D}$. The former case has been throughly examined within the linear spin-wave theory (LSWT) [5–9, 11–14] and we summarize it here briefly. Choosing the spin-quantization axis $\hat{\mathbf{z}}\parallel\mathbf{M}\parallel\mathbf{D}$ one can straightforwardly rewrite (1) as

$$\hat{\mathcal{H}}=-J\sum_{\langle ij\rangle}S^z_iS^z_j-\frac{1}{2}\sum_{\langle ij\rangle}\left(\mathcal{J}\,S^+_iS^-_j+\mathcal{J}^*\,S^-_iS^+_j\right), \quad (2)$$

where $\mathcal{J}=J-iD$ and the DM term provides imaginary component to the spin-flip hoppings. Taking into account lattice geometry, rewriting spin flips as bosons, and diagonalizing the corresponding 3×3 matrix for the kagomé unit cell yields the harmonic-order, LSWT Hamiltonian

$$\hat{\mathcal{H}}^{(2)}=\sum_{\nu,\mathbf{k}}\varepsilon_{\nu,\mathbf{k}}b^\dagger_{\nu,\mathbf{k}}b_{\nu,\mathbf{k}}\,, \quad (3)$$

where the three magnon branches, $\varepsilon_{\nu,\mathbf{k}}$, are depicted in Fig. 1(b) for a representative value of D, see [16] for details. The main outcomes of the DM term are the gaps at the degeneracy points of the DM-free model, $\Delta\propto|\mathbf{D}|$, and the Berry curvature of the bands due to fictitious fields generated by complex hoppings [5, 12, 13]. It is clear that this procedure can be generalized to an *arbitrary* angle θ between $\mathbf{M}$ and $\mathbf{D}$ by simply replacing $D\rightarrow D\cos\theta$ in $\mathcal{J}$ above. This immediately implies that for $\mathbf{M}\perp\mathbf{D}$ the complex hoppings cease completely and magnon bands should become free of the DM interaction, i.e., equivalent to the $D=0$ picture in Fig. 1(b).

A flaw in this reasoning is in the harmonic approximation. Although for $\mathbf{M}\perp\mathbf{D}$ the DM interaction does not contribute to the LSWT, it *does not* disappear. For the quantization axis $\hat{\mathbf{z}}\parallel\mathbf{M}\perp\mathbf{D}$, the DM term becomes

$$\hat{\mathcal{H}}_{\rm DM}=\frac{D}{2}\sum_{\langle ij\rangle}\left[\left(S^+_i+S^-_i\right)S^z_j-S^z_i\left(S^+_j+S^-_j\right)\right], \quad (4)$$

which indeed does not affect the ground state or harmonic theory. However, it gives rise to anharmonic interaction of magnons [20] as it creates/annihilates a spin-flip in a proximity of another spin flip, with contributions from the nearest bonds not canceling out. Thus, transitions are generated between single- and two-magnon states, which can lead to renormalization of the bands and, most importantly, to magnon damping.

With the formal details given in [16], the resultant cubic interaction of magnons obtained from (4) is [21]

$$\hat{\mathcal{H}}^{(3)}_{DM}=\frac{D}{2!}\sqrt{\frac{2S}{N}}\sum_{\mathbf{k},\mathbf{q}}\sum_{\nu\mu\eta}\widetilde{\Phi}^{\nu\mu\eta}_{\mathbf{qk;p}}\,b^\dagger_{\nu,\mathbf{q}}b^\dagger_{\mu,\mathbf{k}}b_{\eta,\mathbf{p}}+\text{H.c.}, \quad (5)$$

with the vertex $\widetilde{\Phi}^{\nu\mu\eta}_{\mathbf{qk;p}}=F^{\nu\mu\eta}_{\mathbf{qkp}}+F^{\mu\nu\eta}_{\mathbf{kqp}}$ and the amplitude

$$F^{\nu\mu\eta}_{\mathbf{qkp}}=\sum_{\alpha\beta}\epsilon^{\alpha\beta\gamma}\cos(q_{\beta\alpha})\,w_{\nu,\alpha}(\mathbf{q})w_{\mu,\beta}(\mathbf{k})w_{\eta,\beta}(\mathbf{p})\,, \quad (6)$$

where $\mathbf{w}_\nu=(w_{\nu,1},w_{\nu,2},w_{\nu,3})$ are the eigenvectors of the 3×3 matrix diagonalized for the harmonic theory. A generalization of this consideration to an arbitrary $\mathbf{M}$–$\mathbf{D}$ angle is achieved by $D\rightarrow D\sin\theta$ in (4) and (5), also keeping in mind that the eigenvectors $\mathbf{w}_\nu$ in (6) change with $D\cos\theta$ according to the diagonalization leading to (3). Thus, the harmonic and the anharmonic Hamiltonians (3) and (5) complement each other for any $\theta\neq0$.

We note that at $T=0$, the four-magnon terms do not directly affect the spectrum of the model (1) as they necessarily have a $b^\dagger b^\dagger b\,b$ form [16, 22].

Kinematics and two-magnon DoS.—Because the anharmonic term (5) provides a coupling of the single-particle branches with the two-magnon continuum, the properties of the latter are of interest. Consider $\mathbf{M}\perp\mathbf{D}$. From the point of view of the harmonic theory, magnon bands are not affected by the DM term, see Fig. 2, with the flat band (mode 1) degenerate with the dispersive band (mode 3) at the Γ point. Crucially, the two-magnon continuum is highly degenerate at this point because of a ubiquitous property of the magnon spectra of ferromagnets on the non-Bravais lattices. Namely, the two dispersive modes are mirror reflections of each other with

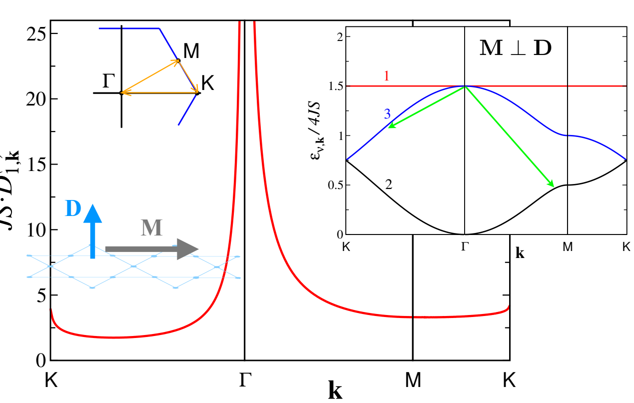

FIG. 2: The LSWT two-magnon DoS (7) for mode 1, $\mathbf{M}\perp\mathbf{D}$. Inset: schematics of a magnon decay from $\mathbf{k}=0$.

respect to their energy at the K-point, which is also precisely one half of the flat mode energy, see Fig. 2. One can easily check that the same structure persists for the pyrochlore and honeycomb lattices [8, 23, 24]. Because of that property, the condition $\varepsilon_1 = \varepsilon_{2,\mathbf{q}} + \varepsilon_{3,-\mathbf{q}}$ is met for *any* value of the momentum $\mathbf{q}$ [24]. This is a much higher degeneracy than the ones leading to more traditional van Hove singularities of the two-magnon continua [20].

A useful quantitative characteristics of the continuum is the on-shell, $\omega = \varepsilon_{\mu,\mathbf{k}}$, two-magnon density-of-states (DoS), which is also a proxy of the on-shell decay rate

$$D^{(2)}_{\mu,\mathbf{k}} = \pi \sum_{\mathbf{q},\nu\eta} \delta\left(\varepsilon_{\mu,\mathbf{k}} - \varepsilon_{\nu,\mathbf{q}} - \varepsilon_{\eta,\mathbf{k}-\mathbf{q}}\right), \qquad (7)$$

shown in Fig. 2 for the flat mode, $\mu = 1$, vs $\mathbf{k}$. It exhibits a strong $1/|\mathbf{k}|$ divergence at $\mathbf{k}\to 0$ due to the high degeneracy in the two-magnon continuum discussed above. The divergent behavior at $\mathbf{k}\to 0$ is identical for $\mu = 3$ [16].

This consideration implies that an *arbitrary weak* coupling of the single-magnon and two-magnon states completely invalidates predictions of the harmonic theory by causing a divergent damping in the optical magnons at $\mathbf{k}\to 0$. As is shown below, a self-consistent treatment regularizes this divergence, but leaves an anomalously large, non-analytic and non-perturbative damping, $\Gamma \propto |\mathbf{D}|$, for both optical magnon modes near the Γ-point and in a broad range of $|\mathbf{k}| \lesssim D/J$ also controlled by D.

Decays and regularization.—One can expect the on-shell decay rate of a magnon due to cubic terms (5)

$$\Gamma_{\mu,\mathbf{k}} = \frac{\pi S D^2}{N} \sum_{\mathbf{q},\nu\eta} \left|\widetilde{\Phi}^{\nu\eta\mu}_{\mathbf{q},\mathbf{k}-\mathbf{q};\mathbf{k}}\right|^2 \delta(\varepsilon_{\mu,\mathbf{k}} - \varepsilon_{\nu,\mathbf{q}} - \varepsilon_{\eta,\mathbf{k}-\mathbf{q}}), \qquad (8)$$

to be small for realistic parameters as it is $\propto D^2/J$. This is indeed the case for the Goldstone branch (mode 2), for which damping is also suppressed kinematically except for large momenta [16]. However, because of the degeneracy of the two-magnon DoS, damping (8) of the mode 3 is divergent as $1/|\mathbf{k}|$, thus suggesting a much stronger effect. The situation is less conspicuous for the flat mode, as the expected similar divergence in (8) is preempted by a subtle cancellation in the vertex, leading to a finite, $O(D^2)$, damping at $\mathbf{k}\to 0$ [16]. However, this cancellation is lifted in the off-shell consideration, which, counterintuitively, leads to a strongly enhanced decay rate of the flat mode in the self-consistent treatment. We note that the real part of the same self-energy [25] also diverges for both optical magnon modes, but its divergence is much weaker [16], $\mathrm{Re}\,\Sigma_{\mu,\mathbf{k}} \propto \ln|\mathbf{k}|$.

A regularization of the divergencies is achieved via a self-consistent solution of the Dyson's equation (DE), which naturally accounts for the damping of the initial-state magnon, $\omega - \varepsilon_{\mu,\mathbf{k}} - \Sigma_{\mu,\mathbf{k}}(\omega^*) = 0$, where $\Sigma_{\mu,\mathbf{k}}(\omega)$ is the self-energy due to cubic terms and the complex conjugate ω^* respects causality, see [26]. The real and imaginary parts of this equation have to be solved together. However, once the initial-state damping is introduced, the weak divergence in the real part will be cut [26]. Therefore, for small $d = D/J$, it will constitute a small energy correction, $\propto d^2 \ln|d|$, neglecting which yields an "imaginary-only" Dyson's equation, which we coin as iDE: $\Gamma_{\mu,\mathbf{k}} = -\mathrm{Im}\,\Sigma_{\mu,\mathbf{k}}(\varepsilon_{\mu,\mathbf{k}} + i\Gamma_{\mu,\mathbf{k}})$, or, explicitly

$$1 = \frac{SD^2}{N} \sum_{\mathbf{q},\nu\eta} \frac{\left|\widetilde{\Phi}^{\nu\eta\mu}_{\mathbf{q},\mathbf{k}-\mathbf{q};\mathbf{k}}\right|^2}{\left(\varepsilon_{\mu,\mathbf{k}} - \varepsilon_{\nu,\mathbf{q}} - \varepsilon_{\eta,\mathbf{k}-\mathbf{q}}\right)^2 + \Gamma^2_{\mu,\mathbf{k}}}. \qquad (9)$$

With the numerical results for the iDE to follow, its key result can be appreciated. At small $|\mathbf{k}|$, the difference of magnon energies in (9) for the divergent decay channels $\mu \to \{2,3\}$ is negligible, giving: $\Gamma_{\mu,\mathbf{k}\to 0} \approx |D|\sqrt{S}$. Physically, the "fuzziness" of the initial-state magnon removes strict energy-momentum conservations in the decay process, regularizing the divergencies.

This constitutes the main result of the iDE regularization. The decay rate of both flat and gapped modes for $\mathbf{M}\perp\mathbf{D}$ at $\mathbf{k}\to 0$ is given by a non-perturbative answer, $\Gamma_{1(3),\mathbf{k}} \propto |D|$, strongly enhanced compared to the perturbative expectations. The $\mathbf{k}$-region in which the broadening is strongly enhanced can be easily estimated as $|\mathbf{k}| \lesssim |\mathbf{k}^*| \propto |D|/J$ with the damping decreasing to the perturbative values, $\Gamma_{3(1),\mathbf{k}} \propto D^2/J$, for $|\mathbf{k}| \gtrsim |\mathbf{k}^*|$.

The numerical solutions of the iDE (9) for damping $\Gamma_{\mathbf{k}}$ for all three magnon modes for $S = 1/2$ and $D/J = 0.3$ are shown in Fig. 3 along the KΓMK path. One can see that, indeed, the damping is strongly enhanced in the $|\mathbf{k}| \lesssim |D|/J$ region around the Γ point for the flat and dispersive optical modes, see also [16] for other values of D/J. The inset shows the full width of magnon spectral lines at half-maximum, $\varepsilon_{\mathbf{k}} \pm \Gamma_{\mathbf{k}}$, to demonstrate effects of the broadening on the magnon spectrum. One can also see that the decay rates of modes 1 and 3 at $\mathbf{k} = 0$ coincide because of the symmetry of the the cubic vertices [16]. Some remnants of the more conventional, logarithmic van Hove singularities [20] can be seen in both Figs. 3 and 4.

Angular dependence.—Since magnetization is not pinned for model (1), one can manipulate its direction. Then, the natural question is: how does one transition from the well-defined excitations with the gap $\propto D$ to the broadened excitations with the widths $\propto D$ as a function of the $\mathbf{M}$–$\mathbf{D}$ angle θ?

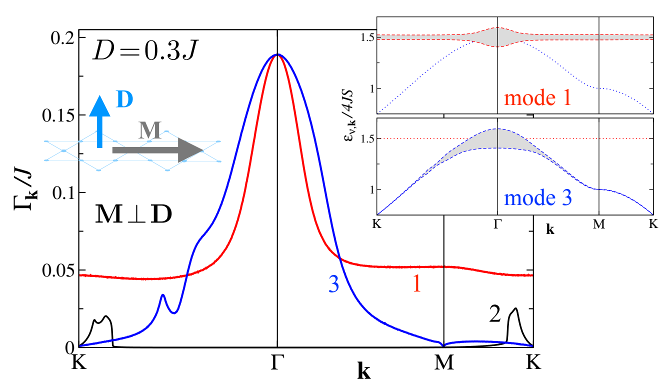

FIG. 3: Solutions of the iDE (9) for $D/J = 0.3$, $S = 1/2$ along the KΓMK path. Inset: the FWHM of spectral lines, $\varepsilon_{\mathbf{k}} \pm \Gamma_{\mathbf{k}}$.

For $\theta < \pi/2$, magnetization is partially along $\mathbf{D}$ and magnon bands split due to complex hoppings ($\sim\cos\theta$), while cubic interaction in (5) is reduced ($\sim\sin\theta$) as described above. The main complication is that, for $\mathbf{M}\not\parallel\mathbf{D}$, the eigenvectors in the vertices (6), $\mathbf{w}_\nu$, are not derivable analytically in a compact form [27], and have to be obtained numerically from diagonalization of the 3×3 matrix [16]. Physically, the band splitting also contributes to regularization of singularities in magnon decays.

In Fig. 4, we provide detailed predictions for the angular dependence of the damping of the optical magnon modes obtained from iDE (9). Fig. 4(a) shows a gradual decrease of the broadening for both modes at $\mathbf{k}=0$ from its maximal value to zero upon the decrease of the angle θ, with the insets showing $\Gamma_{\mu,\mathbf{k}}$ along the KΓM path for several values of the angle. Fig. 4(b) panels present the 2D intensity plots of the broadening of the mode 3 in $\mathbf{k}$-space for three different angles. These results complement the data in Figs. 3 and 4(a) and demonstrate a rather dramatic distribution of the broadening in the Brillouin zone and its nontrivial evolution with the angle. This detailed picture is completed in Fig. 4(c) by the $\mathbf{k}-\theta$ intensity maps of the broadening for both optical modes along the KΓM path. They reveal an interesting contribution of the conventional van Hove singularities of the two-magnon continuum and highlight an unusual evolution of the magnon linewidth.

Our minimal-model consideration may seem to imply that there is always a special direction of $\mathbf{M}$ that can allow one to switch off cubic anharmonic coupling and associated decay effects. However, in a more general and realistic setting, the DM term has both in- and out-of-plane components [8, 9], making magnon decays inevitable. It is, thus, imperative to take their effects into account in a consideration of magnon bands in real materials.

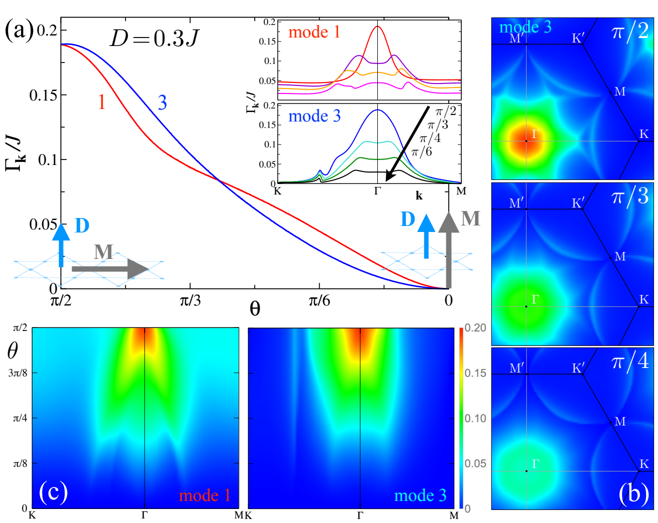

FIG. 4: (a) $\Gamma_{1(3),\mathbf{k}=0}$ vs θ. Insets: $\Gamma_{\mu,\mathbf{k}}$ along the KΓM path for several θ. (b) The 2D intensity distributions of $\Gamma_{3,\mathbf{k}}$ in $\mathbf{k}$-space for $\pi/2$, $\pi/3$, and $\pi/4$. (c) The $\mathbf{k}-\theta$ intensity maps of $\Gamma_{1(3),\mathbf{k}}$ along the KΓM path. $D/J=0.3$, $S=1/2$.

Experiments.—Experimental evidence of the broadening of the flat mode in the vicinity of $\mathbf{k}=0$ has been recently reported for a kagome-lattice ferromagnet with $D_z/J\approx0.15$ [9]. For $\mathbf{M}\perp\mathbf{D}$, the broadening varying from $0.05J$ in external field to $0.13J$ in zero field was suggested, see Supplemental material of [9]. Our consideration yields the broadening of both optical modes of a somewhat lesser value of $0.09J$ in zero field [16]. One can suggest that a larger broadening can be registered due to the overlap of the two modes. Other experimental factors that can affect a direct comparison include averaging of the data over a range of $\mathbf{k}$ and contributions of the in-plane DM components to decays. The close agreement with the available data and our detailed predictions above call for a closer experimental analysis of the suggested dramatic broadening effects. They can be tested by the neutron-scattering, resonant neutron-scattering spin-echo, and by ESR.

Summary.—We demonstrated that the idea of non-interacting topologically nontrivial bands, familiar from fermionic systems, cannot be trivially transplanted to bosonic systems such as ferromagnets on the geometrically frustrated lattices. The key difference is in the particle-non-conserving terms that are generated by the same interactions that are necessary for the sought-after Berry curvature of the bands. These terms, combined with a ubiquitous degeneracy of the two-magnon continuum, produce a substantial broadening of magnon bands precisely in the ranges of $\mathbf{k}$ and ω that are essential for the topological properties to occur, thus potentially undermining the entire free-band consideration. Same phenomena should be common to ultracold atomic, phonon-like, and other bosonic systems. How the topologically-nontrivial properties of the bands can be defined in the presence of a substantial broadening remains an open question.

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[24] This is due to tight-binding form of magnon bands and the traceless form of the nearest-neighbor hopping matrix that connects only different sublattices, which leads to $\sum_\nu \varepsilon_{\nu,\mathbf{k}} = const$, see [16].

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