High-field quantum spin liquid transitions and angle-field phase diagram of Kitaev magnet $\alpha$-RuCl$_3$

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The pursuit of quantum spin liquid (QSL) in the Kitaev honeycomb magnets has drawn intensive attention recently. In particular, $\alpha$-RuCl$_3$ has been widely recognized as a promising candidate for the Kitaev QSL. Although the compound exhibits an antiferromagnetic order under zero field, it is believed to be endowed with fractionalized excitations, and can be driven to the QSL phase by magnetic fields. Here, based on a realistic $K$-$J$-$\Gamma$-$\Gamma'$ model for $\alpha$-RuCl$_3$ [1], we exploit the exponential tensor renormalization group approach to explore the phase diagram of the compound under magnetic fields. We calculate the thermodynamic quantities, including the specific heat, Grüneisen parameter, magnetic torque, and the magnetotropic susceptibility, etc, under a magnetic field with a tilting angle $\theta$ to the $c^*$-axis perpendicular to the honeycomb plane. We find an extended QSL in the angle-field phase diagram determined with thermodynamic responses. The gapless nature of such field-induced QSL is identified from the specific heat and entropy data computed down to very low temperatures. The present study provides guidance for future high-field experiments for the QSL in $\alpha$-RuCl$_3$ and other candidate Kitaev magnets.

I. INTRODUCTION

Quantum spin liquids (QSL) constitute an exotic many-body state without symmetry-breaking spin order, where a number of unconventional properties such as fractionalized excitations and long-range entanglement emerge [2–6]. The celebrated, exactly solvable Kitaev model has attracted enormous attention due to the QSL ground state with localized excitations and long-range entanglement emerge [2–6]. The present study provides guidance for future high-field experiments for the QSL in $\alpha$-RuCl$_3$ and other candidate Kitaev magnets.

On the other hand, in the theoretical studies, the accurate microscopic model description of $\alpha$-RuCl$_3$ is important for understanding the compound, which, however, has been unsettled for a long period [32]. Recently, some of the authors proposed a Kitaev-Heisenberg-Gamma-Gamma' ($K$-$J$-$\Gamma$-$\Gamma'$) model with dominant Kitaev interaction $K = \pm 25$ meV, nearest-neighbor Heisenberg coupling $J = \pm 0.1|K|$, off-diagonal terms $\Gamma = 0.3|K|$ and $\Gamma' = \pm 0.02|K|$, which puts

![Fig. 1. The angle-field phase diagram of the realistic $K$-$J$-$\Gamma$-$\Gamma'$ model for $\alpha$-RuCl$_3$. There are three phases including zigzag (ZZ), quantum spin liquid (QSL), and the polarized (P) states as indicated in the figure. The phase boundaries are determined from the responses in Grüneisen parameters $\Gamma_B$, magnetic torque $\tau$, and the magnetotropic susceptibility $k$ at $T/|K| \approx 0.01$, in consistent with that from the ground-state magnetization curves [31]. We reveal that the phase transitions between ZZ (a ‘solid’ order), QSL (liquid-like phase), and the P (a weakly interacting ‘gas’-like system) phases meet at a tricritical point. The inset illustrates the honeycomb lattice defined on a cylinder of width $W = 4$, where the $x$, $y$, and $z$ bonds with bond-directional Kitaev interactions are marked in blue, green, and red colors, respectively. The in-plane $a$-axis, out-of-plane $c^*$-axis, and the angle $\theta$ of the applied field within the $ac^*$-plane are indicated by the arrows.](image)
the major experimental observations in a coherent picture, and makes a relevant prediction of QSL states induced by high out-of-plane fields [1]. Such a high-field QSL phase is separated from zigzag antiferromagnetic and the polarized phases, through two quantum phase transitions (QPTs) at 35 T and 130 T, respectively. This theoretical prediction is recently confirmed in high pulsed field experiments [31].

In this work, we extend the previous theoretical studies to the angle-field phase diagram of the realistic $K$-$J$-$\Gamma$-$\Gamma'$ model with the thermal tensor network approach [33–35]. Through the finite-temperature simulations of the specific heat $C_m$, Grüneisen parameters $\Gamma_B$, magnetic torque $\tau$, and magnetotropic susceptibility $k$, etc., we find a high-field QSL phase residing between the zigzag antiferromagnetic and the field-polarized phases. We determine the transition fields with prominent thermodynamic responses and offer concrete theoretical proposal for experimental probes of such spin liquid transitions in $\alpha$-RuCl$_3$ and potentially also other Kitaev candidate magnets.

II. MODEL AND METHODS

The effective spin Hamiltonian of $\alpha$-RuCl$_3$ [1] considered in this work reads

$$H = \sum_{\langle i,j \rangle} \left[ K S_i^x S_j^y + J S_i^z S_j^z + \Gamma(S_i^x S_j^x + S_i^y S_j^y) \right] + \Gamma'(S_i^x S_j^y + S_i^y S_j^x + S_i^z S_j^z + S_i^z S_j^z)],$$

where the summation is over the nearest-neighbor (NN) bond $\langle i,j \rangle$, with $\gamma = \{x,y,z\}$ (see inset in Fig. 1). $K$ denotes the bond-dependent Kitaev interactions, $J$ is the Heisenberg term, and $\Gamma, \Gamma'$ are the off-diagonal symmetric couplings with $(\alpha, \beta, \gamma)$ being the three spin components under a cyclic permutation.

The magnetic field $B$ is applied along the direction $[l m n]$ in the spin space $(S^x, S^y, S^z)$, i.e., the Zeeman term is $H_{\text{Zeeman}} = H_{\text{Zeeman}} = \frac{B}{\sqrt{\gamma^2 + \gamma^2}} [S_i^x S_j^x + S_i^y S_j^y] \cdot [l, m, n]$. Therefore, $H_{[110]}$ and $H_{[111]}$ correspond to the fields applied along the $a$- and $c^*$-axis, respectively. The angle between the applied field $H_{[110]}$ and $c^*$-axis within the $ac^*$-plane can be represented by $\theta = \arccos\left(\frac{2 \pi n}{\sqrt{6} + 2 \pi} \right) \times 180^\circ / s$, as depicted in the inset of Fig. 1.

Simulations based on the $K$-$J$-$\Gamma$-$\Gamma'$ model can well reproduce the low-temperature zigzag order [14–16], double-peaked specific heat [16, 36, 37], magnetic anisotropy [14, 15, 36, 38–41], magnetization curves [17, 20, 36, 38], and the prominent M-star dynamical spin structure factors [15, 16] in $\alpha$-RuCl$_3$ (see a brief recapitulation in Appendix A). Besides, one remarkable prediction based on this realistic model is the presence of high-field QSL driven by out-of-plane fields [1], whose nature is still under intensive investigation [42].

Below we employ the exponential tensor renormalization group (XTRG) [34, 35] method and perform finite-temperature calculations on a honeycomb-lattice cylinder with total sites $N = W \times L \times 2$, where the width is fixed as $W = 4$ and length $L$ ranges from 6 to 12, as illustrated in the inset of Fig. 1. We retained up to $D = 400$ bond states with truncation errors $\epsilon \approx 10^{-4}$ down to the lowest temperature $T/|K| \approx 0.0085$, which guarantees well converged results till the lowest temperature (c.f., Appendix B).

III. FINITE-TEMPERATURE CHARACTERISTICS OF QUANTUM SPIN STATES AND TRANSITIONS

A. Specific heat and isentropes

We start with conventional thermodynamic quantities such as the specific heat $C_m$ and magnetic entropy $S/\ln 2$ in the field $[110]$ direction, the double-peaked $C_m$ structure can be observed under a finite range of fields $B/|K| \lesssim 0.22$, with the high-$T$ and low-$T$ peaks correspond to two temperature scales $T_{H}$ and $T_{L}$: the short-range spin correlations establish at $T_{H}$ and the long-range antiferromagnetic zigzag order is formed below $T_{L}$, respectively. When the field $B/|K|$ is increased from 0 to 0.22, the low-$T$ $C_m$ peak moves towards lower temperatures, indicating that the zigzag order gets gradually suppressed by the magnetic fields. On the other hand, as the field exceeds $B/|K| = 0.22$, and below the polarization field, a low-$T$ peak emerges as indicated by $T_{c}^*$, below which there exists a field-induced QSL phase (c.f., Appendix A).

The corresponding isentropes with $\theta = 0.8^\circ$ are shown in Fig. 2(c). The adiabatic $T$-$B$ curves exhibit distinct changes when entering (rapid increase of $T$) and leaving (a dip) the intermediate QSL regime. They clearly signal two QPTs from the zigzag order to the QSL phase then to the field-polarized phases, at $B/|K| \approx 0.22$ and 0.62, respectively. The transition fields determined with density matrix renormalization group (DMRG) calculations on the same geometry [31] are denoted in the $T = 0$ axis with solid dots, where excellent agreements with the present finite-temperature results are seen.

The situation changes dramatically when the field angle increases to $\theta = 5^\circ$. As shown in Fig. 2(b,d), the results suggest that there is only one critical field between the zigzag ordered and field polarized phases, with no intermediate states any more. The behaviors of $C_m$ and $S$ are quite similar to that of the in-plane-field case [1], except that the transition field is higher. Thus we find the intermediate QSL phase very sensitively depends on the angle $\theta$. To accurately determine the phase boundaries in the angle-field phase diagram, below we resort to the thermodynamic, experimentally accessible quantities and parameters.

B. Grüneisen parameter

The magnetic Grüneisen parameter $\Gamma_B$ has been employed to accurately determine the critical in-plane fields in $\alpha$-RuCl$_3$ [43], which, however, poses challenges to many-body calculations. Here with the state-of-the-art XTRG method, we are able to compute this thermodynamic ratio and show the results.
The field-dependent Grüneisen parameters $\Gamma$ calculated by B$_{\text{KFL}}$ liquid (KFL), and paramagnetic (PM) phases are guides for the eye. (c,d) show the isentropes $S/I2$ for two different $\theta$ angles, where the critical fields ($B_{c1,2}$ and $B_c$) are indicated by the dots. These calculations with scanned fields are performed on the YC4 $\times$ 6 $\times$ 2 lattice. (e) The field-dependent Grüneisen parameters $\Gamma_B$ at various $\theta$ angles with fixed initial temperatures $T_i \approx$ 0.015, 0.012, and 0.01. The data are calculated by $\Gamma_B \approx 1/(dT/dH)_{\text{eff}}$, and are shifted vertically by a value of 30 for clarity. For small $\theta$, e.g., 0.8°, 1.4°, and 2.8°, two critical fields $B_{c1}$ and $B_{c2}$ indicated by the red and blue arrows denote the low- and high-field phase transitions, respectively; while only a single phase transition $B_c$ indicated by a black arrow is observed for $\theta \geq 4°$. The segment around each arrow gives the range of errorbar for the determined transition fields.

in Fig. 2(e). The field-dependent $\Gamma_B = 1/T(dT/dH)_{\text{eff}}$ are derived from the simulated isentropes starting from various initial temperatures (and a fixed field). A sign change structure in $\Gamma_B$ can be observed in Fig. 2(e) near the higher transition field $B_{c2}/|K| \approx 0.62$ (indicated by the blue arrows), and it becomes more and more pronounced as temperature lowers, revealing a second-order phase transition from QSL to the polarized phase. On the other hand, in the relatively low-field regime with $B_{c1}/|K| \approx 0.22$, a peak in $\Gamma_B$ is observed (indicated by a red arrow) that corresponds to a first-order QPT between ZZ and the QSL phases.

When the field is rotated within the ac*-plane, the higher transition field shifts from $B/|K| \approx 0.62$ to 0.06 as the angle $\theta$ changes from 0.8° to 20°, which reflects that the polarization field is very sensitive to the angle $\theta$. The first-order QPT stays around $B_{c1}/|K| \approx 0.23$ for small angles and merge to the second-order QPT at around $\theta \approx 4°$, where a tricritical point emerges. In Fig. 1, we gather the transition fields estimated by $\Gamma_B$ and obtain the angle-field phase diagram. As also indicated in Fig. 2(e), the errorbars of the phase boundaries can be estimated as the difference in field strengths of the dips and peaks in $\Gamma_B$.

C. Magnetic torque and magnetotropic susceptibility

The torque magnetometry constitutes a sensitive technique to probe the magnetic anisotropies in quantum materials, and recently been used to study the intricate quantum spin states and transitions in $\alpha$-RuCl$_3$ [26, 44]. However, its numerical results are lacking, partly due to the challenges in its many-body simulations.

With thermal tensor networks, we can compute the magnetic torque and its derivative, magnetotropic susceptibility, with a high accuracy. As the free energy $F$ can be written as $dF = -\delta SdT - PdV - MdB + \tau d\theta$ where $\theta$ is the tilted angle of the magnetic field, the first derivative $\tau = \partial F/\partial \theta$ represents the magnetic torque, which can be measured in $\alpha$-RuCl$_3$ experiments through $M \times B$ [44]. Recently, resonant torsion magnetometry technique is also used to measure the magnetotropic susceptibility $k = \partial^2 F/\partial \theta^2$ (the second derivative of free energy) [26, 45]. Following this line, below we perform XTRG calculations of the $K$-$J$-$\Gamma$-$\Gamma'$ model for $\alpha$-RuCl$_3$, investigate $\tau$ and $k$ at various temperatures and fields, and predict salient features of the two QPTs in the magnetotropic quantities to be checked in future high-field measurements.

In Fig. 3(a), we show the magnetic torque $\tau(\theta/2) = (F_\theta - F_0)/\theta$ (where $F_0$ represents the free energy at zero-field) with
Fig. 3. (a) The calculated magnetic torque $\tau$ (the upside curves with left axis) and the absolute value of its derivative $|d\tau/dB|$ (the downside two with right axis) of $\alpha$-RuCl$_3$ model with fields applied along $\theta = 0.4^\circ$ and $0.7^\circ$ at $T = 0.03$, 0.02 and 0.01. Two transition fields $B_{c1}$ and $B_{c2}$ are identified from the peak positions of $|d\tau/dB|$ indicated by the red and blue arrows, respectively. (b) The static spin-structure factors $S(k)$ (see the main text) for $\theta = 0.8^\circ$ with $k = M$ and $\Gamma$ in the Brillouin zone (shown in the inset). The red arrow denotes a fast drop of $S(M)$, indicating the suppression of the zigzag antiferromagnetic order at low temperatures, while the blue arrow corresponds to the field where both $S(M)$ and $S(\Gamma)$ decrease towards zero. (c) The calculated magnetotropic susceptibility $k$ for $\theta = 0.8^\circ$ at various low temperatures. The sharp dip corresponds to the second-order phase transition denoted by the blue arrow, while a kink occurs at around $B/|K| = 0.19$ signposted by the red arrow as zoomed in in the inset.

$\theta = 0.8^\circ$ and $1.4^\circ$, computed at low temperatures $T/|K| = 0.03$, 0.02 and 0.01. At low fields, $B < B_{c1}$, we find a relatively small value of $\tau$, which is understandable as the torques in two sublattices are expected to cancel each other in the antiferromagnetic ZZ phase, resulting in a nearly zero total net torque value. As fields further increase, the calculated $\tau$ gets enhanced rapidly as the ZZ order is suppressed in the intermediate QSL regime, which eventually drops again to small values at high fields as the system enters to the polarized phase. This can be ascribed to the fact that the angle between induced moments and fields is almost zero. The transition fields can thus be determined from where the torque changes most rapidly by computing the derivatives of $\tau$ with respect to the field $B$, i.e., $d\tau/dB$ shown in Fig. 3(a). The red and blue arrows indicate the transition fields from ZZ to QSL and QSL to polarized phases, respectively.

The behaviors of magnetic torque are also found consistent with the static spin-structure factor results

$$S(k) = \sum_{j\neq i, j \neq 0} e^{ikr_j-\tau} (\langle S_{i \alpha} S_j \rangle - \langle S_{i \alpha}\rangle \langle S_j \rangle),$$

where $i_0$ indicates a central reference site and the results are at relatively low temperature $T/|K| = 0.02$ and 0.01. As shown in Fig. 3(b), the zigzag spin correlations at small fields, e.g., $B < B_{c2}$ can be evidenced by the large $S(M)$ value [with the M as well as $\Gamma$ point indicated in the inset of Fig. 3(b)], which becomes suppressed in the intermediate QSL phase. The enhancement of $S(\Gamma)$ near $B_{c1}$ signals the buildup of uniform magnetization where the torque $\tau$ also increases rapidly in Fig. 3(a). When the system enters the spin polarized phase at $B_{c2}$, the structure factor peaks at M and $\Gamma$ points both vanish as expected [Fig. 3(b)].

The magnetotropic susceptibility $k$ can also be used to sensitively probe the two quantum phase transitions. In Fig. 3(c), we plot the results with $\theta = 0.8^\circ$ at $T/|K| = 0.03$, 0.02, and 0.01. The parameter $k$, second-order derivative of the free energy with respect to the magnetic field orientation $\theta$, has intimate relation to the susceptibility $\chi$ [45] and exhibits discontinuities at second-order phase transitions. In Fig. 3(c), the sharp dip at around $B \approx B_{c2}$ denoted by the blue arrow corresponds to a second-order transition, while the low-field one, as emphasized in the inset, shows a kink at around $B \approx B_{c1}$ which corresponds to a first-order phase transition. From the magnetotropic quantities $\tau$ and $k$, we determine the transition fields at $\theta = 0.7^\circ$ and $0.8^\circ$ and show them also in Fig. 1. Besides, we have also computed the matrix product operator (MPO) entanglement of the system, which provides accurate estimate of transition fields in accordance with the results above (see Appendix C). With these finite-temperature simulations, we show that the high-field torque magnetometry measurements can be used to sensitively detect the two QPTs associated with the intermediate QSL phase in future experimental studies.

IV. GAPLESS NATURE OF THE HIGH-FIELD QSL IDENTIFIED FROM THERMODYNAMICS

As indicated by the dome-like feature in Fig. 2(a,c), there exist an intermediate QSL regime below the emergent low-temperature scale $T_c$.

$T_c$. To further reveal the nature of this in-
termediate phase, we push the calculations of $C_m$ and $S/\ln 2$ to longer YC4 cylinders with $L$ up to 12.

In Fig. 4(a), we find the high- and low-temperature scales $T_H$ and $T'_L$ change only slightly as we elongate the system from $L = 6$ to $L = 12$. The height of the peak at $T'_L$ gets lowered, while the $C_m$ values for $T < T'_L$ are actually enhanced, which gives rise to a shoulder-like structure for the largest system size $L = 12$ as indicated by the grey arrow below $T/K \approx 0.03$. The corresponding entropy curves are shown in Fig. 4(b), where we see that there are considerable amount of low-temperature entropies below $T/K \approx 0.03$, indicating the strong spin fluctuations and large spin excitation density of states. In the inset of Fig. 4(b), we subtract the results of two YC4 lattices with different (adjacent) lengths, e.g., the [8–6] represent results obtained by subtracting YC4×6×2 data from the YC4×8×2. The obtained entropy results reflect the bulk property in the central columns and suffer less severe boundary effects, and a power-law behavior of entropy $S \sim T''$ can be clearly seen, which indicates that the high-field QSL has gapless low-energy excitations and there are considerable entropies released only below the temperature $T \approx 0.03[K]$. Overall, the thermodynamic results here along the tilted angle point to the conclusion of a gapless QSL, consistent with previous DMRG results (restricted to out-of-plane fields) [1].

V. CONCLUSIONS AND DISCUSSIONS

In the present work, we have calculated the experimentally relevant thermodynamic properties, i.e., magnetic specific heat, magnetocaloric effect characterized by the Grüneisen parameters, magnetic torque, and the magnetotropic susceptibility of the primary candidate Kitaev magnet α-RuCl$_3$ based on the realistic $K$-$J$-$Γ$-$Γ'$ model and through highly accurate XTRG method. Recently, a high-field magnetization measurement on α-RuCl$_3$ up to 102 T have witnessed two phase transitions enclosing an intermediate phase [31], in agreement with the prediction based on the model calculations [1]. Here we calculated further thermodynamic properties that provide a comprehensive angle-field phase diagram and useful guide for future experimental studies. For $θ < 4^\circ$, we find two field-induced quantum phase transitions evidenced by various quantities. (i) The diverging Grüneisen parameter $Γ_B$ shows a sign change behavior at high-field transition point $B_{c2}$, suggesting a second-order phase transition. Exactly at the same field, the magnetotropic susceptibility $k$ features a sharp peak. (ii) The hump in $Γ_B$ at around $B_{c1}$ reflects a quantum phase transition possibly of first-order. There is also a peak in $d|S|/dB$ and a kink in $k$, which point to the same conclusion. On the other hand, for large $θ \geq 4^\circ$, only a single phase transition from antiferromagnetic to polarized phase is found, suggesting the absence of an intermediate QSL phase.

Moreover, it is noteworthy that besides the conventional candidate materials with Kitaev interactions, e.g., $X_2$IrO$_3$ (X = Na, Li, Cu) [46–54], $X_2$LiIr$_2$O$_4$ (X = Ag, Cu, H) with Ir$^{4+}$ [55–57], and $X$ $3$ (X = Ru, Yb, Cr, R = Cl, I, Br) [51, 58–73], etc., some newly reported Kitaev family such as rare-earth chalcaholide REChX (RE = rare earth; Ch = O, S, Se, Te; X = F, Cl, Br, I) [74, 75] and cobalt honeycomb oxides Na$_2$Co$_2$TeO$_6$ [76, 77], Na$_2$Co$_2$SbO$_6$ [78], and BaCo$_2$(AsO$_4$)$_2$ [79], etc., also offer a platform exhibiting highly anisotropic, bond-dependent exchange couplings. It would be worthwhile to explore their field-induced quantum spin states along the out-of-plane direction and generally tilted angles in the future, and the present study on angle-field phase diagram of the $K$-$J$-$Γ$-$Γ'$ model provides theoretical guide for experimental explorations in these intriguing quantum magnets.

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Appendix A: Realistic α-RuCl₃ model and the high-field QSL

In the strongly correlated transition metal compounds, α-RuCl₃ is believed to serve as a prototypical candidate material for the Kitaev model [14, 15, 38, 41, 63, 66, 80]. As it undergoes a magnetic transition to antiferromagnetic order at a relatively low temperature, i.e., T ≃ 7 K [14, 15, 36, 38], people have taken efforts to find out the effective spin Hamiltonian of α-RuCl₃, which includes not only the Kitaev term K but also the Heisenberg interactions J, J₂, and J₃, and off-diagonal Γ and Γ' couplings [51, 58–67, 81, 82], which is important for gaining insights into the candidate Kitaev material. The Kitaev interaction in this compound has been widely accepted to be ferromagnetic [16, 41, 51, 58–65, 83]. However, the magnitudes of K and even the signs of non-Kitaev terms were undetermined, and it is very challenging to find a model that can accurately describe the realistic α-RuCl₃ [32].

We focus on the minimal K-J-Γ-Γ' model [1, 81, 84, 85], especially on its field-induced properties. In our previous work [1], we determine the parameters from fitting the thermodynamic properties, i.e., the double-peak feature of specific heat with two temperature scales at around 100 K and 7 K [16, 36, 37], and the anisotropic susceptibilities along α- and c'-axis [15, 39, 40]. The determined parameter set is K = −25 meV, Γ = 0.3|K|, Γ' = −0.02|K|, J = −0.1|K|, with in-plane and out-of-plane Landé factor gₐ = 2.5 and g,c = 2.3, respectively. With this model, the low-temperature zigzag antiferromagnetic order [15] and its magnetization curve can be well reproduced, which are found in quantitative agreement with experiments [36, 38]. The transition fields that suppressed the zigzag order are also in accordance with experimental observations, along the in-plane direction [17, 20, 36, 38] and with a tilted θ = 35° angle [21]. Besides, it was also found that the zigzag order gets suppressed at 35 T under out-of-plane fields (θ = 0°), above which, and below a polarization field of 100 T level, a field-induced QSL phase emerges as evidenced by both density matrix renormalization group (DMRG) and variational Monte Carlo (VMC) method at ground state [1]. Here in this work, we further extend the conclusion that the high-field QSL phase can extend to a finite range of θ angles.

Appendix B: Exponential tensor renormalization group method

The exponential tensor renormalization group (XTRG) method [33, 34] exploited in this work carries out the finite-temperature many-body simulations down to low temperature exponentially fast, which has been shown to be a highly efficient and very powerful tool in solving various 2D spin lattice models [34, 35, 86, 87], realistic quantum magnets [1, 88–90], and correlated fermion system [91, 92]. Below we synthesize the main idea of such method and provide some benchmark results on the realistic K-J-Γ-Γ' model.

In XTRG, we start with the high-temperature density matrix ρ(τ₀) with the initial τ₀ = |K|/T = 0.0025 through a series expansion in thermal tensor networks [33], i.e.,

\[
\rho(τ₀) = e^{-τ₀\hat{H}} \equiv \sum_{n=0}^{N_τ} \frac{(-τ₀)^n}{n!} \hat{H}^{n},
\]

where N_τ is the expansion order (often smaller than 10 in practice) and the ρ(τ₀) could converge to machine precision. Given the density matrix ρ(τ₀) represented in the form of a matrix product operator (MPO), we keep squaring the MPO via tensor network contractions and thus cool down the system exponentially as ρ(τₙ) = ρ(τₙ₋₁) · ρ(τₙ₋₁) where τₙ = 2^nτ₀ (n ≥ 1). Based on this, various thermodynamic properties can be computed, including free energy f, internal energy u, magnetic thermal entropy S, and static spin-structure factors S(k), etc. We parallel perform the simulations with interleaved data points along the temperature axis, and interpolate data between those sampling points.

\[\text{Fig. A1. The low-temperature magnetization curves with field applied along } \theta = 0.8° \text{ of the } \alpha-\text{RuCl}_3 \text{ model. When the temperature is sufficiently low, the results converge to the ground state curve computed with DMRG [31] on the same } \text{YC}_4 \times 6 \times 2 \text{ geometry.} \]

For the YC₄×6×2 geometry considered in the main text, we compare in Fig. A1 the low-temperature magnetization curves (θ = 0.8° case) calculated by XTRG method with the DMRG results [31], where we find T/|K| = 0.0085 data converges well with the DMRG data. This confirms that the XTRG calculations can approach the low-temperature regime in the close vicinity of the ground state.

Appendix C: Matrix product operator entanglement

The phase transitions can be detected sensitively by the entanglement entropy of the matrix product operator
The bipartite MPO entanglement entropies $S_E$ under various fields $B$ applied along $\theta = 0.8^\circ$ and $\theta = 5^\circ$, at a low temperature $T/|K| = 0.0085$. A drop and a peak features can be seen at respectively the low and high transition fields $B_1$ and $B_2$ for $\theta = 0.8^\circ$ (denoted by the red and blue arrows). For the $\theta = 5^\circ$ case, there is only a single peak in the $S_E$ curve located at $B_c$.

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(π) [34]. Regarding the MPO as a supervector, we can take a Schmidt decomposition of the purified “wavefunction” and compute the entanglement entropy $S_E$ between the two parts of the system. Here we study its field-dependent behaviors for $\theta = 0.8^\circ$ and $5^\circ$, on the $YC_4 \times 6 \times 2$ lattice with the calculated results shown in Fig. A2.

The MPO entanglement entropy $S_E$ is expected to diverge at the second-order quantum critical point (QCP) in the low temperature limit. In finite-size calculations, it instead exhibits a peak near the QCP. In Fig. A2, we show the low-temperature $S_E$ vs. magnetic fields $B$, and find for $\theta = 0.8^\circ$ case there exists a peak near $B_{c1}$. This is indicated by the blue arrow, with the determined field value consistent with that obtained from $\Gamma_B$ data in Fig. 2(e). In addition, it can be seen that $S_E$ firstly shows an almost steady behavior at the low-field antiferromagnetic phase, then drops abruptly near $B_{c1}$ as indicated by the red arrow, reflecting that the low-field phase transition is likely of first order. For $\theta = 5^\circ$ case, there is only a single peak for $S_E$ vs. $B$ as shown in Fig. A2, where a prominent peak at $B_c$ clearly signals the QCP between the zigzag and spin polarized phases.
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