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Thermal Hall Effect of Spin Excitations in a Kagome Magnet

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At low temperatures, the thermal conductivity of spin excitations in a magnetic insulator can exceed that of phonons. However, because they are charge neutral, the spin waves are not expected to display a thermal Hall effect. However, in the kagome lattice, theory predicts that the Berry curvature leads to a thermal Hall conductivity \( \kappa_{xy} \). Here we report observation of a large \( \kappa_{xy} \) in the kagome magnet Cu(1,3-bdc) which orders magnetically at 1.8 K. The observed \( \kappa_{xy} \) undergoes a remarkable sign reversal with changes in temperature or magnetic field, associated with sign alternation of the Chern flux between magnon bands. The close correlation between \( \kappa_{xy} \) and \( \kappa_{xx} \) firmly precludes a phonon origin for the thermal Hall effect.

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In a magnetic insulator, experiments on the magnon heat current can potentially yield incisive information on novel quantum magnets. An example is the chiral magnet [1], in which unusual spin textures engender a finite Berry curvature \( \Omega(k) \). \( \Omega(k) \) acts like a magnetic field in \( k \) space. In its presence, a magnon wave packet subject to a potential gradient acquires an anomalous velocity perpendicular to the gradient [2–4]. The most surprising outcome [1,5,6] is that the neutral heat current can be deflected left or right by a physical magnetic field \( H \) as if a Lorentz force were present. The predicted thermal Hall conductivity \( \kappa_{xy} \) was observed in two recent experiments on the ordered magnet Lu2V2O7 [7] and the frustrated quantum magnet Tb2Tl2O7 [8]. However, to test more incisively the role of \( \Omega(k) \) and to exclude a phononic origin [9], we need results that can be compared with microscopic calculations based on \( \Omega(k) \). An interesting prediction based on the Chern number sign alternation between magnon bands is the induced sign change in \( \kappa_{xy} \) when either temperature or field is varied. Here we report measurements on the planar kagome magnet Cu(1,3-benzenedicarboxylate) [or Cu(1,3-bdc)] [10–12] which can be compared with calculations on the same material [13]. The close correlation between \( \kappa_{xy} \) and \( \kappa_{xx} \) precludes identifying the former with phonons.

In magnets with strong spin-orbit interaction, competition between the Dzyaloshinskii-Moriya (DM) exchange \( D \) and the Heisenberg exchange \( J \) can engender canted spin textures with long-range order (LRO). Katsura, Nagaosa, and Lee (KNL) [1] predicted that, in the kagome and pyrochlore lattices, the competition can lead to a state with extensive chirality \( \chi = \mathbf{S}_i \cdot \mathbf{S}_j \times \mathbf{S}_k \) (\( \mathbf{S}_i \) is the spin at site \( i \)) and a large thermal Hall effect. Subsequently, Matsumoto and Murakami (MM) [5,6] amended KNL’s calculation using the gravitational-potential approach [14,15] to relate \( \kappa_{xy} \) directly to the Berry curvature. In the boson representation of the spin Hamiltonian, \( \chi \) induces a complex “hopping” integral \( t = \sqrt{\frac{J}{2} + D^2 e^{i\phi}} \) with tan \( \phi = D/J \) [Fig. 1(a), inset] [1,5,13]. Hence as they hop between sites, the bosons accumulate the phase \( \phi \), which implies the existence of a vector potential \( \mathbf{A}(k) \) permeating \( k \) space. The Berry curvature \( \Omega(k) = \nabla_k \times \mathbf{A}(k) \) imparts an anomalous velocity to magnons, leading to a thermal Hall conductivity \( \kappa_{xy} \). Each magnon band \( n \) contributes a term to \( \kappa_{xy} \) with a sign determined by the integral of \( \Omega(k) \) over the Brillouin zone (the Chern number). Recently, Lee, Han, and Lee (LHL) [13] calculated how \( \kappa_{xy} \) undergoes sign changes as the occupancy of the bands changes with \( T \) or \( B \).

The kagome magnet Cu(1,3-bdc) is comprised of stacked kagome planes separated by \( d = 7.97 \) Å [10–12]. The spin-\( \frac{1}{2} \) Cu\( ^{2+} \) moments interact via an in-plane ferromagnetic exchange \( J = 0.6 \) meV (details in the Supplemental Material [16]).

As we cool the sample in zero \( B \), the thermal conductivity \( \kappa \) (nearly entirely from phonons) initially rises to a very broad peak at 45 K [Fig. 1(a)]. Below the peak, \( \kappa \) decreases rapidly as the phonons freeze out. Starting near 10 K, the spin contribution \( \kappa' \) becomes apparent. As shown in Fig. 1(b), this leads to a minimum in \( \kappa \) near \( T_C (1.85 \) K) followed by a large peak at \( \sim 2T_C \). Factoring out the entropy, we find that \( \kappa/T \) (red curve) increases rapidly below \( T_C \). This reflects the increased stiffening of the magnon bands as LRO is established. Below 800 mK, the increase in \( \kappa/T \) slows to approach saturation. The open black circles represent the phonon conductivity \( \kappa_{ph} \) deduced from the large-\( B \) values of \( \kappa_{xx}(T,H) \) (see below). Likewise, \( \kappa_{ph}/T \) is plotted as open red circles. The
difference $\kappa - \kappa_{\text{ph}}$ is the estimated thermal conductivity of magnons $\kappa^*$ in zero $B$.

Given that Cu(1,3-bdc) is a transparent insulator, it exhibits a surprisingly large thermal Hall conductivity (Fig. 2). Above $T_C$, the field profile of $\kappa_{\text{xy}}$ is nonmonotonic, showing a positive peak at low $B$, followed by a zero crossing at higher $B$ [see curve at 2.78 K in Fig. 2(a)]. We refer to a positive $\kappa_{\text{xy}}$ as “$p$ type.” Below $T_C$, an interesting change of sign is observed (curves at 1.74 and 0.82 K). The weak hysteresis, implying a coercive field $< 1500$ Oe at the lowest temperatures, is discussed in the Supplemental Material [16]. This sign change is investigated in greater detail in sample 3 [we plot $\kappa_{\text{xy}}/T$ in Figs. 2(b) and 2(c)]. The curves of $\kappa_{\text{xy}}/T$ above $T_C$ are similar to those in sample 2. As we cool towards $T_C$, the peak field $H_p$ decreases rapidly, but remains resolvable below $T_C$ down to 1 K [Fig. 2(c)]. However, as $T \to 0.6$ K, the $p$-type response is eventually dominated by an $n$-type contribution. The thermal Hall response in the limit $B \to 0$, measured by the quantity $[\kappa_{\text{xy}}/BT]_0$ plotted in Fig. 2(d), closely correlates with the growth of $\kappa_{\text{xy}}$ below $T_C$.

To relate the thermal Hall results to magnons, we next examine the effect of $B$ on the longitudinal thermal conductivity $\kappa_{\text{xx}}$. As shown in Fig. 3(a), $\kappa_{\text{xx}}$ is initially $B$ independent for $T > 10$ K, suggesting negligible interaction between phonons and the spins. The increasingly strong $B$ dependence observed below 4 K is highlighted in Fig. 3(b). Despite the complicated evolution of the profiles, all the curves share the feature that the $B$-dependent part is exponentially suppressed at large $B$, leaving a $B$-independent “floor” which we identify with $\kappa_{\text{ph}}(T)$ [plotted as open symbols in Fig. 1(b)]. Subtracting the floor allows the thermal conductivity due to spins to be defined as $\kappa_{\text{xx}}(T, H) = \kappa_{\text{xy}}(T, H) - \kappa_{\text{ph}}(T)$. The exponential suppression becomes apparent in the scaled plot of $\kappa_{\text{xx}}/T$ vs $B/T$ [Fig. 3(c)]. The asymptotic form at large $B$ in all curves depends only on $B/T$.

In the interval 0.9 K $\to T_C$, $\kappa_{\text{xx}}$ displays a $V$-shaped minimum at $B = 0$ followed by a peak at the field $H_p(T)$. Since $\kappa^* (at B = 0)$ falls rapidly within this interval due to softening of the magnon bands [see Fig. 1(b)], we associate the $V$-shaped profile with stiffening of the magnon bands by the applied $B$. At low enough $T (< 0.8$ K), this stiffening is unimportant and the curves are strictly monotonic. We find that they follow the same universal form. To show this, we multiply each curve by a $T$-dependent scale factor $s(T)$ and plot them on a semilog scale in Fig. 3(d). In the limit of large $B$, the universal curve follows the activated form

$$\kappa_{\text{xx}}^* \to T e^{-\beta\Delta},$$

with the Zeeman gap $\Delta = g\mu_B B$ where $\beta = 1/k_BT$, $\mu_B$ is the Bohr magneton, and $g$ the $g$ factor. The inferred value of $g$ ($\sim 1.6$) is consistent with the Zeeman gap measured in a recent neutron scattering experiment.

For comparison, we have also plotted $-\kappa_{\text{xy}}/T$ (at 0.47 K) in Fig. 3(d). Within the uncertainty, it also decreases exponentially at large $B$ with a slope close to $\Delta$. Hence, the exponential suppression of the magnon population resulting from $\Delta$ is evident in both $\kappa_{\text{xy}}^*$ and $\kappa_{\text{xy}}$.

LHL [13] have calculated $\kappa_{\text{xy}}(T, B)$ applying the Holstein-Primakoff (HP) representation below and above $T_C$, and Schwinger bosons (SBs) above $T_C$. In the ordered phase, the HP curves capture the sign changes observed in

FIG. 1 (color online). The in-plane thermal conductivity $\kappa$ (in zero $B$) measured in the kagome magnet Cu(1,3-bdc). At 40–50 K, $\kappa$ displays a broad peak followed by a steep decrease reflecting the freezing out of phonons [panel (a)]. The spin excitation contribution becomes apparent below 2 K. The inset is a schematic of the kagome lattice with the LRO chiral state [1]. The arrows on the bonds indicate the direction of advancing phase $\phi = \tan^{-1}D/J$. Panel (b) plots $\kappa$ (black symbols) and $\kappa/T$ (red) for $T < 4.5$ K. Below the ordering temperature $T_C = 1.8$ K, the magnon contribution to $\kappa$ appears as a prominent peak that is very $B$ dependent. Values of $\kappa$ and $\kappa/T$ at large $B$ (identified with the phonon background) are shown as open symbols.
\(\kappa_{xy}(T,H)\): a purely \(n\)-type curve at the lowest \(T\) and, closer to \(T_C\), a sign-change induced by a \(p\)-type term. Moreover, the calculated curves at each \(T\) exhibit the high-field suppression, in agreement with Fig. 3(d). For sample 3, the peak values of \(\kappa_{xy}^{(p)}\) agree with the HP curves (0.04 K at \(T = 0.4\) K; 0.2 K at 4.4 K). In the paramagnetic region, however, our field profiles disagree with the SB curves. Above \(T_C\), \(\kappa_{xy}\) is observed to be \(p\)-type at all \(B\) whereas the SB curves are largely \(n\)-type apart from a small window at low \(B\). The comparison suggests that the HP approach is a better predictor than the SB representation even above \(T_C\).

A weak \(\kappa_{xy}\) was reported in Ref. [9] and identified with phonons. A phonon Hall effect based on the Berry curvature was calculated in Refs. [17,18]. Here, however, the evidence is compelling that \(\kappa_{xy}\) arises from spin excitations. The close correlation between the profiles of \(\kappa_{xy}\) and \(\kappa_x\) vs \(T\) implies that they come from the same heat carriers. Moreover, the plots in Fig. 3(d) and Eq. (1) show that, when a gap opens, both the longitudinal and Hall channels are suppressed at the same rate versus \(B\). To us this is firm evidence for spin excitations—the phonon current cannot be switched off by a gap opening in the spin spectrum (we discuss this further in the Supplemental Material [16]).

In addition to confirming the existence of a large \(\kappa_{xy}\) in the kagome magnet, the measured \(\kappa_{xy}\) can be compared with calculations. For chiral magnets, \(\kappa_{xy}\) is capable of probing incisively the effect of the Berry curvature on transport currents.

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FIG. 3 (color online). The effect of field $B$ on $\kappa_{xx}$ and scaling behavior at low $T$, for sample 3. The curves in panel (a) show that the $B$ dependence of $\kappa_{xx}$ is resolved (in the range $|B| < 14$ T) only at $T \sim 6.5$ K. The expanded scale in panel (b) shows that, near $T_C$ (1.8 K), $\kappa_{xx}$ has a nonmonotonic profile with a $V$-shaped minimum at $B = 0$ (identified with stiffening of the magnon bands by the field). Below 1 K, however, $\kappa_{xx}$ has a strictly monotonic profile that terminates in a sharp cusp peak as $B \to 0$. At each $T < T_C$, the constant “floor” profile at large $B$ is identified with $\kappa_{ph}$. The pattern in panel (b) simplifies when plotted as $\kappa_{xx}/T$ vs $B/T$ [panel (c)]. Multiplying by a scaling factor $s(T)$ collapses all the curves below 1 K to a “universal” curve, shown on log scale in panel (d). The slope at large $B$ gives a Zeeman gap with $g = 1.6$. The Hall curve $-\kappa_{xy}/T$ has a similar slope at large $B$.


