Thermal Hall Effect of Spin Excitations in a Kagome Magnet

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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 κ_{xy}. Here we report observation of a large κ_{xy} in the Kagome magnet Cu(1-3, bdc) which orders magnetically at 1.8 K. The observed κ_{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 κ_{xy} and κ_{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 $\mathbf{\Omega}(\mathbf{k})$ ($\mathbf{\Omega}(\mathbf{k})$ acts like a magnetic field in $\mathbf{k}$ 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 $\mathbf{H}$ as if a Lorentz force were present. The predicted thermal Hall conductivity κ_{xy} was observed in two recent experiments on the ordered magnet $Lu_2V_2O_7$ [7] and the a frustrated quantum magnet $Tb_2Ti_2O_7$ [8]. However, to test more incisively the role of $\mathbf{\Omega}(\mathbf{k})$ and to exclude a phononic origin [9], we need results that can be compared with microscopic calculations based on $\mathbf{\Omega}(\mathbf{k})$. An interesting prediction based on the Chern number sign-alternation between magnon bands is the induced sign-change in κ_{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 κ_{xy} and κ_{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 κ_{xy} directly to the Berry curvature. In the boson representation of the spin Hamiltonian, χ 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 ϕ, which implies the existence of a vector potential $\mathbf{A}(\mathbf{k})$ permeating $\mathbf{k}$-space. The Berry curvature $\mathbf{\Omega}(\mathbf{k}) = \nabla_{\mathbf{k}} \times \mathbf{A}(\mathbf{k})$ imparts an anomalous velocity to magnons, leading to a thermal Hall conductivity κ_{xy}. Each magnon band n contributes a term to κ_{xy} with a sign determined by the integral of $\mathbf{\Omega}(\mathbf{k})$ over the Brillouin zone (the Chern number). Recently, Lee, Han and Lee (LHL) [13] calculated how κ_{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 supplementary information SI).

As we cool the sample in zero B, the thermal conductivity κ (nearly entirely from phonons) initially rises to a very broad peak at 45 K (Fig. 1A). Below the peak, κ decreases rapidly as the phonons freeze out. Starting near 10 K, the spin contribution κ^s becomes apparent. As shown in Fig. 1B, this leads to a minimum in κ 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 κ/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 κ/T slows to approach saturation. The open black circles represent the phonon conductivity κ_{ph} deduced from the large-B values of $\kappa_{xx}(T, H)$ (see below). Likewise, κ_{ph}/T is plotted as open red circles. The difference $\kappa - \kappa_{ph}$ is the estimated thermal conductivity of magnons κ^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 κ_{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.

2A). We refer to a positive κ_{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 SI. This sign-change is investigated in greater detail in Sample 3 (we plot κ_{xy}/T in Figs. 2B,C). The curves of κ_{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. 2C). 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 $[\kappa_{xy}/BT]_0$ plotted in Fig. 2D, closely correlates with the growth of κ_s below T_C.

To relate the thermal Hall results to magnons, we next examine the effect of B on the longitudinal thermal conductivity κ_{xx}. As shown in Fig. 3A, κ_{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. 3B. 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. 1B). Subtracting the floor allows the thermal conductivity due to spins to be defined as $\kappa^s_{xx}(T,H) \equiv \kappa_{xy}(T,H) - \kappa_{ph}(T)$. The exponential suppression becomes apparent in the scaled plot of κ^s_{xx}/T vs. B/T (Fig. 3C). The asymptotic form at large B in all curves depends only on B/T.

In the interval 0.9 K$\to$ T_C, κ^s_{xx} displays a V-shaped minimum at $B = 0$ followed by a peak at the field $H_p(T)$. Since κ^s (at $B = 0$) 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

$$\kappa^s_{xx} \to Te^{-\beta\Delta}, \tag{1}$$

with the Zeeman gap $\Delta = g\mu_B B$ where $\beta = 1/k_BT$, μ_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 Δ. Hence the exponential suppression of the magnon population resulting from Δ is evident in both κ^s_{xx} and κ_{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 κ^{2D}_{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, κ_{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 κ_{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 κ_{xy} arises from spin excitations. The close correlation between the profiles of κ_{xy} and κ_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 κ_{xy} in the Kagome magnet, the measured κ_{xy} can be compared with calculations. For chiral magnets, κ_{xy} is capable of probing incisively the effect of the Berry curvature on transport currents.

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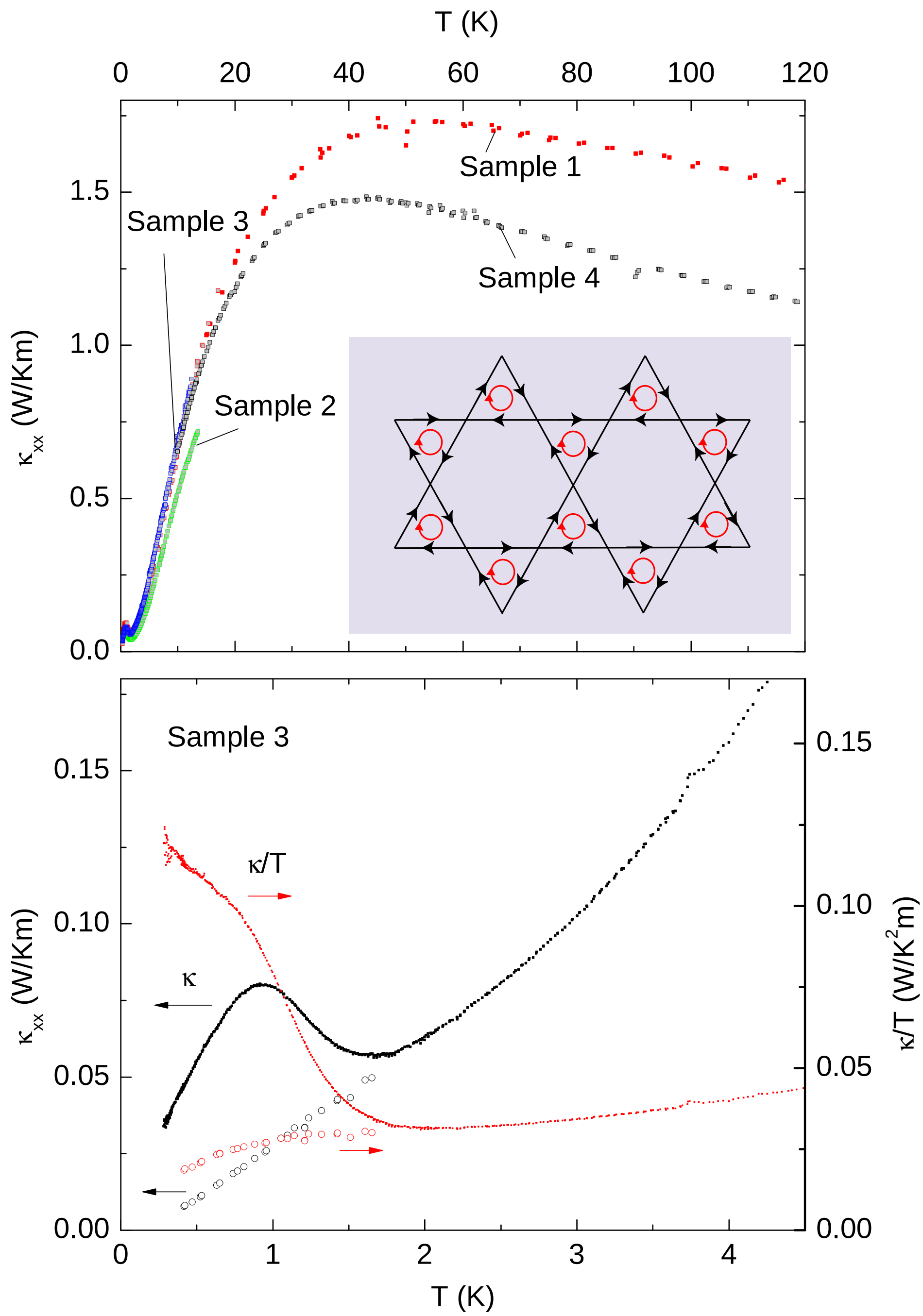

FIG. 1: The in-plane thermal conductivity κ (in zero B) measured in the Kagome magnet Cu(1,3-bdc). At 40-50 K, κ displays a broad peak followed by a steep decrease reflecting the freezing out of phonons (Panel A). The spin excitation contribution becomes apparent below 2 K. The inset is a schematic of 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 κ (black symbols) and κ/T (red) for $T < 4.5$ K. Below the ordering temperature $T_C = 1.8$ K, the magnon contribution to κ appears as a prominent peak that is very B dependent. Values of κ and κ/T at large B (identified with the phonon background) are shown as open symbols.

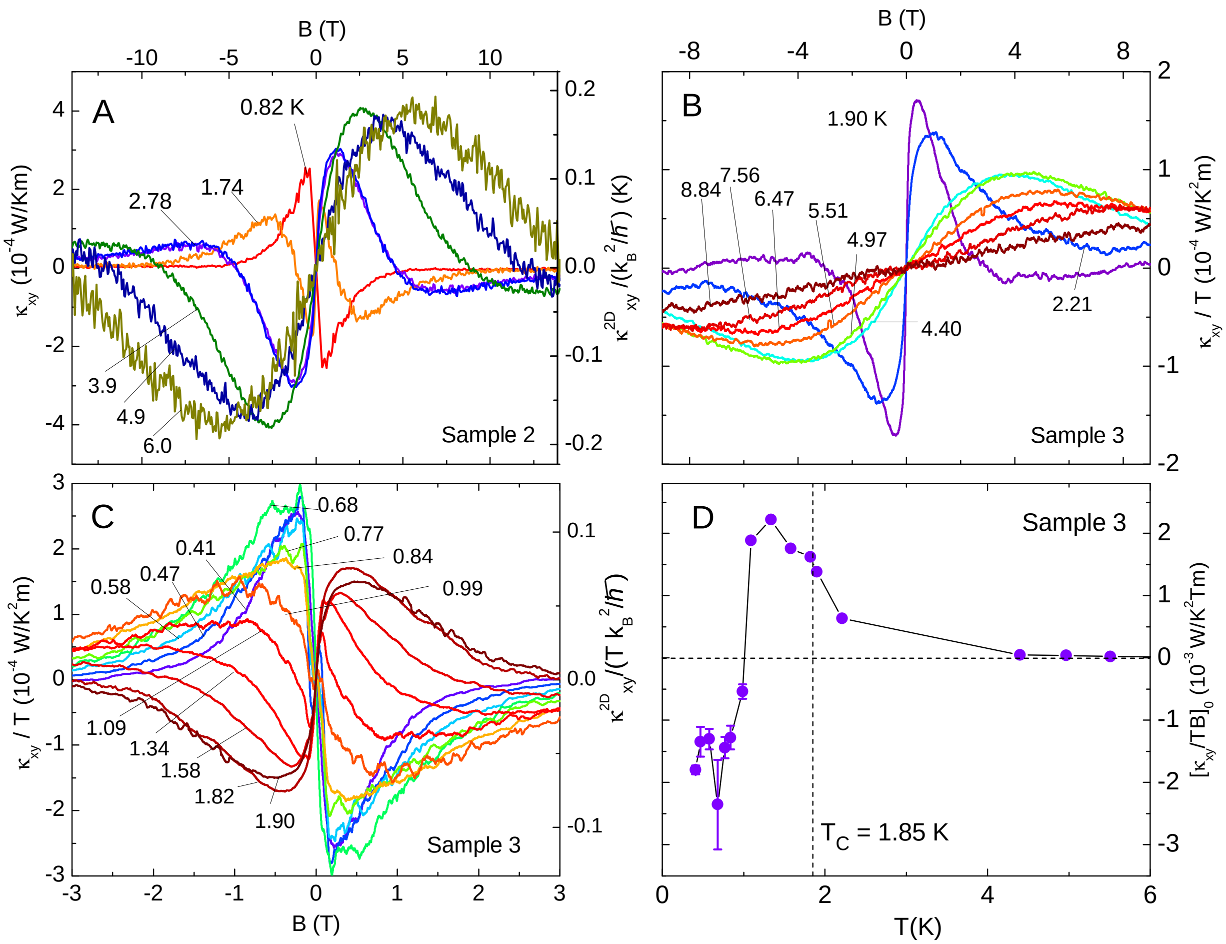

FIG. 2: The thermal Hall conductivity κ_{xy} measured in Cu(1,3-bdc). In Panel A, we plot the strongly non-monotonic profiles of κ_{xy} vs. B in Sample 2. The dispersion-like profile changes sign below ~ 1.7 K. The right scale gives $\kappa^{2D}/(k_B^2/\hbar)$ (per plane) obtained by multiplying κ_{xy} by $d\hbar/k_B^2 = 443.2$ (SI units). Panels B and C show corresponding curves in Sample 3 (now plotted as κ_{xy}/T). Above T_C (Panel B), κ_{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 κ_{xy}/T to n-type at all B. Right scale in C reports $\kappa_{xy}^{2D}/(Tk_B^2/\hbar)$. 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 κ_{xx}^S vs. T (aside from the sign change).

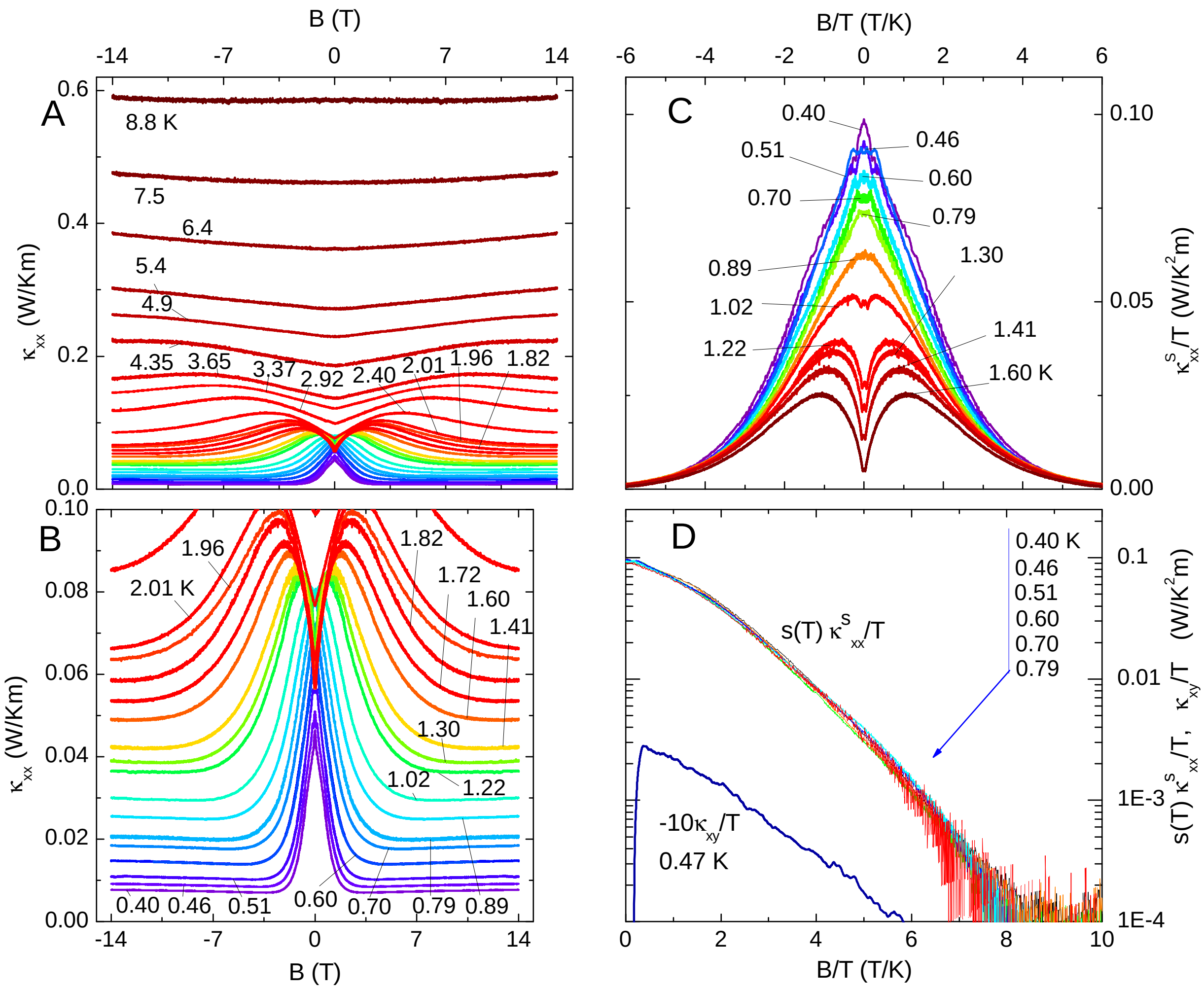

FIG. 3: The effect of field B on κ_{xx} and scaling behavior at low T, for sample 3. The curves in Panel A show that the B-dependence of κ_{xx} is resolved (in the range $|B| < 14$ T) only at $T <\sim 6.5$ K. The expanded scale in Panel B shows that, near T_C (1.8 K), κ_{xx} has a non-monotonic profile with a V-shaped minimum at $B = 0$ (identified with stiffening of the magnon bands by the field). Below 1 K, however, κ_{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 κ_{ph}. The pattern in Panel B simplifies when plotted as κ^s_{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.