Topological magnetic excitations on the distorted kagomé antiferromagnets: Applications to volborthite, vesignieite, and edwardsite

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Abstract – In this letter, we identify a noncoplanar chiral spin texture on the distorted kagomé-lattice antiferromagnets induced by the presence of a magnetic field applied perpendicular to the distorted kagomé plane. The noncoplanar chiral spin texture has a nonzero scalar spin chirality similar to a chiral quantum spin liquid with broken time-reversal symmetry. In the noncoplanar regime we observe nontrivial topological magnetic excitations and thermal Hall response through the emergent field-induced scalar spin chirality in contrast to collinear ferromagnets in which the Dzyaloshinskii-Moriya (DM) spin-orbit interaction is the driving force. Our results shed light on recent observation of thermal Hall conductivity in distorted kagomé volborthite at nonzero magnetic field with no signal of DM spin-orbit interaction, and the results also apply to other distorted kagomé antiferromagnets such as vesignieite and edwardsite.

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Introduction. – A chiral spin liquid (CSL) is a particular type of spin liquids in which time-reversal symmetry (TRS) is broken macroscopically [1–3]. The scalar spin chirality $\chi_{ijk} = \langle S_i \cdot (S_j \times S_k) \rangle$ measures broken TRS and defines the hallmark of a CSL. It is also generally believed that the magnetic excitations in any quantum spin liquid (QSL) are bosonic spin-$1/2$ spinons in which the conventional bosonic spin-$1$ magnons fractionalize. According to Kalmeyer and Laughlin [1] a CSL can be thought of as a bosonic version of the $\nu = 1/2$ Laughlin state in the fractional quantum Hall effect. In recent years, a plethora of theoretical models has been proposed in which a CSL phase can be stabilized. The recent theoretical numerical works show that a CSL can emerge in a kagomé lattice Mott insulator in which a magnetic field induces an explicit spin chirality interaction in a $t/U$ perturbative expansion of the Hubbard model at half-filling [4], hence TRS is broken explicitly. In other numerical works a spontaneously broken TRS CSL is found away from the isotropic kagomé-lattice antiferromagnet by adding additional second- and third-nearest neighbour interactions [5–9] or Ising anisotropy [10].

In real materials, however, a CSL has remained elusive because material synthesis usually comes with defects such as structural distortion and perturbative anisotropy like the intrinsic DM spin-orbit interaction [11,12], which tend to destabilize disordered QSL and induce a magnetic long-range order. For quantum kagomé antiferromagnets (QKAF), various experimentally accessible materials such as iron jarosites [13–16], vesignieite $\text{BaCu}_3\text{V}_2\text{O}_{8-}\text{OH}_2$ [17–22], edwardsite $\text{Cd}_2\text{Cu}_3\text{(SO}_4\text{)}_2(\text{OH})_6\text{.4H}_2\text{O}$ [23], and volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2\text{.2H}_2\text{O}$ [24–27] have long-range magnetic orders below certain temperatures and they are generally attributed to the presence of DM spin-orbit interaction. The recent proposals of QSL materials are herbertsmithite $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ [28,29] and the calcium-chromium oxide $\text{Ca}_{10}\text{Cr}_7\text{O}_{28}$ [30] which show a continuum of spinon excitations in inelastic neutron scattering experiment [30,31]. Nevertheless, an applied magnetic field or pressure destabilizes the QSL nature of these materials and leads to an induced magnetic order at low temperatures [30,32,33].

The emerging experimental technique to probe the nature of magnetic excitations in quantum magnets with possible QSL ground states is the measurement of thermal Hall effect. For quantum magnets with magnetic long-range order the thermal Hall effect has been realized in
the collinear kagomé ferromagnet Cu(1-3, bdc) [34] and a number of collinear pyrochlore ferromagnets [35,36]. In this case the transverse thermal Hall conductivity ($\kappa_{xy}$) can be explained in terms of Berry curvature induced by the DM spin-orbit interaction [37–39] leading to topological magnons [40–48] and Weyl magnons [49,50] similar to spin-orbit coupling electronic systems [51–55]. In a recent experiment [56], a nonzero $\kappa_{xy}$ has been observed in a frustrated distorted kagomé volborthite at a strong magnetic field of 15 T with no signs of the DM spin-orbit interaction and no discernible thermal Hall signal was observed at zero magnetic field [57]. The authors attributed the presence of $\kappa_{xy}$ to nontrivial elementary excitations in the gapless QSL phase; however, a strong field of 15 T causes low-temperature magnetic phases in volborthite [25–27]. Nevertheless, the exact nature of the low-temperature magnetic phases at 15 T is poorly understood, but the intrinsic DM spin-orbit anisotropy suggests a $Q = 0$ coplanar/noncollinear Néel order.

In previous exact diagonalization study, it has been established that the out-of-plane DM spin-orbit anisotropy can induce a quantum critical point (QCP) in spin-(1/2) QKAF at $D^c_\perp/J \sim 0.1$ [58]. A moment free QSL phase is predicted for $D_\perp < D^c_\perp$ and a $Q = 0$ Néel phase exists for $D_\perp > D^c_\perp$. From different experimental inspections, it is generally believed that both edwardsite and vesignieite are located in the ordered regime below a certain temperature with $0.1 < D_\perp/J < 0.16$ [19,20], and there is a possibility that volborthite also belongs to the ordered regime at low temperatures. But herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$ has $D_\perp/J \sim 0.08$ [29], thus remain a QSL. An applied external magnetic field ($H \perp$ 2D plane) can induce noncoplanar spin textures with a nonzero scalar spin chirality. The emergent scalar spin chirality due to noncoplanar spins might be different from the spontaneous scalar spin chirality in the CSL, but their effects on the magnetic excitations should be the same. In this letter we show that a nonzero Berry curvature can be induced by a chiral spin texture in real space rather than the DM spin-orbit interaction. The Berry curvature stems from the emergent scalar spin chirality which measures the solid angle subtended by three noncoplanar spins in a unit triangular plaquette of the kagomé lattice. The Berry curvature of the chiral spin texture is the driving force of the thermal Hall effect in frustrated distorted QKAF with/without magnetic long-range order.

Model. — We consider the microscopic spin Hamiltonian of the distorted QKAF subject to a magnetic field perpendicular to the kagomé plane. The Hamiltonian is given by

$$
\mathcal{H} = \sum_{\langle ij \rangle} [J_{ij} S_i \cdot S_j + D_{ij} (S_i \times S_j)] - H \cdot \sum_i S_i, \quad (1)
$$

where $S_i$ is the magnetic spin moment at site $i$, and the summation runs over nearest-neighbour spins. $J_{ij} = J > 0$ on the diagonal bonds, and $J_{ij} = J' = J \delta$ on the horizontal bonds with $\delta \neq 1$ as shown in fig. 1. The inversion symmetry breaking between the bonds on the kagomé lattice allows a DM vector $D_{ij} = -D_\perp \hat{z}$ and they lie at the midpoints between two magnetic ions as shown in fig. 1. The last term is the out-of-plane external magnetic field $H = h\hat{z}$ and $h = g\mu_B H$. The Hamiltonian (1) is applicable to volborthite [59–63] and other forms of Hamiltonian have also been proposed for volborthite [64–66]. The out-of-plane DM interaction is always present on the kagomé lattice and it stabilizes the coplanar/noncollinear spin configuration [13]. In some kagomé materials an in-plane DM interaction $D_{ij}$ may be present due to the lack of mirror planes. It leads to weak out-of-plane ferromagnetism with small ferromagnetic moment. However, it is usually negligible compared to the out-of-plane component for most spin-(1/2) kagomé antiferromagnetic materials [29,58] and can be removed by gauge transformation. For kagomé volborthite [56], there was no signal of both in-plane and out-of-plane DM interaction on the observed $\kappa_{xy}$. Also, for potassium jarosite with spin-(5/2) the in-plane DM interaction (or the DM interaction in general) does not necessarily induce topological magnetic excitations [16]. Therefore, we will neglect the small in-plane DM component at the moment and comment on its effects in the subsequent sections.

![Fig. 1](image-url) (Color online) The canted noncollinear/coplanar magnetic order with positive vector chirality on the distorted kagomé lattice and the symmetry group of the lattice. The out-of-plane DM interaction lies at the midpoints between two magnetic ions as indicated by small circles. The spin triads are separated by an angle $\vartheta \neq 120^\circ$ on each isosceles triangle with $J \neq J'$ as shown in the top figure.
Let us start from what is known for distorted kagomé antiferromagnets at \( D_{ij} = h = 0 \) [61]. In this case, the Hamiltonian can be written as

\[
\mathcal{H} = \frac{J\delta}{2} \sum_{\Delta} \left( \frac{1}{\delta} S_A + S_B + S_C \right)^2 - \text{const}, \tag{2}
\]

where \( S_{A,B,C} \) form a triad as depicted in fig. 1. It is easily seen that the classical energy is minimized for \( \frac{1}{\delta} S_A + S_B + S_C = 0 \), yielding \( \theta = \arccos(-1/2\delta) \neq 120^\circ \) for \( \delta \neq 1 \). The classical configuration is now a canted coplanar/noncollinear spins for \( \delta > 1/2 \) [61]. However, it also has an extensive degeneracy as in the ideal kagomé Heisenberg antiferromagnets. There are several limiting cases in which the system is either bipartite with collinear magnetic order or nonbipartite with non-collinear magnetic order. The limiting case \( \delta \to 0 \) maps to a decoupled antiferromagnetic chains. For \( \delta \to \infty \) the classical ground state is collinear (up-down-down state) and there is no degeneracy except for a global spin rotation about the \( z \)-axis, and for \( \delta > 1/2 \) the classical ground state is noncollinear but coplanar. The collinear phases are trivial. In the present study, we focus on the canted coplanar/noncollinear regime \( \delta > 1/2 \) as the ground-state properties of volborthite, vesignite, and edwardsite are believed to live in this regime.

**Classical ground state.** – Now, we consider the classical ground state of the full Hamiltonian (1). In the classical limit, the spin operators can be approximated as classical vectors, written as \( S_i = S n_i \), where \( n_i = (\sin \phi \cos \theta_i, \sin \phi \sin \theta_i, \cos \phi) \) is a unit vector and \( \theta_i \) labels the spin oriented angles on the spin triad and \( \phi \) is the magnetic-field–induced canting angle. For \( \delta > 1/2 \) the ground state is the canted coplanar/noncollinear spin configuration in fig. 1. The classical energy is given by

\[
e_0(\phi) = 2J(2 + \delta) \left[ (1 - \cos \theta) \cos^2 \phi + \cos \theta \right] - 4D_1 \sin^2 \phi \sin \theta(1 - \cos \theta) - 3h \cos \phi, \tag{3}
\]

where \( e_0(\phi) = E(\phi)/N S^2 \), and \( N \) is the number of sites per unit cell. The magnetic field is rescaled in unit of \( S \). The minimization of \( e_0(\phi) \) yields the magnetic-field–induced canting angle \( \cos \phi = h/h_s \) where

\[
h_s = \frac{(1 - \cos \theta)}{3} \left[ 4J(2 + \delta) + 8D_1 \sin \theta \right]. \tag{4}
\]

As can be seen from eq. (3) the classical energy depends on the DM interaction as it contributes to the stability of the coplanar/noncollinear spin configuration. In collinear spin configurations, the DM interaction does not contribute to the classical energy, instead it provides TMDs, e.g., in kagomé ferromagnets [34–38,40–45,49,50].

**Topological magnetic excitations.** – The magnetic excitations of frustrated QKAF can be studied in different formalisms. In real materials the intrinsic DM spin-orbit interaction and an external applied magnetic field are very likely to induce a magnetic long-range order in frustrated QKAF [25–27,32,33]. Therefore, we will employ the Holstein-Primakoff (HP) transformation [67] valid in the low-temperature regime. In the Supplemental Material Supplementarymaterial.pdf (SM), we have shown that a magnetic field \((H \perp 2D \text{ plane}) \) can induce a chiral noncoplanar spin texture with an emergent scalar spin chirality

\[
\mathcal{H}_\phi = J_\phi \sum_{i,j,k,\Delta} \chi_{ijk}, \tag{5}
\]

where \( J_\phi = \cos \phi \propto h \). As we previously mentioned, the scalar spin chirality breaks time-reversal (T) symmetry macroscopically and can be spontaneously developed in CSL [1–8]. Its effects on the magnetic excitations should be the same in the ordered and disordered phases. In the magnetic-field–induced noncoplanar regime the DM interaction is not necessarily needed. Its main effect on QKAF is to induce a long-range magnetic order. The system exhibits nontrivial topological effects through real-space Berry curvature of the chiral noncoplanar spin texture. The magnetic excitations at zero magnetic field and zero DM interaction with \( \delta \neq 1 \) still exhibit a zero energy mode \( \omega_0 = 0 \) (not shown). Lattice distortion is unable to lift the zero energy mode. In addition, there are two nondegenerate dispersive modes \( \omega_1 \neq \omega_2 \) (not shown) induced by lattice distortion in contrast to undistorted kagomé antiferromagnets with degenerate modes \( \omega_1 = \omega_2 \). Although the exact parameter values of volborthite are not known, an out-of-plane DM interaction is intrinsic to the kagomé lattice. A moderate DM interaction \((D_{ij}/J = 0.2) \) lifts the zero energy mode and stabilizes the coplanar/noncollinear spin configuration as shown in fig. 2. We find that the lifted zero mode is no longer a constant flat band but varies in the BZ (see SM), that is \( \omega_0(\Gamma) \neq \omega_0(\mathbf{K}) \), where \( \Gamma = (0,0) \) and \( \mathbf{K} = (2\pi/3,0) \), see fig. 3. This is one of the differences between distorted and undistorted kagomé antiferromagnets at finite DM interaction.

The symmetry group of the kagomé lattice is generated by lattice translation \( \mathbf{T} \), 6-fold rotation \( C_6 \) (\( \pi/3 \) rotation

![Fig. 2: (Color online) Magnon dispersion of distorted QKAF at zero magnetic field and several values of distortion with \( D_{ij}/J = 0.2 \).](image-url)
Fig. 3: (Color online) The lifted zero mode $\omega_0$ varies in the Brillouin zone as function of the distortion for zero magnetic field $H_\phi = 0$. The dashed lines connect the isotropic point $\delta = 1$. The coplanar/noncollinear order is valid for $\delta > 0.5$.

about the center of the hexagonal plaquettes), and mirror reflection symmetry $\mathcal{M}$ (see fig. 1). Several combinations of the space group symmetry and $T$-symmetry can lead to identity in the coplanar order phase. For instance, $\mathcal{M}$-symmetry is a good symmetry of the kagomé lattice, but flips the in-plane spins and $T$-symmetry flips the spins again and leaves the magnetic order invariant. Hence $T\mathcal{M}$ is a symmetry of the coplanar/noncollinear magnetic order. Since the lattice distortion and the out-of-plane DM interaction preserve this combined symmetry the system should be topologically trivial as we will show below. Therefore, we expect this model to be topologically nontrivial when either $T$-symmetry or $\mathcal{M}$-symmetry is broken.

The combined symmetry can be broken in two ways:

1) If the kagomé lattice lacks $\mathcal{M}$-symmetry, an in-plane DM component $D_{||}$ can be allowed according to the Moriya rules [12]. The in-plane DM component preserves $T$-symmetry but breaks $\mathcal{M}$-symmetry, hence $T\mathcal{M}$ will be broken. This leads to weak out-of-plane ferromagnetism or noncoplanar spin canting with scalar spin chirality and weak ferromagnetic moment [13,68]. Thus, for kagomé antiferromagnetic materials with $D_\perp \ll D_{||}$ we expect the out-of-plane ferromagnetism to be dominant and hence the (in-plane) DM interaction should be the primary source of topological spin excitations as we already know in collinear ferromagnets with out-of-plane DM interaction. However, most kagomé antiferromagnetic materials have a dominant intrinsic out-of-plane component $D_\perp \gg D_{||}$ and the weak ferromagnetism induced by $D_{||}$ can be negligible. In fact, there was no signal of any DM interaction on the observed $\kappa_{xy}$ in kagomé volborthite [56]. Therefore, there must be another source of topological spin excitations apart from the DM interactions.

2) An alternative way to break $T\mathcal{M}$ symmetry is by applying an external magnetic field perpendicular to the kagomé plane ($H \perp 2D$ plane). A finite out-of-plane magnetic field breaks $T$-symmetry but preserves $\mathcal{M}$-symmetry. It induces a noncoplanar spin texture with a nonzero scalar spin chirality (see SM). The magnon dispersions are also gapped as depicted in fig. 4 similar to the case without magnetic field. However, the gap excitations with and without the magnetic field are different which can be shown by solving for the chiral edge modes. The magnon edge modes for a strip geometry with open boundary conditions along the $y$-direction and infinite along the $x$-direction are shown in fig. 5 at zero magnetic field (top panel). We see that the system does not possess topologically protected gapless edge modes and the Chern number vanishes (see SM). On the other hand, the edge modes at finite magnetic field shown in the bottom panel of fig. 5 are gapless —an indication of a topological system. The magnetic-field–induced noncoplanar chiral spin texture does not require a DM interaction in general as long-range order can be induced through other interactions such as a second-nearest neighbour. Besides, the scalar spin chirality $\chi_{ijk}$ should have the same effects on magnetic excitations in the CSL.

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Fig. 6: (Color online) Low-temperature dependence of the topological thermal Hall conductivity with $D\perp/J = 0.2$.

frustrated magnets has remained an open question. In electronic (metallic) systems there is a concept of the topological Hall effect in which an unconventional anomalous Hall conductivity is induced by the scalar spin chirality resulting from the configuration of the chiral spin textures rather than the spin-orbit interaction as reported in several electronic materials [69–72]. In this section, we will show that this mechanism can be observed also in charge-neutral magnetic excitations on the frustrated QKAF. In this case the appropriate transport measurement is the thermal Hall effect in which a temperature gradient $−\nabla T$ induces a heat current $J_Q$. From linear response theory, one obtains

$$J_Q = −\sum_\beta \kappa_{\alpha\beta} \nabla_\beta T,$$

where $\kappa_{\alpha\beta}$ is the thermal conductivity and the transverse component $\kappa_{xy}$ is associated with the thermal Hall conductivity given explicitly in ref. [39].

Figure 6 shows the low-temperature dependence of $\kappa_{xy}$ (in units of $k_B/\hbar$) for two values of the lattice distortion and the magnetic field. The topological Hall conductivity captures a negative value in both regimes $\delta < 1$ and $\delta > 1$ and the thermal Hall conductivity is suppressed in the latter. As we previously noted the thermal Hall conductivity does not originate from the DM spin-orbit interaction but from the chiral spin texture of the noncoplanar spins induced by the magnetic field. The DM spin-orbit interaction is only necessary to stabilize the coplanar/noncollinear magnetic spin structure. We showed that lattice distortion and DM interaction introduce gap magnetic excitations, but the system remains topologically trivial with neither protected chiral edge modes nor finite thermal Hall conductivity. By turning on an out-of-plane external magnetic field, we showed that a finite scalar spin chirality is induced from the noncoplanar chiral spin texture. This gives rise to a real-space Berry curvature which measures the solid angle subtended by three noncoplanar spins independent of the DM interaction. We note that the real-space Berry curvature of the chiral spin texture should persist in the chiral spin liquid (CSL) phase due to the presence of the spontaneous scalar spin chirality.

The results of this letter suggest that the experimental result of thermal Hall response in volborthite can be best described using a distorted Heisenberg model as opposed to a frustrated alternating spin chain. It is noted that the DM interaction can be removed by a gauge transformation for frustrated one-dimensional (1D) spin chain [74] and an ideal 1D system should not have a chiral noncoplanar magnetic order and scalar spin chirality should be absent. An alternative approach is the Schwinger boson formalism; however, the scalar spin chirality does not appear explicitly in this formalism. Instead the DM interaction generates a magnetic flux leading to topological spin excitations even in the absence of an applied magnetic field [42,75].

Fig. 7: (Color online) Field dependence of the topological thermal Hall conductivity at low temperatures with $D\perp/J = 0.2$.

Conclusion. – In summary, we have studied topological magnetic excitations and thermal Hall effect induced by the real-space Berry curvature of the chiral spin texture on the distorted kagomé antiferromagnets applicable to vesignieite, edwardsite, and volborthite. The lack of inversion symmetry on the kagomé lattice allows a Dzyaloshinskii-Moriya (DM) interaction. However, in contrast to ferromagnets the DM interaction is only necessary to stabilize the coplanar/noncollinear Neél order. Therefore, kagomé antiferromagnetic materials with a weak (negligible) DM interaction can possess gap magnetic excitations, but the system remains topologically trivial with neither protected chiral edge modes nor finite thermal Hall conductivity. By turning on an out-of-plane external magnetic field, we showed that a finite scalar spin chirality is induced from the noncoplanar chiral spin texture. This gives rise to a real-space Berry curvature which measures the solid angle subtended by three noncoplanar spins independent of the DM interaction. We note that the real-space Berry curvature of the chiral spin texture should persist in the chiral spin liquid (CSL) phase due to the presence of the spontaneous scalar spin chirality.

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This is reminiscent of collinear ferromagnets and, thus, sharply in contrast with the present results. These results also call for further investigation of the thermal Hall effect in the kagomé volborthite and other distorted quantum kagomé antiferromagnets such as vesig-nieite [17–22] and edwardsite [23]. Although we captured a negative thermal Hall conductivity ($\kappa_{\text{xy}}$) as seen in the experiment [56], it would be interesting to determine the parameter values of volborthite and also measure the magnetic-field dependence of $\kappa_{\text{xy}}$ and possibly the topological magnetic excitations at various magnetic-field and temperature ranges. Furthermore, experiment should also try to measure the spontaneous or magnetic-field–induced scalar spin chirality that gives rise to topological magnetic excitations and thermal Hall response. We believe that the results of this letter have shed some light on the recent observation of thermal Hall conductivity in volborthite [56].

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