Thermal Hall Effects of Spins and Phonons in Kagome Antiferromagnet Cd-Kapellasite

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Abstract

We have investigated the thermal-transport properties of the kagome antiferromagnet Cd-kapellasite (Cd-K). At low temperatures, the longitudinal thermal conductivity $\kappa_{xx}$ was strongly suppressed in the magnetic field, suggesting a large spin contribution $\kappa_{xx}^{sp}$. Clear thermal Hall signals were observed in the spin liquid phase in all Cd-K samples. We find a large sample dependence of the magnitude of the thermal Hall conductivity $\kappa_{xy}$ of Cd-K, of which the peak increases almost linear to $\kappa_{xx}$. On the other hand, the temperature dependence of $\kappa_{xy}$ is similar in all Cd-K samples and shows a peak at almost the same temperature of the peak of the phonon thermal conductivity $\kappa_{xx}^{ph}$ which is estimated by $\kappa_{xx}$ at 15 T. These results indicate the presence of a phonon thermal Hall $\kappa_{xy}^{ph}$ in Cd-K. In addition to $\kappa_{xy}^{ph}$, we find that the non-linear field dependence of $\kappa_{xy}$ at low temperatures indicates the presence of a spin thermal Hall $\kappa_{xy}^{sp}$ in Cd-K. Remarkably, both $\kappa_{xy}^{ph}$ and $\kappa_{xy}^{sp}$ disappear in the antiferromagnetic ordered phase at low fields, showing that $\kappa_{xy}^{ph}$ alone does not exhibit the thermal Hall effect. A high field above $\sim 7$ T induces $\kappa_{xy}^{ph}$, concomitantly with a field-induced increase of $\kappa_{xx}$ and the specific heat. These results demonstrate that the spin liquid state in the Cd-K harbors both $\kappa_{xy}^{sp}$ and $\kappa_{xy}^{ph}$, and suggest a coupling of the phonons to the field-induced excitations as the origin of $\kappa_{xy}^{ph}$. 

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I. INTRODUCTION

The magnetic ground state of a two-dimensional (2D) kagome structure has been attracting tremendous attention because the strong frustration effect caused by the corner-sharing network of the triangles has been expected to suppress the magnetic order even at the absolute zero temperature. Instead of a long-range ordered state, emergence of a quantum disordered state of spins, termed as a quantum spin liquid (QSL), has been shown in the kagome Heisenberg antiferromagnet (KHA) by various numerical calculations. A lot of QSLs have been theoretically suggested as the ground state of the KHA such as $\mathbb{Z}_2$ spin liquids, topological spin liquids, Dirac spin liquids, and chiral spin liquids. These different QSLs are characterized by different elementary excitations. It is thus an experimental challenge to pin down the QSL realized in KHA by clarifying the elementary excitation.

Thermal-transport measurement is a powerful probe to study the elementary excitations in QSLs because it has the advantage of detecting only the itinerant excitations. Therefore, one can avoid effects of localized excitations caused by impurities which are often inevitable in candidate materials. Moreover, further detail of the elementary excitation can be studied by investigating the thermal Hall effect. It has been shown that the thermal Hall effect in an insulator is given by the Berry curvature of the elementary excitation as

$$\kappa_{xy} = \frac{k_B T}{\hbar V} \sum_k \sum_n c_2[g(\epsilon_{nk})] \Omega_{nk},$$

where $c_2[g(\epsilon_{nk})]$ is a distribution function given by the elementary excitations of energy $\epsilon_{nk}$ and $\Omega_{nk}$ is the Berry curvature of the elementary excitations. Therefore, from $\kappa_{xy}$ measurements, one can study the statics of the elementary excitations (fermions or bosons) as well as the Berry curvature of the corresponding energy bands.

The thermal Hall effect of spins ($\kappa_{xy}^{\text{sp}}$) has been observed in ferromagnetic insulators, which is well understood as a magnon thermal Hall effect. The spin thermal Hall effect has also been reported in paramagnetic states of kagome, spin ice, and Kitaev compounds. In these frustrated magnets, the paramagnetic phase extends well below the temperatures determined by the interaction energy $J$, realizing a spin liquid phase in a wide temperature range $T_N \leq T \ll J/k_B$. For $\kappa_{xy}$ observed in the spin liquid phase of kagome antiferromagnets volborthite and Ca kapellasite (Ca-K), it has been shown that the Schwinger-boson mean field theory (SBMFT) can well reproduce both the temperature dependence and the magnitude of $\kappa_{xy}$ by tuning the two
fitting parameters of the spin interaction energy $J$ and the Dzyaloshinskii-Moriya (DM) interaction $D$ (Ref. [19]). Remarkably, the fitting results of $J$ and $D$, obtained by the SBMFT fitting to $\kappa_{xy}$ of both kagome compounds, are close to the values estimated by the temperature dependence of the magnetic susceptibility and that by the deviation of the $g$ factor, respectively. This excellent agreement suggests that the elementary spin excitations in the KHA can be well described by the bosonic spinons of SBMFT.

In addition to the spin thermal Hall effects, the thermal Hall effects of phonons ($\kappa_{xy}^{ph}$) have been reported in various compounds [26][30]. The origin of the phonon thermal Hall has also been extensively studied theoretically [31][39]. However, the understanding of the phonon thermal Hall effect has been left out in the consideration of spin thermal Hall effect, because the nature of the coupling between phonons and spin fluctuations has remained unclear.

In this Article, we report our thermal-transport measurements of a new kagome compound Cd kapellasite (Cd-K). Previous studies [40][41] have shown that the spin Hamiltonian of Cd-K is well approximated to a KHA with the spin interaction energy of $J/k_B \sim 45$ K. The frustration effect of the kagome structure suppresses the ordering temperature ($T_N \sim 4$ K) well below $J/k_B$, realizing a spin liquid phase in a wide temperature range. We find a large contribution of the spins in $\kappa_{xx}$ which can be essentially turned off by applying a magnetic field. This field-tuning of $\kappa_{xx}^{sp}$ enables us to find both $\kappa_{xy}^{sp}$ and $\kappa_{xy}^{ph}$ in Cd-K. Most remarkably, we find that both $\kappa_{xy}^{sp}$ and $\kappa_{xy}^{ph}$ disappear in the AFM phase at low fields. Further, we find that $\kappa_{xy}^{ph}$ is induced by applying a high field which concomitantly induce additional excitations probed by the specific heat and $\kappa_{xx}$. We conclude that Cd-K is a prominent frustrated magnet in which the spin liquid state shows thermal Hall effects of both spins and phonons. The dual nature of the thermal conductivities prompts us to speculate that a coupling of phonons with the field-induced magnetic excitations is taking place to give rise to $\kappa_{xy}^{ph}$ in this compound.

II. MATERIALS AND METHODS

Cd-Kapellasite CdCu$_3$(OH)$_6$(NO$_3$)$_2$·H$_2$O is a trigonal compound with space group $P\bar{3}m1$ and lattice constants $a = 6.5449$ Å, $c = 7.0328$ Å [42], in which the magnetic Cu$^{2+}$ ions form an undistorted kagome lattice (Fig. [1](a)). Cd-K is a structural polymorph of herbertsmithite [23] in which the Zn ions located between the kagome layer and the site mixings between the Zn and Cu ions [12] allow an inter-layer coupling between the kagome layers. In contrast, in Cd-K, the nonmagnetic
Cd ions are located at the center of the hexagon of the kagome lattice and there is no site mixings in Cd-K because of the larger ionic radii of Cd$^{2+}$ (0.95 Å) than that of Cu$^{2+}$ (0.73 Å)\textsuperscript{43}, realizing a more ideal KHA in Cd-K.

Three magnetic interactions in Cd-K are suggested by the fitting of the magnetic susceptibility as $J/k_B = 45.44$ K, $J_2/J = -0.1$, $J_d/J = 0.18$, where $J$ is the nearest-neighbor interaction, $J_2$ the next-nearest-neighbor interaction, and $J_d$ the diagonal interaction via the non-magnetic Cd ion (see Fig.1(b)). The development of a short-range antiferromagnetic correlation has been shown by the decrease of the magnetic susceptibility below 30 K\textsuperscript{40}. The $g$ factors are estimated as $g_a = 2.28$, $g_c = 2.37$\textsuperscript{41}. The lack of the inversion symmetry allows both the in-plane and out-of-plane DM interactions, which has been suggested to cause a negative vector chiral order below the Neel temperature of $T_N \sim 4$ K\textsuperscript{40}.

The thermal-transport measurements were performed by the steady-state method as described in Refs.\textsuperscript{18-20}. One heater and three thermometers were attached to the sample, and then the temperature difference $\Delta T_x$ ($\Delta T_x = T_{\text{High}} - T_{L1}$) and $\Delta T_y$ ($\Delta T_y = T_{L1} - T_{L2}$) were measured by applying the heat current $Q$ in the kagome plane (Fig.1(c)). The longitudinal thermal conductivity $\kappa_{xx}$ and the thermal Hall conductivity $\kappa_{xy}$ is derived by

$$\begin{pmatrix} Q/wt \\ 0 \end{pmatrix} = \begin{pmatrix} \kappa_{xx} & \kappa_{xy} \\ -\kappa_{xy} & \kappa_{xx} \end{pmatrix} \begin{pmatrix} \Delta T_x/L \\ \Delta T_y/w \end{pmatrix},$$

where $t$ is the thickness of the sample, $L$ is the length between $T_{\text{High}}$ and $T_{L1}$, and $w$ is the sample width between $T_{L1}$ and $T_{L2}$.

We measured $\kappa_{xx}$ and $\kappa_{xy}$ of three Cd-K samples (Sample 1, 2, and 3) by using a variable temperature insert (VTI) (2–60 K, 0–15 T). Measurements of Sample 2 were also done in a dilution refrigerator (DR) (0.1–4 K, 0–14 T). The magnetic field was applied along the $c$ axis of the sample. A typical sample is shown in Fig.1(d). A heat current $Q$ was applied along the direction 1 (\perp $a$-axis, see Fig.1(d)) in Samples 1, 2, and the first run of Sample 3 (denoted as Sample 3-1). In the second run of Sample 3 (Sample 3-2), the direction of $Q$ was change to the direction 2 (\parallel $a$-axis, see Fig.1(d)). The specific heat measurement was performed for multiple single crystals by a thermal relaxation method in a temperature range of 0.1–2 K and a magnetic field range up to 14 T.
FIG. 1. (a) Crystal structure and (b) top-view of a kagome layer of Cd-K. The magnetic interactions between nearest-neighbor, next-nearest-neighbor, and diagonal Cu$^{2+}$ spins are denoted by $J$ (solid black line), $J_2$ (dotted line) and $J_d$ (dashed line), respectively. (c) Schematic illustration of $\kappa_{xx}$ and $\kappa_{xy}$ measurements. A heater and three thermometers ($T_{\text{High}}, T_{L1}, T_{L2}$) were attached to the sample fixed on the LiF heat bath. A heat current $Q$ was applied within the kagome layer and a magnetic field $B$ was applied along the $c$-axis. (d) A typical crystal of Cd-K. The direction of the heat current for the sample 1, 2, and 3-1 (3-2) is shown by the arrow 1 (2).

III. RESULTS

A. Longitudinal Thermal Conductivity

Figure 2 (a) shows the temperature dependence of $\kappa_{xx}$ of all Cd-K samples at zero magnetic field. For reference, $\kappa_{xx}$ of Ca-K is also shown. As shown in Fig. 2 (a), $\kappa_{xx}$ of all Cd-K samples is about one order of magnitude larger than that of Ca-K. Although the magnitude of $\kappa_{xx}$ in different Cd-K samples are different in factor of $\sim 2$, $\kappa_{xx}$ of all Cd-K samples show a similar temperature dependence. The temperature dependence of $\kappa_{xx}$ shows a shoulder-like enhancement around 15 K, which is followed by a hump near $T_N$ and a rapid decrease for $T < T_N$.

The temperature dependence of $\kappa_{xx}$ at different magnetic fields is shown in Figs. 2 (b)–(f). In all Cd-K samples, a decrease of $\kappa_{xx}$ by applying the magnetic field was observed below 30 K. This
FIG. 2. (a) Temperature dependence of the longitudinal thermal conductivity ($\kappa_{xx}$) of all samples of Cd-Kapellasite (Cd-K) and that of Ca-Kapellasite (Ca-K) at 0 T. The longitudinal thermal conductivity of Ca-K is taken from Ref. 19. (b to f) The data of each sample under magnetic fields. In the sample 2, $\kappa_{xx}$ was measured down to 0.1 K. An enlarged view of low-temperature region (0.1–0.3 K) of (c) is shown in (d). The filled and open circles in (c) and (d) show $\kappa_{xx}$ measured by a variable temperature insert (VTI) (2 K < $T$) and a dilution refrigerator (DR) (0.1 < $T$ < 4 K), respectively.

field suppression effect is larger for a sample with a large $\kappa_{xx}$. The hump in the temperature dependence of $\kappa_{xx}$ was observed at lower temperatures as the magnetic field was increased, implying a suppression of the magnetic transition temperature to lower temperatures (see Fig. 3).

Figure 4 shows the magnetic field dependence of $\kappa_{xx}$ of sample 2. The vertical axis is normalized by the zero-field value as $[\kappa_{xx}(B) - \kappa_{xx}(0)]/\kappa_{xx}(0)$. The field dependence of $\kappa_{xx}$ of other samples were essentially the same. Above 40 K, $\kappa_{xx}$ increased linearly by applying the magnetic field (Fig. 4(a)). On the other hand, below 30 K, the suppression of $\kappa_{xx}$ by the magnetic field was observed (Fig. 4(b)). The field suppression effect became larger at lower temperatures and reached the maximum reduction of ~ 70% by 15 T at 2 K. Below 0.3 K, a new peak was observed in the field dependence of $\kappa_{xx}$ at 6–7 T (Fig. 4(c)).
FIG. 3. Temperature dependence of $\kappa_{xx}$ of Sample 3-1 near the Neel temperature at different fields. The red arrows point the field where $\kappa_{xx}$ deviates from the linear temperature dependence (dash line) due to magnetic order.

B. Thermal Hall Conductivity

Figure 5(a) shows the magnetic field dependence of $\Delta T_y/Q$ of Sample 2 in the spin liquid phase. As shown in Fig. 5(a), the field dependence of $\kappa_{xx}$ is dominated by the symmetric longitudinal component caused by the misalignment effect. To extract the asymmetric thermal Hall effect, the
field dependence of $\Delta T_y/Q$ is antisymmetrized with respect to the field direction as $\Delta T_y^{\text{Asym}}(B) = (\Delta T_y(+B) - \Delta T_y(-B))/2$. The field dependence of $\Delta T_y^{\text{Asym}}(B)$ of sample 2 is shown in Fig. 5(b). As shown in Fig. 5(b), $\Delta T_y^{\text{Asym}}/Q$ shows a linear magnetic field dependence at high temperatures.

The field dependence of $\kappa_{xy}$ is determined by $\Delta T_y^{\text{Asym}}$ in accordance with Eq. 1, and is plotted in Fig. 6. From this field dependence, $\kappa_{xy}/TB$ is estimated by the linear fitting of $\kappa_{xy}$ as shown by the solid lines in Fig. 6. As shown in Fig. 6, the linear field dependence of $\kappa_{xy}$ observed at 20 K becomes non-linear at lower temperatures, resulting in a large error in estimating $\kappa_{xy}/TB$ at lower temperatures.

This thermal Hall signal disappears in the AFM phase at low fields. Figure 5(c) shows the field dependence of $\Delta T_y^{\text{Asym}}$ of sample 2 measured in a dilution refrigerator. As shown in Fig. 5(c), the thermal Hall effect was absent at low fields, which is followed by an increase above $\sim 7$ T. Thus, for the measurements done in the dilution refrigerator, we determine $\kappa_{xy}/TB$ by $\Delta T_y^{\text{Asym}}(B)$ at 14 T (open symbols in Fig. 8). A similar non-linear field dependence is confirmed in all Cd-K samples done at the lowest temperature of the VTI measurement (2 K) as shown in Fig. 7.

Figure 8(a) shows the temperature dependence of the thermal Hall conductivity of all Cd-K samples. As shown in Fig. 8(a), the thermal Hall conductivity $\kappa_{xy}/TB$ of all Cd-K samples shows a similar temperature dependence with a peak around 8 K. In Sample 2, $\kappa_{xy}/TB$ obtained in the ordered phase by the DR measurements is also shown by open symbols, which seems to be
FIG. 6. The field dependence of $\kappa_{xy}$ of all Cd-K samples for 4–20 K. The data above 4 K is shifted for clarity. The solid lines show a linear fit of the data to estimate $\kappa_{xy}/TB$ at each temperatures.

FIG. 7. Magnetic field dependence of the asymmetrized transverse temperature difference divided by the heat current ($\Delta T_{y}^{\text{Asym}}/Q$) at 2 K.

smoothly connected to the data measured by VTI (filled symbols). This temperature dependence is also similar to that of Ca-K\textsuperscript{19} and volborthite \textsuperscript{18} (Fig. 8 (b)). On the other hand, as shown in Fig. 8 (b), the peak temperature of $\kappa_{xy}/TB$ is clearly shifted to a lower temperature in Cd-K.
FIG. 8. Temperature dependence of $\kappa_{xy}/TB$. (a) Comparison of $\kappa_{xy}/TB$ of three Cd-K samples. The filled (open) symbols represent $\kappa_{xy}/TB$ calculated from the linear fitting of $\kappa_{xy}$ in the VTI measurements (from the values at 14 T in the DR measurements). (b) Comparison of $\kappa_{xy}/TB$ of Cd-K sample 2, Ca-K$^{19}$ (open gray pentagon) and volborthite$^{18}$ (open gray hexagon). For clarity, $\kappa_{xy}/TB$ of volborthite is multiplied by −1. Error bars corresponding to 1 standard deviation. The inset shows an enlarged view of the low temperature data of $\kappa_{xy}/TB$ of sample 2. The dashed line shows a guide to the eye.

C. Specific Heat

Figure 9 (a) shows the temperature dependence of the specific heat ($C$) measured in multiple single crystals. The zero-field data shows a good agreement with the previous data$^{40}$ shown as open circles in Fig. 9. The specific heat under magnetic fields increases below 0.5 K owing to the nuclear Schottky anomaly ($C_{Ncl}$). The magnetic field dependence of the specific heat at 0.5 K is shown in Fig. 9 (b). After the specific heat was decreased by applying the magnetic field, the specific heat was increased by applying the magnetic field above 7 T. We note that this field increase of $C$ above 7 T is much larger than that expected by $C_{Ncl}$ (dotted line in Fig. 9 (b)) which is estimated as $\sim 2$ mJ K$^{-1}$ mol$^{-1}$ at 0.5 K and 10 T from the fit of $C_{Ncl} \propto H^2/T^2$ for the data shown in Fig. 9 (a).
FIG. 9. (a) Temperature dependence of the specific heat divided by the temperature \(C/T\) measured in multiple single crystals. The zero-field data from the previous report is also shown by open circles. (b) Magnetic field dependence of \(C\) at 0.5 K. The red dotted line shows an estimation of the magnetic field dependence of the nuclear Schottky specific heat \(C_{\text{Ncl}}\) at 0.5 K. The data of \(C_{\text{Ncl}}\) is shifted to compare the amount of the field increase.

IV. DISCUSSION

A. Longitudinal Thermal Conductivity

First, we discuss the sample dependence of \(\kappa_{xx}\) (Fig. 2 (a)) in terms of the sample quality. The longitudinal thermal conductivity of an insulator is given by the sum of the contribution of the phonons \(\kappa_{xx}^{ph}\) and that of the spins \(\kappa_{xx}^{sp}\). Considering \(J/k_B \sim 45\) K, it can be expected that \(\kappa_{xx}\) above 45 K is almost given by \(\kappa_{xx}^{ph}\), which is consistent with the field dependence of \(\kappa_{xx}\). It is known that \(\kappa_{xx}^{ph}\) increases in the magnetic field because the spin-phonon scatterings are reduced under magnetic field by suppressing spin fluctuations. In fact, as shown in Fig. 4 (a), the increase of \(\kappa_{xx}\) by applying the magnetic field is observed above 40 K, showing a dominant \(\kappa_{xx}^{ph}\) in \(\kappa_{xx}\) at high temperatures.

The phonon thermal conductivity \(\kappa_{xx}^{ph}\) is given by a product of the specific heat \(C_{ph}\), the mean free path \(\ell_{ph}\), and the velocity \(v_{ph}\) of phonons, as \(\kappa_{xx}^{ph} = (1/3)C_{ph}\ell_{ph}v_{ph}\). Since \(C_{ph}\) and \(v_{ph}\) are
common in all Cd-K samples, the difference in the magnitude of $\kappa_{xx}$ shown in Fig. 2(a) reflects the difference in $\ell_{\text{ph}}$ of each sample. Therefore, a sample with a larger $\kappa_{xx}$ is a better crystal with less impurities. Also, the larger $\kappa_{xx}$ of Cd-K than that of Ca-K shows that $\ell_{\text{ph}}$ of Cd-K is much longer than that of Ca-K because $C_{\text{ph}}$ and $\nu_{\text{ph}}$ of Cd-K are similar to those of the isostructural Ca-K. This longer $\ell_{\text{ph}}$ of Cd-K than that of Ca-K indicates that Cd-K has a more ideal kagome structure without the randomness of ions or the lattice defects found in Ca-K. We note that the difference of $\kappa_{xx}$ of Sample 3-1 and that of Sample 3-2 might be caused by the ambiguity in estimating the sample size (up to 10%) owing to the irregular shape of the sample (see Fig. 1(d)).

Next, we discuss the field suppression effect on $\kappa_{xx}$ observed below 30 K (Fig. 4(b)). One of the field-suppression mechanisms of $\kappa_{xx}^{\text{ph}}$, which normally increases in the magnetic field, is a resonance scattering of phonons being absorbed by impurity free spins. This resonant scattering is most effective when the spin Zeeman gap ($g\mu_B H$) coincides with the phonon peak ($\sim 4k_B T$) given by the Debye distribution, where $\mu_B$ is the Bohr magneton. Therefore, this resonance scattering produces a suppression peak of $\kappa_{xx}$ at 5.4 T for 2 K as observed in volborthite. However, as shown in Fig. 4(b), $\kappa_{xx}$ at 2 K decreases monotonically with increasing magnetic field up to 15 T without the expected suppression peak. Therefore, the field suppression effect of $\kappa_{xx}$ cannot be explained by the resonance scattering effect on $\kappa_{xx}^{\text{ph}}$. Therefore, the field suppression effect is caused by the decrease of $\kappa_{xx}^{\text{sp}}$ under magnetic fields. A similar field suppression effect on $\kappa_{xx}^{\text{sp}}$ has also been observed in the spin liquid state of the spin-chain compound, volborthite, and Ca-K. In volborthite, a field suppression effect up to $\sim 30\%$ at 15 T was observed together with the resonance scattering effect. Compared to the field suppression effects in volborthite and Ca-K, the field suppression of $\kappa_{xx}$ in Cd-K is much larger (more than 70\% at 2 K), showing a dominant contribution of $\kappa_{xx}^{\text{sp}}$ in $\kappa_{xx}$ at low temperatures.

Here, we consider the hump-like increase of $\kappa_{xx}$ observed near $T_N$. This increase is caused by the increase of $\kappa_{xx}^{\text{ph}}$ by a reduction of spin fluctuations and/or the appearance of a magnon contribution in the ordered state. In the former case, as observed in $\alpha$-RuCl$_3$ (Ref. 47), the increase of $\kappa_{xx}$ at $T_N$ should be larger under higher fields because the spin fluctuations are more strongly suppressed under higher fields. However, as shown in Fig. 3, the increase becomes smaller at higher fields. This is consistent with the field suppression effect on $\kappa_{xx}^{\text{sp}}$. In addition, the large field suppression of $\kappa_{xx}$ at low temperatures suggests a dominant contribution of $\kappa_{xx}^{\text{sp}}$. Therefore, the increase of $\kappa_{xx}$ below $T_N$ is likely attributed to the magnon contribution. The increase of $\kappa_{xx}$ below $T_N$ was observed larger in a better crystal with a larger $\kappa_{xx}$. We note that a similar sample depen-
idence of magnon thermal conduction has been observed in volborthite\textsuperscript{29}, which also supports the presence of a magnon contribution below $T_N$.

A new field-induced peak is observed in the magnetic field dependence of $\kappa_{xx}$ around 7 T below 0.3 K (Fig. 4 (c)). Below 0.3 K, the resonance scattering effect on phonons takes place below 1 T. Also, the critical field of the AFM phase is much larger than 7 T because the hump-like increase of $\kappa_{xx}$ is still observed above 7 T (Fig. 3). Therefore, this magnetic field dependence of $\kappa_{xx}$ cannot be explained by an increase of $\kappa_{xx}^{\text{ph}}$ by a reduction of the spin fluctuations. The magnon contribution is also excluded for the field-induced increase because the magnon contribution is suppressed by fields as observed as the decrease of the hump-like increase (Fig. 3). Therefore, this increase of $\kappa_{xx}$ around 7 T indicates an appearance of some field-induced spin excitations.

The thermal conduction of spin is also given by $\kappa_{xx}^{\text{sp}} = C_{\text{sp}} v_{\text{sp}} \ell_{\text{sp}}/3$, where $C_{\text{sp}}$, $v_{\text{sp}}$, and $\ell_{\text{sp}}$ is the specific heat, the velocity and the mean free path of the spin excitations, respectively. As shown in Fig. 5 (b), the increase of $C_{\text{sp}}$ is also observed around 7 T. This also supports the appearance of the field-induced excitations. Further, in the ordered phase, a finite thermal Hall effect is observed only above 7 T (Fig. 5 (c)), implying that the thermal Hall effect is caused by the field-induced excitations observed in the field dependence of $\kappa_{xx}$ and $C$. As shown in Figs. 8, $\kappa_{xy}/TB$ observed in the ordered phase above 7 T shows a good agreement with the data in the spin liquid phase, implying that the field-induced excitations in the ordered phase are similar to those in the spin liquid phase. This field dependence of the thermal Hall effect in the ordered phase of Cd-K is in sharp contrast to that observed in Ca-K\textsuperscript{19} where a finite $\kappa_{xy}$ in the ordered phase is observed only in a low-field and is absent above $\sim 6$ T. This difference implies a difference in the ordered state of Cd-K from that in Ca-K.

\section*{B. Thermal Hall Conductivity}

We first discuss the temperature dependence of $\kappa_{xy}/TB$ of all three kagome compounds of Cd-K, Ca-K\textsuperscript{19} and volborthite\textsuperscript{18}. As shown in Fig. 8 (b), $\kappa_{xy}/TB$ of these kagome antiferromagnets shows a similar temperature dependence. As reported in Ref. 19, both the temperature dependence and the magnitude of $\kappa_{xy}/TB$ of Ca-K and volborthite show a good agreement with a simulation based on the SBMFT\textsuperscript{25,48}. In the SBMFT framework, the kagome Heisenberg Hamiltonian with a Zeeman term and a DM interaction is diagonalized by taking a mean-field value of the bond operator of Schwinger bosons. From the energy bands and the Berry curvature calculated by the
SBMFT, $\kappa_{\text{SBMFT}}^{xy}$ is calculated by Eq. (1) and is expressed by a dimensionless function $f_{\text{SBMFT}}$ as

$$\frac{\kappa_{\text{SBMFT}}^{xy}}{T} = \frac{k_B^2}{\hbar} D g \mu_B B \frac{J^2}{f_{\text{SBMFT}}(k_B T / J^2)}. \quad (3)$$

To compare this SBMFT calculation, the thermal Hall conductivity per one 2D kagome layer is estimated from the experimental data by $\kappa_{\text{2D}}^{xy} = \kappa_{\text{xy}} d$, where $d = 7.0328 \text{ Å}$ for Cd-K\textsuperscript{40} is the distance between the kagome layers. We then compare $\kappa_{\text{2D}}^{xy}$ with $f_{\text{SBMFT}}$ by normalizing $\kappa_{\text{2D}}^{xy}$ as

$$\frac{\kappa_{\text{2D}}^{xy}}{T} = \frac{k_B^2}{\hbar} D g \mu_B B \frac{J^2}{f_{\text{exp}}}, \quad (4)$$

where $J$ and $D$ are the fitting parameters.

![Fig. 10](image.png)

**FIG. 10.** Normalized thermal Hall conductivity $f_{\text{exp}}$ of kagome lattice antiferromagnets fitted by the parameters listed in Table I. The solid line shows a numerical calculation of $f_{\text{SBMFT}}$ at $D/J = 0.1$ by the Schwinger-boson mean field theory (SBMFT)\textsuperscript{19}. The data of Ca-K and that of volborthite is taken from Ref.\textsuperscript{19} and Ref.\textsuperscript{18} respectively.

We have adopted this analysis for Cd-K and fitted $\kappa_{\text{2D}}^{xy}$ by tuning the fitting parameters of $J$ and $D$. Figure 10 shows the results of the fitting by the fitting parameters listed in Table I. As shown in Fig. 10, all the data well converges to one single curve given by the SBMFT (solid line in Fig. 10). However, $J = 30$ K used for the fit of Cd-K is considerably smaller than that estimated by the temperature dependence of $\chi$ ($J = 45$ K\textsuperscript{41}). More importantly, the magnitude of $D$ used to
TABLE I. Values of $J$ and $|D/J|$ used to fit $\kappa_{xy}^{2D}$ to the SBMFT simulation (Fig. 10) for kagome lattice antiferromagnets. The data of Ca-K and that of volborthite is taken from Ref.[19] and Ref.[18], respectively.

| Material          | Sample No. | $J/k_B$ (K) | $D/J$  |
|-------------------|------------|-------------|--------|
| Cd-Kapellasite    | 1          | 30          | 0.28   |
|                   | 2          | 30          | 0.09   |
|                   | 3-1        | 29          | 0.65   |
|                   | 3-2        | 28          | 0.6    |
| Ca-Kapellasite    | 66         | 0.12        |
| Volborthite       | 60         | −0.07       |

The fit of $\kappa_{xy}$ of Cd-K differs in a factor of 7 among the Cd-K samples owing to the very different magnitudes of $\kappa_{xy}$. This large difference of $D$ in Cd-K samples is too large to explain it by the ambiguity in estimating the sample dimensions. In addition, the largest value of $D/J = 0.65$ is unphysically larger than the value of $D/J \sim 0.19$ estimated from the deviation of the $g$ factor from 241. This is in sharp contrast to the analysis done for Ca-K in which both $J$ and $D$ determined by the SBMFT fit of $\kappa_{xy}$ well coincide with the value estimated from the temperature dependence of $\chi$ and that from the deviation of the $g$ factor, respectively. These results indicate that the origin of the thermal Hall effect in Cd-K is different from the spin thermal Hall effect observed in Ca-K.

To investigate the mechanism of the thermal Hall effect in Cd-K, we check the dependence of the maximum of $|\kappa_{xy}^{2D}|/TB$ on $\kappa_{xx}/T$ at the peak temperature of $|\kappa_{xy}^{2D}|/TB$ for Cd-K, Ca-K and volborthite (Fig. 11). As shown in Fig. 11, the maximum of $|\kappa_{xy}^{2D}|/TB$ is looked constant for $\kappa_{xx}/T < 0.15 \text{ W K}^{-2} \text{ m}^{-1}$, which is followed by a linear increase for $\kappa_{xx}/T > 0.15 \text{ W K}^{-2} \text{ m}^{-1}$. This $\kappa_{xx}$ dependence bears similarity to that of the anomalous Hall effect (AHE) in ferromagnetic metals. In the AHE, it has been known that the dominant mechanism of AHE depends on the magnitude of the longitudinal conductivity; the intrinsic mechanism by the Berry curvature of the energy bands is dominant in a moderate dirty metal whereas the extrinsic mechanism by skew scatterings is dominant for a super-clean metal. This good analogy between the thermal Hall effect in the kagome materials and the AHE in ferromagnetic metals implies a presence of another mechanism and/or carriers for the thermal Hall effect in a good insulator.

In the AHE in ferromagnetic metals, both the longitudinal and the transverse conductions are carried by electrons. In contrast, $\kappa_{xx}$ of Cd-K is given by a sum of $\kappa_{xx}^{ph}$ and $\kappa_{xx}^{sp}$. Therefore, $\kappa_{xy}$ of
Cd-K can also be given a phonon contribution $\kappa_{xy}^{ph}$ in addition to the spin contribution $\kappa_{xy}^{sp}$ which is given by the SBMFT for Ca-K. As discussed in section IV A, a larger $\kappa_{xx}$ of Cd-K reflects a larger $\kappa_{xx}^{ph}$ given by a longer $\ell_{ph}$ in a better-quality sample. We therefore, from the positive correlation between $\kappa_{xy}$ and $\kappa_{xx}$ observed in Cd-K (Fig. 11), tentatively conclude that an extrinsic part of the thermal Hall conduction in Cd-K is phonon-mediated.

Thermal Hall effects of phonons has been reported in various compounds. In the nonmagnetic insulator SrTiO$_3$, in which only phonons are responsible for the thermal transport, $\kappa_{xy}$ of phonons is found to show a peak at the same temperature of the peak in $\kappa_{xx}$. We thus checked this relation for Cd-K. In Cd-K, $\kappa_{xx}$ of Cd-K contains a spin contribution $\kappa_{xx}^{sp}$ which becomes dominant at lower temperatures. On the other hand, as shown in Fig. 4, a magnetic field suppresses a large portion of $\kappa_{xx}^{sp}$ whereas it slightly increases $\kappa_{xx}^{ph}$. Therefore, $\kappa_{xx}^{ph}$ can be estimated by $\kappa_{xx}$ at 15 T.

Figure 12 shows the temperature dependence of $\kappa_{xx}/T$ at 15 T (left axis) and that of $\kappa_{xy}/T$ (right axis) of all Cd-K samples. As shown in Fig. 12 $\kappa_{xx}/T$ at 15 T shows a peak at almost the same temperature of the peak of $\kappa_{xy}/T$, which resembles the case of the phonon thermal Hall effect observed in SrTiO$_3$ (Ref. 29). Therefore, this temperature dependence showing a peak at a similar
temperature indicates that $\kappa_{xy}$ of Cd-K contains a dominant phonon contribution. We also find that this is clearly not the case for Ca-K. As shown in Figs. 12(e) and (f), $\kappa_{xx}/T$ at 15 T peaks at a much lower temperature than that of $\kappa_{xy}/T$, which is consistent with the spin origin of $\kappa_{xy}$ in Ca-K.

![Graphs showing temperature dependence of $\kappa_{xx}$ and $\kappa_{xy}$](image)

FIG. 12. Temperature dependence of $\kappa_{xx}/T$ at 15 T (left axis) and that of $\kappa_{xy}/T$ (right axis) of all Cd-K samples (a–d), and Ca-K (e, f). The data of Ca-K is taken from Ref. 19.

The energy scale of the phonon thermal Hall effect should be given by the Debye temperature, which is 180 K for Cd-K. Since this energy scale is order of magnitude larger than that of magnetic field of 15 T, $\kappa_{xy}^{ph}$ is expected to have a linear field dependence. On the other hand, as shown in Fig. 6, $\kappa_{xy}$ at low temperatures shows a non-linear field dependence that the slope of $\kappa_{xy}/B$ is larger at lower fields. As discussed in section IV A, $\kappa_{xy}^{sp}$ shows a similar field suppression effect which becomes larger at lower temperatures. Therefore, the non-linear field dependence of $\kappa_{xy}$ suggests a presence of a spin contribution $\kappa_{xy}^{sp}$ which is related to $\kappa_{xx}^{sp}$. We thus conclude that the thermal Hall effect in Cd-K contains both $\kappa_{xx}^{ph}$ and $\kappa_{xy}^{sp}$, and that $\kappa_{xy}^{sp}$ becomes larger at lower temperatures as $\kappa_{xx}^{sp}$ does.

In the AFM phase of Cd-K, no thermal Hall effect is observed below ~ 7 T (Fig. 5(c)), showing
that both $\kappa_{xy}^{\text{ph}}$ and $\kappa_{xy}^{\text{sp}}$ disappear in the AFM phase at low fields. This field dependence is quite different from that of Ca-K$^{19}$ where $\kappa_{xy}^{\text{sp}}$ is observed only below $\sim 6$ T. This contrasting field dependence of $\kappa_{xy}^{\text{sp}}$ in Cd-K implies that the ordered state in Cd-K has a different magnon spectrum from that of Ca-K, possibly by the larger $D$ in Cd-K.

The temperature dependence of the high-field $\kappa_{xy}$ below $T_N$ well follows that at the spin liquid phase (the inset of Fig. 8(b)). Given that $\kappa_{xy}^{\text{ph}}$ is dominant at 15 T, this high-field $\kappa_{xy}$ below $T_N$ is also given by phonons. Therefore, the field dependence of $\kappa_{xy}$ in the AFM phase (Fig. 5(c) and Fig. 7) shows that the linear field dependence of $\kappa_{xy}^{\text{ph}}$ disappears at low fields whereas it comes back at high fields. This absence of $\kappa_{xy}^{\text{ph}}$ in the low-field AFM phase demonstrates that the phonons cannot alone exhibit the thermal Hall effect, putting a strong constraint on the origin of the phonon Hall effect. In other words, $\kappa_{xy}^{\text{ph}}$ requires the field-induced excitations observed in the field dependence of $\kappa_{xx}$ (Fig. 4(c)) and that of $C$ (Fig. 7(b)).

Thermal Hall effects by phonons have been theoretically studied with respect to various aspects.$^{31-39}$ The absence of the stand-alone phonon thermal Hall effect in Cd-K is inconsistent with intrinsic mechanism, but rather points to extrinsic origins with microscopic couplings between phonons and the field-induced excitations. Such microscopic coupling has also been suggested to play an important role in the thermal Hall effect observed in multiferroics$^{31}$ where a large thermal Hall effect is observed in the ferrimagnetic phase despite the absence of the conventional magnon Hall effect. It has been pointed out that a magnon-phonon coupling induces a thermal Hall effect even in the system where neither phonons nor magnons alone show a thermal Hall effect.$^{35}$ Therefore, the absence and the presence of $\kappa_{xy}^{\text{ph}}$ in the AFM phase of Cd-K suggests that the phonon thermal Hall effect in Cd-K has an extrinsic origin requiring a coupling with the field-induced excitations. At present, the details of the magnetic structure of Cd-K have not been known. Further studies, including NMR or neutron scattering experiments to clarify the magnetic excitations in the AFM phase under the low and the high magnetic fields, will be important to reveal the origins of the thermal Hall effects in Cd-K.

One clearly has to wonder why, despite the dual origin (spins and phonons) of the thermal Hall effects in the Cd-K compounds, the scaling fit derived entirely from the SBMFT works so well as shown in Fig. 10. This is not an unreasonable conclusion, however, provided we further assume that $\kappa_{xy}^{\text{sp}}$ is proportional to $\kappa_{xy}^{\text{ph}}$, due to the fact that the two excitations are microscopically coupled.
V. SUMMARY

We have investigated $\kappa_{xx}$ and $\kappa_{xy}$ of three Cd-K samples. From the large field suppression effect on $\kappa_{xx}$ at low temperatures, we find a dominant contribution of $\kappa_{xx}^{sp}$ in $\kappa_{xx}$ at low temperatures. Below $T_N$, we find a new peak in the field dependence of $\kappa_{xx}$ at $\sim 7$ T. Above 7 T, we also find a field-induced increase both in the specific heat and in the thermal Hall effect.

Clear thermal Hall effects have been observed in the spin liquid states of all Cd-K samples. We find that the temperature dependence of $\kappa_{xy}$ shows a very similar temperature dependence with a peak at almost the same temperature. On the other hand, the magnitude of $\kappa_{xy}$ depends on $\kappa_{xx}$. This $\kappa_{xy}$ dependence is inconsistent with the spin thermal Hall effect observed in the related compound in Ca-K. We conclude that a phonon thermal Hall effect observed in the related compound from the positive correlation between $\kappa_{xy}$ and $\kappa_{xx}$ (Fig. 11) and the similar temperature dependence of $\kappa_{xy}$ and $\kappa_{xx}$ at 15 T (Fig. 12). We further find that the non-linear field dependence of $\kappa_{xy}$ at low temperatures (Fig. 6) shows the presence of a spin thermal Hall $\kappa_{xy}^{sp}$ at low temperatures and at low fields. These results demonstrate that the spin liquid state of Cd-K harbors thermal Hall effects of both spins and phonons.

Remarkably, both $\kappa_{xy}^{ph}$ and $\kappa_{xy}^{sp}$ disappear in the AFM phase at low fields. At high fields above 7 T, we find that $\kappa_{xy}^{ph}$ is induced concomitantly with the field induced excitations observed in the field dependence of $\kappa_{xx}$ and $C$. These results suggest that field-induced excitations give rise to a recovery of the phonon thermal Hall effect of Cd-K. We conclude that the phonons alone do not exhibit the thermal Hall effect and require to merge with the field-induced excitations to appear.

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