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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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.

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.

The difference $\kappa - \kappa_{ph}$ 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 $\kappa_{xy}$ 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 $\kappa_{xy}$ 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 $\kappa_{xy}/T$ in Figs. 2(b) and 2(c)]. The curves of $\kappa_{xy}/T$ 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 $T \rightarrow 0.6$ K, the $p$-type response is eventually dominated by an $n$-type contribution. The thermal Hall response in the limit $B \rightarrow 0$, measured by the quantity $[\kappa_{xy}/BT_0]_0$ plotted in Fig. 2(d), closely correlates with the growth of $\kappa$ below $T_C$.

To relate the thermal Hall results to magnons, we next examine the effect of $B$ on the longitudinal thermal conductivity $\kappa_{zx}$. As shown in Fig. 3(a), $\kappa_{zx}$ 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 $\kappa_{ph}(T)$ [plotted as open symbols in Fig. 1(b)]. Subtracting the floor allows the thermal conductivity due to spins to be defined as $\kappa_{zx}(T, H) = \kappa_{xy}(T, H) - \kappa_{ph}(T)$. The exponential suppression becomes apparent in the scaled plot of $\kappa_{zx}/T$ 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 $\rightarrow T_C$, $\kappa_{zx}$ 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

$$
\kappa_{zx}^* \rightarrow T e^{-\beta \Delta},
$$

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

For comparison, we have also plotted $-\kappa_{xy}/T$ (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 $\kappa_{zx}$ and $\kappa_{xy}$.

LHL [13] have calculated $\kappa_{xy}(T, B)$ 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
The thermal Hall conductivity $\kappa_{xy}$ measured in Cu(1,3-bdc). In panel (a), we plot the strongly nonmonotonic profiles of $\kappa_{xy}$ vs $B$ in sample 2. The dispersionlike profile changes sign below $\sim$1.7 K. The right scale gives $\kappa^{2D}/(kB/h)$ (per plane) obtained by multiplying $\kappa_{xy}$ by $\hbar/k_B = 443.2$ (SI units). Panels (b) and (c) show corresponding curves in sample 3 (now plotted as $\kappa_{xy}/T$). Above $T_C$ [panel (b)], $\kappa_{xy}/T$ is $p$ type. The behavior below 1.90 K is shown in panel (c). At 1.09 K, the $n$-type contribution appears in weak $B$, and eventually changes $\kappa_{xy}/T$ to $n$-type at all $B$. Right scale in (c) reports $\kappa^{2D}/(Tk_B/h)$. In panel (d), we plot the $T$ dependence of the quantity $|\kappa_{xy}/TB_0|$ which measures the thermal Hall response in the limit $B \to 0$. The $T$ dependence of $|\kappa_{xy}/TB_0|$ closely correlates with $\kappa_{xx}$ vs $T$ (aside from the sign change).

$\kappa_{xy}(T, H)$: 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 $\kappa_{xy}^{2D}$ 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 $\kappa_s$ 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 $B$.

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