

Sign Structure of Thermal Hall Conductivity and Topological Magnons for In-Plane Field Polarized Kitaev Magnets

Li Ern Chern[✉], Emily Z. Zhang[✉], and Yong Baek Kim

Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada

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The appearance of half-quantized thermal Hall conductivity in α -RuCl₃ in the presence of in-plane magnetic fields has been taken as a strong evidence for the Kitaev spin liquid. Apart from the quantization, the observed sign structure of the thermal Hall conductivity is also consistent with predictions from the exact solution of the Kitaev honeycomb model. Namely, the thermal Hall conductivity changes sign when the field direction is reversed with respect to the heat current, which is perpendicular to one of the three nearest neighbor bonds on the honeycomb lattice. On the other hand, the thermal Hall conductivity is almost zero when the field is applied along the bond direction. Here, we theoretically demonstrate that such a peculiar sign structure of the thermal Hall conductivity is a generic property of the polarized state in the presence of in-plane magnetic fields. In this case, the thermal Hall effect arises from topological magnons with finite Chern numbers, and the sign structure follows from the symmetries of the momentum space Berry curvature. Using a realistic spin model with bond-dependent interactions, we show that the thermal Hall conductivity can have a magnitude comparable to that observed in the experiments. Hence, the sign structure alone cannot make a strong case for the Kitaev spin liquid. The quantization at very low temperatures, however, will be a decisive test as the magnon contribution vanishes in the zero temperature limit.

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Introduction.—The Kitaev honeycomb model [1], in which nearest neighbor $S = 1/2$ moments are coupled to each other by bond-dependent Ising interactions, is one of the few exactly soluble spin models that leads to an unusual ground state known as a quantum spin liquid. In the Kitaev spin liquid, the $S = 1/2$ moments fractionalize into Majorana fermions [2] coupled to a \mathbb{Z}_2 gauge field. The Kitaev interaction is proposed to have a dominant presence in systems with $4d$ or $5d$ transition metal elements [3,4] such as Na₂IrO₃ [5–7] and α -RuCl₃ [8]. However, there exist other interactions as well [9], which pave the way for a zigzag magnetically ordered state [10–13] instead of the desired quantum spin liquid. A dramatic twist in the materialization of the Kitaev spin liquid came with the observation of half-quantized thermal Hall conductivity in α -RuCl₃ under an external magnetic field, which has both finite in-plane and out-of-plane components [14]. Since the half-quantization is a signature of Majorana fermions [1,15–17], the experiment strongly hints at a field induced Kitaev spin liquid in α -RuCl₃ [18–26]. If the half-quantization is confirmed, this would be a smoking gun that quantum spin liquid does exist in nature, not merely being a theoretical concept.

More recently, a similar thermal transport measurement with in-plane magnetic fields was performed [27]. It was reported that the half-quantization of thermal Hall conductivity can still occur even when the field is completely in plane. Compared to the usual textbook example of

two-dimensional conductors where the Hall effect only takes place under out-of-plane fields, the sizable thermal Hall signal—not to mention the additional fact that it is half-quantized—in Ref. [27] is in some sense anomalous. The experimental setup is described as follows. The two independent in-plane directions are conventionally chosen to be (i) the a direction, which is perpendicular to one of the three nearest neighbor bonds on the honeycomb lattice, and (ii) the b direction, which is parallel to a nearest neighbor bond and perpendicular to the a direction (see Fig. 1). In the experiment, the heat current is always applied along the a direction, while the magnetic field is applied along the a , b , or $-a$ direction. The corresponding thermal Hall conductivity is observed to be positive, zero, or negative, assuming

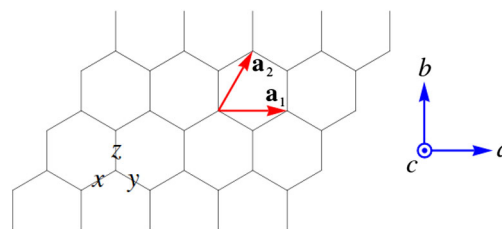


FIG. 1. The nearest neighbor bond types x , y , and z in the KTT' model, the primitive lattice vectors \mathbf{a}_1 and \mathbf{a}_2 on the honeycomb lattice, and the crystallographic directions a (in plane), b (in plane), and c (out of plane). Measured in the cubic basis, according to which the spin components in the KTT' model are defined, the a , b , and c directions are $[11\bar{2}]$, $[\bar{1}10]$, and $[111]$, respectively.

that the transverse temperature gradient is measured along the b direction [28]. Such a sign structure fits into the theory of a non-Abelian spin liquid, which is stabilized in the Kitaev model under a magnetic field. The half-quantized thermal Hall conductivity is observed along the a and $-a$ directions within a certain range of temperatures and field strengths.

An independent measurement [29] confirmed such a sign structure of the thermal Hall conductivity but did not quite observe the half-quantization plateau. Rather, the thermal Hall conductivity looks more like a smooth function across a wide range of temperatures (including the suspected spin liquid regime) and vanishes rapidly when the temperature approaches zero. This discovery suggests the existence of a state that may be different from the non-Abelian spin liquid but able to produce the same sign structure of the thermal Hall conductivity.

In this Letter, we theoretically demonstrate that the peculiar sign structure of the thermal Hall conductivity κ_{xy} is a generic property of the polarized state in Kitaev magnets under in-plane magnetic fields. In this case, the thermal Hall effect arises from topological magnons [30–34] with finite Chern numbers $C = \pm 1$, while the sign structure of κ_{xy} is a consequence of the symmetries of the momentum space

Berry curvature. Instead of the Kitaev model, we consider a more realistic $K\Gamma\Gamma'$ model subjected to in-plane magnetic fields as in the experiments [27,29]. We derive analytically the following theorems concerning the sign structure of the thermal Hall conductivity due to magnons in the linear spin wave theory of the polarized state, which are consistent with the experimental observations.

Theorem 1: κ_{xy} in the polarized states under magnetic fields along the a and $-a$ directions differ by a minus sign.

Theorem 2: κ_{xy} in the polarized state under a magnetic field along the b direction is zero.

Concise proofs will be presented later in the main text, with details relegated to the Supplemental Material [35]. Theorem 1 only tells us the relative sign of κ_{xy} in the a and $-a$ polarized states, not their absolute signs. Also, Theorem 1 allows for $\kappa_{xy} = 0$, so it does not guarantee a finite thermal Hall effect. It is thus important to assume reasonable values of K , Γ , and Γ' , which minimally model α - RuCl_3 , and perform numerical calculations of κ_{xy} to check its magnitude and sign. We find that, with dominant $K < 0$ and $\Gamma > 0$ [36], κ_{xy} is indeed finite and positive (negative) in the polarized state along the a ($-a$) direction, and essentially zero in the polarized state along the b direction [see Figs. 2(a)–2(f)]. Moreover, the size and trend

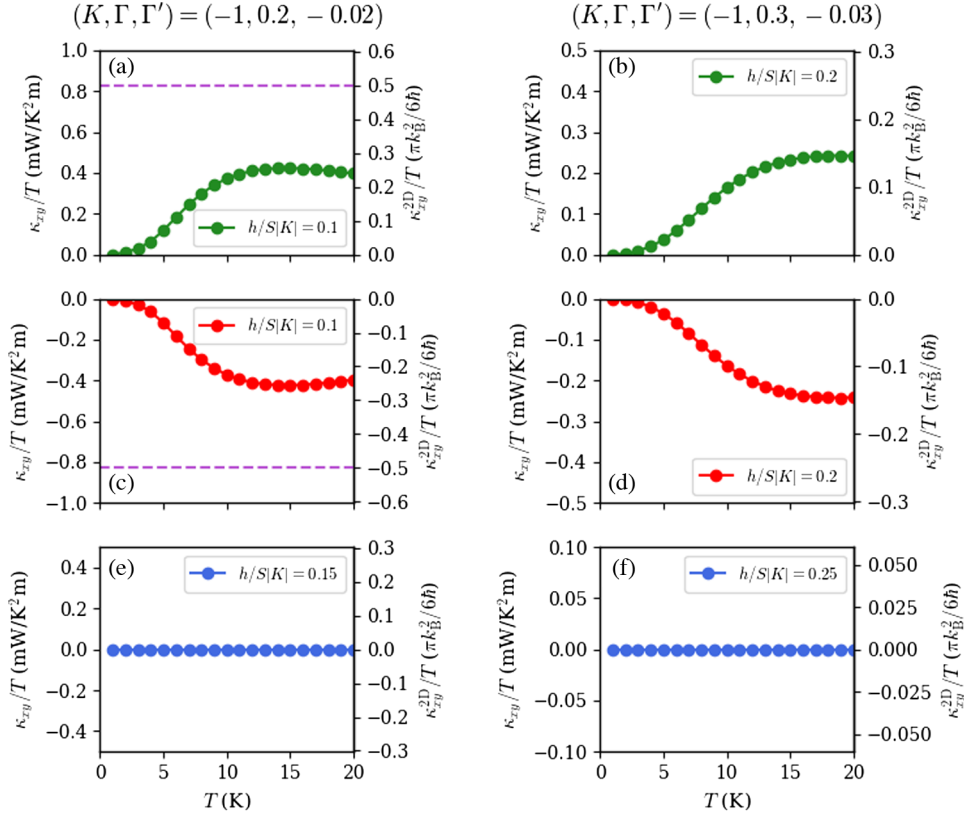


FIG. 2. Thermal Hall conductivity κ_{xy}/T due to magnons in various polarized states as a function of temperature T , for the parameterizations $(K, \Gamma, \Gamma') = (-1, 0.2, -0.02)$ and $(-1, 0.3, -0.03)$, shown in the left and right panels, respectively. The magnetic field is applied along the a direction in (a) and (b), the $-a$ direction in (c) and (d), and the b direction in (e) and (f). The corresponding values of $\kappa_{xy}^{2D}/T \equiv \kappa_{xy} d/T$ are also indicated. The purple dashed line in (a) and (c) represents the half-quantized thermal Hall conductivity.

of κ_{xy} are also comparable to those measured experimentally. Using the linear spin wave theory, we also calculate the dynamical spin structure factor of the a polarized state [35], which compares well with the inelastic neutron scattering experiment [22] at about the same fields. Our result suggests that the observed thermal Hall conductivity, in case the half-quantization is absent, may originate from the polarized state with magnons as heat carriers.

Model.—The $K\Gamma\Gamma'$ model, which minimally describes α - RuCl_3 , under a magnetic field, is given by $H = \sum_{\lambda \in \{x,y,z\}} \sum_{\langle ij \rangle \in \lambda} \mathbf{S}_i^T H_\lambda \mathbf{S}_j - \sum_i \mathbf{h} \cdot \mathbf{S}_i$, where

$$H_x = \begin{pmatrix} K & \Gamma' & \Gamma' \\ \Gamma' & 0 & \Gamma \\ \Gamma' & \Gamma & 0 \end{pmatrix}, \quad H_y = \begin{pmatrix} 0 & \Gamma' & \Gamma \\ \Gamma' & K & \Gamma' \\ \Gamma & \Gamma' & 0 \end{pmatrix},$$

$$H_z = \begin{pmatrix} 0 & \Gamma & \Gamma' \\ \Gamma & 0 & \Gamma' \\ \Gamma' & \Gamma' & K \end{pmatrix}.$$

We apply the linear spin wave theory [37,38] to a field polarized state in the $K\Gamma\Gamma'$ model. First, we rotate the coordinate frames of all spins uniformly such that the z axes align with the spins $\mathbf{S}_i = R\tilde{\mathbf{S}}_i$. Let the orientation of polarized spins in the original frame be parameterized by two angles θ and ϕ as $\mathbf{S}_i = S(\sin\theta \cos\phi, \sin\theta \sin\phi, \cos\theta)$. We choose the rotation matrix to be [38]

$$R = \begin{pmatrix} \cos\theta \cos\phi & -\sin\phi & \sin\theta \cos\phi \\ \cos\theta \sin\phi & \cos\phi & \sin\theta \sin\phi \\ -\sin\theta & 0 & \cos\theta \end{pmatrix} \in SO(3). \quad (1)$$

Notice that the columns of R are mutually orthonormal, and they satisfy the right-hand rule of cross product. Then, we apply the Holstein Primakoff transformation [37,38] to $\tilde{\mathbf{S}}_i$ and neglect the terms of third and higher order in the bosonic operators b and b^\dagger . Upon a Fourier transformation, we arrive at the linear spin wave Hamiltonian in momentum space $H/S = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger D_{\mathbf{k}} \Psi_{\mathbf{k}}$, where $D_{\mathbf{k}}$ is a four-dimensional Hermitian matrix and $\Psi_{\mathbf{k}} = (b_{1\mathbf{k}}, b_{2\mathbf{k}}, b_{1-\mathbf{k}}^\dagger, b_{2-\mathbf{k}}^\dagger)$. $D_{\mathbf{k}}$ has to be diagonalized by a Bogoliubov transformation [39] in order to preserve the commutation relation of bosons. Once we obtain the linear spin wave dispersion $\varepsilon_{n\mathbf{k}}$, we can calculate the thermal Hall conductivity due to magnons [40–42] as

$$\kappa_{xy} = -\frac{k_B^2 T}{\hbar V} \sum_n \sum_{\mathbf{k} \in \text{FBZ}} \left\{ c_2[g(\varepsilon_{n\mathbf{k}})] - \frac{\pi^2}{3} \right\} \Omega_{n\mathbf{k}}, \quad (2)$$

where FBZ denotes the first Brillouin zone, $c_2(x) = (1+x)\{\ln[(1+x)/x]\}^2 - (\ln x)^2 - 2\text{Li}_2(-x)$, g is the

Bose-Einstein distribution, and $\Omega_{n\mathbf{k}}$ is the Berry curvature of the n th band at momentum \mathbf{k} . The term $-\pi^2/3$ will be dropped in subsequent discussions because the summation of all Chern numbers is zero [43].

Proof of Theorem 1.—The sets of angles $\{\theta, \phi\}$ in Eq. (1) for the polarized states along the a and $-a$ directions are, respectively, $\{\cos^{-1}(-\sqrt{2/3}), \pi/4\}$ and $\{\cos^{-1}(\sqrt{2/3}), 5\pi/4\}$. After some algebra [35], one can show that the linear spin wave Hamiltonians of the a and $-a$ polarized states are related by $D_{\mathbf{k}}^{\bar{a}} = (D_{-\mathbf{k}}^a)^*$. Suppose that $T_{\mathbf{k}}^a$ is the Bogoliubov transformation of $D_{\mathbf{k}}^a$, then $T_{\mathbf{k}}^{\bar{a}} = (T_{-\mathbf{k}}^a)^*$ is the Bogoliubov transformation of $D_{\mathbf{k}}^{\bar{a}}$,

$$(T_{\mathbf{k}}^{\bar{a}})^\dagger D_{\mathbf{k}}^{\bar{a}} T_{\mathbf{k}}^{\bar{a}} = [(T_{-\mathbf{k}}^a)^\dagger D_{-\mathbf{k}}^a T_{-\mathbf{k}}^a]^* = \mathcal{E}_{-\mathbf{k}}^a = \mathcal{E}_{\mathbf{k}}^{\bar{a}}, \quad (3a)$$

$$T_{\mathbf{k}}^{\bar{a}} \sigma^3 (T_{\mathbf{k}}^{\bar{a}})^\dagger = [T_{-\mathbf{k}}^a \sigma^3 (T_{-\mathbf{k}}^a)^\dagger]^* = \sigma^3, \quad (3b)$$

where $\sigma^3 = \text{diag}(1, 1, -1, -1)$. Equation (3a) says that the energy eigenvalues at \mathbf{k} when $\mathbf{h} \parallel -a$ are the same as those at $-\mathbf{k}$ when $\mathbf{h} \parallel a$. Next, we relate the Berry curvatures in the two polarized states,

$$\begin{aligned} \Omega_{n-\mathbf{k}}^{\bar{a}} &= i \left[\sigma^3 \left(\frac{\partial T_{-\mathbf{k}}^{\bar{a}}}{\partial k_x} \right)^\dagger \sigma^3 \frac{\partial T_{-\mathbf{k}}^{\bar{a}}}{\partial k_y} - (x \leftrightarrow y) \right]_{nn} \\ &= - \left\{ i \left[\sigma^3 \left(\frac{\partial T_{\mathbf{k}}^a}{\partial k_x} \right)^\dagger \sigma^3 \frac{\partial T_{\mathbf{k}}^a}{\partial k_y} - (x \leftrightarrow y) \right]_{nn} \right\}^* \\ &= -\Omega_{n\mathbf{k}}^a. \end{aligned} \quad (4)$$

Therefore, the thermal Hall conductivities are related by

$$\begin{aligned} \kappa_{xy}^{\bar{a}} &= -\frac{k_B^2 T}{\hbar V} \sum_{n\mathbf{k}} c_2[g(\varepsilon_{n-\mathbf{k}}^{\bar{a}})] \Omega_{n-\mathbf{k}}^{\bar{a}} \\ &= \frac{k_B^2 T}{\hbar V} \sum_{n\mathbf{k}} c_2[g(\varepsilon_{n\mathbf{k}}^a)] \Omega_{n\mathbf{k}}^a \\ &= -\kappa_{xy}^a. \end{aligned} \quad (5)$$

In other words, the thermal Hall conductivities are same in magnitude but opposite in sign. This completes the proof of Theorem 1.

Proof of Theorem 2.—The set of angles $\{\theta, \phi\}$ in (1) for the polarized state along the b direction is $\{\pi/2, 3\pi/4\}$. After some algebra [35], one can show that the linear spin wave Hamiltonian of the b polarized state $D_{\mathbf{k}}^b$ depends on k_x and k_y only through the combination $\cos(k_x a/2) \exp(-i\sqrt{3}k_y a/2)$ and its complex conjugate. Therefore, the linear spin wave Hamiltonian is an even function in k_x , $D_{(-k_x, k_y)}^b = D_{(k_x, k_y)}^b$. Consequently, the matrix of energy eigenvalues and the Bogoliubov transformation are even in k_x , $\mathcal{E}_{(-k_x, k_y)}^b = \mathcal{E}_{(k_x, k_y)}^b$ and $T_{(-k_x, k_y)}^b = T_{(k_x, k_y)}^b$. Since the derivative of an even function is an odd function,

$$\frac{\partial T_{(-k_x, k_y)}^b}{\partial k_x} = -\frac{\partial T_{(k_x, k_y)}^b}{\partial k_x}. \quad (6)$$

In addition, we have

$$\begin{aligned}\frac{\partial T_{(-k_x, k_y)}^b}{\partial k_y} &= \lim_{\epsilon \rightarrow 0} \frac{T_{(-k_x, k_y + \epsilon)}^b - T_{(-k_x, k_y)}^b}{\epsilon} \\ &= \lim_{\epsilon \rightarrow 0} \frac{T_{(k_x, k_y + \epsilon)}^b - T_{(k_x, k_y)}^b}{\epsilon} \\ &= \frac{\partial T_{(k_x, k_y)}^b}{\partial k_y}.\end{aligned}\quad (7)$$

Therefore, the Berry curvatures at $\mathbf{k} = (\pm k_x, k_y)$ are related by

$$\begin{aligned}\Omega_{n(-k_x, k_y)}^b &= i \left[\sigma^3 \left(\frac{\partial T_{(-k_x, k_y)}^b}{\partial k_x} \right)^\dagger \sigma^3 \frac{\partial T_{(-k_x, k_y)}^b}{\partial k_y} - (x \leftrightarrow y) \right]_{nn} \\ &= -i \left[\sigma^3 \left(\frac{\partial T_{(k_x, k_y)}^b}{\partial k_x} \right)^\dagger \sigma^3 \frac{\partial T_{(k_x, k_y)}^b}{\partial k_y} - (x \leftrightarrow y) \right]_{nn} \\ &= -\Omega_{n(k_x, k_y)}^b,\end{aligned}\quad (8)$$

which leads to zero thermal Hall conductivity because the first Brillouin zone (a hexagon centered at $\mathbf{k} = 0$ in the reciprocal space) is symmetric about $k_x = 0$. This completes the proof of Theorem 2. Equation (8) also implies that the Chern numbers $C_n \sim \sum_{\mathbf{k}} \Omega_{n\mathbf{k}}$ of the magnon bands in the b polarized state are zero.

The fact that $D_{\mathbf{k}}^b$ is even in k_x can be argued more heuristically without scrutinizing its explicit form, as follows. When the field is applied along the b direction, the spin Hamiltonian possesses a C_2 rotational symmetry about the b axis [44,45]. In principle, C_2 acts on both the spatial coordinates and the spins, $C_2: \mathbf{S}_i \rightarrow C_2^{-1} \mathbf{S}_{C_2(i)}$. However, in the b polarized state, the spin rotation part is effectively an identity operator since all spins lie exactly along the axis of rotation. In other words, C_2 only affects the spatial coordinates. In the reciprocal space, the C_2 symmetry translates into the invariance of $D_{\mathbf{k}}^b$ under $k_x \rightarrow -k_x$, and the rest of the proof follows.

Corollary.—Theorems 1 and 2 still hold if Heisenberg interactions, for instance $J(J_3)$ between the (third) nearest neighbors, are added to the KIT' model, due to the following reasons: (i) the Heisenberg interaction is proportional to an identity matrix, which is left invariant under a global rotation of spins; and (ii) the Heisenberg interaction is isotropic, namely it is the same along all bond directions. This indicates the robustness of Theorems 1 and 2 against the choice of spin model for α -RuCl₃, which is suggested to be a $JKT'\Gamma$ model [46] or a $JKTJ_3$ model [47–49].

Numerical Results.—Using two sets of interaction parameters $(K, \Gamma, \Gamma') = (-1, 0.2, -0.02)$ and $(-1, 0.3, -0.03)$ that are relevant to α -RuCl₃, we numerically evaluate the thermal Hall conductivity due to magnons, Eq. (2), in the a , b , and $-a$ polarized states. We first use classical simulated annealing [50,51] to obtain the critical fields to the polarized

states along the a , b , and $-a$ directions. We set the spin magnitude to be $S = 1/2$ in the linear spin wave theory and assume the strength of the Kitaev interaction to be $|K| = 80$ K [46,52] in the calculation of the thermal Hall conductivity. We also use the interlayer distance $d = 5.72$ Å of α -RuCl₃ [13,14,27,52].

We plot the thermal Hall conductivity as a function of temperature for the three polarized states in Figs. 2(a)–2(f). The field strengths h are chosen such that the system is indeed in the corresponding polarized states according to the classical model. We make two important observations from the results. First, κ_{xy} is positive, zero, and negative when the field is along the a , b , and $-a$ directions, respectively, which matches the experimentally observed signs of the thermal Hall conductivity. The sign structure is also consistent with Theorems 1 and 2. Second, in the a and $-a$ polarized states, κ_{xy}/T is of the order 0.1×10^{-3} W/K² m, which is comparable in magnitude to the thermal Hall signals experimentally measured at low temperatures [14,27,29]. For instance, the maximum value of κ_{xy}^{2D}/T for $(K, \Gamma, \Gamma') = (-1, 0.2, -0.02)$ is about $0.25 \times (\pi/6)(k_B^2/\hbar)$, which is half of the half-quantized value. While there is always a sign difference between the magnon thermal Hall conductivities of the a and $-a$ polarized states according to Theorem 1, the sign of κ_{xy} for each of these field directions really depends on the choice of the interaction parameters, such that κ_{xy} of the

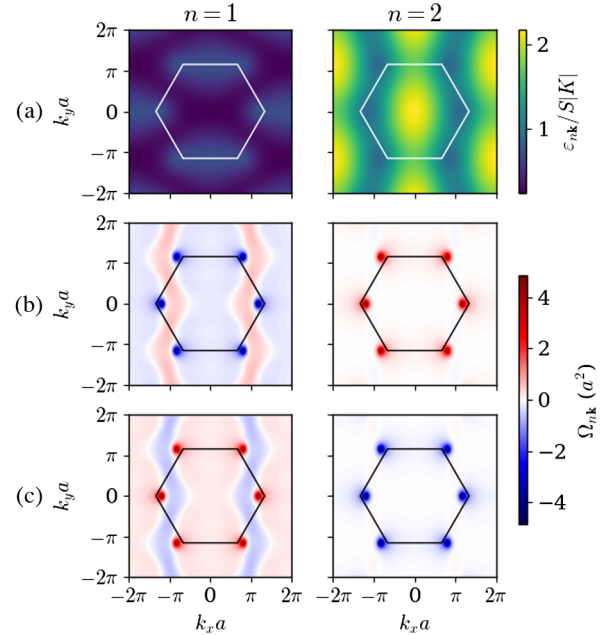


FIG. 3. (a) Linear spin wave dispersion $\epsilon_{n\mathbf{k}}$ of the a and $-a$ polarized states, and Berry curvature $\Omega_{n\mathbf{k}}$ in (b) the a polarized state and (c) the $-a$ polarized state, as functions of momentum \mathbf{k} . $n = 1$ and 2 are the band indices. The interaction parameters are chosen to be $K = -1$, $\Gamma = 0.2$, and $\Gamma' = -0.02$. The first Brillouin zone is indicated by a hexagon. The symbol a that appears in the units of \mathbf{k} and $\Omega_{n\mathbf{k}}$ is the lattice constant, not to be confused with the a direction.

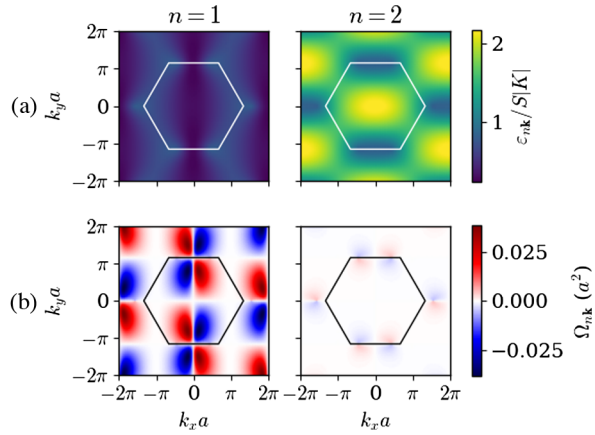


FIG. 4. (a) Linear spin wave dispersion ε_{nk} and (b) Berry curvature Ω_{nk} of the b polarized state as functions of momentum \mathbf{k} . The interaction parameters and notations used are same as in Figs. 3(a)–3(c).

a ($-a$) polarized state can be negative (positive) in a different parameter regime.

Furthermore, we numerically verify the symmetries discussed in the proofs of Theorems 1 and 2, by plotting the linear spin wave dispersion and the Berry curvature as functions of momentum [53–55] in the polarized states. When the field is flipped from the a direction to the $-a$ direction, the dispersion remains the same, as shown in Fig. 3(a). However, the Berry curvature changes sign, as shown in Figs. 3(b) and 3(c). By Eq. (2), the thermal Hall conductivity gains an overall minus sign. When the field is along the $\pm a$ direction, the Chern numbers of the lower ($n = 1$) and upper ($n = 2$) magnon bands are ∓ 1 and ± 1 , respectively, signifying their topological nontriviality. On the other hand, when the field is applied along the b direction, the linear dispersion is symmetric about $k_x = 0$, as shown in Fig. 4(a). However, the Berry curvature is antisymmetric about $k_x = 0$, as shown in Fig. 4(b), which results in cancellations of the summands in Eq. (2), eventually leading to a zero thermal Hall conductivity. The magnon bands are topologically trivial, i.e., they have zero Chern numbers.

Conclusion.—In this Letter, we have shown both analytically and numerically that the thermal Hall conductivity due to magnons in the field polarized states along the a , b , and $-a$ directions is positive, zero, and negative, respectively, which agrees with the experimentally observed sign structure [27,29] in the Kitaev material α - RuCl_3 . If the half-quantization plateau is present (absent), then the ground state may be the non-Abelian spin liquid (polarized state) with Majorana fermions (magnons). Most importantly, the thermal Hall effect does occur in both the non-Abelian spin liquid and the polarized state, and their thermal Hall conductivities have the same sign structure. Therefore, the sign structure alone cannot serve as a strong evidence for the non-Abelian spin

liquid. The ultimate test for the non-Abelian spin liquid will be the half-quantization of the thermal Hall conductivity at very low temperatures, where the magnon contribution vanishes.

Finally, we have only considered in-plane magnetic fields in this Letter. In the future, it will be interesting to investigate the implications of magnons to the thermal transport in Kitaev magnets under tilted and out-of-plane magnetic fields, where experiments also suggest the existence of unusual quasiparticles [14,27,52,56].

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