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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 \(\text{Cu}(1,3\text{-bdc})\) 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 \(\text{Lu}_2\text{V}_2\text{O}_7\) \([7]\) and the a frustrated quantum magnet \(\text{Tb}_2\text{Ti}_2\text{O}_7\) \([8]\). 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 \(\text{Cu}(1,3\text{-benzenedicarboxylate})\) \([\text{Cu}(1,3\text{-bdc})]\) \([10–12]\) 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 \(\chi = \mathbf{S}_i \cdot \mathbf{S}_j \times \mathbf{S}_k\) \((\mathbf{S}_i\) 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 \(t = \sqrt{t^2 + D^2} \cdot e^{i\phi}\) 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 \(\mathbf{A}(k)\) 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 \(\text{Cu}(1,3\text{-bdc})\) is comprised of stacked Kagome planes separated by \(d = 7.97\) Å \([10–12]\). The spin-\(\frac{1}{2}\) \(\text{Cu}^{2+}\) 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 \(\kappa/\kappa_{ph}\) at \(T_C\) \((1.85\) K) followed by a large peak at \(\sim 1.2 T_C\). 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 \(\text{Cu}(1,3\text{-bdc})\) 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.
resulting from $\Delta$ is evident in both the exponential suppression of the magnon population exponentially at large $K$ in Fig. 3D. Within the uncertainty, it also decreases in a recent neutron scattering experiment. The $g$-factor is the Bohr magneton, and $\mu_B$ is the absolute constant for 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. 2D. 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 $\kappa_{xx}$ 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 $\kappa_{xy}$ 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 $\kappa_{xx}$ and $\kappa_{xy}$ 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 $\kappa_{xy}^{2D}/(k_B^2/\hbar)$ (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 $\kappa_{xy}^{2D}/(T k_B^2/\hbar)$. 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 $B \rightarrow 0$. 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 $\kappa_{xx}^S/T$ 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 $B$. 