Magnetic thermal conductivity far above the Néel temperatures in the Kitaev-magnet candidate $\alpha$-RuCl$_3$

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We have investigated the longitudinal thermal conductivity of $\alpha$-RuCl$_3$, the magnetic state of which is considered to be proximate to a Kitaev honeycomb model, along with the spin susceptibility and magnetic specific heat. We found that the temperature dependence of the thermal conductivity exhibits an additional peak around 100 K, which is well above the phonon peak temperature ($\sim 50$ K). The higher-temperature peak position is comparable to the temperature scale of the Kitaev couplings rather than the Néel temperatures below 15 K. The additional heat conduction was observed for all five samples used in this study, and was found to be rather immune to a structural phase transition of $\alpha$-RuCl$_3$, which suggests its different origin from phonons. Combined with experimental results of the magnetic specific heat, our transport measurements suggest strongly that the higher-temperature peak in the thermal conductivity is attributed to itinerant spin excitations associated with the Kitaev couplings of $\alpha$-RuCl$_3$. A kinetic approximation of the magnetic thermal conductivity yields a mean free path of $\sim 20$ nm at 100 K, which is well longer than the nearest Ru-Ru distance ($\sim 3$ Å), suggesting the long-distance coherent propagation of magnetic excitations driven by the Kitaev couplings.

Introduction

Quantum spin liquid is a phase of magnetic insulator in which frustration or quantum fluctuation prohibits magnetic order while keeping spin correlation$^{1,2}$. It induces rich physical phenomena depending on the types of spin liquids, which cannot be realized in standard ordered magnets. Several quantum-spin models have been proposed as possible realizations of quantum spin liquids along with candidate frustrated magnets such as $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ (Refs. 3,4), ZnCu$_3$(OH)$_6$Cl$_2$ (Ref. 5), Na$_3$Ir$_3$O$_8$ (Refs. 6,7) and $\alpha$-RuCl$_3$ (Ref. 8).

Among them, the Kitaev honeycomb model is unique in that the ground state is exactly calculated and known to be a two-dimensional (2D) quantum spin liquid$^{12,13}$. In this model, spin-1/2 moments, $S_x$, sit on a honeycomb lattice and interact via the bond-dependent Ising couplings; different spin components interact via Ising coupling for the three different bonds of the honeycomb lattice. These anisotropic couplings, called the Kitaev couplings $J_\alpha S_x^\alpha S_y^\beta$ ($\alpha = x, y, z$) (Ref. 9), yield frustration and induce a quantum spin liquid. As a result, the localized spins break down into two fractionalized excitations, called itinerant Majorana fermions and gauge fluxes$^{10,11}$. Several physical quantities of the Kitaev honeycomb model can be captured in terms of these fractionalized particles. For example, magnetic entropy release was shown to occur at two distinct temperatures using a Monte Carlo simulation$^{12}$; the higher-temperature release is attributed to the onset of Fermi degeneracy of itinerant Majorana fermions, and the lower-temperature one to thermal fluctuations of gauge fluxes.

Originally, the Kitaev honeycomb model was introduced as a toy model possessing nontrivial topological properties$^{15}$. However, the model was predicted to be realized as a low-energy spin system that describes magnetism of spin-orbit coupled Mott insulators$^{16}$. Moreover, a successive theoretical study showed that the spin-liquid ground state survives even when a small Heisenberg interaction is added to the Kitaev model$^{17}$. Following these theoretical results, actual magnetic compounds have been explored intensively$^{15–25}$ for the Kitaev quantum spin liquid along with further theoretical studies, which unraveled various physical properties of the Kitaev model and its variants$^{26–37}$.

One of the candidate materials is the Mott insulator $\alpha$-RuCl$_3$ (Refs. 9,10,12,22). In this material, Ru$^{3+}$ has an effective spin-1/2 owing to the strong spin-orbit coupling. Ru$^{3+}$ ions are located on an almost perfect honeycomb lattice by sharing the edges of the octahedral RuCl$_6$ in the $ab$ plane, as shown in Fig. 1a. The Kitaev couplings are predicted to arise from superexchange couplings in such a special network of Cl$^-$ ions. In fact, the low-temperature magnetic ordering below about 15 K is attributed to proximity to the Kitaev spin liquid$^{23,37}$. Even above the ordering temperature, properties proximate to the Kitaev spin liquid were reported for $\alpha$-RuCl$_3$ in recent experiments such as Raman and inelastic neutron scattering measurements$^{23–25}$. From these measurements, the strength of the Kitaev couplings was estimated to be about 100 K. In addition, measurement of the specific heat$^{23}$ suggested magnetic-entropy release at about 90 K, which appears consistent with the theoretical prediction for itinerant Majorana fermions in the Kitaev spin liquid$^{12,35}$.
Various experimental techniques have been applied to α-RuCl₃; however, its transport properties have not been well explored despite the fact that transport experiments are often used for detecting signatures of spin liquid states and their fractionalized excitations. In this paper, we experimentally study the thermal conductivity \( \kappa \) in α-RuCl₃. The temperature dependence of \( \kappa \) reveals an anomalous sub-peak that is distinguished from a main peak due to phonons. Importantly, the anomalous heat conduction is concomitant with the growth of the magnetic specific heat\(^{22} \) and is maximized at about 100 K; the same energy scale of the Kitaev couplings reported for α-RuCl₃. Our observations suggest that the sub-peak in \( \kappa \) versus \( T \) is due to the propagation of magnetic excitations driven by the Kitaev couplings.

**Experimental details**

Single crystals of α-RuCl₃ were grown by a vertical Bridgman method, which was exactly the same as reported by one of the present authors.\(^{21} \) The grown single crystals were cleaved along the \( ab \) plane, and cut into a cuboid that was typically 5mm long, 1mm wide, and 0.5mm thick with the largest surface in the \( ab \) plane. The magnetization of the grown single crystals was measured with a Magnetic Property Measurement System (Quantum Design, Inc.).

The thermal conductivity of α-RuCl₃ was measured by a standard steady-state method\(^{22} \). We used five samples for the thermal conductivity measurement, which were selected from two batches grown separately under the same condition: sample #1 was selected from one batch while samples #2-5 from the other. We used two types of glue to attach the samples to a copper block: silver-filled epoxy adhesive (H20E, EPO-TEK) for samples #1-3, which was dried by heating at 120 °C for an hour; GE varnish for samples #4 and #5, which was air-dried without being heated. In the following, samples #1-3 are collectively called the heated group while samples #4-5 the unheated group. As described below, we found that heating α-RuCl₃ samples modifies the magnetic susceptibility, probably attributable to crystal stacking faults, but does not affect the thermal conductivity. A temperature gradient was generated within the \( ab \) plane, or along the longitudinal direction of the single crystal by using a chip resistance heater (1 kOhm) attached to the end of the single crystal. The resulting temperature difference was measured using two cernox sensors (CX-1050-BG-HT, 0.75×1.0×0.25 mm\(^3 \)) attached to the top surface with 5×1 mm\(^2 \). The magnitude of temperature differences was set to less than five percent of the lower temperature of the cernox sensors. The measurement was performed in the temperature range between 2.5 K and 300 K under zero magnetic field in a high vacuum (<10⁻⁵ Torr) using a Physical Property Measurement System (Quantum Design, Inc.).

**Results and discussion**

We first discuss the effect of heating an α-RuCl₃ sample on interlayer stacking faults before the thermal conductivity measurement (see also the section of experimental details). It is known that crystal stacking faults form easily along the \( c \)-axis because of the weak interlayer coupling, and affect the structural and magnetic properties of α-RuCl₃ (Refs. \(^{20} \)\(^{22} \)). In Figs. 1(b) and 1(c), we show the temperature \( T \) dependence of the magnetic susceptibility \( \chi \) for a magnetic field perpendicular to the \( ab \) plane, investigated after the thermal conductivity measurement. We found that the behavior of \( \chi \) around a Néel temperature of \( \sim 8 \) K is different between heated and unheated groups [see insets to Figs. 1(b) and 1(c)]; for the heated group, \( \chi \) decreases smoothly on decreasing \( T \) across 8 K while it increases rather abruptly for the unheated group. Additionally, we found another change in a much higher-\( T \) region. For the unheated group [Fig. 1(c)], a hysteresis loop with a width of \( \sim 10 \) K is clearly resolved between 120 K and 165 K, which arises from
a structural phase transition via the strong spin-orbit coupling. In contrast, such a clear loop disappears for the heated group, replaced with the much smaller and broader loop [Fig. 1(b)]. Such sample dependence of the magnetic properties has been reported, and attributed to the different proportion of crystal stacking faults.

In the present study, the structural phase transition is blurred in the heated group, probably by a larger region of crystal stacking faults introduced under the heating procedure.

Having observed different hysteresis loops of the structural phase transition between the heated and unheated groups, we now turn to thermal conductivity results. Figure 2(a) shows the \( T \) dependence of \( \kappa \) of the heated group. \( \kappa \) increases monotonically with decreasing \( T \) from 300 K to 180 K. This is attributed to the growing mean free path of phonons; the rate of Umklapp scattering decreases with decreasing \( T \) in this temperature range. With decreasing \( T \) further, however, the \( T \) dependence of \( \kappa \) deviates from a known function of the phonon thermal conductivity, and \( \kappa \) begins increasing more rapidly from 180 K. As a result, a sub-peak structure forms around 110 K, which is reminiscent of the spin thermal conductivity in low-dimensional quantum spin systems.

\( \kappa \) then reaches a main peak at \( \sim 50 \) K and decreases strongly upon decreasing \( T \) further. The main peak is simply due to phononic heat conduction, the position of which \( \sim 50 \) K results from competition between the increasing mean free path and the decreasing number of phonons with decreasing \( T \) (Refs. 46,50).

Crucially, the thermal conductivity \( \kappa \) of the unheated group, shown in Fig. 2(b), exhibits a sub-peak in the same position as that for the heated group despite the clear hysteresis loop in the magnetic susceptibility data [see also Fig. 1(b)]. The result indicates a low correlation of \( \kappa \) with the structural phase transition. This is also supported by comparing \( \kappa \) measured while increasing and decreasing \( T \). The inset to Fig. 2(a) compares \( \kappa \) around 150 K measured in such scans of \( T \). A hysteresis loop is not observed over the sub-peak of \( \kappa \) within the error range of 0.2 W K\(^{-1}\) m\(^{-1}\) for sample #1 and 0.1 W K\(^{-1}\) m\(^{-1}\) for samples #2-5. The observation shows that the sub-peak is insensitive to the structural phase transition, which is consistent with the same \( T \) dependence of the sub-peak shared by the heated and unheated groups. This means that a possible change in the phonon thermal conductivity caused by the structural phase transition is too small to be observed. Accordingly, the phonon thermal conductivity may be approximated by the conventional one that exhibits a single peak as a function of \( T \) in a practical analysis; a different origin from phonons is needed to explain the sub-peak structure. We note that a sharp peak of the phonon thermal conductivity begins growing from 8 K [see also the inset to Fig. 2(b)], which is comparable with a Néel temperature shown in Fig. 1. This increase in the phonon thermal conductivity may originate from the suppression of phonon scattering off paramagnetic fluctuations.

We extract the sub-peak component of \( \kappa \) by assuming the 2D Debye model for the phonon component. Using an elementary kinetic theory, the phonon thermal conductivity is given by \( \kappa_{\text{ph}} = C_{\text{ph}} v_{\text{ph}} l_{\text{ph}}/2 \), where \( C_{\text{ph}} \) is the lattice specific heat, \( v_{\text{ph}} \) is the velocity, and \( l_{\text{ph}} \) is the mean free path of the phonons.

![FIG. 2: (a), (b) Temperature \( T \) dependence of the thermal conductivity \( \kappa \) for samples #1-3 (a) and #4-5 (b). The inset to (a) shows a magnified view of a temperature region for sample #1 in which the structural phase transition occurs. Red and sky-blue data points were taken while increasing and decreasing \( T \), respectively. The inset to (b) shows a magnified view of a low-temperature region for samples #1-5.](image-url)
maximum reported for the $T$ dependence of the magnetic specific heat $C_{\text{mag}}$ of an $\alpha$-RuCl$_3$ sample grown by the same method$^{21}$, as shown in Fig. 3(c). Since the broad maximum of $C_{\text{mag}}$ without magnetic phase transitions is qualitatively consistent with a theoretical calculation based on itinerant Majorana fermions$^{12,36}$, the agreement between $\Delta \kappa$ and $C_{\text{mag}}$ suggests that the itinerant quasiparticles carry heat. It should be noted that the agreement is confirmed for both heated and unheated groups along with the similar magnitude of $\Delta \kappa$, despite the different $T$ dependence of $\chi$ between these two groups. This suggests that the itinerant quasiparticles are rather immune to crystal stacking faults, and to the resulting variation in interlayer magnetic couplings relevant to three-dimensional Néel orders; the dynamics of the quasiparticles are expected to be governed by 2D magnetic couplings that are much stronger than the interlayer ones. We also emphasize that conventional spin waves in magnetically ordered states are irrelevant to the peak around $T_p = 110 \text{ K}$ as $T_p$ is much higher than the ordering temperatures (< 15 K) of $\alpha$-RuCl$_3$. Instead, $T_p$ corresponds to the strength of the 2D Kitaev couplings reported for $\alpha$-RuCl$_3$ (∼ 100 K). In this high-$T$ range, a continuum of Raman scattering$^{21}$, persistent short-range spin correlation$^{23,25}$, as well as fermionic response$^{25}$ were reported and attributed to 2D Kitaev coupling.

We are in a position to characterize $\Delta \kappa$ presumably related with the Kitaev couplings of $\alpha$-RuCl$_3$. Below, we assume that a paramagnetic state of $\alpha$-RuCl$_3$ is proximate to the Kitaev spin liquid and that the broad peak in $\Delta \kappa$ originates from itinerant Majorana fermions. The assumption allows us to use the 2D elementary kinetic theory for the magnetic thermal conductivity $\kappa_{\text{mag}}$, given by $\kappa_{\text{mag}} = C_{\text{mag}} v_{\text{mag}} l_{\text{mag}} / 2$ to analyze $\Delta \kappa$. Here, $v_{\text{mag}}$ and $l_{\text{mag}}$ are, respectively, the velocity and the mean free path of the magnetic excitations. We note that the quasi-particle picture is supported also by a Fermi-liquid behavior identified in the numerical calculation in Ref. [12]. Microscopic models have not been established yet to quantitatively capture the magnetic properties of $\alpha$-RuCl$_3$; to estimate $l_{\text{mag}}$, therefore, we simply assume that $\kappa_{\text{mag}}$ is driven by the Kitaev couplings $J^\alpha \sim 100 \text{ K}$ ($\alpha = x, y, z$) alone and that $v_{\text{mag}}$ is given by averaging the Majorana-fermion group velocity of the Kitaev honeycomb model$^{10,33,53}$ in a certain region of the Brillouin zone$^{22}$. The velocity $v_{\text{mag}}$ is then found to be 1,620 m s$^{-1}$ by setting the Kitaev couplings to be 100 K. By putting $\Delta \kappa = \kappa_{\text{mag}}$ and using the experimental data of $C_{\text{mag}}$ and $\kappa_{\text{mag}}$, $l_{\text{mag}}$ was found to be 10 ~ 20 nm between 60 K and 110 K, into which the data for all the samples fit as shown in the inset to Fig. 3(c). The result implies that the excitations can carry entropy over a distance up to 60 times as long as the nearest Ru-Ru distance (∼ 3 Å). Although our estimation is reliable only in a qualitative level, it suggests that the anomalous heat conduction is due to the coherent propagation of itinerant spin excitations around $T_p$, which has not been revealed by measurements of the magnetic susceptibility nor the specific heat performed so far. Further evidence for the magnetic origin may be derived from investigating the purely phononic heat conduction in a nonmagnetic analogue of $\alpha$-RuCl$_3$, for example ScCl$_3$, that has a similar honeycomb lattice$^{54}$.

**Conclusion**

In this study, we have investigated the temperature dependence of the thermal conductivity of the Mott insulator $\alpha$-RuCl$_3$, the magnetism of which is related with a Kitaev honeycomb model. The thermal conductivity was observed to exhibit a sub-peak structure around 110 K that is insensitive to interlayer crystal stacking faults. Compared with the temperature dependence of the magnetic specific heat, the broad peak in the thermal conductivity was attributed to itinerant spin excitations. Applying a kinetic approximation to the magnetic thermal conductivity yielded a long mean free path that was up to 60 times longer than the nearest inter-spin distance between 60 K and 110 K. This result suggests the coherent propagation of itinerant spin excitations due to the Kitaev couplings, possibly itinerant Majorana fermions.

**Note added.**

After submitting our original manuscript, we became aware of a similar study$^{55}$, which was published after our work was submitted.
aware of experimental work by Ian A. Leahy et al. and Richard Hentrich et al., who both focused on the high-field dependence of the thermal conductivity of $\alpha$-RuCl$_3$ at low temperatures. Their results obtained at zero field are well consistent with ours, especially a kink around 8 K. In addition, Seung-Hwan Do et al. recently reported the temperature dependence of the magnetic specific heat of $\alpha$-RuCl$_3$, and observed a broad peak structure at about 100 K, consistent with our result. We also became aware of theoretical work by Joji Nasu et al. Their computation with the use of the Kitaev model showed that itinerant Majorana fermions contribute to the longitudinal thermal conductivity and the magnetic specific heat within the same temperature range, which qualitatively reproduces our result.

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\[
\left( \Delta k \right)^{-1} \int_{\Delta k} k \, dk \frac{\partial E}{\partial k} = \frac{1}{2} \frac{3}{\pi} \frac{1}{\hbar}. 
\]
Here, \( \Delta k \) denotes the distance between the \( \Gamma \) point and a \( K \) point in wavenumber space.
55 For comparison, we note that the mean free path of one-dimensional spinons in \( \text{Sr}_2\text{CuO}_3 \) reaches 50 to 100 nm at 100 K, depending on the purity of the primary chemicals.
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