Evidence of a Phonon Hall Effect in the Kitaev Spin Liquid Candidate α-$RuCl_3$

É. Lefrançois,[1,†] G. Grissonnanche,[1] J. Baglo,[1] P. Lampen-Kelley,[2,3] J.-Q. Yan,[2] C. Balz,[4,*] D. Mandrus,[2,3] S. E. Nagler,[4] S. Kim,[5] Young-June Kim,[5] N. Doiron-Leyraud,[1] and Louis Taillefer[1,6,‡]

[1]*Institut Quantique, Département de physique and RQMP, Université de Sherbrooke, Sherbrooke, Québec, Canada*
[2]*Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA*
[3]*Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee, USA*
[4]*Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA*
[5]*Department of Physics, University of Toronto, Toronto, Ontario, Canada*
[6]*Canadian Institute for Advanced Research, Toronto, Ontario, Canada*

(Received 11 November 2021; revised 2 February 2022; accepted 22 February 2022; published 29 April 2022)

The material α-$RuCl_3$ has been the subject of intense scrutiny as a potential Kitaev quantum spin liquid, predicted to display Majorana fermions as low-energy excitations. In practice, α-$RuCl_3$ undergoes a transition to a state with antiferromagnetic order below a temperature $T_N \approx 7$ K, but this order can be suppressed by applying an external in-plane magnetic field of $H_\parallel = 7$ T. Whether a quantum spin liquid phase exists just above that field is still an open question, but the reported observation of a quantized thermal Hall conductivity at $H_\parallel > 7$ T by Kasahara and co-workers [Nature (London) **559**, 227 (2018)] has been interpreted as evidence of itinerant Majorana fermions in the Kitaev quantum spin liquid state. In this study, we reexamine the origin of the thermal Hall conductivity κ_{xy} in α-$RuCl_3$. Our measurements of $\kappa_{xy}(T)$ on several different crystals yield a temperature dependence very similar to that of the phonon-dominated longitudinal thermal conductivity $\kappa_{xx}(T)$, for which the natural explanation is that κ_{xy} is also mostly carried by phonons. Upon cooling, κ_{xx} peaks at $T \simeq 20$ K, then drops until T_N, whereupon it suddenly increases again. The abrupt increase below T_N is attributed to a sudden reduction in the scattering of phonons by low-energy spin fluctuations as these become partially gapped when the system orders. The fact that κ_{xy} also increases suddenly below T_N is strong evidence that the thermal Hall effect in α-$RuCl_3$ is also carried predominantly by phonons. This implies that any quantized signal from Majorana edge modes would have to come on top of a sizable—and sample-dependent—phonon background.

DOI: 10.1103/PhysRevX.12.021025

Subject Areas: Condensed Matter Physics, Magnetism
Strongly Correlated Materials

I. INTRODUCTION

The quasi-2D Mott insulator α-$RuCl_3$ has attracted much interest since it was proposed as a promising material for realizing a Kitaev spin liquid state, thanks to its honeycomb lattice with anisotropic bond-directional spin interactions [1,2]. The Kitaev model predicts the existence of mobile Majorana fermions as quasiparticles that would manifest as topologically protected heat carriers on the edges of the sample. In principle, such Majorana edge modes could be detected by measuring the thermal Hall effect [3–5], and their signature would be a half-integer quantized thermal Hall conductivity per plane $\kappa_{xy}^{2D} = \kappa_{xy}\, d$ at low temperature (d is the honeycomb interlayer distance). However, because α-$RuCl_3$ orders antiferromagnetically at low temperature, below $T_N = 7$ K, in order to access the putative spin liquid phase one must apply an in-plane magnetic field in excess of a critical field $H_\parallel = 7$ T to suppress the magnetic order. The possibility thus arises that a spin liquid phase could emerge immediately above that critical field. An inelastic neutron scattering study finds a magnetically disordered state at $T \to 0$, sandwiched between the ordered phase below $H_\parallel = 7$ T and a field-polarized phase above $H_\parallel \simeq 9$ T, which is reminiscent of a quantum spin

*Present address: ISIS Neutron and Muon Source, STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, United Kingdom.
†etienne.lefrancois@usherbrooke.ca
‡louis.taillefer@usherbrooke.ca

liquid [6]. A few studies of heat transport have reported the observation of a quantized κ_{xy} in some samples of α-$RuCl_3$, in a narrow range of in-plane fields ($6 < H_{\parallel} < 9$ T) and temperatures ($3 < T < 6$ K) [7–9]. However, since then, a number of studies have reported κ_{xy} signals in various insulators and attributed them to phonons [10–14]. The κ_{xy} response can have either sign, and the magnitude of κ_{xy} is easily as large as that found in α-$RuCl_3$, often larger. Typically, in those studies that attribute the thermal Hall effect to phonons, one finds that the magnitude of κ_{xy} scales roughly with the magnitude of the phonon thermal conductivity κ_{xx}, with a ratio $|\kappa_{xy}/\kappa_{xx}|$ of order 10^{-3}. These studies raise the possibility that phonons might also generate a thermal Hall effect in α-$RuCl_3$.

In this paper, we explore that possibility. We measured the thermal conductivity κ_{xx} and thermal Hall conductivity κ_{xy} in several samples of α-$RuCl_3$, coming from two different sources. We find a substantial variation in the magnitude of κ_{xx} and κ_{xy} from sample to sample, but the same qualitative behavior. Upon cooling from 80 K, κ_{xx} and κ_{xy} exhibit similar behaviors, namely a peak in κ/T versus T at roughly the same temperature $T \simeq 20$ K, followed by a dip toward T_N. If the magnetic field is applied along the c^* direction, both κ_{xx}/T and κ_{xy}/T exhibit a rapid rise below T_N. If a magnetic field is applied such that its in-plane component is $H_{\parallel} = 7$ T, thereby suppressing the magnetic order, both κ_{xx}/T and κ_{xy}/T continue their monotonic decrease as $T \to 0$. The fact that $\kappa_{xy}(T)$ mimics the phonon-dominated $\kappa_{xx}(T)$ is strong evidence that the thermal Hall signal is also dominated by phonons. Moreover, we find that $\kappa_{xy}/\kappa_{xx} \simeq 10^{-3}$ at low temperature, a magnitude typical of the phonon Hall effect in other insulators. Finally, the fact that there is a sizable κ_{xy} signal below T_N, in the antiferromagnetic phase, shows that it does not simply arise from the excitations of a pure spin liquid phase. All this calls into question any interpretation of prior data exclusively in terms of Majorana fermions.

II. METHODS

A. Samples

We measured a variety of samples coming from two different groups: Oak Ridge National Laboratory (ORNL) and the University of Toronto (UofT). Single crystals from ORNL were grown using the vapor transport method on α-$RuCl_3$ powder coming from Furuya Metal Co., Ltd; the growth technique is detailed elsewhere [15]. Single crystals from UofT were grown using the same method but using powder coming from Sigma-Aldrich. The powder used was composed of 45%–55% ruthenium and was sealed in a quartz tube under vacuum. The latter was placed inside a two-zone tube furnace using a temperature gradient of 70 °C (warmest side was 850 °C). The powder was annealed over two days, followed by a 4 °C/h cooldown while maintaining the temperature gradient (see Ref. [16] for more details). Here we report data on four samples from ORNL (labeled O1, O2, O3, and O4) and one sample from UofT (labeled T1). The samples from ORNL were cut into thin rectangular platelets of typical dimensions 1×1 mm, with thicknesses ranging from 20 to 140 μm, whereas the sample T1 from UofT is as grown (not cut). Specific heat and magnetization curves versus T in high-quality crystals from the same batch as sample T1 show no trace of extra phases. Sample T1 was handled very carefully, so as to not induce any strain in the mounting process. The magnetic field was only applied parallel to the c^* axis, thereby producing no torque. The higher conductivity κ_{xx} of sample T1 compared to the other samples [Fig. 2(a)] may, as a result, reflect a higher structural quality. Note, however, that all our findings are qualitatively consistent across all samples, leading to the same conclusions. The contacts on the samples were made by gluing thin silver leads with silver paste.

B. Measurement technique

Measurements were performed by a steady-state method using a standard four-terminal technique, with the thermal current applied along the length of the sample within the honeycomb layers (perpendicular to the Ru-Ru bonds; $J||a$). The thermal conductivity κ_{xx} was measured by employing a standard one-heater–two-thermometers method, using a 5 kΩ resistor and two *in situ* calibrated bare chip CX-1050 Cernox sensors. A constant heat current J was injected at one end of the sample, while at the other end the sample is well heat sunk to a copper block referenced at a temperature T_0 (see Fig. 1). The heat current was generated by sending an electrical current through a 5 kΩ strain gauge whose resistance is marginally dependent on temperature and magnetic field. A longitudinal thermal gradient $\Delta T_x = T^+ - T^-$ is measured at two points along the length of the sample, separated by a distance l. The longitudinal thermal

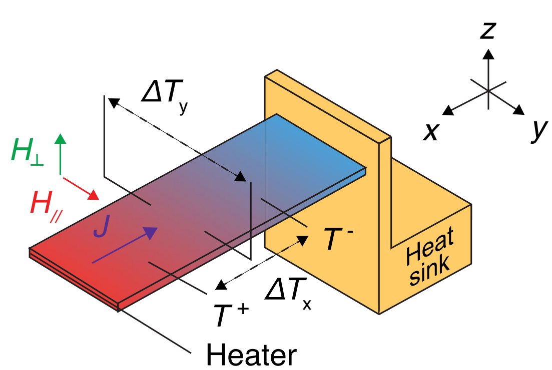

FIG. 1. Sketch of the thermal Hall conductivity measurement setup. Directions of both thermal current and external magnetic field are shown with colored arrows; the latter can be decomposed into two components $H_{\parallel}$ (red) and $H_{\perp}$ (green) if the total field H makes an angle with the sample's basal plane.

conductivity is given by $\kappa_{xx} = J/(\Delta T_x \alpha)$, where $\alpha = wt/l$ is the geometric factor of the sample (width w, thickness t).

Under a magnetic field applied perpendicular to the basal plane, a transverse thermal gradient ΔT_y resulted (see Fig. 1). This transverse gradient was measured using a differential type-E thermocouple. The field can also be tilted at an angle, such that it will have a component in the plane ($H_{\parallel}$), as well as normal to the plane ($H_{\perp}$). When the field direction is between the plane and the c^* direction, a torque is applied to the sample, which can sometimes detach the sample from its mount or bend the sample. We performed several attempts with a field at 45°, using different samples. For the measurement reported here at 45° out of the basal plane, the experiment worked well: the sample was glued firmly enough to withstand the torque, and was robust enough not to be bent by the external magnetic field. It emerged unaffected from the experiment and the data are reliable.

The thermal Hall conductivity is then given by $\kappa_{xy} = \kappa_{yy}(\Delta T_y/\Delta T_x)(l/w)$ after having antisymmetrized the thermal Hall gradient via $\Delta T_y(H) = [\Delta T_y(T,H) - \Delta T_y(T,-H)]/2$ (we take $\kappa_{yy} = \kappa_{xx}$). The error bars on the absolute values of thermal coefficients come mostly from the uncertainty in estimating the dimensions (l, w, and t) of the samples, approximately $\pm 20\%$. The applied current was chosen such that $\Delta T/T \simeq 5\%$–10%; the resulting κ_{xx} was independent of ΔT, indicating that there was no heat loss. Any contamination of $\Delta T_y(H)$ coming from the copper heat sink was ruled out by a previous study that compared heat sinks made of Cu versus LiF (see Supplemental Information in Ref. [14]). Moreover, ΔT_y data obtained with thermocouples were found to be in good agreement with ΔT_y data obtained with Cernox sensors applied to the same sample.

III. RESULTS

In Fig. 2(a), we show the thermal conductivity of α-$RuCl_3$ measured in five crystals, plotted as κ_{xx}/T versus T. We see that there is a considerable variation in the magnitude of κ_{xx} among samples, by a factor 3 or so, with the largest $\kappa_{xx}(T)$ value seen in sample T1. Prior data by Leahy *et al.* [17] and Hentrich *et al.* [18] fall within the range of magnitudes of our own samples. Kasahara *et al.* [19] find a $\kappa_{xx}(T)$ that is 2–3 (7–8) times larger than our data on sample T1 (O2). This variation in magnitude is attributed to different levels of disorder, perhaps structural (associated with domains that form upon cooling through the structural transition at 130 K). Despite this quantitative variation, the qualitative behavior of $\kappa_{xx}(T)$ is the same in all samples. There is a peak in κ_{xx}/T versus T at $T \simeq 20$ K, below which κ_{xx}/T drops as $T \to T_N$. As argued by Hentrich *et al.* [18], the dominant carriers of heat in α-$RuCl_3$ are phonons, and these become increasingly scattered by low-energy antiferromagnetic spin fluctuations upon cooling below 80 K. It is this scattering that causes κ_{xx}/T to drop as $T \to T_N$ (from above). Application of a magnetic field in the plane gaps the low-energy spin fluctuation spectrum, causing κ_{xx} at $T = 10$ K ($> T_N$) to increase rapidly for $H > 10$ T [18].

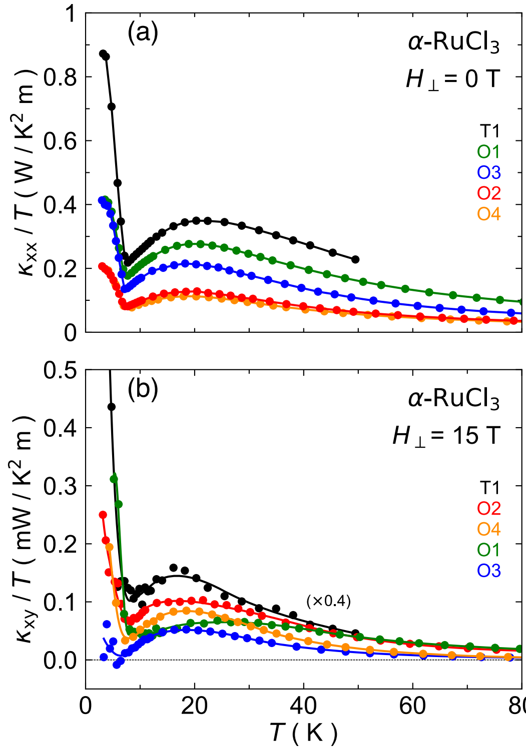

FIG. 2. (a) Thermal conductivity of α-$RuCl_3$ in zero magnetic field, plotted as κ_{xx}/T versus T, for our five different samples: T1 (black), O1 (green), O2 (red), O3 (blue), and O4 (gold). (b) Thermal Hall conductivity of the same five samples, measured in a magnetic field of $H = 15$ T applied normal to the honeycomb layers (ab plane), plotted as κ_{xy}/T versus T. The κ_{xy} data for sample T1 (black) have been multiplied by a factor 0.4. Lines are a guide to the eye.

As we cool below T_N, at $T < 7$ K ($< T_N$), we see that κ_{xx}/T (in zero field) shoots up immediately in all samples [Fig. 2(a) and Refs. [17–19]], presumably because the magnetic order also causes a gapping of the low-energy spin fluctuation spectrum. This V-shaped dependence of κ_{xx}/T versus T at $H = 0$ T is mimicked by a similar V-shaped dependence of κ_{xx} versus H at $T < 1$ K, with the minimum at the critical field of 7 T [20]. In summary, the thermal conductivity of α-$RuCl_3$ at low temperature ($T < 50$ K) can be understood essentially in terms of phonons scattered by spin fluctuations.

In Fig. 2(b), we show the thermal Hall conductivity of α-$RuCl_3$ measured in the same 5 samples, plotted as κ_{xy}/T versus T for a field $H = 15$ T applied normal to the plane ($H \| c^*$). There is considerable variation in the magnitude of κ_{xy} across samples, even larger than that seen in κ_{xx}. Prior data by Hentrich *et al.* [21] (with $H = 16$ T) yield a $\kappa_{xy}(T)$ curve very similar, quantitatively and qualitatively, to the curve for our sample O2 (for which $H = 15$ T). Kasahara *et al.* [19] find a $\kappa_{xy}(T)$ curve that is 3–4 times larger than that. Despite the quantitative variation, the qualitative behavior of $\kappa_{xy}(T)$ is the same in all samples from all groups—at least for $T > T_N$. The thermal Hall signal is positive, and there is a peak in κ_{xy}/T versus T at $T \simeq 20$ K, below which κ_{xy}/T drops as $T \to T_N$. In other words, $\kappa_{xy}(T)$ mimics the phonon-dominated $\kappa_{xx}(T)$.

Below T_N, we observe an increase in $\kappa_{xy}(T)$ upon cooling [Fig. 2(b)], in all our samples. Although $\kappa_{xy}(T)$ dips down to a minimum at T_N, in one case getting close to zero (sample O4), it always remains positive. Data by Hentrich *et al.* [21] on one sample show a $\kappa_{xy}(T)$ curve that dips to zero at T_N, becoming perhaps very slightly negative; however, no data were reported for $T < 7$ K. The data by Kasahara *et al.* [19] do extend below T_N, where they report a negative κ_{xy} signal, in contrast to our positive signal. It is not clear where this discrepancy comes from. We note that Kasahara *et al.* also report that κ_{xy} goes from positive to negative when the temperature is raised above $T \simeq 55$ K, a sign change that is seen neither by Hentrich *et al.* nor by us, in any sample [Fig. 2(b)]. In summary, our $\kappa_{xy}(T)$ data from all five samples show a clear similarity with the phonon-dominated $\kappa_{xx}(T)$ data, not only above T_N, but also below T_N.

In Fig. 3, we compare these two curves for one sample (O2), where the similarity between $\kappa_{xx}(T)$ [Fig. 3(a)] and $\kappa_{xy}(T)$ [Fig. 3(b)] is striking. In Fig. 3(c), we plot the ratio κ_{xy}/κ_{xx} versus T, measured at $H = 15$ T, and see that κ_{xy} is approximately 1000 smaller than κ_{xx}, at $T \simeq 20$ K. The same is true for sample T1, whose κ_{xy} amplitude is 3–4 times larger (at $T \simeq 20$ K; see Fig. 2). For our five samples, the ratio at $T = 20$ K and $H = 15$ T ranges from 0.03% to 0.10%. This is consistent with prior data by Hentrich *et al.* [21] and Kasahara *et al.* [19], where $\kappa_{xy}/\kappa_{xx} \simeq 0.05\%$. As we discuss below, a ratio of this magnitude is typical for a phonon thermal Hall effect.

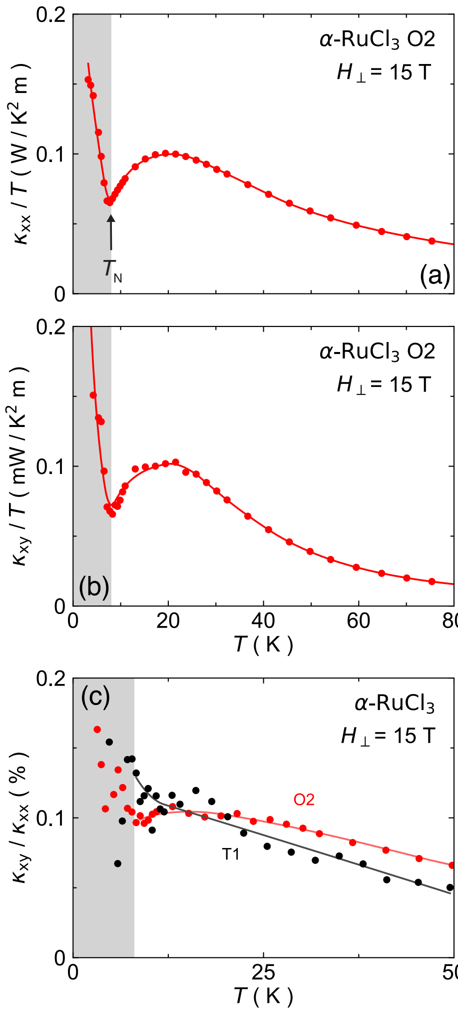

FIG. 3. (a) Thermal conductivity of sample O2, plotted as κ_{xx}/T versus T, for $H||c^* = 15$ T. (b) Thermal Hall conductivity of the same sample, plotted as κ_{xy}/T versus T, under the same conditions. (c) Ratio κ_{xy}/κ_{xx} (sample O2, red): data in (b) over data in (a). Corresponding data for sample T1 are shown in black. The gray shaded area delineates the region below T_N (arrow) where antiferromagnetic order is present. Lines are a guide to the eye.

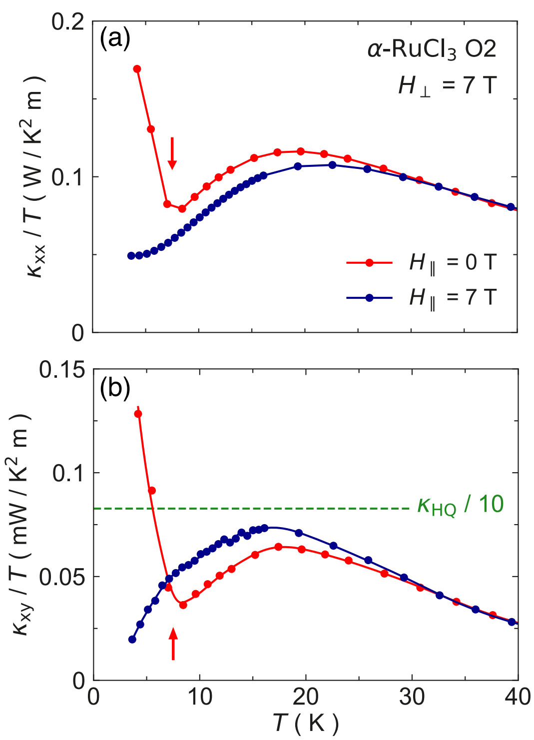

FIG. 4. (a) Thermal conductivity κ_{xx} of sample O2, plotted as κ_{xx}/T versus T, for an applied magnetic whose component normal to the honeycomb layers is $H_\perp = 7$ T and whose component parallel to the layers is either $H_\parallel = 0$ T (red) or $H_\parallel = 7$ T (blue). (b) Same as in (a), for κ_{xy}. The horizontal dashed line marks the quantized value (κ_{HQ}) expected for Majorana edge modes, divided by 10. The arrows mark T_N. Lines are a guide to the eye.

In Fig. 4, we show the effect of applying a component of the field parallel to the honeycomb layers, so that the antiferromagnetic order is suppressed. The red curves are for $H_\perp = 7$ T and no in-plane field ($H_\parallel = 0$ T). Both curves—$\kappa_{xx}(T)$ [Fig. 4(a)] and $\kappa_{xy}(T)$ [Fig. 4(b)]—are very similar to those in Fig. 3, where $H_\perp = 15$ T (and $H_\parallel = 0$ T). The only difference is quantitative: the magnitude of κ_{xy} is down roughly by a factor 7 T/15 T (the reduction in $H_\perp$). Adding an additional 7 T field component in the plane (blue curves) has only a small effect above 7 K, but a dramatic one below 7 K: now $\kappa_{xx}(T)$ and $\kappa_{xy}(T)$ no longer suddenly increase below 7 K ($= T_N$) but continue to decrease smoothly through 7 K, in the absence of ordering. This makes sense for phonons, which continue to be scattered by low-lying spin fluctuations that remain ungapped down to the lowest temperature. So again, the striking similarity between $\kappa_{xy}(T)$ and the phonon-dominated $\kappa_{xx}(T)$ argues for a thermal Hall signal carried by the phonons.

IV. DISCUSSION

Traditionally, the thermal Hall effect from phonons has been considered very small [22]. First observed in 2005, its magnitude in $Tb_3Ga_5O_{12}$ was detected to be $\kappa_{xy} \simeq 0.02$ mW/K m at $H = 3$ T and $T = 5$ K [23]. This is 2 orders of magnitude smaller than the signal first reported in α-$RuCl_3$, in 2017: $\kappa_{xy} \simeq 4$ mW/K m at $H = 6$ T and $T = 20$ K [19]. So it was perhaps natural to rule out phonons in 2017, although that year a large thermal Hall effect was reported in the antiferromagnetic insulator $Fe_2Mo_3O_8$ and attributed to phonons (and their coupling to spins), with $\kappa_{xy} \simeq 30$ mW/K m at $H = 14$ T and $T = 35$ K [24], one order of magnitude larger than in α-$RuCl_3$. Since then, it has become abundantly clear that phonons can carry a sizable thermal Hall conductivity in various insulators, whether magnetic, as in the antiferromagnetic cuprate Mott insulator La_2CuO_4 [10,13], or nonmagnetic, as in the quantum paraelectric $SrTiO_3$ [12]. The magnitude of κ_{xy} (at $H = 15$ T and $T = 20$ K) can vary from $|\kappa_{xy}| \simeq 1$ mW/K m in $Tb_2Ti_2O_7$ [11,22] to $|\kappa_{xy}| \simeq 200$ mW/K m in Nd_2CuO_4 [14] and become as large as $|\kappa_{xy}| \simeq 1000$ mW/K m in Cu_3TeO_6 [25]. Although the mechanisms by which phonons acquire a handedness (become chiral) in a magnetic field are still unclear in these materials, the cumulative evidence that phonons are the carriers of the thermal Hall effect in these insulators is strong: although $|\kappa_{xy}|$ varies by 3 orders of magnitude, the ratio of κ_{xy} over the phonon-dominated κ_{xx} is roughly the same, namely, $|\kappa_{xy}/\kappa_{xx}| \simeq 2$–$5 \times 10^{-3}$ in all cases (see Table 1 in Ref. [25]). In other words, what really varies from material to material is the ability of phonons to conduct heat, i.e., the magnitude of κ_{xx}.

Being now aware of these more recent studies, it seems very likely that phonons must also generate a sizable κ_{xy} signal in the antiferromagnetic insulator α-$RuCl_3$, especially given the measured ratio $|\kappa_{xy}/\kappa_{xx}| \simeq 1 \times 10^{-3}$ [at $H = 15$ T and $T = 20$ K; see Fig. 3(c)]. [The ratio κ_{xy}/κ_{xx} varies somewhat from sample to sample, namely, from $\simeq 0.03\%$ in samples O1 and O3 to $\simeq 0.1\%$ in samples O2, O4 and T1, at $T = 20$ T and $H = 15$ T. Note that the phonon conductivity κ_{xx} also varies by a factor 3 or so [Fig. 2(a)], due to varying degrees of disorder.] We emphasize that the ratio κ_{xy}/κ_{xx} has the same magnitude ($\simeq 0.1\%$ in sample O2) inside the quantum spin liquid regime (at $H_\parallel = 7$ T and $T \simeq 5$ K) (Fig. 4) and outside that regime (at $H_\perp = 15$ T, $H_\parallel = 0$ T, and $T \simeq 20$ K) (Fig. 3). The immediate implication of having a sizable phonon contribution to κ_{xy} in α-$RuCl_3$ is that the total measured value of κ_{xy} in any sample would include a sizable phonon background, to which any contribution from Majorana fermions would add. In this context, the reported observation of a plateau in κ_{xy}/T, over a small range of in-plane fields ($6 < H_\parallel < 9$ T) and temperatures ($3 < T < 6$ K) [7], with a measured total value of κ_{xy}/T having precisely the half-quantized value of $\pi k_B^2/12\hbar$ per plane expected theoretically for Majorana edge modes [3], can only be meaningful if the phonon background is precisely zero. As we have argued, this condition is unlikely to be satisfied in α-$RuCl_3$, at least when there is a component of the magnetic field normal to the plane (and to the heat current), which was the case in the study of Ref. [7]. It is not clear at this stage whether phonons in α-$RuCl_3$ could produce a transverse thermal gradient even when the field is applied entirely in the plane ($H_\perp = 0$), e.g., along the a axis. This open question requires new theoretical work, but it has been suggested, for instance, that the anisotropic g factor of α-$RuCl_3$ could cause such a planar thermal Hall effect (where $J \| H \| a$). It may also be that the thermal Hall conductivity of α-$RuCl_3$ is not in fact half-quantized, as recently argued on the basis of new data [26]. To investigate the same configuration as in Ref. [7], we turn to our data in a tilted field, displayed in Fig. 4 (blue curves). If the entire signal is attributed to Majorana fermions, it is difficult to understand why our value of κ_{xy}/T at $H_\parallel = 7$ T and $T = 6$ K is only $\kappa_{xy}/T \simeq 0.04$ mW/K^2 m [Fig. 4(b)], while the value reported in Ref. [7] is $\kappa_{xy}/T \simeq 0.8$ mW/K^2 m, a factor 20 larger. On the other hand, if the signal is attributed to phonons, then the likely reason for the much smaller κ_{xy} in our sample is simply that its κ_{xx} is much smaller, with $\kappa_{xx} \simeq 0.5$ W/K m at $T = 10$ K and $H_\parallel = 7$ T [Fig. 4(a)], while the value reported in Ref. [7] is $\kappa_{xx} \simeq 5$ W/K m—a factor of 10 larger. In other words, a phonon scenario solves the puzzle of why a thermal Hall conductivity that exceeds the half-quantized value, $\kappa_{xy}/T = \kappa_{HQ} = 0.8$ mW/K^2 m, has only been observed in samples that have the largest values of κ_{xx} [7–9,27]: a larger value of κ_{xy} is what we expect from phonons that conduct better (and thus produce a larger κ_{xx}), as nicely demonstrated by the highly conductive samples of Cu_3TeO_6 [25].

V. SUMMARY

We have measured the thermal conductivity κ_{xx} and the thermal Hall conductivity κ_{xy} of α-$RuCl_3$, for a heat current within the honeycomb layers (perpendicular to the Ru-Ru bond; $J||a$) in five different single crystals, from two separate sources. Although the magnitude of κ_{xx} and κ_{xy} vary significantly from sample to sample, presumably because of varying degrees of structural disorder, we find the same qualitative behavior in all samples. Upon cooling, both $\kappa_{xx}(T)$ and $\kappa_{xy}(T)$ are found to have the same temperature dependence, namely a broad peak located at the same temperature $T \simeq 20$ K, followed by a decrease until $T = 7$ K, whereupon $\kappa_{xx}(T)$ and $\kappa_{xy}(T)$ either rapidly rise in tandem upon entering the antiferromagnetic phase at $T_N = 7$ K, when the field is applied normal to the layers, or both continue their decrease, when the field has an in-plane component sufficient to remove the antiferromagnetic order. The fact that $\kappa_{xy}(T)$ mimics the phonon-dominated $\kappa_{xx}(T)$ so well leads us to conclude that the thermal Hall effect in α-$RuCl_3$ is carried predominantly by phonons. This interpretation is supported by the fact that the magnitude of κ_{xy} in various samples of α-$RuCl_3$ roughly scales with the magnitude of the phonon-dominated κ_{xx}. Moreover, the ratio κ_{xy}/κ_{xx} has a magnitude comparable to that found in several other insulators where phonons have been shown or argued to cause the Hall effect, namely, $|\kappa_{xy}/\kappa_{xx}| \simeq 1 \times 10^{-3}$ (see Table 1 in Ref. [25]). If phonons contribute significantly to the κ_{xy} signal of α-$RuCl_3$ samples, the experimentally measured κ_{xy} cannot be attributed directly and exclusively to theoretically predicted Majorana fermions, as has been done in some reports.

ACKNOWLEDGMENTS

We thank L. Balents, K. Behnia, B. Flebus, C. Hess, H.-Y. Kee, Y. B. Kim, S. A. Kivelson, A. H. MacDonald, N. Perkins, J. A. Quilliam, S. Sachdev, L. Savary, and R. Valentí for stimulating discussions. We thank S. Fortier for his assistance with the experiments and M. Dion for his assistance with sample orientation. L. T. acknowledges support from the Canadian Institute for Advanced Research (CIFAR) as a CIFAR Fellow and funding from the Institut Quantique, the Natural Sciences and Engineering Research Council of Canada (NSERC; PIN:123817), the Fonds de Recherche du Québec–Nature et Technologies (FRQNT), the Canada Foundation for Innovation (CFI), and a Canada Research Chair. This research was undertaken thanks in part to funding from the Canada First Research Excellence Fund. C. B. and S. E. N. were supported by the U.S. Department of Energy (DOE), Basic Energy Sciences, Scientific User Facilities Division. S. E. N. also acknowledges support from the Quantum Science Center (QSC), a National Quantum Information Science Research Center of the U.S. DOE. Materials synthesis by J. Q. Y. was supported by the U.S. DOE, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division. D. M. acknowledges support from the Gordon and Betty Moore Foundation's EPiQS Initiative, Grant No. GBMF9069. Work at the University of Toronto was supported by NSERC (RGPIN-2019-06449 and RTI-2019-00809), CFI, and Ontario Ministry of Research and Innovation.

[1] G. Jackeli and G. Khaliullin, *Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models*, Phys. Rev. Lett. **102**, 017205 (2009).

[2] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, α-$RuCl_3$*: A Spin-Orbit Assisted Mott Insulator on a Honeycomb Lattice*, Phys. Rev. B **90**, 041112(R) (2014).

[3] J. Nasu, J. Yoshitake, and Y. Motome, *Thermal Transport in the Kitaev Model*, Phys. Rev. Lett. **119**, 127204 (2017).

[4] M. Ye, G. B. Halász, L. Savary, and L. Balents, *Quantization of the Thermal Hall Conductivity at Small Hall Angles*, Phys. Rev. Lett. **121**, 147201 (2018).

[5] Y. Vinkler-Aviv and A. Rosch, *Approximately Quantized Thermal Hall Effect of Chiral Liquids Coupled to Phonons*, Phys. Rev. X **8**, 031032 (2018).

[6] C. Balz, P. Lampen-Kelley, A. Banerjee, J. Yan, Z. Lu, X. Hu, S. M. Yadav, Y. Takano, Y. Liu, D. A. Tennant, M. D. Lumsden, D. Mandrus, and S. E. Nagler, *Finite Field Regime for a Quantum Spin Liquid in* α-$RuCl_3$, Phys. Rev. B **100**, 060405(R) (2019).

[7] Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, S. Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi, and Y. Matsuda, *Majorana Quantization and Half-Integer Thermal Quantum Hall Effect in a Kitaev Spin Liquid*, Nature (London) **559**, 227 (2018).

[8] T. Yokoi, S. Ma, Y. Kasahara, S. Kasahara, T. Shibauchi, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, C. Hickey, S. Trebst, and Y. Matsuda, *Half-Integer Quantized Anomalous Thermal Hall Effect in the Kitaev Material Candidate* α-$RuCl_3$, Science **373**, 568 (2021).

[9] J. A. N. Bruin, R. R. Claus, Y. Matsumoto, N. Kurita, H. Tanaka, and H. Takagi, *Robustness of the Thermal Hall Effect Close to Half-Quantization in* α-$RuCl_3$, Nat. Phys. **18**, 401 (2022).

[10] G. Grissonnanche, A. Legros, S. Badoux, E. Lefrançois, V. Zatko, M. Lizaire, F. Laliberté, A. Gourgout, J.-S. Zhou, S. Pyon, T. Takayama, H. Takagi, S. Ono, N. Doiron-Leyraud, and L. Taillefer, *Giant Thermal Hall Conductivity in the Pseudogap Phase of Cuprate Superconductors*, Nature (London) **571**, 376 (2019).

[11] Y. Hirokane, Y. Nii, Y. Tomioka, and Y. Onose, *Phononic Thermal Hall Effect in Diluted Terbium Oxides*, Phys. Rev. B **99**, 134419 (2019).

[12] X. Li, B. Fauqué, Z. Zhu, and K. Behnia, *Phonon Thermal Hall Effect in Strontium Titanate*, Phys. Rev. Lett. **124**, 105901 (2020).

[13] G. Grissonnanche, S. Thériault, A. Gourgout, M.-E. Boulanger, E. Lefrançois, A. Ataei, F. Laliberté, M. Dion, J.-S. Zhou, S. Pyon, T. Takayama, H. Takagi, N. Doiron-Leyraud, and L. Taillefer, *Chiral Phonons in the Pseudogap Phase of Cuprates*, Nat. Phys. **16**, 1108 (2020).

[14] M.-E. Boulanger, G. Grissonnanche, S. Badoux, A. Allaire, E. Lefrançois, A. Legros, A. Gourgout, M. Dion, C. H. Wang, X. H. Chen, R. Liang, W. N. Hardy, D. A. Bonn, and L. Taillefer, *Thermal Hall Conductivity in the Cuprate Mott Insulators* Nd_2CuO_4 *and* $Sr_2CuO_2Cl_2$, Nat. Commun. **11**, 5325 (2020).

[15] A. F. May, J. Yan, and M. A. McGuire, *A Practical Guide for Crystal Growth of van der Waals Layered Materials*, J. Appl. Phys. **128**, 051101 (2020).

[16] J. A. Sears, Y. Zhao, Z. Xu, J. W. Lynn, and Y.-J. Kim, *Phase Diagram of* α-$RuCl_3$ *in an In-Plane Magnetic Field*, Phys. Rev. B **95**, 180411(R) (2017).

[17] I. A. Leahy, C. A. Pocs, P. E. Siegfried, D. Graf, S.-H. Do, K.-Y. Choi, B. Normand, and M. Lee, *Anomalous Thermal Conductivity and Magnetic Torque Response in the Honeycomb Magnet* α-$RuCl_3$, Phys. Rev. Lett. **118**, 187203 (2017).

[18] R. Hentrich, A. U. B. Wolter, X. Zotos, W. Brenig, D. Nowak, A. Isaeva, T. Doert, A. Banerjee, P. Lampen-Kelley, D. G. Mandrus, S. E. Nagler, J. Sears, Y.-J. Kim, B. Büchner, and C. Hess, *Unusual Phonon Heat Transport in* α-$RuCl_3$*: Strong Spin-Phonon Scattering and Field-Induced Spin Gap*, Phys. Rev. Lett. **120**, 117204 (2018).

[19] Y. Kasahara, K. Sugii, T. Ohnishi, M. Shimozawa, M. Yamashita, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi, and Y. Matsuda, *Unusual Thermal Hall Effect in a Kitaev Spin Liquid Candidate* α-$RuCl_3$, Phys. Rev. Lett. **120**, 217205 (2018).

[20] Y. J. Yu, Y. Xu, K. J. Ran, J. M. Ni, Y. Y. Huang, J. H. Wang, J. S. Wen, and S. Y. Li, *Ultralow-Temperature Thermal Conductivity of the Kitaev Honeycomb Magnet* α-$RuCl_3$ *across the Field-Induced Phase Transition*, Phys. Rev. Lett. **120**, 067202 (2018).

[21] R. Hentrich, M. Roslova, A. Isaeva, T. Doert, W. Brenig, B. Büchner, and C. Hess, *Large Thermal Hall Effect in* α-$RuCl_3$*: Evidence for Heat Transport by Kitaev-Heisenberg Paramagnons*, Phys. Rev. B **99**, 085136 (2019).

[22] M. Hirschberger, R. Chisnell, Y. S. Lee, and N. P. Ong, *Thermal Hall Effect of Spin Excitations in a Kagome Magnet*, Phys. Rev. Lett. **115**, 106603 (2015).

[23] C. Strohm, G. L. J. A. Rikken, and P. Wyder, *Phenomenological Evidence for the Phonon Hall Effect*, Phys. Rev. Lett. **95**, 155901 (2005).

[24] T. Ideue, T. Kurumaji, S. Ishiwata, and Y. Tokura, *Giant Thermal Hall Effect in Multiferroics*, Nat. Mater. **16**, 797 (2017).

[25] L. Chen, M.-E. Boulanger, Z.-C. Wang, F. Tafti, and L. Taillefer, *Large Phonon Thermal Hall Conductivity in a Simple Antiferromagnetic Insulator*, arXiv:2110.13277.

[26] P. Czajka, T. Gao, M. Hirschberger, P. Lampen-Kelley, A. Banerjee, N. Quirk, D. G. Mandrus, S. E. Nagler, and N. P. Ong, *The Planar Thermal Hall Conductivity in the Kitaev Magnet* α-$RuCl_3$, arXiv:2201.07873.

[27] M. Yamashita, J. Gouchi, Y. Uwatoko, N. Kurita, and H. Tanaka, *Sample Dependence of Half-Integer Quantized Thermal Hall Effect in the Kitaev Spin-Liquid Candidate* α-$RuCl_3$, Phys. Rev. B **102**, 220404(R) (2020).