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The 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 a frustrated quantum magnet Tb2Ti2O7 [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 confront 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, Nagoasa and Lee (KNL) [1] predicted that, in the Kagome and pyrochlore lattices, the competition can lead to a state with extensive chirality \( \chi = S_i \cdot S_j \times S_k \) (\( S_i \) 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 \( t = \sqrt{J^2+D^2} \cdot e^{i\phi} \) with \( \tan \phi = D/J \) (Fig. 1A, inset) [1, 5, 13]. Hence as they hop between sites, the bosons accumulate the phase \( \phi \), which implies the existence of a vector potential \( A(k) \) permeating \( k \) space. The Berry curvature \( \Omega(k) = \nabla_k \times A(k) \) 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 \( d = 7.97 \) A [10–12]. The spin-\( \frac{1}{2} \) Cu\( ^{2+} \) moments interact via an in-plane ferromagnetic exchange \( J = 0.6 \) meV (details in supplementary information SI).

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. 1A). Below the peak, \( \kappa \) decreases rapidly as the phonons freeze out. Starting near 10 K, the spin contribution \( \kappa^s \) becomes apparent. As shown in Fig. 1B, this leads to a minimum in \( \kappa \) near \( T_C \) (1.85 K) followed by a large peak at \( \sim \frac{1}{2} T_C \). 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 \( \kappa_{xy}(T, H) \) (see below). Likewise, \( \kappa_{ph}/T \) is plotted as open red circles. The difference \( \kappa - \kappa_{ph} \) is the estimated thermal conductivity of magnons \( \kappa^s \) 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 non-monotonic, showing a positive peak at low \( B \), followed by a zero-crossing at higher \( B \) (see curve at 2.78 K in Fig.
resulting from $\Delta$ is evident in both $\kappa$ the exponential suppression of the magnon population $B$ exponentially at large $s$ in a recent neutron scattering experiment. The value of $g$ is the Bohr magneton, and $T$ is the temperature.

Holstein-Primakoff (HP) representation below and above $B$. In the limit of large-$g\mu_B$, we associate the magnon bands by the applied $H_p$. Since $B/T$ is falling rapidly within this interval due to softening of the magnon bands (see Fig. 1B), 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 semilog scale in Fig. 3D. In the limit of large-$B$, the universal curve follows the activated form

$$\kappa_{xx}^s \rightarrow T e^{-\beta\Delta},$$

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 $g$ ($\sim 1.6$) 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. 3D. 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 $\kappa_{xx}^s$ 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 (SB) above $T_C$. In the ordered phase, the HP curves capture the sign changes observed in $\kappa_{xy}(T, H)$: 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. 3D. For Sample 3, the peak values of $\kappa_{xy}^{sp}$ 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. [16, 17]. Here, however, the evidence is compelling that $\kappa_{xy}$ arises from spin excitations. The close correlation between the profiles of $\kappa_{xy}$ and $s$ vs. $T$ implies that they come from the same heat carriers. Moreover, the plots in Fig. 3D 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 SI).

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. 1: 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. 2: The thermal Hall conductivity $\kappa_{xy}$ measured in Cu(1,3-bdc). In Panel A, we plot the strongly non-monotonic profiles of $\kappa_{xy}$ vs. $B$ in Sample 2. The dispersion-like profile changes sign below $\sim 1.7$ K. The right scale gives $\kappa^{2D}/(k_B^2/\hbar)$ (per plane) obtained by multiplying $\kappa_{xy}$ by $d\hbar/k_B^2 = 443.2$ (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 $\kappa^{2D}_{xy}/(Tk_B^2/\hbar)$. 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 $\kappa_{xx}^S$ vs. $T$ (aside from the sign change).
FIG. 3: 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 non-monotonic 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 $B$. 