Abstract

Dispersionless ‘zero energy mode’ is one of the hallmarks of frustrated kagomé antiferromagnets (KAFMs). It points to extensive classically degenerate ground-states. The ‘zero energy mode’ can be observed experimentally when lifted to a flat mode at finite energy by a strong intrinsic magnetic anisotropy. In this letter, we study the effects of irradiation of laser light on the KAFMs. We adopt the magnon picture without loss of generality. It is shown that circularly or linearly polarized light lifts the ‘zero energy mode’, stabilizes magnetic order, and induces energy gaps in the KAFMs. We find that the circularly polarized light-induced anisotropies have similar features as the intrinsic in-plane and out-of-plane Dzyaloshinskii–Moriya interaction in KAFMs. The former stabilizes long-range magnetic order and the latter induces spin canting out-of-plane with nonzero scalar spin chirality. The Floquet thermal Hall effect shows that the synthetic magnetic excitation modes in the case of circularly polarized light are topological, whereas those of linearly polarized light are not.

Geometrically frustrated KAFMs have been an intensively active field of research due to their exotic properties such as the possibility of quantum spin liquids (QSLs) [1–3]—states where spin frustration forbids long-range magnetic order down to the lowest temperatures. Classically, the ideal Heisenberg KAFMs have an extensive ground-state degeneracy [4, 5], resulting in a ‘zero energy mode’ [6]—a signature that no classical ground-state configuration is favoured. However, order–by–disorder phenomenon [4, 5] is believed to lift the extensive classically degenerate ground-states and selects a specific magnetic order, usually the $q = 0$ spin configuration in which three spins on the triangular plaquette of the kagomé lattice are oriented at $120^\circ$ apart. Another possibility of lifting the extensive classically degenerate ground-states is by adding further neighbouring interactions [5]. Moreover, the geometry of the kagomé lattice lacks an inversion centre and by the Moriya’s rules a Dzyaloshinskii–Moriya interaction (DMI) [7, 8] is allowed. The out-of-plane DMI is capable of inducing long-range magnetic order in the frustrated KAFMs [9]. Hence, the ‘zero energy mode’ can be observed experimentally as a flat mode with finite energy [10].

In realistic quantum kagomé materials, however, further neighbouring interactions can be perturbatively small and negligible, therefore the out-of-plane DMI is usually the dominant anisotropy in real materials. Nevertheless, magnetic order in quantum spin-1/2 KAFMs appears beyond a certain quantum critical point (QCP) of the out-of-plane DMI ($D_z/f \approx 0.1$) [11]. Below the QCP it is believed that QSL persists. For instance, herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$ has dominant out-of-plane DMI just below the QCP ($D_z/f \approx 0.08$) [12] and thus remains a QSL [13]. The kagomé calcium–chromium oxide Ca$_{10}$Cr$_7$O$_{28}$ has also been proposed as a QSL material and there is no evidence of a strong DMI [14]. In the current study, we shall consider an alternative source of inducing magnetic order in frustrated KAFMs. The approach will be based on irradiation of laser light on the magnetic insulators. This approach has attracted considerable attention as a possible mechanism to engineer synthetic topological systems [15–33].

In this letter, we formulate the theory of laser-irradiated KAFMs based on the Holstein–Primakoff magnon picture. However, the basic idea can also be extended to charge-neutral bosonic spinons in a similar fashion. The main results of this letter are as follows. First, we show that circularly or linearly polarized laser light is capable of lifting the ‘zero energy mode’ in the KAFMs, thereby inducing magnetic order. The associated magnetic...
excitation modes exhibit gaps at various points in the Brillouin zone. By inspection of measured spin waves in iron jarosites [10], we are able to conclude that the circularly polarized laser-induced symmetry breaking interactions possess distinctive features that are similar to the effects of intrinsic in-plane and out-of-plane DMI on kagomé magnetic insulators [9]. By studying the Floquet–Bloch thermal Hall effect close to thermal equilibrium, we further establish that the magnetic excitation modes in circularly polarized light are topological, whereas those of linearly polarized light are not. These results suggest that laser-irradiation can be considered as one of the possible ways to induce magnetic order and nontrivial topological magnetic excitation modes in KAFMs. Second, we apply the theory of laser-irradiation to KAFMs with an intrinsic out-of-plane DMI. The results show that radiation can also tune the out-of-plane DMI in KAFMs and induce a possible synthetic noncoplanar spin configuration with nonzero scalar spin chirality.

Let us consider the simple Hamiltonian for frustrated KAFMs, which is given by

$$\mathcal{H} = J \sum_\langle ij \rangle \mathbf{S}_i \cdot \mathbf{S}_j + \mathcal{H}_{\text{ani}},$$  

(1)

where $\mathbf{S}_i$ are the spin magnetic moments at the lattice sites $i$ located at $\mathbf{r}$, and $J > 0$ is an antiferromagnetic interaction between nearest-neighbour sites. Here, $\mathcal{H}_{\text{ani}}$ is a small perturbative anisotropy to the Heisenberg exchange term, which is dominated by the out-of-plane DMI given by $\mathcal{H}_{\text{ani}} = \sum_\langle ij \rangle \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j$, where $\mathbf{D}_{ij} = \pm D_z \mathbf{z}$ is an intrinsic out-of-plane DMI component due to inversion symmetry breaking on the kagomé lattice at the midpoint connecting two magnetic sites, and the $\pm$ sign alternates between the triangular plaquettes of the kagomé lattice as shown in figure 1.

In the absence of the out-of-plane DMI the classical ground states of the ideal Heisenberg KLAFM (i.e., first term in equation (1)) are the $\mathbf{q} = 0$ spin configurations shown in figure 1. However, they are infinitely degenerate. A direct application of linear spin wave approximation about this classical spin configurations leads to a’zero energy mode’ [6], which points to the fact that no particular ground-state spin configuration is favoured. Application of a static Zeeman magnetic field partially lifts the degeneracy but does not remove it entirely. In the quantum limit this would imply that the system is disordered [34]. In the later sections, we will consider the possibility of lifting the ‘zero energy mode’ and inducing magnetic order by laser-irradiation.

As previously shown the out-of-plane DMI induces and stabilizes the $\mathbf{q} = 0$ classical spin configurations [9]. The sign of the out-of-plane DMI determines which vector chirality of this long-range magnetic order is selected. In this letter, we consider the positive vector chirality $D_z > 0$ with the minus sign. This form of the out-of-plane DMI respects the symmetries of the kagomé lattice in figure 1. In particular, the combination of time-reversal symmetry (TRS) $T$ and mirror reflection symmetry $\mathcal{M}$ (i.e. $\mathcal{M} T$ or $\mathcal{M}_y T$), is a good symmetry of the coplanar $\mathbf{q} = 0$ spin configuration. Therefore, we expect the underlying magnetic excitations to be protected by this symmetry and there should be a possibility of Dirac point in the Brillouin zone. As we will show later irradiation by laser light will modify this spin structure.

The concept of laser-driven magnetic insulators rely on the magnetic dipole moments of the underlying magnetic excitations as recently introduced in quantum ferromagnets [36]. This is due to the fact that charge-neural bosonic quasi-particles do not interact with an electromagnetic field except through their magnetic dipole moment. Therefore, this concept applies to both magnons and spinons. We take the magnetic dipole moment to be along the in-plane ordering direction $\mu_r = -g \mu_B \mathbf{r}$, where $\mu_B$ is the Bohr magneton and $g$ is the spin $g$-factor. Now, we suppose that an in-plane laser light with dominant electric field components $E(t)$ is

Figure 1. (a) Schematic of the kagomé lattice with out-of-plane DMI (dotted and crossed circles) and a coplanar $\mathbf{q} = 0$ spin configuration (arrows). We also show the mirror reflection symmetry $\mathcal{M}_y$ about the $y$-axis and $\mathcal{M}_x$ about the $x$-axis. The primitive vectors are $\mathbf{a}_1 = (1, 0)$, $\mathbf{a}_2 = (1/2, \sqrt{3}/2)$, and $\mathbf{a}_3 = \mathbf{a}_2 - \mathbf{a}_1$. (b) First Brillouin zone of the kagomé lattice with indicated paths.
irradiated on the kagomé lattice. In the background of the time-varying electric field the hopping of charge-neutral bosonic quasi-particles will lead to a time-dependent Aharonov–Casher phase [37], which is given by

$$\theta_{ij}(t) = \frac{\hbar}{\epsilon} \int_{t_i}^{t_j} \mathbf{A}(t) \cdot d\mathbf{r},$$

(2)

where \( \mathbf{A}(t) = A_0 (\sin \omega t, \sin (\omega t + \phi), 0) \) is the vector potential with amplitude \( A_0 \) due to the electric field \( \mathbf{E}(t) = -\frac{\partial \mathbf{A}(t)}{\partial t} \). Note that circularly polarized laser light corresponds to \( \phi = \pi/2 \) and linearly polarization corresponds to \( \phi = 0 \) or \( \pi \). In the following we take the units \( \hbar = c = \frac{\gamma}{\mu_B} = 1 \).

Next, we perform the standard spin wave analysis of the coplanar/noncollinear spin configuration on the kagomé lattice [6, 35]. The basic procedure follows by rotating the coordinate axis about the \( z \)-axis by the spin orientation angles:

$$\mathcal{R}_z(\theta_i) = \begin{pmatrix} \cos \theta_i & -\sin \theta_i & 0 \\ \sin \theta_i & \cos \theta_i & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

(3)

where \( \theta_i = 0, \pm 2\pi/3 \). Hence the spin transforms as \( \mathbf{S}_i = \mathcal{R}_z(\theta_i) \mathbf{S}_i \). Next, we implement the Holstein–Primakoff transformation: \( S^+_i \to S - a_i^\dagger a_i, \ S^+_{-i} \to \sqrt{2S} a_i \) (where \( S^\pm = S^x \pm iS^y \)) and \( a_i^\dagger (a_i) \) are the bosonic creation (annihilation) operators. The corresponding time-dependent magnon tight-binding Hamiltonian is given by

$$\mathcal{H}(t) = JS \sum_{(ij)} [G^0_{ij}(a_i^\dagger a_j + a_j^\dagger a_i) + G^1_{ij}(a_i^\dagger a_j e^{i\theta_{ij}(t)} + \text{h.c.}) + G^2_{ij}(a_i^\dagger a_j^\dagger e^{i\theta_{ij}(t)} + \text{h.c.})],$$

(4)

where \( G^0_{ij} = (1 + D_i)/2; \ G^1_{ij} = (1 - D_i)/4; \ G^2_{ij} = (3 + D_i)/4 \) and \( D_i = \sqrt{3} \Delta_i/J \). Note that the hopping terms have now acquired a time-dependent phase by virtue of the Peierls substitution. In the momentum space we have

$$\mathcal{H}_k(t) = 2JS \left( \mathbf{G}^0 \cdot \mathbf{G}(t) \mathbf{G}^2(t) \right),$$

(5)

where \( \mathbf{G}^0 = (1 + D_i) \mathbf{I}_x + s, \mathbf{G}(t) = (1 - D_i) \mathbf{A}(t)/4, \) and \( \mathbf{G}^2(t) = (3 + D_i) \mathbf{A}(t)/4. \)

$$\mathbf{A}(t) = \begin{pmatrix} 0 & \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_1] & \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_2] \\ \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_1] & 0 & \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_3] \\ \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_2] & \cos[(\mathbf{k} + \mathbf{A}(t)) \cdot \mathbf{a}_3] & 0 \end{pmatrix}.$$  

(6)

Note that equations (4) and (5) have off-diagonal terms which do not exist in ferromagnets [36]. However, the general formalism of Floquet theory applies to any time-dependent tight-binding Hamiltonian.

The basic idea of Floquet theory is to transform a time-dependent Hamiltonian such as equation (4) or (5) into an effective static Hamiltonian. To proceed, we write the Floquet–Bloch wave function that obeys time-dependent Schrödinger equation as \( \psi_{\mathbf{k}, \alpha}(t) = e^{i\epsilon_{\mathbf{k}, \alpha}(t) t} \phi_{\mathbf{k}, \alpha}(t) \), where \( \phi_{\mathbf{k}, \alpha}(t) = \Phi_{\mathbf{k}, \alpha}(t + T) \) is a periodic function that denotes the Floquet–Bloch states for band \( \alpha \) with period \( T = 2\pi/\omega \) and \( \epsilon_{\mathbf{k}, \alpha}(t) \) is the quasi-energy. The periodic function can be expanded in Fourier space: \( \Phi_{\mathbf{k}, \alpha}(t) = \sum_{\omega m} e^{im\omega t} \Phi_{\mathbf{k}, \alpha}^m \). We define the Floquet Hamiltonian operator as \( \hat{\mathcal{H}}_k(t) = \mathcal{H}_k(t) - i\hbar \partial/\partial t. \) Then the states \( \Phi_{\mathbf{k}, \alpha}^m \) leads to a time-independent Floquet energy eigenvalue equation

$$\sum_{m} \left[ \mathcal{H}^\mathbf{G}^{m-m} + m \omega \delta_{m,0} \right] \Phi_{\mathbf{k}, \alpha}^m = \epsilon_{\mathbf{k}, \alpha} \Phi_{\mathbf{k}, \alpha}^m,$$

(7)

where \( \mathcal{H}^\mathbf{G} = \int_{0}^{T} dt e^{-i\omega t} \mathcal{H}_k(t). \)

The Floquet formalism is reliable in the high frequency limit \( \omega \gg J \). Therefore, we work in this limit and consider the truncation \( T = 0, \pm 1. \) At this juncture the Floquet Hamiltonian contains the zeroth order Bessel function \( J_0(\xi) \) and the first order Bessel function \( J_1(\xi) \), where \( \xi \) depends on the amplitude \( A_0 \) and phase \( \phi. \) However, the resulting Hamiltonian is a big matrix comprising numerous Floquet–Bloch side-bands.

Therefore, analytical analysis is unfeasible we therefore resort to numerical analysis. We first consider the limit of zero out-of-plane DMI \( (D_z/J = 0) \). In this case a ‘zero energy mode’ is inevitable [6]. In figure 2 we have shown the Floquet–Bloch magnon bands in the presence of linearly (a) and circularly (b) polarized light for \( D_z/J = 0 \) and \( A_0 = 1.5 \) along the Brillouin zone paths [38] in figure 1. In both cases, it is evident that the ‘zero energy mode’ is lifted to nearly flat mode at nonzero energy. We also observe that all the bands have a finite energy gap at the \( \Gamma \)-point as well as the \( \mathbf{K} \)-point.

It is believed that circularly polarized laser light breaks TRS, but it is very crucial to identify the form of the symmetry breaking interaction induced by the laser light. Interestingly, the magnon bands in figure 2 (b) for circularly polarized light exhibit all the important features reported in kagomé iron jarosites [10]. They include gapped magnetic excitations at various points in the Brillouin zone, which were theoretically identified as a
Whereas the latter which points inside the triangular plaquettes breaks mirror reflection symmetry and rotational invariance and it is responsible for spin canting out-of-plane and leads to a non-coplanar umbrella spin configuration with nonzero scalar spin chirality. The in-plane DMI is the primary source of the energy gaps at the $\Gamma$ and $K$ points. Therefore we conclude that irradiation by laser light can induce long-range magnetic order in highly frustrated magnets in the same way as the intrinsic DMIs [9]. As we will discuss in the following the laser-induced anisotropy in the case of linearly polarized light (figure 2(a)) does not break TRS.

Next, we consider the limit of nonzero out-of-plane DMI ($D_z/f = 0$). As shown in figure 2(c) the intrinsic out-of-plane DMI lifts the ‘zero energy mode’ in the undriven system and stabilizes the conventional coplanar spin configuration leading to lifted ‘zero energy mode’ as shown in figure 1. The gapped excitations at the $\Gamma$-point suggests that the induced synthetic anisotropy is an in-plane DM component which obviously breaks rotational invariance and modifies the conventional $q = 0$ spin configuration as previously mentioned.

Now, we would like to compute an experimentally observable quantity in DM models on the kagomé lattice. The gapped excitations at the $K$-point suggests that the Floquet–Bloch magnon bands will be topologically nontrivial with finite Berry curvature. The effects of topologically nontrivial magnetic excitations are believed to manifest in the study of thermal Hall effect. Recently, the study of thermal Hall effect of charge-neutral bosonic quasi-particles has attracted a lot of interest in ferromagnets [40–49] and spin liquid magnetic insulators [50, 51]. In the former, the theory of Berry curvature induced by the DMI can explain the observed thermal Hall conductivity. In the latter, however, the origin of thermal Hall conductivity remains an open question, but the scalar spin chirality could play a vital role [52].

In the current study, synthetic TRS breaking interactions are induced by circularly polarized laser light and they are identified as the in-plane and out-of-plane DMIs. Thus, the theory of thermal Hall effect can be applied in a similar way. However, the Floquet theory leads to nonequilibrium distribution of the quasi-particles and the Bose function $n_b(\epsilon_{ak})$ will depend on detail properties of the system. In the following we focus on the case in which the Bose function is close to thermal equilibrium and write the linear response thermal Hall conductivity as $\kappa_{xy} = -k_b^2 T \int_{BZ} \frac{d^2 k}{(2\pi)^2} \sum_{\alpha=1}^{N} \epsilon_{2}(\epsilon_{ak}) \Omega_{ak}$, where $\Omega_{ak} \equiv \frac{n_b(\epsilon_{ak})}{\epsilon_{ak}} = (e^{\epsilon_{ak}/k_b T} - 1)^{-1}$ is the Bose function close to thermal equilibrium. $\epsilon_2(x) = (1 + x) \left( \ln \frac{1+x}{x} \right)^2 - (\ln x)^2 - 2Li_2(-x)$, and $Li_2(x)$ is a polylogarithm. The Berry curvature is given by $\Omega_{ak} = (\nabla \times A_{\nu ak})_\nu$, where $A_{\nu ak} = i \langle \theta_{ak} | \nabla | \theta_{ak} \rangle$ is the Berry connection.

We note that $\Omega_{ak}$ vanishes in the undriven system with only out-of-plane DMI due to an effective TRS, and hence $\kappa_{xy}$ is zero. For laser-driven system by circularly polarized light the Chern number defined as the integration of the Berry curvature over the Brillouin zone is proportional to the synthetic scalar-chirality of the noncoplanar spin configurations induced by the synthetic in-plane DMI. A nonzero thermal Hall conductivity
implies that the underlying magnetic excitations are topologically nontrivial. In figure 3 we have confirmed that, indeed, the magnetic excitations are topologically nontrivial in the case of circularly polarized light ($\phi = \pi/2$) with nonzero $k_{xy}$ for $D_2/J = 0$ (a) and $D_2/J = 0.2$ (b). Furthermore, we have checked numerically that $k_{xy}$ vanishes for linearly polarized light ($\phi = 0$ or $\pi$)—an indication that the laser-induced anisotropy in this case does not break TRS.

In summary, we have shown that the dispersionless 'zero energy mode' in frustrated KAFMs can be lifted by laser-irradiation and long-range magnetic order can be induced. The possible circularly polarized laser-induced synthetic interactions are identified as the in-plane and out-of-plane DMIs, which play the same role in frustrated KAFMs with low crystal symmetry [9]. In the present case, however, their strength can be tuned by the laser light. We also showed that the system possessed transport properties that can be experimentally accessible. We believe that these results are within experimental reach and can be accessible with the current terahertz frequency using ultrafast terahertz spectroscopy [53]. Thus far, induced magnetic order in frustrated magnets is limited to applying an external magnetic field or pressure. However, these methods usually do not lead to an induced in-plane and out-of-plane DMI. Therefore, experiments can now look for how laser irradiation modifies the properties of frustrated magnets. Most importantly, the laser-induced topological magnetic excitation should be the primary motive as it quantifies the effects of laser irradiation on the system. Then thermal Hall effect can be measured by applying a temperature gradient. Currently, a lifted 'zero energy mode' has only been seen clearly in iron jarosite [10], but there are numerous frustrated kagomé antiferromagnets. Therefore, another experimental task would be the possibility of inducing lifted 'zero energy mode' by laser irradiation on frustrated kagomé antiferromagnets such as Herbertsmithite $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$, which is a known kagomé antiferromagnet with QSL properties. In the future, it would be interesting to extend the current approach to spinons by the Schwinger boson formalism. This will pave the way for optical manipulation of strongly correlated materials including chiral spin liquids [54].

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**Figure 3.** Floquet thermal Hall conductivity versus temperature for circularly polarized light $\phi = \pi/2$ and two laser amplitudes with $\omega/J = 10$. 
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