Unusual Phonon Heat Transport in α-$RuCl_3$: Strong Spin-Phonon Scattering and Field-Induced Spin Gap

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The honeycomb Kitaev-Heisenberg model is a source of a quantum spin liquid with Majorana fermions and gauge flux excitations as fractional quasiparticles. Here we unveil the highly unusual low-temperature heat conductivity κ of α-$RuCl_3$, a prime candidate for realizing such physics: beyond a magnetic field of $B_c \approx 7.5$ T, κ increases by about one order of magnitude, both for in-plane as well as out-of-plane transport. This clarifies the unusual magnetic field dependence unambiguously to be the result of severe scattering of phonons off putative Kitaev-Heisenberg excitations in combination with a drastic field-induced change of the magnetic excitation spectrum. In particular, an unexpected, large energy gap arises, which increases linearly with the magnetic field, reaching remarkable $\hbar\omega_0/k_B \approx 50$ K at 18 T.

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Topological quantum spin liquids (QSLs) are characterized by the massive quantum entanglement of states and constitute peculiar states of matter where quantum fluctuations are so strong that even in the ground state, magnetic long-range ordering is suppressed. Amazingly, despite the inherent quantum disorder, QSLs are conjectured to possess well-defined quasiparticles. These are highly nontrivial, because unlike classical systems, the QSLs' quasiparticles arise from the fractionalization in a ground state with topological degeneracy and may have anyonic statistics [1–3]. Since QSL ground states are experimentally elusive, the detection and rationalization of these quasiparticles appear as the natural path towards identifying a QSL system.

Heat conductivity experiments constitute one of the few probes to study such quasiparticle physics, because they provide information on the quasiparticles' specific heat, their velocity, and their scattering [4,5]. In fact, such experiments have been very revealing in clarifying the unconventional ballistic heat-transport characteristics of spinon excitations, the fractional excitations of the spin-1/2 chain [6,7], and signatures of unconventional spin heat transport in QSL candidate materials which realize spin-1/2 triangular lattices [8,9] or spin-ice systems [10,11].

Experimental realizations of QSLs generally are rare. In the quest of finding a pertinent material to experimentally investigate their physics, α-$RuCl_3$ recently emerged as a prime candidate for hosting a proximate Kitaev QSL with Majorana fermions and gauge flux excitations as new kinds of fractional quasiparticles [12–16].

In this material, strong spin-orbit coupling and an edge-sharing configuration of $RuCl_6$ octahedra yield a honeycomb lattice of $j_{\mathrm{eff}} = 1/2$ states with dominant Kitaev interaction [15,17–19]. Long-range magnetic order at $T_N \approx 7$ K occurs in as-grown samples of α-$RuCl_3$ without stacking faults [15,19–22]. Remarkably, a moderate in-plane magnetic field of $\sim$8 T, which is far away from full polarization [23], is sufficient to completely suppress the long-range magnetic order [21,23–26].

In order to probe the emergence of unusual quasiparticles in this putative Kitaev QSL, we have measured the thermal conductivity κ of α-$RuCl_3$ single crystals in magnetic fields up to 18 T. Overall, four samples (labeled I to IV) from different crystal growth laboratories have been scrutinized. All samples were of high crystalline quality, evidenced by the onset of magnetic long-range order in the range $T_N = 7.0$ K (sample II) to $T_N = 7.4$ K (sample I); see the Supplemental Material for details [27]. For fields applied parallel to the

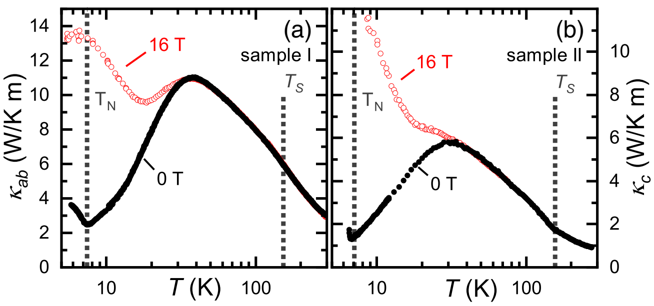

FIG. 1. T dependence of the heat conductivity of α-$RuCl_3$ at zero magnetic field and at $B = 16$ T. The heat current was aligned (a) parallel to the ab direction for sample I (κ_{ab}) and (b) perpendicular to it for sample II (κ_c). In both cases, the field was applied parallel to the ab planes and perpendicular to the heat current. The onset of long-range magnetic order at $T_N \approx 7$ K and the structural transition at $T_s \approx 155$ K are indicated.

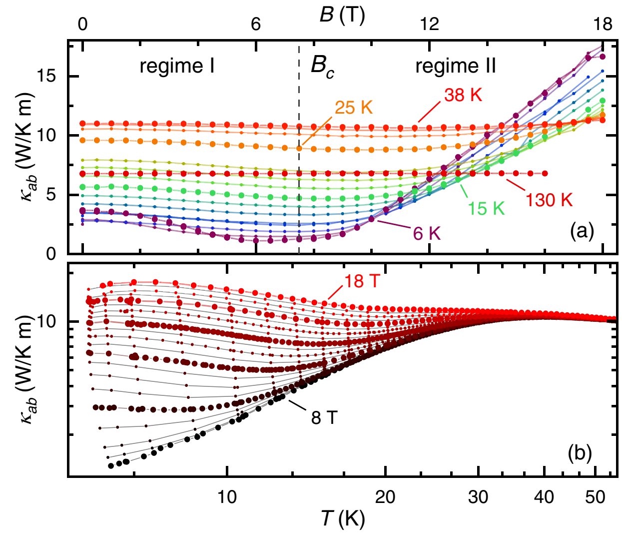

FIG. 2. B and T dependence of the heat conductivity of α-$RuCl_3$ (sample I) with the heat current and the magnetic field B parallel to the ab planes. (a) Isothermal field-dependent heat conductivity $\kappa_{ab}(B)$ for selected T. (b) $\kappa_{ab}(T)$ for $B > 7.5$ T, measured in steps of 0.5 T.

honeycomb planes, we observe a strong impact of the magnetic field on the T dependence of κ, which upon exceeding $B_c \approx 7.5$ T, i.e., in the absence of magnetic order, exhibits a qualitatively new behavior: a low-T peak arises which grows with magnetic field. The analysis of our data unambiguously reveals a primarily phononic origin of the heat transport in α-$RuCl_3$, the strong field-induced enhancement of which implies a radical change in the low-energy spectrum of magnetic excitations. In particular, for $B > B_c$, an energy gap opens which increases approximately linearly with the magnetic field, in agreement with recent NMR results [25].

The left panel of Fig. 1 shows representative data of the in-plane thermal conductivity κ_{ab} of α-$RuCl_3$ as a function of T in zero magnetic field (see the Supplemental Material for κ_{ab}'s of other single crystals with essentially the same T dependence [27]) and at $B = 16$ T, applied parallel to the ab planes. In zero magnetic field, upon cooling from 300 K down to the base temperature (5.5 K) of our setup, κ_{ab} increases steadily up to a distinct maximum at around 40 K, and decreases steeply at lower T. A kink around 7.5 K coincides roughly with the onset of long-range magnetic order [22].

At first glance, these zero-magnetic-field data of $\kappa_{ab}(T)$ at $T > T_N$ with a single peak structure resemble that of a conventional phononic heat conductor [28]: Here, the phononic heat conductivity, which can coarsely be estimated as $\kappa_{\mathrm{ph}} \sim c_V v l$, increases strongly at low T, where the phononic velocity v and mean free path l are essentially T independent, with the phononic specific heat c_V. Towards higher T, phonon umklapp processes increasingly limit l, resulting in a broad peak in κ_{ph} followed by a fast decline. For antiferromagnetic insulators it is well known that the scattering of phonons off paramagnon fluctuations of the incipient long-range order may give rise to a significant suppression of κ_{ph} above and a recovery below the Néel ordering temperature, respectively [29–33]. The whole $\kappa_{ab}(T)$, including the observed kink at T_N, seems perfectly in line with such a scenario.

Strikingly, the application of a large in-plane magnetic field of $B = 16$ T, at which magnetic order is absent, dramatically changes κ_{ab}, and thereby challenges such a rather conventional interpretation: κ_{ab} is drastically enhanced at low T where a second, large peak emerges at around 7 K which even exceeds the one at higher T.

The unconventional nature of the field-induced double-peak structure is further confirmed by a detailed mapping of $\kappa_{ab}(T, B)$ up to $B = 18$ T, as is shown in Fig. 2. Apparently, the impact of the magnetic field on the heat transport is profoundly different for fields smaller and fields larger than a critical field $B_c \approx 7.5$ T, clearly defining two field regimes (hereinafter labeled I and II, respectively). In regime I, as is evident from panel (a) of Fig. 2, κ_{ab} slightly decreases for all $T < 40$ K upon increasing the field from zero to B_c. This suppression is most pronounced at $T = 6$ K, where it reflects the suppression of long-range magnetic order. A dramatically different field dependence occurs upon further increasing the field (regime II), where κ_{ab} strongly increases with increasing field. Remarkably, for $T \lesssim 15$ K, this increase is essentially linear in magnetic field up to 18 T.

Without any doubt, the most prominent feature of the present data is the large field-induced low-T peak in κ_{ab} in regime II, which grows with the field [Fig. 2(b)]. Note that the critical field B_c, which marks the onset of this regime, coincides with the complete field-induced suppression of long-range magnetic order which governs the lowest-temperature physics in regime I but is absent in regime II [21,23–25]. Thus, the low-T peak in regime II must be of a qualitatively different origin than the low-T upturn in regime I below T_N, which is closely related to spin fluctuations in the system.

A priori, two very different scenarios can be invoked for explaining the nature of a double-peak structure in $\kappa(T)$ of an electrical insulator which hosts a fluctuating spin system. On the one hand, this could be the signature of magnetic heat transport that, in turn, leads to a pertinent contribution to the (otherwise conventional phononic) heat conductivity. Such a mechanism is common in low-dimensional systems such as spin chains, ladders, and planes [6,7,34–37]. In these cases, phonons and magnetic excitations yield two independent transport channels. On the other hand, a double-peak structure is also known to occur in purely phononic heat transport, resulting from the heat-carrying phonons scattering off another degree of freedom, such as a spin excitation with a well-defined excitation energy $\hbar\omega_0$ [38,39]. Such scattering affects the phononic heat transport over a large temperature range, but it has its strongest impact in the temperature regime where the energy of the majority of heat carrying phonons coincides with $\hbar\omega_0$.

An unambiguous signature of low-dimensional magnetic heat transport is its anisotropy: spin heat is transported only along crystal directions along which a significant energy dispersion of the spin excitations exists; i.e., the magnetic heat transport is absent along directions without a significant magnetic exchange interaction [6,7,34–37]. We therefore investigated the heat conductivity of α-$RuCl_3$ perpendicular to the planes (κ_c), where the magnetic exchange interaction is negligible [sample II, see Fig. 1(b)]. Remarkably, apart from minor differences in details (see Fig. S2 in the Supplemental Material [27]), we observe practically the same T and B dependence as for κ_{ab}. The most important finding is the direct comparability of κ_c and κ_{ab} at $B = 0$ and 16 T, with κ_c exhibiting the same low-T enhancement, i.e., the presence of a high-B, low-T peak. Hence, we can exclude the possibility that transport by the emergent elementary excitations of the spin system gives rise to the low-T peak in regime II. This means unambiguously that the field-induced low-T peak of κ_{ab} is primarily phononic, and its peculiar T and B dependence arises from an unusual field-dependent scattering process of the phonons.

After having established this essential finding, we move on to rationalizing the B and T dependence of the heat conductivity more thoroughly. Without further analysis and by invoking the magnetic scattering scenario of phonons, the presence of the double-peak structure in regime II with a clear minimum at $T_{\min}$ implies that an energetically well-defined magnetic mode with energy $\hbar\omega_0$ exists which scatters primarily the heat-carrying phonons of that energy. This can be understood from the fact that the energy of the phonons which predominantly carry heat, $\hbar\tilde{\omega}$, is strongly T dependent [28]: The two peaks at lower and higher T correspond to $\tilde{\omega} < \omega_0$ and $\tilde{\omega} > \omega_0$, whereas $\tilde{\omega} \approx \omega_0$ at

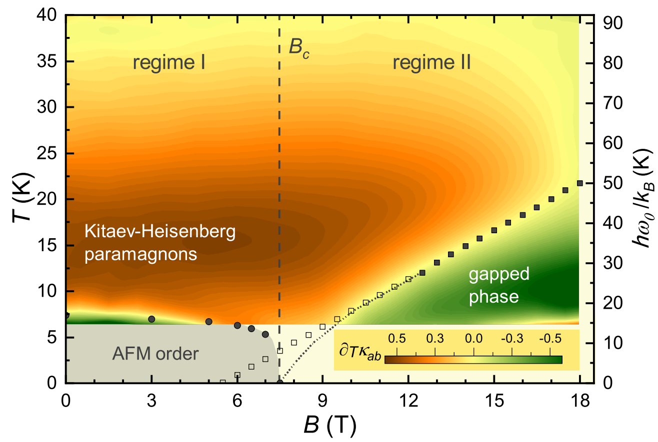

FIG. 3. False-color representation of the T derivative $\partial\kappa_{ab}(T, B)/\partial T$ (sample I) together with the gap energy $\hbar\omega_0/k_B$ (solid squares) extracted from the phononic fits. The color scale is in units of W/K^2m. The left ordinate shows the temperature T of the measurement, while the right ordinate shows $\hbar\omega_0/k_B$. Note that for $B \leq 12$ T, an unambiguous extraction of ω_0 cannot be obtained from fitting $\kappa_{ab}(T)$, because the $T_{\min}$ is too close to the lower limit of the measurement range. Nevertheless, good fits to the data are obtained if $\omega_0(B)$ for $B > 12$ T is extrapolated towards smaller fields (open squares) and subsequently used to fit $\kappa_{ab}(T)$ at the corresponding fields (see the Supplemental Material [27]). These extrapolated ω_0's should be regarded as an upper limit only. The data can be described similarly well with a somewhat smaller $\omega_0(B)$ (dotted line).

$T_{\min}$. This immediately suggests that a rough quantitative estimate of the scattering magnetic mode energy $\hbar\omega_0$ can be obtained by reading $T_{\min}$ off the data. We therefore plot the T derivative $\partial\kappa_{ab}/\partial T$ in false-color representation in Fig. 3. At $B \gtrsim 11$ T, the minimum position $T_{\min}$ depends about linearly upon B. At smaller fields, however, $T_{\min}(B)$ attains a steeper slope and rapidly moves out of the measured temperature window, suggestive of an approximate extrapolation towards B_c at $T = 0$ (dotted line in Fig. 3). Thus, while the dominant magnetic scattering mode energy $\hbar\omega_0$ seems to be very close to zero for $B \lesssim B_c$, it rapidly develops a substantial size at higher magnetic fields.

One can exploit $T_{\min}(B)$ further and estimate the field dependence of the magnetic mode energy $\hbar\omega_0(B)$ quantitatively by considering that for a conventional isotropic phononic system, the majority of heat-carrying phonons possess an energy of about $\alpha k_B T$ with $\alpha \approx 3.8$ [28]. For more anisotropic phononic systems, as one might expect for α-$RuCl_3$, simple dimensional considerations (see the Supplemental Material [27]) suggest a reduced α (in particular, $\alpha \approx 2.6$ for a hypothetical purely two-dimensional phononic system). By translating $T_{\min}(B)$ into the phonon energy which is affected most strongly by the magnetic scattering, one can directly extract the field dependence of the magnetic mode energy as $\hbar\omega_0(B) \approx \alpha T_{\min}(B)$. Thus, for $B > 10$ T, the magnetic excitation energy scales linearly with the applied magnetic field, with the factor $\alpha = 2.3$ indeed indicating a deviation from the case of 3D isotropic phonons [40]. In fact, the low-T specific heat of α-$RuCl_3$ has been reported to follow T^2 rather than T^3 [22], which indicates a significant anisotropy of the phonons. Still, the anisotropy of the heat transport is only moderate (see Fig. 1) as compared to prototype quasi-two-dimensional phononic systems such as graphite [41], implying significant interlayer lattice coupling, and consequently only a moderate anisotropy for the phononic system, in contrast to the two-dimensional nature of the magnetic system.

A further corroboration of the scenario of magnetic phonon scattering can be obtained by analyzing $\kappa(T, B)$ in terms of a phononic model which takes the magnetic scattering into account. We follow the approach of Callaway [42,43] and express κ in terms of an energy-dependent relaxation time τ_c which takes various scattering processes into account (see the Supplemental Material [27]). For conventional phononic systems, good descriptions of the T dependence of κ can be achieved if the standard expressions for the phonon relaxation times describing umklapp, point defect, and boundary scattering are comprised in τ_c.

As one can already conjecture in view of the unconventional double-peak structure of $\kappa_{ab}(T, B)$ at $B > B_c$, a standard Callaway-type fit to our data fails. However, a qualitatively reasonable fit is indeed possible if we adapt the model to the previously sketched magnetic scattering scenario by introducing an additional relaxation time τ_{mag}. More specifically, we assume the phonons to scatter within an energetically broad magnetic excitation spectrum (mimicking the theoretically predicted [14,44] and experimentally observed [45] character of the excitation spectrum of the Kitaev model and α-$RuCl_3$, respectively) from a reservoir which is dominated by an energetically sharp and magnetic-field-dependent low-energy mode at $\hbar\omega_0$ (see the Supplemental Material [27]).

Despite the simplicity of this model, it is possible to simultaneously fit the data in regime II with a field-independent parameter set for the usual phonon scattering terms and a field-dependent magnetic scattering term $\tau_{\mathrm{mag}}(B)$; see Fig. 4. The values for $\omega_0(B)$ extracted thereby are plotted in Fig. 3 as solid squares. Obviously, they display the same linear field dependence as the position of $T_{\mathrm{min}}(B)$, and are in the same energy range as expected from the above considerations with respect to T_{min}. We emphasize that a qualitatively similar result is reached upon analyzing κ_c with the same procedure, which further corroborates this analysis (see the Supplemental Material [27]).

At magnetic fields smaller than B_c, the heat conductivity of α-$RuCl_3$ is always significantly smaller than in the high-field phase. This straightforwardly implies that in regime I the phonon scattering off the magnetic excitation spectrum is even stronger than in regime II (see Fig. 1). In view of the vanishing ω_0 at around B_c, this suggests that in regime I the scattering magnetic modes are at relatively low energy if not gapless. Evidently, the field-induced changes of κ_{ab} in this phase are relatively small (see Fig. 2). This small field dependence then naturally is consistent with more subtle field-induced changes of the magnetic spectrum as in regime II, connected to the gradual suppression of long-range magnetic order. These are more subtle than those apparently induced by high magnetic fields.

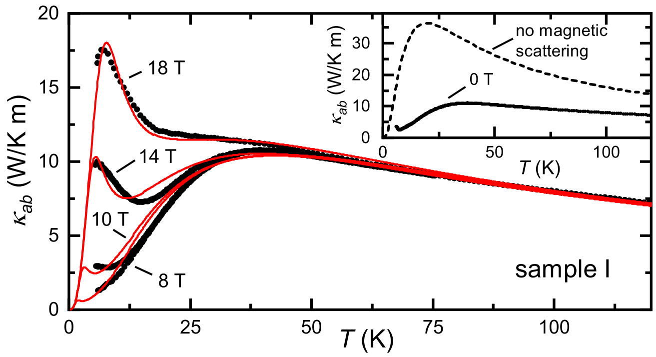

FIG. 4. κ_{ab} data of sample I (solid circles) and fits (solid lines) to the Callaway model for selected magnetic fields. The fits have been obtained by incorporating magnetic scattering of the phonons (see the Supplemental Material [27]). Inset: κ_{ab} data of sample I (solid circles) at zero field as compared to the hypothetical phononic heat conductivity described by the model if the magnetic scattering mechanism is switched off (dashed line). This highlights that the magnetic scattering affects the phonon heat conductivity in a very large temperature range, where the field-induced changes occur essentially at $T < 50$ K.

In conclusion, our investigations clearly show that the heat transport of α-$RuCl_3$ is primarily of phononic type. More specifically, the field-induced low-T peak in the heat conductivity cannot be explained by the expected exotic excitations of a putative Kitaev-Heisenberg QSL carrying heat. This is in contrast to the results of a recent study [46] where the field-induced low-T peak is interpreted as the signature of in-plane heat transport by massless so-called *proximate Kitaev spin excitations*. Nevertheless, the magnetic excitations of α-$RuCl_3$ dramatically impact the phononic heat transport along all directions through scattering of the phonons. This scattering is particularly strong in regime I, which at first glance seems to be consistent with the incipient long-range magnetic order. However, the magnitude of the low-T increase of κ in the ordered phase is relatively small as compared to the dramatic enhancement in regime II. This suggests that even in the magnetically ordered phase, considerable magnetic degrees of freedom exist, which is in line with the significantly reduced ordered magnetic moment observed in inelastic neutron scattering [15]. These residual degrees of freedom scatter the phonons and are likely to remain disordered down to zero energy due to quantum fluctuations. Since the phonon heat conductivity at low T primarily is carried by acoustic phonons with small momenta $k \sim 0$, it seems natural to conclude that the low-energy paramagnons relevant for the scattering possess small momenta as well [47]. On the other hand, the dramatic enhancement of the heat conductivity at higher fields in regime II ($B > B_c$) implies that these low-energy excitations are increasingly gapped out; i.e., the strongest field-induced change of the excitation spectrum concerns the excitations close to the Γ point. One might thus speculate that the field-induced phase at $B > B_c$ is governed by new physics, where the emergent quasiparticles are indeed different from those of the Kitaev-Heisenberg paramagnons at $B < B_c$.

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