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## Theory of the Thermal Hall Effect in Quantum Magnets

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We present a theory of the thermal Hall effect in insulating quantum magnets, where the heat current is totally carried by charge-neutral objects such as magnons and spinons. Two distinct types of thermal Hall responses are identified. For ordered magnets, the intrinsic thermal Hall effect for magnons arises when certain conditions are satisfied for the lattice geometry and the underlying magnetic order. The other type is allowed in a spin liquid which is a novel quantum state since there is no order even at zero temperature. For this case, the deconfined spinons contribute to the thermal Hall response due to Lorentz force. These results offer a clear experimental method to prove the existence of the deconfined spinons via a thermal transport phenomenon.

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The ground state and low-energy excitations of correlated electronic systems are the subject of recent intensive interest, and the possible quantum liquid states are especially the focus both theoretically and experimentally [1–5]. For the quantum magnets, magnetic susceptibility, neutron scattering, and specific heat are the experimental tools to study this issue. In the conducting systems, on the other hand, charge transport properties also offer important clues to the novel electronic states such as the non-Fermi liquid or the quantum Hall liquid. Therefore, a natural question is whether there are any transport properties in insulating quantum magnets which provide insight into the ground state. To answer this question, we study in this Letter the thermal Hall effect theoretically and find several different mechanisms leading to the classification of the quantum magnets.

For a finite Hall response, time-reversal symmetry must be broken due to the magnetic field and/or magnetic ordering. The Hall effect in itinerant magnets, where the spin structure and conduction electron motion are coupled, has been studied extensively. In this case, in addition to the usual Lorentz force, the scalar (spin) chirality defined for three spins as  $\vec{S}_i \cdot (\vec{S}_j \times \vec{S}_k)$  plays an important role [6–8]. The scalar chirality acts as a fictitious magnetic flux for the conduction electrons and gives rise to a nontrivial topology of the Bloch wave functions, leading to the Hall effect. It is natural to expect that a similar effect occurs even in the localized spin systems for, e.g., the spin current [9]. Another important tool to detect charge-neutral modes is the thermal transport measurement. In low-dimensional magnets, the ballistic thermal transport property was predicted from the integrability of the one-dimensional Heisenberg model [10] and has been experimentally observed in  $\text{Sr}_2\text{CuO}_3$  [11]. In  $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$ , one of the possible candidates for two-dimensional quantum spin liquids [1], the thermal transport measurement was used as a probe to unveil the nature of low-energy spin excita-

tions [12]. The measurements have been limited to the longitudinal thermal conductivity so far. In this Letter we predict a nonzero thermal Hall conductivity, i.e., the Righi-Leduc effect, which will provide important information as described below.

First, we need to consider the influence of the external magnetic field on localized spin systems. In addition to the Zeeman coupling, we have the ring exchange process leading to the coupling between the scalar chirality and external magnetic fields. This coupling is derived from the  $t/U$  expansion for the Hubbard model at half filling with on-site Coulomb interaction  $U$  and complex hopping  $t_{ij} = te^{iA_{ij}}$  [13,14] and its explicit form is given by

$$H_{\text{ring}} = -\frac{24t^3}{U^2} \sin\Phi \vec{S}_i \cdot (\vec{S}_j \times \vec{S}_k), \quad (1)$$

where  $\Phi$  is the magnetic flux through the triangle formed by the sites  $i$ ,  $j$ , and  $k$  in a counterclockwise way. Since the coefficient is proportional to  $t^3/U^2$ , it is expected to be small. In the vicinity of the Mott transition, however, this coupling is not negligible. We first examine the effect of  $H_{\text{ring}}$  within the spin-wave approximation. Then we find that if the lattice geometry and the magnetic order satisfy certain conditions, the magnons can experience the fictitious magnetic field and there occurs the intrinsic thermal Hall effect, i.e., the thermal Hall conductivity  $\kappa^{xy}$  due to the anomalous velocity of the magnons. In this case,  $\kappa^{xy}$  is independent of the lifetime of magnons ( $\tau$ ), whereas the longitudinal one  $\kappa^{xx}$  depends on  $\tau$  [15,16]. It can be regarded as a bosonic analogue of the quantum Hall effect with zero net flux [17]. We also derive a TKNN-type formula [18] of the thermal Hall conductivity for a general free-bosonic Hamiltonian. It should be possible to apply this formula to the recently found phonon thermal Hall effect [19]. Finally, we consider the effect of  $H_{\text{ring}}$  in quantum spin liquids. Since there is no magnetic order in such a system, it has been proposed that deconfined fermi-

onic spinons exist. In contrast to the magnons, the spinons, which are the gauge dependent object, can feel the vector potential  $\vec{A}$  just as in the case of electrons, leading to the Landau level formation [14]. We propose a novel way to detect the spinon deconfinement via the thermal Hall effect measurement in a candidate of quantum spin liquid,  $\kappa$ -(ET)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub>.

*No-go theorem for the coupling to magnetic flux.*—Let us first consider the spin-wave expansion of Eq. (1) to find a system in which the intrinsic thermal Hall effect occurs. We consider the collinear ground state spin configurations. The fluctuation of the scalar chirality up to the second order in  $\delta\vec{S} \equiv \vec{S} - \langle\vec{S}\rangle$  is written as

$$\langle\vec{S}_i\rangle \cdot (\delta\vec{S}_j \times \delta\vec{S}_k) + \langle\vec{S}_j\rangle \cdot (\delta\vec{S}_k \times \delta\vec{S}_i) + \langle\vec{S}_k\rangle \cdot (\delta\vec{S}_i \times \delta\vec{S}_j).$$

Note here that the linear order terms in  $\delta\vec{S}$  vanish since  $\langle\vec{S}_i\rangle \times \langle\vec{S}_j\rangle = \vec{0}$ .

As an example, we consider the ferromagnetic Heisenberg model on a triangular lattice shown in Fig. 1(a) with an ordered moment  $S_0$  along  $\hat{y}$ . In this case, the quadratic terms in  $\delta\vec{S}$  from Eq. (1) always cancel. To explain this, let us focus on the edge  $\langle jk \rangle$ . From the upper triangle, this edge gives  $S_0(\delta\vec{S}_j \times \delta\vec{S}_k)^y$ . On the other hand, from the lower triangle, it gives  $S_0(\delta\vec{S}_k \times \delta\vec{S}_j)^y$ , which cancels out the former one. Since such a cancellation occurs on any edge,  $H_{\text{ring}}$  in Eq. (1) does not contribute to the spin-wave Hamiltonian to quadratic order. This observation leads us to conclude that such a cancellation occurs for any ferromagnetic model where each edge is shared by the equivalent cells such as plaquettes and triangles. A similar cancellation occurs for certain antiferromagnetic systems. An example is shown in Fig. 1(b). In this example, there are several different types of ring exchange processes, but again the cancellation between the cells sharing a link occurs for the collinear antiferromagnetic configuration. Finally, for noncollinear spin structures, we considered the 120° magnetic order on a

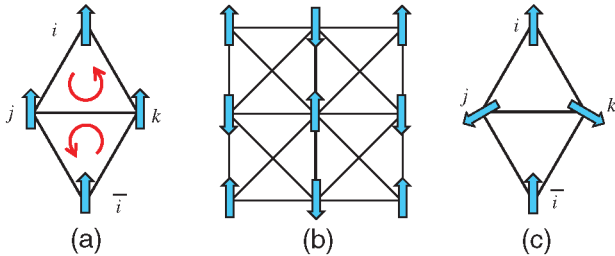


FIG. 1 (color online). (a) A portion of the triangular lattice with ferromagnetic order. The thick blue arrows indicate the directions of spins. The counterclockwise rotations (red arrows) indicate the order of sites in  $H_{\text{ring}}$ . We use this convention throughout this Letter. (b) Antiferromagnetic model on a square lattice. The interaction along the diagonal edge is smaller than that along the vertical or horizontal edge. (c) A portion of the triangular lattice with the 120° order.

triangular lattice [Fig. 1(c)] and conclude that the cancellation again occurs since ordered components of  $\vec{S}_i$  and that of  $\vec{S}_j$  are the same as shown in Fig. 1(c).

*Intrinsic thermal Hall effect in the spin-wave approximation.*—Once we understand the principles of the cancellation, it is rather easy to find an example where it does not occur, namely, when the link is shared by inequivalent cells. An example is the ferromagnetic model on the kagome lattice. In this case, the spin-wave Hamiltonian is influenced by the magnetic flux  $\Phi$ . We will develop below a theoretical formalism to calculate the thermal Hall conductivity in terms of the Kubo formula. For this purpose, we consider a general Hamiltonian for noninteracting bosons which can be regarded as a spin-wave Hamiltonian:

$$H = \sum_{j,\alpha} h(\vec{R}_{j\alpha}), \quad h(\vec{R}_{j\alpha}) = \frac{1}{2} \sum_{\vec{\delta}_\alpha} t_{\vec{\delta}_\alpha} b_{\vec{R}_{j\alpha} + \vec{\delta}_\alpha}^\dagger b_{\vec{R}_{j\alpha}} + \text{H.c.},$$

where  $b_{\vec{R}_{j\alpha}}$  annihilates a boson at the  $\alpha$ th site in the  $j$ th unit cell and  $\vec{\delta}_\alpha$  are vectors connecting  $\vec{R}_{j\alpha}$  and its neighboring sites. The hopping  $t_{\vec{\delta}_\alpha}$  is in general complex. In momentum space, the Hamiltonian is written as  $H = \sum_{\vec{k}} b_\alpha^\dagger(\vec{k}) \mathcal{H}_{\alpha,\beta}(\vec{k}) b_\beta(\vec{k})$ , where  $b_\alpha(\vec{k})$  is a Fourier transform of  $b_{\vec{R}_{j\alpha}}$  and repeated indices are summed over. The average energy current density is defined by  $\vec{j}_E \equiv i[H, \sum_{j,\alpha} \vec{R}_{j\alpha} h(\vec{R}_{j\alpha})]/V$ , where  $V$  is the total volume [20]. Using the Fourier transform of  $\sum_{\alpha} h(\vec{R}_{j\alpha})$  defined by  $h(\vec{q}) = \sum_{j,\alpha} e^{i\vec{q} \cdot \vec{R}_{j\alpha}} h(\vec{R}_{j\alpha})$ , the energy current density is rewritten as  $\vec{j}_E = \partial_{\vec{q}} [h(0), h(\vec{q})]_{\vec{q}=0}/V$ . Using this fact, the following convenient expression for  $\vec{j}_E$  is obtained:

$$\vec{j}_E = \frac{1}{2V} \sum_{\vec{k}} b_\alpha^\dagger(\vec{k}) (\partial_{\vec{k}} \mathcal{H}(\vec{k})^2)_{\alpha\beta} b_\beta(\vec{k}), \quad (2)$$

where the differential operator  $\partial_{\vec{k}}$  acts only on  $\mathcal{H}(\vec{k})^2$ . We introduce the spin-wave basis  $|u_\alpha(\vec{k})\rangle$  which diagonalizes  $\mathcal{H}(\vec{k})$  with eigenvalues  $\omega_\alpha(\vec{k})$ . It is important to note that even in this basis  $\vec{j}_E$  is not diagonal. In addition to the expected diagonal term  $\frac{d\omega_\alpha}{dk} \omega_\alpha(\vec{k})$ , there are off-diagonal terms which can be thought of as arising from anomalous velocities. As we see below, these terms are responsible for  $\kappa^{xy}$ , just as in the case of the intrinsic anomalous Hall effect in metals [21].

Starting from the Kubo formula, the following expression analogous to the TKNN formula [18] can be obtained for the thermal Hall conductivity  $\kappa^{xy}$ :

$$\kappa^{xy} = -\frac{1}{2T} \text{Im} \sum_{\alpha} \int_{\text{BZ}} \frac{d^2k}{(2\pi)^2} n_\alpha(\vec{k}) \times \langle \partial_{k_x} u_\alpha(\vec{k}) | (\mathcal{H}(\vec{k}) + \omega_\alpha(\vec{k})^2) | \partial_{k_y} u_\alpha(\vec{k}) \rangle, \quad (3)$$

for the noninteracting spin waves (free bosons) where the integral is over the Brillouin zone (BZ), and  $n_\alpha(\vec{k}) = (e^{\beta\omega_\alpha(\vec{k})} - 1)^{-1}$  is the Bose distribution function. The in-

tegrand in Eq. (3) is independent of a  $\vec{k}$ -dependent phase change of the spin-wave basis functions  $|u_\alpha(\vec{k})\rangle$  in a similar way as for the orbital magnetization [22].

*Thermal Hall effect in kagome ferromagnet.*—We now apply the above formula to the ferromagnetic model on the kagome lattice. The Hamiltonian is given by  $H = \sum_{\Delta, \nabla} h_{\Delta} + h_{\nabla}$  with

$$h_{\Delta/\nabla} = -J(\vec{S}_i \cdot \vec{S}_j + \vec{S}_j \cdot \vec{S}_k + \vec{S}_k \cdot \vec{S}_i) - \frac{K}{S} \vec{S}_i \cdot (\vec{S}_j \times \vec{S}_k),$$

where  $-J$  is the ferromagnetic exchange coupling,  $K$  is proportional to  $\sin\Phi$  according to Eq. (1), and the sum is taken over all the triangles in the kagome lattice [see Fig. 2(a)]. Again, note that the sites  $i$ ,  $j$ , and  $k$  form a triangle ( $\Delta$  or  $\nabla$ ) in a counterclockwise way.

Using the Holstein-Primakoff transformation [ $S_j^+ = (2S - n_j)^{1/2} b_j$ ,  $S_j^- = b_j^\dagger (2S - n_j)^{1/2}$ ,  $S_j^z = S - n_j$  with  $n_j = b_j^\dagger b_j$ ], we obtain the spin-wave Hamiltonian as

$$H_{\text{SW}} = 4JS \sum_j n_j - S \sqrt{J^2 + K^2} \sum_{\langle j,k \rangle} (e^{-i\phi/3} b_j^\dagger b_k + \text{H.c.}),$$

where  $\tan(\phi/3) = K/J$  and the sum is taken over all the nearest neighbor bonds. The Fourier transform of the Hamiltonian is given by  $\mathcal{H}(\vec{k}) = 4JS - 2JS[\cos(\phi/3)]^{-1} \Lambda(\vec{k}, \phi)$  with

$$\Lambda(\vec{k}, \phi) = \begin{pmatrix} 0 & \cos k_1 e^{-i\phi/3} & \cos k_3 e^{i\phi/3} \\ \cos k_1 e^{i\phi/3} & 0 & \cos k_2 e^{-i\phi/3} \\ \cos k_3 e^{-i\phi/3} & \cos k_2 e^{i\phi/3} & 0 \end{pmatrix},$$

where  $k_i = \vec{k} \cdot \vec{a}_i$  with  $\vec{a}_1 = (-1/2, -\sqrt{3}/2)$ ,  $\vec{a}_2 = (1, 0)$ , and  $\vec{a}_3 = (-1/2, \sqrt{3}/2)$  as shown in Fig. 2(a). The dispersions of three bands [ $0 \leq \omega_1(\vec{k}) \leq \omega_2(\vec{k}) \leq \omega_3(\vec{k})$ ] for  $\phi = \pi/3$  are shown in Fig. 2(b). In the limit of low temperature and weak magnetic field, the dominant contri-

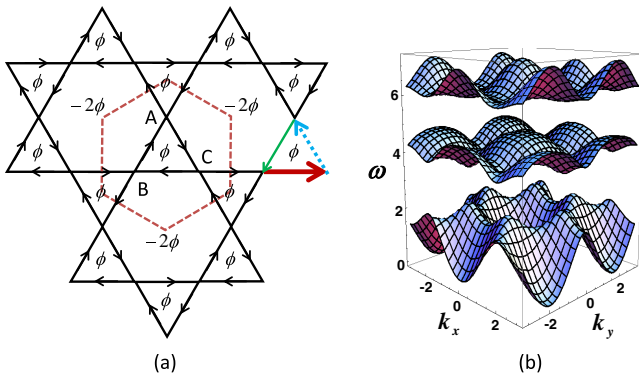


FIG. 2 (color online). (a) The unit cell of the kagome lattice (the area enclosed by the dotted line). The vectors  $\vec{a}_i$  ( $i = 1, 2$ , and  $3$ ) correspond to green (thin line), red (thick line), and blue (dotted line), respectively. The arrows on the edges indicate the sign of the phase factor  $e^{i\phi/3}$  (see main text). The fictitious fluxes through triangle and hexagon are  $\phi$  and  $-2\phi$ , respectively. (b) Dispersions of bands in the case of  $\phi = \pi/3$  with  $JS = 1$ .

bution to the integral in Eq. (3) comes from  $\alpha = 1$  (lowest band) and small  $|\vec{k}|$  due to the Bose factor  $n_\alpha(\vec{k})$ . By an explicit calculation [23], we find that  $\langle \partial_{k_x} u_1(\vec{k}) | \partial_{k_y} u_1(\vec{k}) \rangle \sim -i\phi |\vec{k}|^2 / (27\sqrt{3})$  around  $\vec{k} = 0$  and obtain

$$\kappa^{xy} \sim \frac{(6JS)^2}{2T} \int_0^\infty dk \frac{k}{2\pi e^{\beta JS k^2} - 1} \left( \frac{\phi k^2}{27\sqrt{3}} \right) = \frac{\pi\phi}{36\sqrt{3}} T, \quad (4)$$

where we have replaced the integration over the BZ with that over all  $\vec{k}$ . In this way, a nonzero thermal Hall conductivity is indeed realized by the coupling between the scalar chirality and the magnetic field in the ferromagnetic kagome lattice.

*Thermal Hall effect in quantum spin liquids.*—As discussed above, the spin Hamiltonian contains the magnetic flux  $\Phi$ , and the spin waves or magnons are influenced by the magnetic field only through the off-diagonal matrix elements of the thermal currents, corresponding to the intrinsic anomalous Hall effect. This is in sharp contrast to the case of electrons which is coupled to the vector potential  $\vec{A}$ , and the usual Hall effect due to the Lorentz force occurs there. In this respect, it is interesting to note that the spin operator  $\vec{S}_i$  can be represented by the fermion operators  $f_{i\sigma}^\dagger$  and  $f_{i\sigma}$  (called spinons with spin 1/2) as  $\vec{S}_i = \sum_{\sigma, \sigma'} f_{i\sigma}^\dagger \vec{\sigma} f_{i\sigma'}$  [ $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ : Pauli matrices,  $\sigma, \sigma' = \uparrow, \downarrow$ ] with the constraint  $\sum_{\sigma} f_{i\sigma}^\dagger f_{i\sigma} = 1$ . In this representation, the exchange interaction  $J\vec{S}_i \cdot \vec{S}_j$  can be written as  $-J\chi_{ij}^\dagger \chi_{ij}/2$  with  $\chi_{ij} = \sum_{\sigma} f_{i\sigma}^\dagger f_{j\sigma}$ . This  $\chi_{ij}$  is called the order parameter of the resonating valence bond, which describes the singlet formation between the two spins  $\vec{S}_i$  and  $\vec{S}_j$ . In the mean field approximation for  $\chi_{ij}$ , the free fermion model for  $f_{i\sigma}^\dagger, f_{i\sigma}$  emerges [24]. The phase  $a_{ij}$  of the order parameter  $\langle \chi_{ij} \rangle = |\langle \chi_{ij} \rangle| e^{ia_{ij}}$  and the Lagrange multiplier  $a^0$  to impose the constraint above constitutes the gauge field, which is coupled to the spinons. In the confining phase of this gauge field, two spinons are bound to form a magnon. On the other hand, in the deconfining phase, the spinons behave as nearly free quasiparticles. The latter case is realized in some of the quantum spin-liquid states [24]. A similar state has been obtained also for the Hubbard model [25], which contains the gapped charge excitations. This charge degrees of freedom are represented by the U(1) phase factor  $e^{i\theta_i}$ ; i.e., the electron operator  $c_{i\sigma}$  is decomposed into the product  $f_{i\sigma} e^{-i\theta_i}$ , which is coupled to  $a_{ij} - A_{ij}$  where  $A_{ij}$  is the vector potential (Peierls phase) corresponding to the magnetic flux  $\Phi$  [25]. Then, the Maxwell term  $\mathcal{L}_g = (1/g) \times \int dr \sum_{\mu\nu} (\mathcal{F}_{\mu\nu} - F_{\mu\nu})^2$  ( $g$ : coupling constant,  $\mathcal{F}_{\mu\nu} = \partial_\mu a_\nu - \partial_\nu a_\mu$ ,  $F_{\mu\nu} = \partial_\mu A_\nu - \partial_\nu A_\mu$ ) is generated by integrating over the charge degrees of freedom. To summarize, the spinons are described by the Lagrangian

$$\mathcal{L} = \sum_{j,\sigma} f_{j\sigma}^\dagger (\partial_\tau - ia_j^0 - \mu) f_{j\sigma} - \sum_{j,k} t_{jk} e^{ia_{jk}} f_{j\sigma}^\dagger f_{k\sigma} + \mathcal{L}_g.$$

Following the previous works [14,25], we take the spinon metal with a Fermi surface as a candidate for the 2D quantum spin liquid realized in  $\kappa$ -(ET)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub> [12]. In a magnetic field  $F_{xy} = B_z$ , the average of the gauge flux  $\langle \mathcal{F}_{xy} \rangle = cF_{xy}$  is induced with  $c$  a constant of the order of unity because of the coupling between  $\mathcal{F}_{xy}$  and  $F_{xy}$  in  $\mathcal{L}_g$  [14,24]. Therefore, the spinons are subject to the effective magnetic field  $\langle \mathcal{F}_{xy} \rangle$  and to the Lorentz force.

Let us first estimate the spinon lifetime  $\tau$  from the recent thermal transport measurements in that material. The longitudinal thermal conductivity is obtained from the Wiedemann-Franz law by assuming a Fermi liquid of spinons:

$$\kappa_{\text{sp}}^{\text{xx}} = 2 \frac{\pi^2}{3} \left( \frac{\varepsilon_F}{\hbar} \tau \right) \frac{k_B^2 T}{h} \frac{1}{d}, \quad (5)$$

where  $\varepsilon_F$  is the Fermi energy and  $d \sim 16 \text{ \AA}$  is the inter-layer distance. After a subtraction of the phonon contribution,  $\kappa_{\text{sp}}^{\text{xx}}$  is estimated to be  $\sim 0.02 \text{ W K}^{-1} \text{ m}^{-1}$  at  $T = 0.3 \text{ K}$  [12]. We obtain  $\varepsilon_F \tau / \hbar = 56.5$  and with  $\varepsilon_F = J \sim 250 \text{ K}$ , we estimate  $\tau \sim 1.72 \times 10^{-12} \text{ s}$ . Next we examine  $\kappa_{\text{sp}}^{\text{xy}}$ . As has been shown in [14], the gauge flux for spinons is comparable to the applied magnetic flux and hence  $\kappa_{\text{sp}}^{\text{xy}} \sim (\omega_c \tau) \kappa_{\text{sp}}^{\text{xx}}$ , where  $\omega_c = eB/m_c$  is the cyclotron frequency with the effective mass of spinon  $m_c$ . Estimating  $m_c \sim \hbar^2 / (Ja^2)$  with assuming the lattice spacing  $a \sim 10 \text{ \AA}$ , we obtain  $\omega_c \tau \sim 0.086B$  with  $B$  being measured in tesla. Therefore, the thermal Hall angle  $\kappa_{\text{sp}}^{\text{xy}} / \kappa_{\text{sp}}^{\text{xx}} \sim \omega_c \tau$  becomes of the order of 0.1, which is easily measurable, with a weak magnetic field  $B \sim 1 \text{ T}$  such that the spin-liquid ground state is not disturbed. Also note that compared with the intrinsic thermal Hall effect discussed above, the magnitude of this Lorentz-force driven thermal Hall conductivity is much larger by the factor of  $\sim (\varepsilon_F \tau / \hbar)^2$ . Therefore, the observation of the thermal Hall effect is a clear signature of such deconfined spinons in the spin liquid, and experiments on  $\kappa$ -(ET)<sub>2</sub>Cu<sub>2</sub>(CN)<sub>3</sub> are highly desirable.

Another important difference between the spinon contribution and the intrinsic term is that the spinons are diffusive and see the field  $\vec{A}$ . Thus, in a small sample one can expect mesoscopic effects such as universal conductance fluctuations of the thermal conductivity as a function of  $B$ . Using the Wiedemann-Franz law, we expect the relative fluctuation in  $\kappa^{\text{xx}}$  and  $\kappa^{\text{xy}}$  to be of order  $\hbar / (\varepsilon_F \tau)$  for each coherent volume with dimension  $\sqrt{\ell \ell_{\text{in}}}$  where  $\ell = v_F \tau$  and  $\ell_{\text{in}} = v_F \tau_{\text{in}}$  and  $\tau_{\text{in}}$  is some inelastic scattering time much longer than  $\tau$  at low temperatures. The fluctuation is reduced by  $\sqrt{N}$  if the sample contains  $N$  coherent volumes. We estimate the elastic mean free path  $\ell$  to be  $400 \text{ \AA}$ , so that at low temperatures this effect may be observable in micron-scale samples.

In conclusion, we have studied theoretically the thermal Hall effect in the quantum spin systems induced by the external magnetic field. There are three cases: i.e., (i) no

thermal Hall effect, (ii) intrinsic thermal Hall effect by the magnons, and (iii) large thermal Hall effect due to the Lorentz force. Case (i) corresponds to most of the conventional (anti)ferromagnets on triangular, square, and cubic lattices, while case (ii) corresponds to the magnets on a particular lattice structure such as kagome, and case (iii) corresponds to the spin liquid with deconfined spinons. Therefore, the thermal Hall effect offers a unique experimental method to gain an important insight on the ground state or low-energy excitations of the quantum magnets.

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