

Half-integer quantized anomalous thermal Hall effect in the Kitaev material α -RuCl₃

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Abstract

Heat transport mediated by Majorana edge modes in a magnetic insulator leads to a half-integer thermal quantum Hall conductance, which has recently been reported for the two-dimensional honeycomb material α -RuCl₃. While the conventional electronic Hall effect requires a perpendicular magnetic field, we find that this is not the case in α -RuCl₃. Strikingly, the thermal Hall plateau appears even for a magnetic field with no out-of-plane components. The field-angular variation of the quantized thermal Hall conductance has the same sign structure of the topological Chern number, which is either ± 1 , as the Majorana band structure of the pure Kitaev spin liquid. This observation of a half-integer anomalous thermal Hall effect firmly establishes that the Kitaev interaction is primarily responsible and that the non-Abelian topological order associated with fractionalization of the local magnetic moments persists even in the presence of non-Kitaev interactions in α -RuCl₃.

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The discovery of the integer quantum Hall effect (QHE) in a two-dimensional (2D) electron gas has opened a window for exploring topological phases of quantum matter. When topology is combined with strong correlations, even more interesting phenomena can arise, such as the emergence of collective excitations having fractional quantum numbers with respect to the elementary particles. One of the most prominent examples is the fractional QHE, in which the constituent particles are electrons but the quasiparticles carry fractions of the electron charge. Another venue for the interplay of topology and correlations are quantum spin liquids in magnetic insulators [1], where fluctuations prevent the formation of magnetic order down to $T=0$ and instead lead to the emergence of macroscopic entanglement. A paradigmatic example is the Kitaev spin liquid [2], which forms for strongly interacting spins in a magnetic field and exhibits anyonic quasiparticles in the form of massive Z_2 vortices that bind a Majorana zero mode. The non-Abelian character of these Ising anyons in the Kitaev spin liquid is identical to that of certain topological superconductors [3] and the $\nu = 5/2$ fractional quantum Hall state [4].

Direct experimental evidence for the formation of a topological state with non-Abelian anyons has long been sought-after. In fact, it required the observation of yet another QHE – a quantized *thermal* Hall effect, reported last year in experiments for both the $\nu = 5/2$ fractional quantum Hall state [5] and the spin-orbit entangled Mott insulator α - RuCl_3 [6]. In both experiments the transverse thermal Hall conductivity (per 2D sheet) $\kappa_{xy}^{2\text{D}}$ exhibits a quantized plateau at half integer values,

$$\kappa_{xy}^{2\text{D}} = \frac{1}{2} C_h \cdot K_0, \quad (1)$$

where $K_0 = (\pi^2/3)(k_B^2/h)T$ is the quantum thermal conductance, in analogy to the quantum electronic conductance $2e^2/h$ for the electronic QHE, and C_h is an integer related to the number of chiral edge modes ($C_h = 1$ for α - RuCl_3 and $C_h = 5$ for the $\nu = 5/2$ state). The half-integer quantization is, in both cases, direct evidence of charge neutral Majorana fermions carrying heat along the sample edges, and as such direct evidence for the non-Abelian topological nature of the bulk state. Here, we show that the fundamental nature of the QHE in the two systems, however, must be clearly distinguished. For the two-dimensional electron gas, the QHE arises from the electrons experiencing a Lorentz force and the formation of *Landau levels* in a perpendicular magnetic field and the subsequent splitting of the degenerate states of partially filled Landau levels through Coulomb interactions. In this case, C_h is related to the number of upstream/downstream edge modes and its sign is determined purely by the magnetic field perpendicular to the sample. In the insulator α - RuCl_3 , we show that a different physical mechanism must be at play, consistent with a

Kitaev spin liquid (KSL) that forms in the presence of a magnetic field. In this scenario the local magnetic moments interact via bond-directional Ising-type interactions, which at low temperatures leads to a fractionalization into itinerant Majorana fermions and static Z_2 vortices located on the hexagons of the honeycomb lattice. In the absence of a magnetic field, the Majorana fermions form a semi-metal with a Dirac cone dispersion. Applying a magnetic field, the Majorana spectrum gaps out into a *Chern insulator*, i.e. a band insulator whose bands are endowed with a non-trivial Berry curvature, with the resulting Chern number ± 1 for each band. The non-trivial topology of this time-reversal symmetry breaking insulator also gives rise to chiral Majorana modes at the edges, with the Chern number identified with C_h above, and thus a half-integer quantized thermal QHE with the sign structure of κ_{xy}^{2D} reflecting the non-trivial bulk topology.

Here, we report the observation of an *anomalous* thermal QHE in α -RuCl₃ that gives rise to a half-integer quantized thermal Hall conductance plateau even in the *absence* of an out-of-plane magnetic field, thereby confirming that the thermal QHE in α -RuCl₃ does indeed arise from a topologically non-trivial Chern insulator of Majorana fermions. Our experiments build on recent work identifying the spin-orbit coupled Mott insulator α -RuCl₃ [7] as a Kitaev material [8–10], in which local $j_{\text{eff}} = 1/2$ pseudospins are almost coplanar within the honeycomb lattice [11], see Fig. 1A. Significant bond-directional Kitaev interactions $J_K/k_B \approx 80$ K have been reported [12, 13], which dominate over all other non-Kitaev interactions (such as Heisenberg and off-diagonal exchange). The latter induce antiferromagnetic (AFM) ordering with a zigzag magnetic structure at $T_N \approx 7.5$ K [14]. A double peak structure in the temperature dependence of the specific heat [15, 16], fermionic excitations in Raman scattering spectra [17, 18], and a broad energy continuum in inelastic neutron scattering [15, 19–22] above T_N have been interpreted as signatures of fractionalization.

The AFM order in α -RuCl₃ is strongly suppressed by applying an in-plane magnetic field \mathbf{H} parallel to the 2D honeycomb planes, leading to a field-induced paramagnetic state (Fig. 1B). To probe the emergence of fractional quasiparticles in this regime, thermal transport measurements are a powerful technique [23], as the emergent charge neutral Majorana fermions do not respond to electric fields but can carry heat via a subtle coupling to the phonons [24, 25]. Such non-trivial magnetic excitations can be detected, in particular, via thermal Hall conductivity κ_{xy} measurements [26, 27] transverse to a heat current \mathbf{q} . Very recently, a thermal Hall plateau quantized at a half-integer value of κ_{xy} has been observed [6] for a field tilted away from the crystallographic c axis, proving the spin liquid nature of the field-induced phase. The observation of the half-

integer thermal QHE immediately raises several fundamental questions about the nature of the spin liquid state. In fact, the thermal QHE is observed at *intermediate* magnetic field strengths and in proximity to a magnetically ordered state – an experimental scenario that cannot be captured by the pure Kitaev model, in which the KSL originates in the small-field limit from a gapless spin liquid. Moreover, the influence of non-Kitaev interactions, which are inevitably present in α -RuCl₃ [12, 13], on the KSL has been an issue under intense debate. To demonstrate the unaltered nature of the observed spin liquid state at intermediate field strengths from the theoretically well understood KSL, we here determine the field-angular variation of the thermal conductance plateau, which provides stringent experimental evidence for a Chern insulator that closely mimics the angular dependence of the KSL.

We measured κ_{xy} of five single crystals from the same batch with $\mathbf{q} \parallel \mathbf{a}$ in \mathbf{H} applied to a tilted direction in the ac plane with $\theta = -60^\circ$, where θ is the polar angle measured from the c axis (Figs. S1 A-C). The quantized thermal Hall plateau at one half of K_0 was observed in three crystals (#1–#3), while it was absent in one crystal (#4) with low longitudinal thermal conductivity κ_{xx} and in one crystal (#5), which shows a partial magnetic order at 14 K due to stacking faults. These results indicate that high-quality single crystals are necessary for the half-integer thermal QHE. The magnetic field strengths at which the quantization occurs is slightly sample dependent. Here crystal #3 is used for the measurements. Figure 1B shows the phase diagram of α -RuCl₃ in \mathbf{H} applied parallel to the a axis (zigzag direction of a honeycomb lattice). A critical magnetic field H_{AF} , at which AFM order disappears, is determined by the minimum of $\kappa_{xx}(H)$ (Fig. S2). The determined H_{AF} is close to those determined by other methods [22] but is smaller than that reported by magnetic torque measurements [28].

Varying the field direction, we depict the H -dependence of κ_{xy} at 4.8 K ($\mu_0 H_{AF} \approx 6.5$ T) for \mathbf{H} applied antiparallel to the a axis by the red circles in Figs. 2A and 2B. What is remarkable is that, even for an in-plane magnetic field with *no* out-of-plane component, a finite κ_{xy} with positive sign is observed in the spin liquid regime (left figure of Fig. 2C). In the AFM phase, κ_{xy} is absent below ~ 5 T within the resolution and increases with approaching H_{AF} . Upon entering the spin liquid state, κ_{xy} shows an upward increase. On the right axes of Figs. 2A and 2B, thermal Hall conductance per 2D layer, $\kappa_{xy}^{2D} = \kappa_{xy}d$, where $d=0.57$ nm is the interlayer distance, is plotted in units of K_0 . Taking into account the ambiguity of measuring the contact distances, κ_{xy}^{2D} exhibits a quantized plateau at one half of K_0 , within an error bar of $\pm 10\%$, above ~ 9.7 T (Fig. 2B). Above ~ 11.5 T, κ_{xy}^{2D} is strongly suppressed from the half quantized value. Weakly H -dependent κ_{xy}

between $\mu_0 H = 7$ and 8.5 T is unrelated to the topological property, because κ^{xy}/T in this regime is temperature dependent (Fig. S3). On the other hand, κ_{xy} is strongly suppressed or vanishes when \mathbf{H} is applied parallel to the b axis (armchair direction of a honeycomb lattice) and a half-integer thermal QHE is not observed as shown by blue circles in Figs. 2A and 2B (right figure of Fig. 2C). The absence of the transverse signal for $\mathbf{H} \parallel b$ is expected from the crystal structure, which possesses a two-fold symmetry around the b axis. In contrast, no two-fold symmetry exists around the a axis, because the Ru sites form two sublattices.

A rapid reduction of κ_{xy}^{2D} at low temperatures, followed by a half quantized plateau similar to the present $\mathbf{H} \parallel -a$ results, has also been reported in previous measurements [6] for \mathbf{H} applied in a *tilted* direction in the ac plane with $\theta = -60^\circ$ and -45° . Figure 2D and its inset show T -dependence of κ_{xy}/T at $\mu_0 H = 10.5$ T where the half-integer thermal QHE is observed for the in-plane field. The thermal QHE persists up to ~ 5 K, above which κ_{xy}/T is enhanced from the quantized value, peaks at around 15 K and decreases. A similar T -dependence has also been reported for $\theta = -45^\circ$ and -60° [6]. Thus the thermal Hall response in the spin liquid state for $\mathbf{H} \parallel -a$ is essentially the same as that for $\theta = -45^\circ$ and -60° including the Hall sign. The enhancement from the quantized value at high temperature has been discussed in terms of the signature of a topological phase transition [29]. What sets the current experiments apart is that the observation of a half-integer thermal QHE with no out-of-plane field unambiguously demonstrates that the quantization effect is unrelated to the Lorentz force or the formation of Landau levels, but in fact the manifestation of an anomalous thermal QHE originating from the formation of a Chern insulator of Majorana fermions.

Further varying the field direction, Figs. 3A and 3B depict κ_{xy} at $\theta = \pm 60^\circ$ at 4.3 K. Finite κ_{xy} is observed both in the AFM and spin liquid regimes. The half-integer quantized thermal Hall conductance with positive sign followed by the rapid reduction at high field is observed at $\theta = -60^\circ$, as reported previously. The half-integer quantized thermal Hall conductance is also observed at $\theta = +60^\circ$ but, strikingly, its sign is negative in the spin liquid state. Moreover, this sign change vanishes in the AFM phase below 6 T for which κ_{xy} at $\theta = \pm 60^\circ$ coincide within the experimental resolution. We note that the positive κ_{xy} for $\theta = -90^\circ$ ($\mathbf{H} \parallel -a$, see Fig. 2) means, as required by crystal symmetry, a negative κ_{xy} for $\theta = 90^\circ$. Taken together these out-of-plane results demonstrate a *varied* non-trivial sign structure of κ_{xy} in the spin liquid regime with respect to the angle θ of the c axis within the ac plane. In sharp contrast, such a sign change for $\theta = \pm 60^\circ$ is absent well inside the AFM phase, demonstrating that the mechanism behind the thermal Hall

effect in the spin liquid state is fundamentally different from that in the AFM phase. The finite κ_{xy} in the AFM phase might be attributed to a magnon thermal Hall effect (arising from the Berry curvature of magnon bands [30]). As the magnons are bosonic quasiparticles, κ_{xy}^{2D} is not quantized [31]. The absence of a sign change of κ_{xy} between $\theta = \pm 60^\circ$ and the absence of κ_{xy} for $\mathbf{H} \parallel a$ well inside the AFM phase imply that such a magnon thermal Hall effect is dominated by a finite out-of-plane magnetic field. The eventual splitting of κ_{xy} in the AFM phase upon approaching the transition at H_{AF} for both $\theta = -90^\circ$ (green area in Fig. 1B) and -60° is likely due to fluctuation effects in the proximity of the phase transition.

Both the half-integer thermal QHE without an out-of-plane magnetic field and the nontrivial sign change of κ_{xy} with respect to the c axis are in stark contrast to the conventional Hall effect of the 2D electron gas, but can be naturally explained for the KSL. In the pure Kitaev model, adding a weak Zeeman field generates, at third order in perturbation theory, a three-spin interaction term $\propto -\frac{h_x h_y h_z}{J^2} \sum_{\langle j, k, \ell \rangle} \sigma_j^x \sigma_k^y \sigma_\ell^z$ (where $\langle j, k, \ell \rangle$ stands for three neighboring sites). Crucially, this term breaks time reversal symmetry and opens up a Majorana gap in the bulk. It manifests as a next nearest neighbor hopping of Majorana fermions and gives rise to a non-trivial topological band structure, with an associated topologically protected chiral Majorana edge current [2]. The Chern number C_h of the gapped Majoranas can be determined as

$$C_h = \text{sgn}(h_x h_y h_z), \quad (2)$$

where h_x , h_y and h_z are the field components with respect to the spin axes S^x , S^y and S^z (Fig. 1A), respectively. We note that the directions of the spin axes (x , y and z) are determined by both the spin-orbit interaction and the crystal structure, and are different from the directions of the crystal axes (a , b , and c). Experimentally, C_h can be readily determined by the sign of κ_{xy}^{2D} in the half-integer thermal QHE regime. A measurement of the field-angular dependence of the half-integer thermal QHE thus provides crucial information on the topology of the Majorana fermion band structure, and a crucial test of whether the Kitaev interaction is responsible for the thermal QHE.

The field-angular variation of the Chern number for the pure Kitaev model, with respect to the spin and crystal axes, is shown in Figs. 4A-C. Note that C_h is finite even in the absence of an out-of-plane magnetic field; e.g. $C_h = -1(+1)$ for \mathbf{H} parallel (antiparallel) to the a axis (see purple arrow in Fig. 4C). When the applied magnetic field has no out-of-plane component, one might suspect that the net chirality of any resultant edge states cannot be determined uniquely. However, this is only true when the field is applied along a high-symmetry axis with a two-fold

rotation symmetry. Such a symmetry would reverse the chirality of the edge states, ensuring their absence when the symmetry is present. This guarantees that the thermal conductance vanishes for a field along the b axis (see dashed purple arrow in Fig.4C), which does possess a two-fold rotation symmetry, while there is a finite response for a field along the a axis, which does not (a two-fold rotation would swap the A and B sublattices, as shown by black and white circles in Fig. 4D). The small but finite κ_{xy} experimentally observed for $\mathbf{H} \parallel b$ is likely attributed to a misalignment of the magnetic field direction from the b axis.

In the pure Kitaev model, when the magnetic field is rotated within the ac plane C_h changes its sign at $\theta \approx 35^\circ$ and $C_h=1$ and -1 for $\theta = -60^\circ$ and $\theta = 60^\circ$, respectively (see dashed orange and orange arrows in Fig. 4B). For a real material, it is natural to wonder whether this sign change can persist in the presence of non-Kitaev interactions. Generically such interactions will give rise to a *linear* in field contribution to the Chern number as $C_h = \text{sgn}\{c_1(h_x + h_y + h_z) + c_3h_xh_yh_z + \dots\} = \pm 1$. For in-plane fields, the sum $(h_x + h_y + h_z)$ vanishes, leaving the third order Kitaev contribution as the leading term. As a result, any additional interactions are not expected to play a significant role. For out-of-plane fields however the linear contribution survives and could in principle dramatically alter the sign structure of the Chern number, and hence the thermal Hall conductance. The fact that a sign change of κ_{xy} between $\pm 60^\circ$ is experimentally observed places constraints ([32], section 2) on the ratio of $c_1/c_3 \lesssim 0.2J_K^2$. We thus conclude that the contribution of non-Kitaev interactions to the observed half-quantized thermal QHE is vastly outnumbered by those of the Kitaev exchange.

The sign structure of the observed half-integer thermal QHE clearly shows that the non-trivial topology of the Majorana fermion bands in $\alpha\text{-RuCl}_3$ is consistent with that of the pure Kitaev spin liquid, identifying $\alpha\text{-RuCl}_3$ as the host of a new quantum phase of matter – a Chern insulator of neutral Majorana fermions. The present results demonstrate that the Kitaev interaction is responsible for the quantization of the thermal Hall effect and that the expected non-Abelian topological order persists even in the presence of non-Kitaev interactions.

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Data and materials availability: All the data in the manuscript main text are available as .csv ASCII files in the supplementary materials.

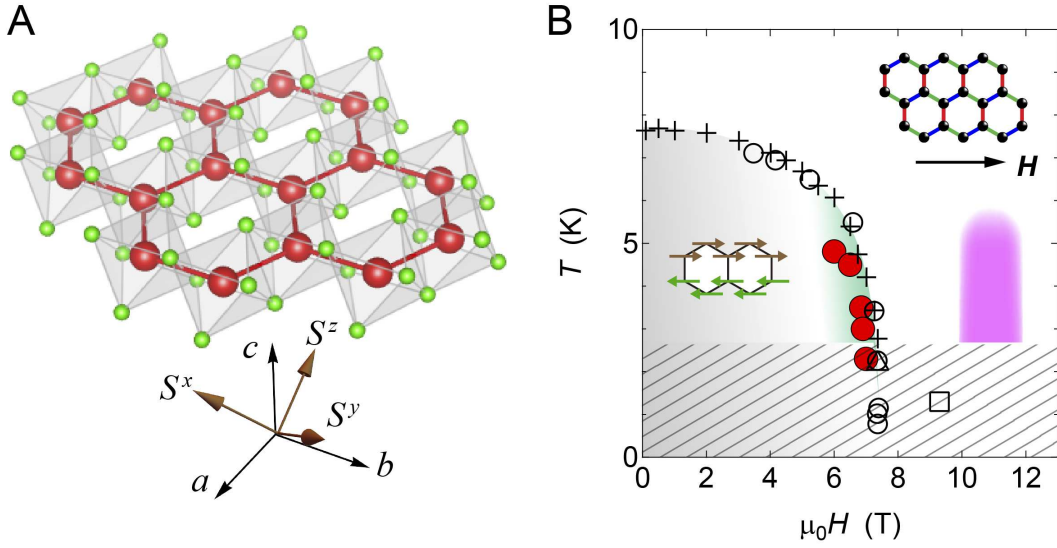


FIG. 1. **(A)** Upper figure illustrates the crystal structure of α -RuCl₃ in the ab plane. α -RuCl₃ is composed of very weakly coupled 2D honeycomb layers of edge-sharing RuCl₃ octahedra. Red and green circles represent Ru [Ru³⁺(4d⁵), $j_{\text{eff}} = 1/2$] and Cl⁻ ions. a (b) axis corresponds to zigzag (armchair) direction in a honeycomb lattice. The lower figure shows spin x , y and z axes (brown arrows), which are determined both by the spin-orbit interaction and crystal structure, crystal a , b and c axes (black arrows). Crystal a , b and c directions correspond to $(1, -1, 2)$, $(1, -1, 0)$ and $(1, 1, 1)$ directions in the spin axis, respectively. **(B)** H - T phase diagram in magnetic field applied parallel to the a axis. Pink area represents the regime where half-integer thermal QHE is observed. The gray shaded area represents the region of zigzag antiferromagnetic order, where κ_{xy} strongly suppressed or vanishes. The green shaded area represents the regime where finite κ_{xy} appears. Red circles indicate antiferromagnetic transition field H_{AF} determined by the longitudinal thermal conductivity (Fig.S2). Crosses, open circles, triangle and square indicate H_{AF} determined by magnetic susceptibility, magnetocaloric effect, neutron elastic scattering [22] and magnetic torque [28], respectively. Hatched area is the region where thermal Hall effect could not be measured.

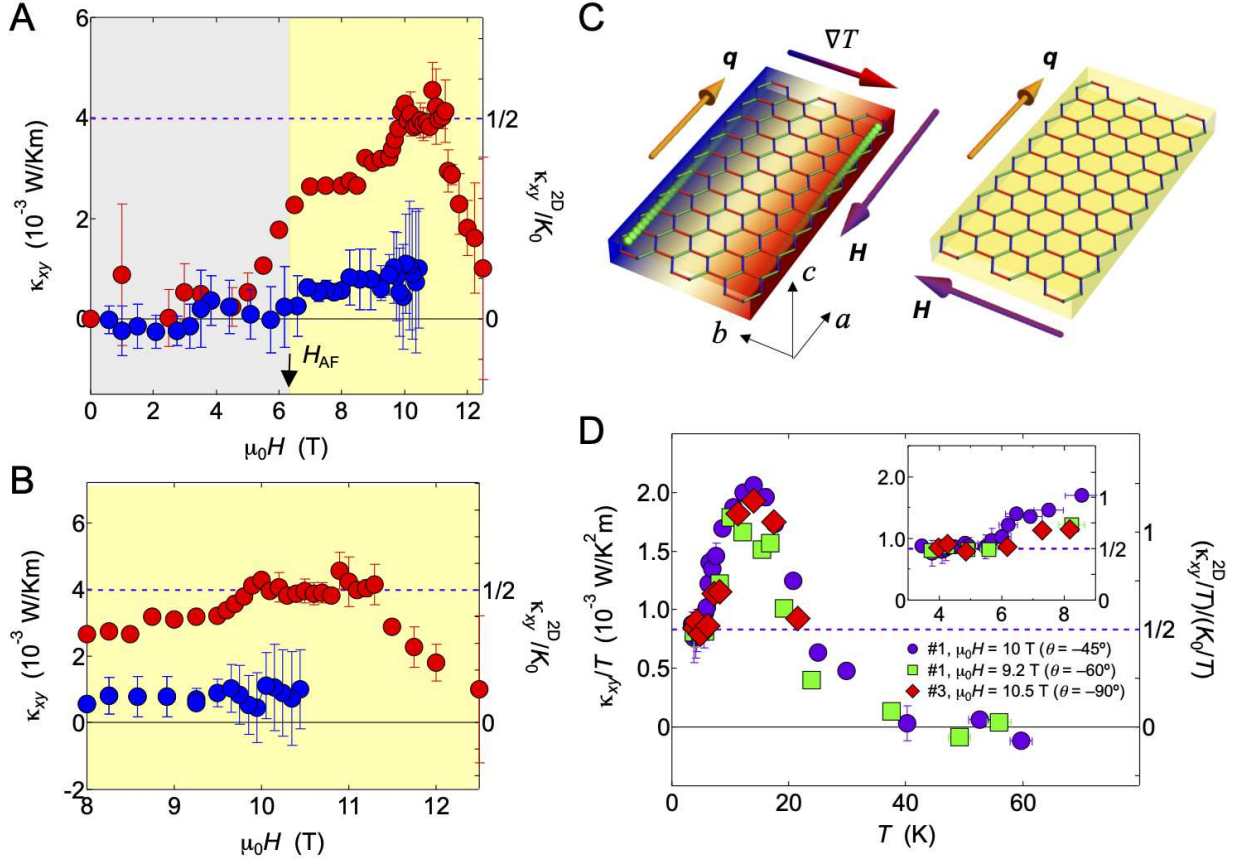


FIG. 2. (A) Thermal Hall conductivity κ_{xy} in the antiferromagnetic (gray shaded area) and spin liquid (yellow) states for $\mathbf{H} \parallel -a$ axis (red circles) and $\mathbf{H} \parallel b$ axis (blue circles) at 4.8 K. In the right axis, thermal Hall conductance per 2D layer κ_{xy}^{2D} is plotted in units of the quantum thermal conductance $K_0 = (\pi^2/3)(k_B^2/h)T$. The small but finite κ_{xy} experimentally observed for $\mathbf{H} \parallel b$ is likely attributed to a misalignment of the magnetic field direction from the b axis. (B) The same data in high field spin liquid region. (C) Schematics of the transverse temperature gradient generated by thermal current \mathbf{q} applied along a axis. Left: When \mathbf{H} is applied along $-a$ axis, temperature gradient ∇T appears along b axis. The red (blue) area indicates high (low) temperature region. Green circles represent the edge current of itinerant Majorana particles. Right: When \mathbf{H} is applied along b axis, no transverse temperature gradient appears along the b axis. (D) Temperature dependence of κ_{xy}/T for $\mathbf{H} \parallel -a$ at the magnetic field where the thermal QHE is observed. The right axis shows κ_{xy}^{2D}/T normalized by K_0/T . The results for $\theta = -60^\circ$ and -45° of different crystal reported previously are also plotted. The inset shows the same data at low temperature range.

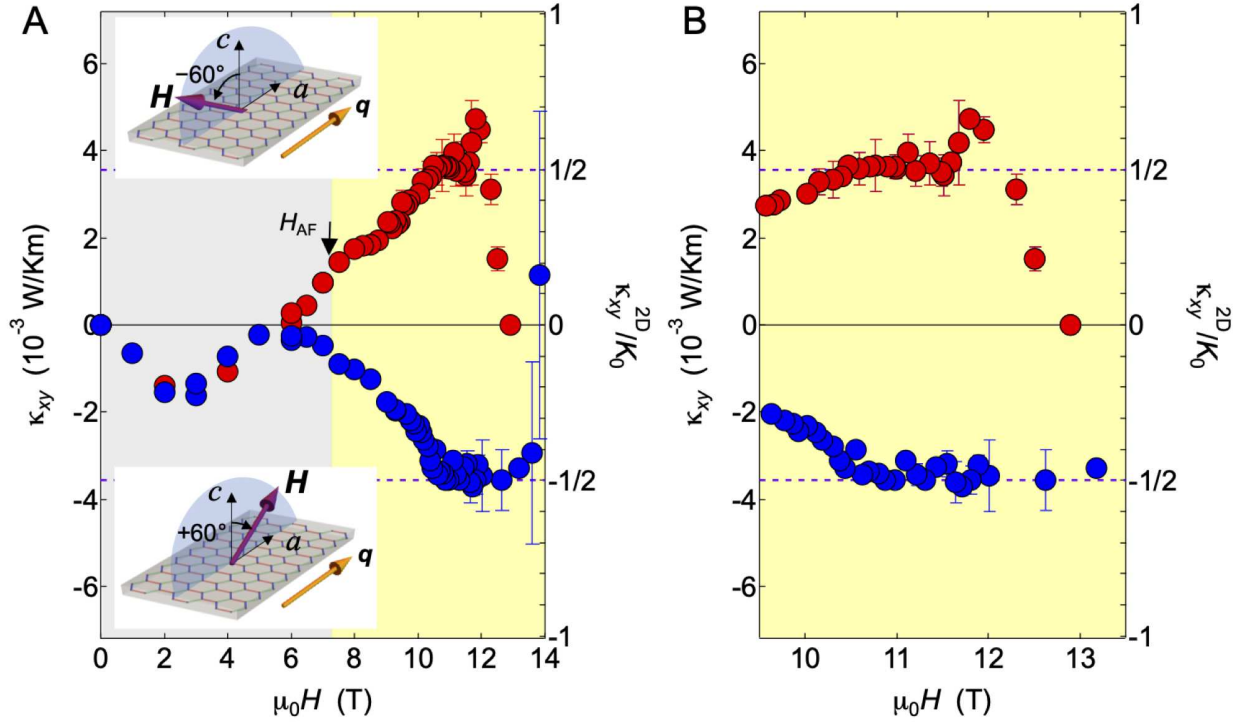


FIG. 3. (A) Field dependence of κ_{xy} at 4.3 K in tilted field of $\theta = -60^\circ$ (red circles) and 60° (blue circles) away from the c axis in the ac plane. Gray and yellow shaded areas represent the AFM ordered and spin liquid states, respectively. Thermal current \mathbf{q} is applied along the a axis. The antiferromagnetic transition field determined by the minimum of $\kappa_{xx}(H)$ is $H_{AF}=7.0$ T (B) The same data in the high field spin liquid state.

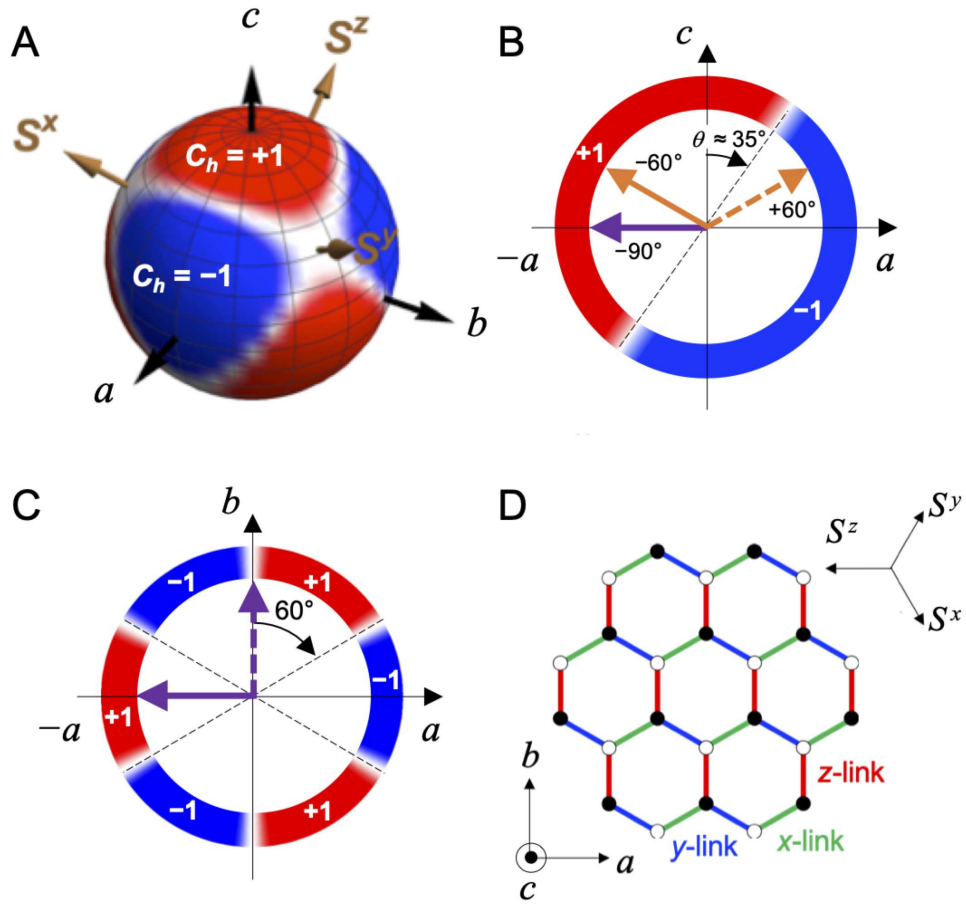


FIG. 4. (A) Theoretical results for the field-angular variation of Chern number C_h , which is ± 1 , for the pure Kitaev model with respect to the spin and crystal axes. (B) Variation of C_h within the ac plane. Orange and orange dashed arrows indicate the field direction at $\theta = -60^\circ$ and $+60^\circ$ from the c axis, respectively. Purple arrow indicates the field direction along $-a$ axis ($\theta = -90^\circ$). (C) Variation of C_h within the ab plane. Purple and dashed purple arrows indicate the field direction along $-a$ and b axis, respectively. (D) Schematic of honeycomb lattice and its directions. Two triangular sublattices comprise the honeycomb lattice. Black and white circles indicate Ru atoms on different sublattice. All links are labeled as x -, y - and z -links according to their orientations. Spin axes are perpendicular to these links.