Universal thermodynamics in the Kitaev fractional liquid

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In the Kitaev honeycomb model, the quantum spin fractionalizes into itinerant Majorana fermion and gauge flux upon cooling, leading to rich experimental ramifications at finite temperature and an upsurge of research interest. However, accurate modelings of the Kitaev materials by including realistic couplings beyond the pure Kitaev model constitute a major challenge for the community. With recently developed exponential tensor-network approach, we clear the pathway and perform finite-temperature simulations of the extended Kitaev model with additional interactions common in real materials. At intermediate temperature, we find an emergent Curie law of magnetic susceptibility and a stripy spin-structure factor characterizing the robust Kitaev fractional liquid. With this insight, we revisit the susceptibility measurements of Na_2IrO_3 and $\alpha\text{-RuCl}_3$ and find evidence of ferromagnetic Kitaev coupling and finite-temperature fractionalization. Bridging the gap between theories and experiments with the state-of-the-art tensor-network simulations, our findings provide guidance for future experimental exploration of the spin liquids in Kitaev materials by thermodynamic measurements and spin-resolved structure factor probes.

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I. INTRODUCTION

Quantum spin liquids realize a novel class of quantum states of matter [1–3] which represent the nonmagnetic states with emergent long-range entanglement and fractionalized excitations in frustrated magnets [4,5]. After extensive research for decades, the exactly soluble Kitaev model of the honeycomb lattice [6], which has three types of bond-directional Ising couplings in the trivalent lattice, was found as a prototypical system that can realize both gapped and gapless spin liquid states with spins fractionalized into itinerant and localized Majorana fermions [6,7]. In particular, in the gapless spin liquid phase, time-reversal symmetry-breaking perturbations such as a magnetic field can immediately induce a gapped non-Abelian chiral spin liquid, which has potential application for topological quantum computation [8].

The pursuit of the materialization of the intriguing Kitaev spin liquid (KSL) states has triggered a competition in the search for crystalline Kitaev-like materials [9–11]. Fortunately, there are Mott insulators with strong spin-orbit

coupling and an effective pseudospin $\frac{1}{2}$ on the honeycomb lattice that can realize the highly anisotropic couplings. These materials have been intensively studied in recent years, including the iridate family, such as A_2IrO_3 ($\text{A} = \text{Li}$ and Na) [12–18] and $\text{H}_3\text{LiIr}_2\text{O}_6$ [19], and the ruthenate family, such as $\alpha\text{-RuCl}_3$ [20–24]. Nonetheless, due to the couplings beyond the Kitaev model such as the Heisenberg, off-diagonal Γ , and further-neighbor interactions in real materials, semiclassical magnetic orders appear at low temperature T in most of these KSL candidate compounds. Surprisingly, signatures of fractionalization in quantum spins can nevertheless be detected at intermediate temperature in some Kitaev materials [17,22–24], even when the low- T states are classically ordered. These exciting observations and the spin liquid phase found in the presence of magnetic fields strongly suggest that these materials should be in proximity to the KSL phase and imminently call for systematic understanding of the thermodynamic properties of these Kitaev materials.

For the pure Kitaev model, the finite- T fractionalization of spins has been well established via the Majorana-based quantum Monte Carlo (QMC) simulations [25–27], which found two temperature scales and an intermediate regime with unconventional properties [25–29]. This provides an understanding of the exotic equilibrium [24] and transport measurements [30] in the Kitaev materials. Nonetheless, the gap between experiments and theoretical understanding is still apparent. Since the non-Kitaev interactions and magnetic field [31,32] spoil the exact solution and restrict QMC simulations to a relatively high temperature [33], little is known about the fractionalization of spins in the extended Kitaev models.

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In this work we employ the exponential tensor renormalization group (XTRG) [34–37] at finite temperature to explore the extended Kitaev model with additional Heisenberg and Γ interactions, which denies QMC simulations. We perform accurate thermodynamic calculations down to very low temperature and find an emergent Curie law of magnetic susceptibility below a high- T scale T_H (and above the low- T scale T_L), due to the fluctuating gauge fluxes and the predominant bond-directional nearest-neighbor (NN) spin correlations. In addition, the well-recognized characteristic features at intermediate temperature, including the fractional thermal-entropy plateau and the linear- T specific-heat scaling, also clearly persist in the extended Kitaev model.

Through exploring these robust finite- T features, we find a generic scenario in the extended Kitaev models relating to the realistic Kitaev materials: A fractionalization of the quantum spin takes place as temperature decreases, and half of the spin degrees of freedom, i.e., the itinerant Majorana fermions, release a large part of the entropy below T_H , accompanied by a saturated short-range spin correlation; meanwhile, the other half, the gauge fluxes, fluctuate strongly until near the much lower temperature T_L . In between the two temperature scales resides a unique finite- T quantum state dubbed the *Kitaev fractional liquid*, which constitutes a precursor of the quantum spin liquids.

Based on our XTRG simulations, we reveal that the fractional liquid regime does exist at intermediate temperature, for an extended range of external fields or non-Kitaev terms including the Heisenberg or Γ couplings, even when the KSL ground state is altered. These results therefore shed light on the exotic finite- T quantum states and intriguing properties of the Kitaev materials. In particular, with insight from the universal susceptibility scaling, we revisit the magnetic susceptibility data of Na_2IrO_3 and $\alpha\text{-RuCl}_3$ and find an evident signature of Curie susceptibility scaling in the two prominent Kitaev materials, suggesting the existence of the fractional liquid therein. We also propose the spin-correlation diagnostics of the fractional quantum liquid, which is available through spin-resolved neutron or x-ray diffraction scatterings.

The rest of the paper is organized as follows. In Sec. II we introduce the extended Kitaev model and the XTRG method for finite- T simulations. Sections III–V present our main results on the universal thermodynamic properties and spin-correlation characterization of the intermediate- T Kitaev fractional liquid that is robust against additional interactions. In Sec. VI we discuss the relevance of universal susceptibility scaling in two Kitaev materials, Na_2IrO_3 and $\alpha\text{-RuCl}_3$, and propose a spin-structure diagnosis of the fractional liquid. In Sec. VII we summarize our main conclusions and discuss their implications in future studies of the Kitaev model and materials.

II. MODEL AND METHODS

We study the celebrated Kitaev honeycomb model with various couplings

$$H = \sum_{\langle i,j \rangle_\gamma} [K_\gamma S_i^\gamma S_j^\gamma + J \mathbf{S}_i \cdot \mathbf{S}_j + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)] + \sum_i h S_i^z, \quad (1)$$

FIG. 1. Cylindrical honeycomb lattice of size $YC 4 \times 6 \times 2$ illustrated with the snakelike quasi-1D mapping path (underlying gray lines), adopted in the XTRG calculations. Three types of Kitaev bonds K_x , K_y , and K_z are marked in blue, green, and red, respectively. The $\mathbb{Z}_2$ flux $W_p = \sigma_1^z \sigma_2^x \sigma_3^y \sigma_4^z \sigma_5^x \sigma_6^y = +1$ (hexagon in white) or -1 (purple), with $\sigma_i^\gamma = 2S_i^\gamma$, can be pair flipped by applying a magnetic field $h \parallel [001]$ on the spin-space cubic axis. Typical nearest and next-nearest bonds d_x , d_z , d_{x+y} , and d_{x+z} are indicated by the dashed lines.

where $\mathbf{S}_i = \{S_i^x, S_i^y, S_i^z\}$ represent the spin- $\frac{1}{2}$ vector operator at site i . The three cubic coordinates $\{\alpha, \beta, \gamma\}$ represent $\{x, y, z\}$ up to a cyclic permutation and $\langle i, j \rangle_\gamma$ denotes the NN γ bond (see Fig. 1) with an Ising coupling K_γ . The non-Kitaev terms include the NN isotropic Heisenberg coupling J and an off-diagonal Γ interaction. Moreover, we apply an external field h coupled to the S^z component, which is along the $[001]$ spin-space direction and tilts an angle of 45° from the honeycomb ab plane (see Fig. 1) in typical Kitaev materials.

For the pure Kitaev model, spin can be represented as localized (b_i^γ) and itinerant Majorana fermions (c_i) through the parton construction $S_i^\gamma = \frac{i}{2} b_i^\gamma c_i$ [8]. The itinerant Majorana fermions can be related to the building up of spin correlation $\langle S_i^\gamma S_j^\gamma \rangle = u_{ij}^\gamma \langle c_i c_j \rangle$, and $u_{ij}^\gamma = b_i^\gamma b_j^\gamma = \pm i$ represents a $\mathbb{Z}_2$ gauge connection. The product of u_{ij}^γ on the six edges of a hexagon gives rise to the flux $W_p = \pm 1$ as illustrated in Fig. 1, which is conserved in the pure Kitaev case.

For the typical Kitaev materials Na_2IrO_3 and $\alpha\text{-RuCl}_3$, the predominant Kitaev interactions have been widely accepted to be ferromagnetic (FM) [10,24,33,38–40]. Thus, we choose $K_\gamma < 0$ henceforth and set $K_z = -1$ as the fixed energy scale. The Γ term is suggested to be of next-to-leading order and with $\Gamma > 0$ in $\alpha\text{-RuCl}_3$ [41,42]. On the other hand, in Na_2IrO_3 the Heisenberg coupling is believed to play an important role and is proposed to be antiferromagnetic (AF) [38,43]. Thus, in our simulations below we introduce an additional AF Heisenberg or Γ term with $J, \Gamma > 0$ and focus on the finite- T quantum states of the extended Kitaev model.

We use the XTRG method [34,36] to study the extended Kitaev model in this work. To start the procedure, we exploit a quasi-one-dimensional (quasi-1D) mapping and express the Hamiltonian of the lattice model as a compact matrix product operator (MPO). As shown in Fig. 1, the cylinder is mapped, via a snakelike path, into a quasi-1D system illustrated by the gray lines in the shadow. Given the MPO representation of the Hamiltonian H , we can represent the high-temperature density matrix also as an MPO via the series expansion (see details in Appendix A). After that, in the course of the

exponential cooling procedure, we keep squaring the density matrices to reach various lower temperatures. The free energy as well as other thermodynamic quantities can be computed at each temperature point via the thermofield double formalism, through a bilayer tensor trace [44].

We work on the cylindrical lattice YC $W \times L \times 2$ (see Fig. 1), with width W (up to 4) and length L (up to 6), thus total site $N = 2WL$, which reproduces results in excellent agreement with benchmark exact diagonalization (ED) on small system sizes as well as large-scale Majorana QMC simulations (see Appendix A). From comprehensive benchmark results, we see that finite-size effects are in most cases modest, at least for the intermediate- T regime of main focus in the present study. This is in contrast with the ground-state calculations and can be ascribed to the extremely-short-range correlations at intermediate temperature, even under non-Kitaev interactions (see Appendix B).

Moreover, as there are two characteristic temperature scales in the Kitaev model that are typically separated by more than one order of magnitude, our XTRG approach that cools down the system exponentially in the temperature scale turns out to be very efficient to address this problem. Therefore, we can exploit the XTRG method that is free of the sign problem to simulate the extended Kitaev model down to extremely low temperature $T \simeq 0.005$. In practical calculations we retain bond dimension up to $D = 600$ that is capable of describing the thermal quantum states accurately and produces well-converged thermodynamic results (Appendix C). We have also employed the density matrix renormalization group (DMRG) method to simulate the ground state, confirming the XTRG results at very low temperature.

III. KITAEV FRACTIONAL LIQUID

A. Pure ferromagnetic Kitaev model at finite temperature

It has been well established that the itinerant and localized Majorana fermions can give rise to the two temperature scales T_H and T_L , respectively. At intermediate temperature $T_L \lesssim T \lesssim T_H$, an exotic fractional quantum liquid emerges. To start with, we simulate finite- T properties of the isotropic FM Kitaev model ($K_{x,y,z} = -1$) by XTRG on a YC $4 \times 6 \times 2$ cylinder. In Fig. 2(a) we plot the spin correlation $\langle S_i^\gamma S_j^\gamma \rangle$ measured along the NN bonds d_z and d_x (d_γ always leads to the same results as d_x and thus is not shown) and next-nearest ones d_{z+y} and d_{x+y} (as depicted in Fig. 1). From the zero-field XTRG results, we find that only the nearest- d_z -bond correlations is established below T_H and the rest remain small down to the lowest temperature. Thus the spin correlations $\langle S_i^\gamma S_j^\gamma \rangle$ are nonzero only on the NN γ bond, i.e., they are extremely short range and bond oriented, in full agreement with the analytical results [45,46].

In fact, the itinerant Majorana fermions release most of their entropy and the NN spin correlation is established at around T_H [Fig. 2(a)]. On the other hand, the rest half of the spins, $\mathbb{Z}_2$ fluxes, strongly fluctuate until the low- T scale T_L , as evidenced by small W_p above T_L in Fig. 2(c). Correspondingly, in Fig. 2(d) the thermal entropy exhibits a quasiplateau with $S \simeq \frac{1}{2} \ln 2$, which constitutes a thermodynamic signature of the intermediate- T Kitaev fractional liquid.

FIG. 2. Spin correlations $\langle S_i^\gamma S_j^\gamma \rangle$ with (a) $h = 0$ and (b) $h = 0.045$ on bond $\langle i, j \rangle = d_x, d_z, d_{x+y}$, and d_{x+z} as depicted in Fig. 1, where the NN spin correlations are established at $T_H \simeq 0.373$ and converge to the ground-state (GS) DMRG results (closed symbols) at low temperature. In (b) the mean magnetization squared $\langle M \rangle^2$ has been subtracted in the correlation data (see the text). (c) Thermal average of flux W_p and (d) thermal entropy S results under various fields. As temperature decreases, the flux increases monotonically for $h \lesssim h_c$ and builds up rapidly at around $T_L \simeq 0.017$, while it becomes nonmonotonic for $h \gtrsim h_c$ and starts to decrease below a certain low temperature after the initial stage of increasing. (d) Entropy with $h \lesssim h_c$ exhibits a stable $S \simeq \frac{1}{2} \ln 2$ quasiplateau, above which the curves almost coincide; for $h \gtrsim h_c$, the entropy leaves a shoulderlike structure at intermediate temperature.

B. Kitaev fractional liquid at finite fields

Recently, the magnetic field effects on the Kitaev materials have raised great interest [30–33,47]. For example, the thermal quantum Hall signature has been detected in the field-induced spin liquid phase of α -RuCl₃ [30]. Given the fractional liquid scenario in the pure Kitaev model, it is also interesting to further study how it evolves under external fields [33]. In the following, we mainly focus on the field direction $h \parallel [001]$; the effects of external fields in other directions are discussed in Appendix D, where qualitatively the same results are seen.

The finite-field entropy results are contour plotted in Fig. 3(a), with the S vs T curves shown explicitly in Fig. 2(d). The two isentropic lines with $S/\ln 2 = 0.75$ and 0.25 in Fig. 3(a) correspond to the crossover temperatures T_H and T_L (or T'_L) very well. The intermediate- T regime, which corresponds to the $\frac{1}{2} \ln 2$ entropy plateau and signifies the fractional liquid, extends to a rather wide field range regardless of whether the low- T phase is an asymptotic KSL or not. The two-step release of entropy gives rise to the double-peaked specific-heat curves as illustrated in Figs. 3(b) and 4.

While T_H remains stable as the magnetic field h increases, the low- T crossover temperature T_L drops at $h = h_c \simeq 0.02$ and increases again as h further increases [T'_L in Fig. 3(b)]. As a matter of fact, T'_L represents a different low- T scale since for $h > h_c$ the ground state is no longer a KSL but a topologically trivial field-induced FM phase. This can be seen evidently in Figs. 3(c) and 3(d), where the KSL is maintained

FIG. 3. Contour plot of (a) thermal entropy $S/\ln 2$ and (b) specific heat of the Kitaev model under magnetic fields h , where two crossover temperature scales and an intermediate Kitaev fractional liquid (KFL) regime are evident. The T_H and T_L (T'_L) determined from (b) coincide with results from Ref. [33] (the white dots, available for $T \geq 0.05$ for $h \neq 0$). (c) Flux W_P and (d) magnetization M are shown vs various h and T . Three finite- T ($T = 0.07$) spin-structure factors at $h = 0, 0.03$, and 0.06 are presented in the inset of (c) (whose intensity color bar ranges from 0.2 to 0.5). A structure peak emerges at the center (Γ point) of the extended Brillouin zone due to the induced FM moment M at $h > 0$, together with a stripy background. (e) The field-induced FM phase is clearly indicated and the black dots represent the spins parallel to the $[001]$ field in the inset.

for $h \lesssim h_c$ [$W_P \neq 0$; see also Fig. 2(c)], while the field-induced moment $M = (\frac{1}{N} \sum_i \langle S_i^z \rangle) \neq 0$ becomes significant only for $h \gtrsim h_c$. Thus the transition field $h_c \simeq 0.02$ is in very good agreement with previous studies [48,49], although $[111]$ fields are usually considered.

C. Spin correlation and structure factor

In the robust fractional liquid regime, we again observe extremely-short-range and bond-directional spin correlations that characterize this exotic finite- T quantum state even under external fields. In Fig. 2(b) we plot the spin correlations

FIG. 4. Specific heat C_V under various magnetic fields h . The linear- T dependence of C_V at intermediate temperature is evident as indicated by the black dashed line, which also underlines the quasiplateau in the C_V/T curves plotted in the inset.

$\langle S_i^z S_j^z \rangle - M^2$ with the field-induced uniform background subtracted. Although the longer-range correlations rise up at lower temperature and spoil the extremely short correlation length inherited from the pure Kitaev model, we nevertheless can observe that the d_z correlation is predominant over a very wide range of intermediate temperatures.

Correspondingly, we compute the static spin-structure factor from the equal-time correlation function

$$S^\gamma(q) = \sum_j \langle S_{i_0}^\gamma S_j^\gamma \rangle_\beta e^{iq(r_j - r_0)}, \quad (2)$$

where r_0 is the position vector of the fixed reference site (i_0) and j (and thus vector r_j) runs over the lattice sites. We observe in Fig. 3(c) a stripy structure factor at intermediate temperature for both $h = 0$ and $h \neq 0$, resembling that in the KSL ground state [17,50,51]. For $h > 0$, there exists an additional bright spot at the Γ point, which represents the field-induced FM background in the system and can be observed clearly at intermediate temperature. Besides the two-point correlation, from Figs. 2(c) and 3 we observe that the $\mathbb{Z}_2$ flux operator W_P remains small at intermediate temperature, suggesting that the gauge fluxes can still flip freely under various fields in the fractional liquid regime.

D. Linear- T specific heat in the Majorana metal

In the Kitaev fractional liquid, the itinerant Majorana fermions couple to the thermally fluctuating fluxes and lead to a finite density of states emerging at the Fermi level, when averaged over the disordered flux sectors, and can be characterized by a linear $T</$

FIG. 5. (a) Uniform magnetic susceptibility χ under various fields h , with $1/\chi$ plotted in the inset. The Curie-Weiss fittings in the high- and intermediate- T regimes are indicated by the red and blue dashed lines, respectively. The high- T behavior can be fitted well by $1/(4T - J)$, and immediately below $T \simeq T_H$ there emerges a second Curie behavior. (b) Zero-field susceptibility of the anisotropic model with $K_x = K_y \neq K_z$, where the emergent Curie behaviors are observed in a wider temperature range. Also presented are the corresponding (c) $1/\chi$ fittings and (d) spin correlations on the d_z bond.

the intermediate- T specific-heat data and thus mask the expected universal linear- T behavior.

Therefore, an in-depth analysis of other readily accessible thermodynamic measurements in experiments is called for that reflects more of the intrinsic magnetic properties of the system, i.e., the susceptibility χ . From the analysis, we reveal that a universal Curie-law behavior emerges in the intermediate- T susceptibility that is robust against non-Kitaev terms and originates from the fractionalization of spins.

A. Magnetic susceptibility in the isotropic and anisotropic Kitaev models

As a direct characterization of unconventional paramagnetism of the fractional liquid, in Fig. 5(a) we show the susceptibility of the isotropic FM Kitaev model under various magnetic fields. For small fields $h \lesssim h_c$, the susceptibility data collapse into a universal Curie form $\chi = \frac{C}{T}$ in the fractional liquid regime, further consolidated by plotting $1/\chi$ vs T in the inset of Fig. 5(a). This unexpected universal Curie behavior extends across the fractional liquid regime, spanning a very wide temperature range of about one order of magnitude ($T \sim 0.03$ – 0.3), and constitutes an emergent phenomenon distinct from conventional paramagnetism at high temperature. The

latter corresponds to a Curie constant $C = S(S + 1)/3 = \frac{1}{4}$ related to the intrinsic moment of spin $S = \frac{1}{2}$, while the emergent Curie constant $C' \approx \frac{1}{3}$ [see Fig. 5(a)] in the fractional liquid regime, suggesting the presence of spin correlations that renormalize the effective magnetic moments.

Besides the isotropic case, we turn to the anisotropic Kitaev model and also find emergent Curie behaviors in Figs. 5(b) and 5(c) that can extend down to even lower temperatures. The Curie-Weiss fittings give $C' \approx 0.50$ for $K_{x,y} = -0.1$ and -0.25 , whose susceptibility is already very close to the ensemble of isolated FM spin pairs [red lines in Figs. 5(b) and 5(c)], and the Curie constant is close to twice a single spin $\frac{1}{2}$, i.e., $C = \frac{1}{2}$. The emergent Curie susceptibility there can extend down to temperatures significantly lower than T_L (of the isotropic case), due to the reduction of flux gap. Therefore, starting from the trivial $K_{x,y} = 0$ limit, as we turn on the FM coupling $K_{x,y}$ in the system, the Curie behavior remains, although the temperature range gets narrowed and the Curie constant becomes modified. For example, with $K_{x,y} = -0.5$, there also exists a Curie-law regime with C' reduced to 0.42, as indicated by the blue dashed lines in Figs. 5(b) and 5(c). This universal Curie scaling extends all the way to the isotropic point, implying that spins can be pair flipped nearly freely in the intermediate- T regime.

In fact, the emergent Curie behaviors can be observed as long as the spin correlations $\langle S_i^z S_j^z \rangle$ are mostly restricted within the d_z bond [see Figs. 2(a) and 2(b)] and in a quasi-plateau vs T at intermediate temperature shown in Fig. 5(d). We note that, according to the perturbation theory, the $\mathbb{Z}_2$ fluxes remain approximately conserved and spin correlations beyond the d_z bonds only appear at the h_c^2 order. At the same time, $\mathbb{Z}_2$ fluxes should be flippable in a nearly free fashion, which can be observed as $W_P \ll 1$ in Fig. 2(c). These conditions are satisfied even when perturbative external fields are applied, which gives rise to the intermediate- T Curie law and will be elaborated in Sec. IV B.

B. Fluctuating fluxes and diverging susceptibility

The diverging susceptibility at intermediate temperature reflects the strongly fluctuating fluxes in the fractional liquid below T_H , and deviation occurs when the flux starts to freeze-out at about T_L . To see this, we exploit the Kubo formula of susceptibility (see Appendix E) and assume that $M = 0$ in the fractional liquid for the sake of discussion,

$$\chi = \sum_j \int_0^\beta \langle S_{i_0}^z(\tau) S_j^z \rangle_\beta d\tau, \quad (3)$$

where $S_{i_0}^z(\tau) = e^{\tau H} S_{i_0}^z e^{-\tau H}$ and j runs over NN sites of i_0 by the d_z bond (as well as i_0 itself) in the fractional liquid regime due to the extremely-short-range correlations. As illustrated in Fig. 1, the spin operator S_j^z flips a pair of fluxes on two neighboring hexagons (in purple) and changes the flux configuration of energy eigenstate. After evolving along the imaginary time of τ , $S_{i_0}^z$ can flip the $\mathbb{Z}_2$ fluxes back and thus restore the flux configurations, which results in a nonzero correlation.

We denote the energy eigenstates by $|E_{\{W_P\}}^n\rangle$, where $\{W_P\}$ represents the flux configurations and n labels the individual

states within the $\{W_P\}$ sector. By inserting the orthonormal basis, we arrive at the Lehmann spectral representation

$$\langle S_{i_0}^z(\tau) S_j^z \rangle_\beta = \sum_{\{W_P\}, n} \sum_{n'} e^{-\beta E_{\{W_P\}}^n} e^{-\tau \Delta_{n, \{W_P\}; n', \{W_P'\}}} \times \langle E_{\{W_P\}}^n | S_{i_0}^z | E_{\{W_P'\}}^{n'} \rangle \langle E_{\{W_P'\}}^{n'} | S_j^z | E_{\{W_P\}}^n \rangle, \quad (4)$$

where $|E_{\{W_P'\}}^{n'}\rangle$ represents a state n' in the flux-flipped sector $\{W_P'\}$. Since the itinerant Majorana fermions are weakly coupled to the flux at intermediate temperature [26] and thus the excitation energy is close to the flux gap, $\Delta_{n, \{W_P\}; n', \{W_P'\}} = |E_{\{W_P'\}}^{n'} - E_{\{W_P\}}^n| \sim T_L \ll T \equiv 1/\beta$. Therefore, the decay factor $e^{-\tau \Delta_{n, \{W_P\}; n', \{W_P'\}}} \simeq 1$, which is not surprising as different flux sectors are nearly degenerate in the fractional liquid regime, as evidenced by $W_P \ll 1$ [see, e.g., Fig. 2(c)].

Consequently, $\langle S_{i_0}^z(\tau) S_j^z \rangle_\beta$ is virtually τ independent at intermediate temperature and can be approximated as $\langle S_{i_0}^z S_j^z \rangle_\beta$ in Eq. (3), thus recovering the conventional susceptibility formula

$$\chi = \frac{1}{T} \sum_j \langle S_{i_0}^z S_j^z \rangle_\beta, \quad (5)$$

which holds only for $T \gtrsim T_L$. As observed in Figs. 2(a) and 5(d), the correlation $\langle S_{i_0}^z S_j^z \rangle_\beta$ exhibits a quasiplateau vs temperatures between T_L and T_H , resulting in a $1/T$ -divergent susceptibility that accounts for the emergent Curie law in the fractional liquid regime. On the other hand, in the KSL regime at $T \lesssim T_L$, although the correlation length remains extremely short there, the (flux) excitation energy $\Delta_{n, \{W_P\}; n', \{W_P'\}} \sim T_L \gtrsim T$ leads to an exponentially suppressive factor in τ and thus a nondivergent susceptibility χ below T_L in Fig. 5.

Above we discussed the emergent susceptibility scaling in the FM Kitaev model and below we briefly discuss the case of the AF model, with the details deferred to Appendix F. For the AF Kitaev model, the spin correlations are also extremely short ranged and their absolute values are identical to those of the FM case, but with opposite sign (AF correlation). Therefore, the emergent Curie law is absent in the uniform susceptibility χ of the AF Kitaev model, while it appears instead in the stagger susceptibility. This can be understood intuitively as follows: One can flip simultaneously the spins on one of the two sublattices on the honeycomb lattice, turning the AF Kitaev model into an FM one, and the staggered susceptibility χ_s corresponds to the uniform χ exhibiting universal scaling at intermediate temperature. Such an interesting correspondence between the FM Kitaev model under uniform field and the AF model under staggered field has been noticed in other studies under different circumstances [52]. Our results indicate a sensitive way to determine the sign of Kitaev coupling via analyzing the susceptibility measurements.

V. THERMAL TENSOR NETWORK SIMULATIONS OF THE EXTENDED KITAEV MODELS

Now we switch to the XTRG simulations of the extended Kitaev models with the Heisenberg and Γ interactions, which hinders other quantum many-body methods like QMC by the sign problem. The Heisenberg coupling is proposed to

be relevant in the Kitaev material Na_2IrO_3 [13,15] and the Kitaev-Heisenberg model has thus been extensively studied for both the ground-state [13,16,48,50,53] and finite- T properties [54,55]. On the other hand, in $\alpha\text{-RuCl}_3$ there exists a symmetric off-diagonal exchange, the so-called Γ term [41,49,51,56], which turns out to be important non-Kitaev interactions. In this section we explore the Kitaev-Heisenberg (on YC $4 \times 6 \times 2$ cylinders) and Kitaev- Γ (YC $4 \times 4 \times 2$) models [see Eq. (1)] at finite temperature, from which we see that although the low-temperature fate of the Kitaev model may be drastically affected by the Heisenberg and Γ terms, the behavior at high and intermediate temperatures, especially in the Kitaev fractional liquid regime, remains qualitatively unchanged.

A. Fractional thermal entropy and specific-heat behaviors

We start with the thermal entropy and specific-heat calculations of the Kitaev-Heisenberg and Kitaev- Γ models and show the results in Figs. 6 and 7. Between the two entropy-release steps [Figs. 7(c) and 7(d)], there exists an extended regime with thermal entropy $S \simeq \frac{1}{2} \ln 2$ [Figs. 6(a) and 6(b)] for both models, which indicates the existence of Kitaev fractional liquid at intermediate temperature. The $\frac{1}{2}$ -entropy quasiplateau remains evident until Heisenberg coupling $J \lesssim 0.16$ and for a Γ interaction of $\Gamma \lesssim 0.06$. Beyond this Γ or J range, a shoulderlike feature with slow entropy release can still be recognized in both models. In Figs. 6(a) and 6(b) the crossover temperature scales T_H and T_L (T_L') can be determined from the isentropic lines of $S/\ln 2 = 0.75$ and 0.25 , which coincide with the higher and lower crossover temperatures in the double-peaked specific-heat curves [see Figs. 7(a) and 7(b) and also Appendix G].

In Figs. 7(a) and 7(b) we show that the linear- T behavior of specific heat also survives in the presence of a finite J or Γ interaction, which also can be recognized from the C_V/T plots in the insets. This universal specific-heat behavior suggests the emergence of Majorana-metal states in the fractional liquid regime. Nevertheless, similar to what was observed in Fig. 4, the J and Γ interactions can shift the flux excitations as well as other low-energy spin fluctuation modes towards higher temperatures and thus may mask the linear- T C_V , leaving eventually only a shoulderlike remnant in the insets of Figs. 7(a) and 7(b).

The finite- T properties can sensitively reflect the quantum phase transition in the ground state. In Fig. 6(a), T_L (T_L') is non-monotonic vs J : For $J \lesssim J_c \simeq 0.12$, T_L represents a crossover from the KSL to the fractional liquid, while for $J > J_c$, the stripe antiferromagnetic (SAF) order sets in below T_L' , where correlations along d_x , d_{x+y} , etc, also increase rapidly as shown in Fig. 6(c). Therefore, we infer from the finite- T results that the Kitaev-Heisenberg model undergoes a transition from the KSL to SAF phase at around $J_c \simeq 0.12$, consistent with previous ground-state studies [13,53,56].

B. Bond-directional spin correlation and stripy structure factor

From the spin-correlation data shown in Figs. 6(c) and 6(d), we emphasize that the two models share common features in the Kitaev fractional liquid regime. First, the spin

FIG. 6. Phase diagram of (a) the Kitaev-Heisenberg and (b) the Kitaev- Γ model obtained by entropy $S/\ln 2$, where the white diamonds indicate the crossover temperature determined from the double-peaked specific heat. The insets show the $T = 0.07$ static structure factors of (a) $J = 0.12$ and (b) $\Gamma = 0.06$ in the extended Brillouin zone. The XTRG results of the correlations $\langle S_i^z S_j^z \rangle$ along four NN bonds with (c) $J = 0.08$ and 0.16 and (d) $\Gamma = 0.03$ and 0.07 all converge at the lowest temperature to the GS values calculated by the DMRG method. For the Kitaev-Heisenberg case with $J = 0.08$ and the Kitaev- Γ case, d_z correlations arise at around T_H and stay predominate across the whole temperature range; for the $J = 0.16$ case, all correlations increase abruptly at the low-temperature scale, indicating the onset of SAF order. (e) and (f) Flux W_p with various J or Γ and (g) and (h) magnetic susceptibility χ and the Curie-Weiss fittings for two models. The χ data are computed under a small uniform field $h = 0.002$.

correlation $\langle S_i^z S_j^z \rangle$ remains extremely short ranged and mostly present on the d_z bonds. This is consistent with a perturbation-theory analysis: Except on the d_z bonds, spin correlations are suppressed because $\mathbb{Z}_2$ fluxes are approximately conserved. Furthermore, such correlations appear at the first order in J but only at the second order in Γ , consistent with the fact that J has a larger effect on spin correlations than Γ , as shown in Figs. 6(c) and 6(d). This prominent spin-correlation feature consequently leads to the stripy structure factor in the insets of Figs. 6(a) and 6(b).

Under the Heisenberg interactions, we observe in the inset of Fig. 6(a) a clear structure peak at the X point, which indicates the enhanced short-range SAF correlation, with the classical spin pattern shown on the right-hand side. Besides the SAF peak, we notice that there exists a stripy structure background which is related to the predominant bond-directional NN correlations at intermediate temperature. On the other hand, in Fig. 6(b), for the Kitaev- Γ model, the low-temperature scale T_L changes only slightly. The KSL-like stripy spin structure can be observed with no clear signature of spin ordering, which, together with the nonzero fluxes $W_p \neq 0$, at low temperature in Fig. 6(f), suggests that the low- T quantum state remains in the asymptotic KSL phase at least for $\Gamma \lesssim 0.1$, in agreement with recent ground-state studies of the Kitaev- Γ model [49,56].

C. Universal susceptibility scaling

At intermediate temperature, we again observe bond-directional spin correlations that are mostly restricted within the NN sites [see Figs. 6(c) and 6(d) and discussion above], as well as the strongly fluctuating flux [Figs. 6(e) and 6(f)] under an extended range of J or Γ interactions. Following the discussion in the pure Kitaev model, we expect an emergent Curie law of susceptibility in the fractional liquid, and this is indeed observed in Figs. 6(g) and 6(h). Although the temperature window gradually narrows as J or Γ increases, the universal susceptibility scaling can be very clearly identified across the intermediate- T regime, regardless of whether the ground state is a KSL or semiclassically ordered.

Nevertheless, there are still some differences in the susceptibility scalings between the Kitaev-Heisenberg and Kitaev- Γ models. At the temperature above T_L , the susceptibility curves in the Kitaev- Γ model collapse into the same universal behavior, with $C' \approx \frac{1}{3}$ (thus $C'/C \approx 1.33$, the same as in the pure Kitaev model) and with the same small θ' . This is in contrast to the case of the Kitaev-Heisenberg model, where although $C' \approx \frac{1}{3}$ remains, the Curie-Weiss temperature θ' grows as J increases, which suggests that the fluxes are no longer free but are coupled with each other since the Heisenberg coupling J introduces longer-range correlations to the system [Fig. 6(c)]. Accordingly, in Fig. 6(e), the gauge flux for $J \lesssim J_c$ behaves very similarly to that of the pure Kitaev model, i.e., W_p freezes

FIG. 7. Specific heat C_V vs T for the (a) Kitaev-Heisenberg and (b) Kitaev- Γ models with various coupling strengths. The black dashed lines indicate the linear T dependence of C_V at intermediate temperature and the corresponding C_V/T data are shown in the insets. With Heisenberg interactions in (a), the C_V/T plateau is apparent for $J \lesssim 0.08$ and leaves a shoulderlike structure until $J_c \approx 0.12$; for the Γ terms this plateau and its remnant signature are clear for $\Gamma \lesssim 0.04$. (c) and (d) Thermal entropy results of both models. Around the stable high- T scale T_H , the entropy curves almost collapse for various J and Γ strengths and consequently leave a quasiplateau with $S \approx \frac{1}{2} \ln 2$.

at around T_L , while it gets suppressed at low temperature for $J > J_c$.

Overall, we observe that the emergent Curie law of susceptibility scaling appears to be robust against the couplings beyond the pure Kitaev model at intermediate temperature, which is found to be suppressed only when the finite flux gap or the longer-range correlation induced by the non-Kitaev terms takes effect at some low temperature. Therefore, such universal susceptibility scaling can serve as a useful tool probing the free $\mathbb{Z}_2$ flux and thus fractionalization in the Kitaev materials.

VI. DIAGNOSIS OF THE KITAEV FRACTIONAL LIQUID

A. Kitaev paramagnetism in the Kitaev materials

Thermodynamic features, including the double-peaked specific-heat curve, the linear- T behavior, and the fractional thermal entropy S integrated from C_V/T , have been exploited in detecting finite- T fractionalization in the Kitaev materials [18,24,57–59]. However, the magnetic response to external fields, the susceptibility χ , has been much less analyzed, except for the high- T behaviors in the Kitaev materials. With insight from the emergent intermediate- T Curie susceptibility from the above model studies, we revisit the experimental χ data of two prominent Kitaev materials, i.e., Na_2IrO_3 [14] and $\alpha\text{-RuCl}_3$ [57], which are believed to realize the Kitaev model under the Heisenberg and/or Γ interactions.

In Fig. 8 we analyze the in-plane and out-of-plane susceptibility data of both materials, where the emergent

FIG. 8. Experimental Na_2IrO_3 [14] and $\alpha\text{-RuCl}_3$ susceptibility data [57] and their Curie-Weiss fittings at both (a) high and (b) intermediate temperatures under fields $H \parallel ab$ (orange circles) and $H \parallel c$ (blue squares), with inverse susceptibility plotted in the insets. (c) Fitted Curie constants (C for high and C' for intermediate T) and the Curie temperatures (corresponding θ and θ'), for both the c and ab directions, are listed in the table.

Curie behaviors can be observed right below certain high-temperature scales, i.e., $T_H^{\text{Ir}} \approx 120$ K and $T_H^{\text{Ru}} \approx 110$ K for Na_2IrO_3 and $\alpha\text{-RuCl}_3$, respectively. Here T_H^{Ir} and T_H^{Ru} are in agreement with the higher crossover temperatures in the magnetic specific-heat curves of two materials. For example, in Na_2IrO_3 the higher specific-heat peak is located at around 110–120 K [17,18], which is in very good agreement with the T_H^{Ir} in Fig. 8(a). Similarly, the high- T entropy is rapidly released at around 100–110 K in $\alpha\text{-RuCl}_3$ [24,57,59], also coinciding with T_H^{Ru} in Fig. 8(b).

Through Curie-Weiss fittings of the intermediate- T susceptibility results in Fig. 8, we find the ratios between intermediate- and high- T Curie constants $C'/C \sim 1.3$ in both materials (and either within the ab plane or parallel to the c axis), suggesting the existence of the Kitaev fractional liquid in these two Kitaev materials. The emergent diverging susceptibility at intermediate temperature is very sensitive to the sign of the Kitaev interaction K , and for the AF case ($K > 0$) a convex uniform susceptibility curve is exhibited without any emergent Curie behavior at intermediate temperature (Appendix F). Therefore, the analysis in Fig. 8 suggests the FM Kitaev interactions in both compounds Na_2IrO_3 and $\alpha\text{-RuCl}_3$, in agreement with previous theoretical [10,38,60,61] and experimental [40] estimates.

Meanwhile, in Fig. 8 we observe that the intermediate Curie-Weiss behaviors deviate at the lower temperature $T_L^{\text{Ir}} \approx 50$ –55 K and $T_L^{\text{Ru}} \approx 60$ –65 K. Therefore, in these two materials there are fractional liquid regimes stretching appreciable temperature ranges as wide as 65–70 K (Na_2IrO_3) and 45–50 K ($\alpha\text{-RuCl}_3$), respectively. In addition, by comparing Fig. 8(a) to Fig. 6(g), we find a wider intermediate- T range for the emergent Curie-Weiss in Na_2IrO_3 . This resembles that of the Kitaev-Heisenberg model, where universal scaling

FIG. 9. Static spin-structure factors $S^\gamma(q)$ calculated from the equal-time spin-correlation functions [see Eq. (2)] at an intermediate temperature $T \simeq 0.15$: (a)–(d) pure Kitaev model results of three spin-structure factors $S^{x,y,z}(q)$ as well as their sum $S(q)$ and (e)–(h) results of the Kitaev-Heisenberg model with $J = 0.14$.

deviates at around the susceptibility peak temperature. This observation suggests that indeed the Heisenberg-type interaction can play an important role in this material [13]. On the other hand, in Fig. 6(g) the Γ term deflects the Curie-Weiss behavior way before the susceptibility peaks; this bears a resemblance to the case of α -RuCl₃ shown in Fig. 8(b), supporting thus the proposal that the Γ term (together with other interactions) is particularly relevant in this material [42,62].

Overall, through analysis of the magnetic susceptibility data of Na₂IrO₃ and α -RuCl₃, we find a universal susceptibility scaling that may be ubiquitous in other members of the intriguing family of Kitaev materials and can serve as a sensitive thermodynamic signature of the Kitaev fractional liquid. In addition, the susceptibility scaling indicates the existence of ferromagnetic Kitaev interactions, together with other non-Kitaev couplings, in the two prominent Kitaev materials.

B. Spin-correlation diagnostics of the Kitaev fractional liquid

The short-range correlated Kitaev fractional liquid leaves unique features in the spin correlations and thus structure factors. At intermediate temperature, the nonzero correlation $\langle S_i^\gamma S_j^\gamma \rangle$ is restricted within the NN γ bond, with the rest of the correlations negligibly small even under magnetic fields and non-Kitaev interactions, as shown in Figs. 2(a), 2(b), 6(c), and 6(d). Correspondingly, in Figs. 3(c), 6(a), and 6(b) we see that this peculiar bond directional correlation pattern results in a stripylike background in the z -direction spin-structure factors $S^z(q)$.

In fact, this stripy spin structure also appears in the structure factors $S^\gamma(q)$ with $\gamma = x, y$, and the direction of the stripe coincides with the associated spin components γ . As shown in Figs. 9(a) and 9(e), the $S^x(q)$ stripe extends from the top left to the bottom right, which rotates counterclockwise by 120° when switched to $S^y(q)$ [Figs. 9(b) and 9(f)], followed by another 120° rotation to $S^z(q)$ in Figs. 9(c) and 9(g). Note that the spin-structure factor results in Fig. 9 are

evaluated at an intermediate temperature $T \simeq 0.15$ for the pure Kitaev model [Figs. 9(a)–9(d)] and the Kitaev-Heisenberg model [Figs. 9(e)–9(h)]. In the latter Kitaev-Heisenberg case with $J = 0.14$, a structure factor peak emerges, representing the semiclassical SAF correlation, which also rotates as the spin component switches. Such a rotation of the short-range correlation peak was previously observed in the spin-resolved magnetic x-ray measurements right above the ordering transition temperature ($T \simeq 17$ K) [17], which provides direct evidence of the dominant bond-directional interactions in the Kitaev material Na₂IrO₃. Therefore, our findings in Figs. 9(e)–9(g) confirm that this experimental observation is indeed related to the presence of Kitaev interactions in the material, although the SAF order involved here is different from the zigzag type in Na₂IrO₃.

Moreover, our results in Figs. 9(d) and 9(h) also shed light on the renowned starlike spin-structure factor observed in inelastic neutron scattering measurement of α -RuCl₃ [23,24]. From the full structure factor $S(q) = \sum_\gamma S^\gamma(q)$, we observe in the pure Kitaev model that the superposition of the three stripes leads to an approximately rotational symmetric shape in Fig. 9(d). By adding bright spots representing short-range SAF correlations on top of that, we instead see a sixfold symmetric $S(q)$ in Fig. 9(h). This clearly suggests that the star-shape structure factor (with sixfold symmetry) observed in inelastic neutron scatterings on α -RuCl₃ can be ascribed to the coexistence of the Kitaev bond-directional correlation and short-range zigzag order. As the temperature/energy scale gradually increases, it approaches asymptotically the rotationally symmetric spin structure in Fig. 9(d), which has also been observed in experiments.

In general, we propose that the bond-directional spin correlation and related intermediate- T stripy feature provide a useful tool for determining the Kitaev materials. Specifically, diffuse x-ray (for Na₂IrO₃) or spin-resolved neutron (for α -RuCl₃) measurement at intermediate temperature, say, $T = 50$ –100 K, can be exploited to observe the stripy spin

structures $S^\gamma(q)$ that serves as direct evidence of the Kitaev fractional liquid.

VII. DISCUSSION AND OUTLOOK

In conventional spin models and magnetic materials, a “crystallization” of spins occurs from the high- T gaslike paramagnetic to the low- T phase with long-range order. Interestingly, in the Kitaev model and related Kitaev materials, an alternative scenario is proposed where a fractionalization of spins takes place upon cooling. This gives rise to an intermediate- T Kitaev fractional liquid and eventually to the low- T quantum spin liquid phase with long-range entanglement. Although the finite-temperature fractionalization of quantum spins has been established [27] in the pure Kitaev model, its relevance in real Kitaev materials remains an open question. The main challenge in the way is how to perform efficient finite-temperature simulations of the extended Kitaev model with realistic interactions.

Here we clear the obstacle with the precise XTRG method and investigate the extended Kitaev model with external magnetic fields and Heisenberg and Γ terms. A Kitaev fractional liquid regime is found at intermediate temperature and is robust to non-Kitaev interactions, regardless of whether the ground state is in a spin liquid phase or not. The exotic finite- T quantum states can be characterized by universal thermodynamics, including the fractional thermal entropy, the linear- T specific heat, and in particular an emergent Curie law in magnetic susceptibility. Remarkably, this universal Curie susceptibility that reflects the thermally free gauge fluxes, together with the previously recognized metallic specific-heat behaviors of itinerant Majorana fermions, each revealing one aspect of the spin fractionalization, constitutes a comprehensive thermodynamics characterization of the exotic Kitaev fractional liquid. In light of this and from analyzing the experimental susceptibility data, we find evidence of the Kitaev fractional liquid in Na_2IrO_3 and $\alpha\text{-RuCl}_3$, which supports the ferromagnetic Kitaev interactions in the two materials and their proximity to quantum spin liquids.

As a matter of fact, the diverging susceptibility is a robust thermodynamic feature related also to the peculiar spin correlations in the fractional liquid regime. They are extremely short ranged and bond directional, thus corresponding to the stripy spin-structure factor that rotates with the spin components. Therefore, besides the magnetic susceptibility measurement, our findings also encourage further material exploration of the fractionalization using spin-resolving neutron and resonant x-ray scatterings. Finally, our present work paves the way to the precise determination of microscopic model parameters in the Kitaev materials by analyzing thermodynamic data, which we leave for future study.

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APPENDIX A: EXPONENTIAL TENSOR RENORMALIZATION GROUP METHOD APPLIED TO THE EXTENDED KITAEV MODEL

The exponential tensor renormalization group method reaches low- T thermal states exponentially fast, which has been shown to be highly efficient in both critical quantum chains and various 2D lattice systems [34–36] and is exploited to study the Kitaev model of interest. To start the XTRG procedure, we need to invoke a quasi-1D mapping of the lattice model and encode the long-range interaction information in a compact matrix product operator form. For the pure Kitaev model on the YC4 geometry, the MPO has a bond dimension of $D_H = 7$, which increases to $D_H = 14$ for the Kitaev-Heisenberg and Kitaev- Γ Hamiltonians.

Given the compact MPO representation of the Hamiltonian H , we can represent $\rho(\tau)$, initiated at a very small initial $\tau \sim 10^{-3}$ – 10^{-4} , also as an MPO conveniently and accurately (up to machine precision) via the series expansion

$$\rho(\tau) = e^{-\tau\hat{H}} = \sum_{n=0}^{N_c} \frac{(-\tau)^n}{n!} H^n. \quad (\text{A1})$$

Through this expansion, H^n can also be expressed as an MPO using tensor compression techniques [63], followed by a sum-and-compression procedure that finally leads to the compact MPO form of $\rho(\tau)$.

Following the XTRG idea of the exponential cooling procedure, we square the density matrix and obtain at the n th step

$$\rho(2^{n+1}\tau) = \rho(2^n\tau)\rho(2^n\tau), \quad (\text{A2})$$

with which we can compute the free energy at $\beta \equiv 2^{n+2}\tau$ via the thermofield double formalism

$$f(\beta) = -\frac{1}{\beta} \ln \text{Tr}[\rho(2^{n+1}\tau)\rho(2^{n+1}\tau)] \quad (\text{A3})$$

by a bilayer tensor trace [44]. Given the free energy, other thermodynamic quantities including the specific heat, thermal entropy, magnetic susceptibility, etc., can also be computed.

We first benchmark the XTRG results with ED on a 12-site cluster (with the same width and boundary condition as YC4 geometry but with only three sites along the zigzag edge). In Fig. 10(a) the relative error of the free energy $\delta f/f$ decreases as the retained bond dimension D increases, reaching a very high accuracy with $D = 600$. On top of that, we show the XTRG results of specific heat computed with $D = 600$, of pure Kitaev, Kitaev-Heisenberg, and Kitaev- Γ models, and show them in Fig. 10(b), where we also find excellent agreement with ED results.

As shown in Figs. 10(c)–10(f), we compute the thermal entropy, flux expectation, specific heat, spin correlations, and magnetic susceptibility and compare our XTRG results with the QMC data taken from Refs. [27,29]. Despite the very different system sizes, i.e., the YC4 cylinder vs the $L = 20$ torus, excellent agreement between two methods can be observed at high and intermediate temperatures, covering the fractional

FIG. 10. (a) Relative errors $\delta f/f$ between the ED and XTRG results of a isotropic pure Kitaev model on the $N = 12$ (YC4) cluster, with retained bond dimensions $D = 200, 400$ and 600 . (b) Corresponding specific-heat results of the pure Kitaev model (blue), the Kitaev-Heisenberg model with $J = 0.2$ (red), and the Kitaev- Γ model with $\Gamma = 1$ (yellow). Comparisons between the large-scale XTRG and QMC simulations [27] are presented for (c) the thermal entropy $S/\ln 2$ (orange) and flux W_P (blue), (d) specific heat C_V , and (e) spin correlations $\langle S_i^z S_j^z \rangle$. We also compare the XTRG susceptibility results to the QMC data on the $L = 20$ torus [29], where the high- and intermediate- T fittings are also performed on the latter (dashed lines). We show in (c)–(f) both the YC $4 \times 6 \times 2$ results (solid curves) and the YC4 data (dotted curves) obtained by subtracting length $L = 6$ by $L = 4$ results. The XTRG results of susceptibility are computed under the $[001]$ field, which agree excellently with the QMC results in (f) above the low temperature T_L , although the latter is along the $[111]$ direction.

liquid regime. The modest deviation of the XTRG results from QMC data near the lower-temperature scale T_L can be ascribed to the cylindrical boundary condition and the finite width, which nevertheless does not lead to any qualitative difference for our discussion.

Moreover, we have also analyzed the large-scale QMC susceptibility data in Fig. 10(f), and from the fitting we find again an emergent Curie behavior clearly at intermediate temperature. The Curie constant $C' \simeq \frac{1}{3}$ is also very close to the YC4 XTRG results in Fig. 5(a).

APPENDIX B: SPIN CORRELATIONS IN THE FRACTIONAL LIQUID

The real-space spin correlations of the extended Kitaev models are computed at a high ($T \simeq 1$) and an intermediate temperature ($T \simeq 0.1$). In Figs. 11(a) and 11(b) we provide $\langle S_i^z S_j^z \rangle$ along two different (zigzag and armchair) paths, under the influence of external fields, from which we find that the correlations are bond directional and extremely short

FIG. 11. High- and intermediate- T spin correlations of the extended Kitaev model, along the (a), (c), and (e) zigzag and (b), (d), and (f) armchair paths, with the reference site marked in red and the hexagon edge length as the unit of distance. Note that in (a) and (b) we have subtracted the (field-induced) mean magnetization squared $\langle M \rangle^2$ from the spin correlations.

ranged. To be specific, $\langle S_i^z S_j^z \rangle$ vanishes along the zigzag path in Fig. 11(a), since there are only x and y bonds there. On the other hand, in Fig. 11(b) the armchair correlations decay instantaneously for distances exceeding one and the nearest z -bond correlation is enhanced as temperature decreases from $T \simeq 1$ to $T \simeq 0.1$. For the Kitaev-Heisenberg and Kitaev- Γ models in Figs. 11(c)–11(f), we observe real-space correlation features very similar to those in Figs. 11(a) and 11(b), except for the curves with $J = 0.16$ and $T \simeq 0.1$ in Fig. 11(d). There the system enters the SAF-ordered phase, where the zigzag correlations develops a small value and the signs alternate following the SAF pattern as shown in the inset of Fig. 6(a).

From the results in Fig. 11, we find spatially anisotropic and extremely-short-ranged spin correlations at intermediate temperature. This, together with the distinct stripy spin-structure factors in the main text, provide the signatures of fraction liquid.

APPENDIX C: MATRIX PRODUCT OPERATOR ENTANGLEMENT

In the MPO form of the density matrix $\rho(\beta \equiv 1/T)$, we compute the bipartite entanglement entropy $S_E(\beta) = -\text{Tr}(\tilde{\rho}_A \ln \tilde{\rho}_A)$, where $\tilde{\rho}_A$ is the reduced matrix of subsystem A (chosen as half the system) in the superstate $|\rho(\beta)\rangle$ purified from the thermal density matrix $\rho(\beta)$. In the XTRG method, $S_E(\beta)$ can be used to measure the required computational resource, since the bond dimension scales as $D \sim e^{S_E(\beta)}$ for an accurate simulation down to a temperature $T \equiv 1/\beta$. For

a gapped system, S_E typically saturated as T is below the excitation gap Δ , and thus one can in principle simulate the low- T properties even down to the ground state with a sufficiently large, but finite, bond dimension D . The logarithmic entanglement scaling $S_E(\beta) \sim \eta \ln \beta$ requires $D \propto \beta^\eta$, with η an algebraic exponent. The logarithmic entanglement scaling at low temperature usually corresponds to gapless excitations. Some typical examples include the low- T thermal states near the 1+1D conformal critical point and in the 2D Heisenberg model with Goldstone modes [34,36]. In the former case, the exponent has been found to be universal, $\eta = c/3$, with c the central charge of the conformal field theory [34].

In Fig. 12(a) we show the bipartite entanglement entropies S_E across the center of the system under various magnetic fields h , which exhibits a logarithmic scaling as $S_E \sim \frac{2}{3} \ln \beta$ in the intermediate- T regime, as indicated by the black dashed line. In Figs. 12(b) and 12(c) we have also computed the MPO entanglement for the Kitaev-Heisenberg and Kitaev- Γ models, where similar logarithmic behaviors are also observed in the fractional liquid regime. As the coupling strength increases, for all three cases in Figs. 12(a)–(c), the entanglement S_E deviates from the logarithmic behavior, exhibits a shoulder/peak, and eventually converges to a constant at sufficiently low temperature.

The exponent $\eta \approx 2/3$ suggests that the required computational resource for simulating the Kitaev models at intermediate temperature is similar to a conformal critical point with central charge $c = 2$. In addition, in Fig. 12(a) the maximal value of S_E appears at the lowest temperature and is less than 3.5, which reveals the accessibility of the YC $4 \times 6 \times 2$ systems by retaining not too many bond states. In fact, we have plotted in Fig. 12(a) the $D = 400$ and 600 results, where a very good convergence can be seen.

APPENDIX D: MAGNETIC SUSCEPTIBILITY ALONG VARIOUS FIELD DIRECTIONS

In Fig. 13 we show the χ data along various field directions, as well as the case $h \parallel [001]$ considered in the main text. We label the field direction $l\hat{h}_x + m\hat{h}_y + n\hat{h}_z$ by $[l, m, n]$, where $\hat{h}_x$, $\hat{h}_y$, and $\hat{h}_z$ are three cubic axes in the spin space; therefore, the Zeeman term reads

$$H_f = - \sum_i \frac{h}{\sqrt{l^2 + m^2 + n^2}} (lS_i^x + mS_i^y + nS_i^z). \quad (\text{D1})$$

First, we apply an in-plane field $h \parallel [-101]$, whose direction is shown in Fig. 13(a), and compute the susceptibility. The results are shown in Figs. 13(b) and 13(c), where an intermediate Curie behavior can be clearly identified. For the out-of-plane field h perpendicular to the ab plane, i.e., $h \parallel [111]$, a Curie susceptibility also emerges [see Fig. 10(f)], as already discussed in Appendix A. As for more field directions, we perform ED calculations on systems of very limited size, $N = 12$ clusters, and show the results in Fig. 13(d). A diverging susceptibility behavior near the high- T scale T_H is also evident, confirming that the emergent Curie-law susceptibility is robust and ubiquitous in the Kitaev model. This universal susceptibility scaling can be observed under various fields of different directions, on different lattice sizes/geometries,

FIG. 12. Bipartite entanglement S_E cut at the center of the MPO representation of density matrices for (a) the Kitaev model with fields, (b) the Kitaev-Heisenberg model, and (c) the Kitaev- Γ model. At intermediate temperature, S_E roughly scales as $\sim 2/3 \ln \beta$, which characterizes the required computational resource of the simulations.

and by different methods including the ED, QMC, and XTRG approaches.

APPENDIX E: KUBO FORMULA OF MAGNETIC SUSCEPTIBILITY

Here we derive the Kubo formula of the magnetic susceptibility, in terms of imaginary-time dynamical correlations. Suppose the magnetization of the system is uniform and the magnetic moment per spin, under a perturbative magnetic field h along the z axis, is the expectation value of S^z on an

FIG. 13. (a) Illustration of the magnetic fields h along $[001]$ and $[-101]$ (parallel to the honeycomb ab plane). Also shown are (b) the magnetic susceptibility χ and (c) its inverse $1/\chi$, calculated with a small field $h = 0.002$. (d) Susceptibility results along various field directions, obtained by ED calculations on the $N = 12$ cluster.

arbitrary site i_0 ,

$$M = \langle S_{i_0}^z \rangle_\beta = \frac{1}{Z(\beta)} \text{Tr} [S_{i_0}^z e^{-\beta H}], \quad (\text{E1})$$

where $H(K, h) = H_0(K) - h S_{\text{tot}}^z$, $H_0(K)$ is the pure Kitaev Hamiltonian, and $S_{\text{tot}}^z = \sum_j S_j^z$ is the total magnetization operator. The magnetic susceptibility $\chi = \partial M / \partial h$ can be obtained by taking the derivative of Eq. (E1). Note that since $[H, S_{\text{tot}}^z] \neq 0$, the derivative of the exponent $-\beta \partial H / \partial h = -\beta S_{\text{tot}}^z$ cannot be taken straightforwardly from the exponential, i.e.,

$$\frac{\partial}{\partial h} e^{-\beta H} \neq -\beta \frac{\partial H}{\partial h} e^{-\beta H}. \quad (\text{E2})$$

Instead, for an operator $O(u)$ as a function of u ,

$$\frac{\partial}{\partial u} e^O = \int_0^1 e^{\lambda O} \frac{\partial O}{\partial u} e^{(1-\lambda)O} d\lambda. \quad (\text{E3})$$

This can be proved via the decomposition $e^O = (e^{O/N})^N$, and for sufficiently large N it satisfies $\partial e^{O/N} / \partial u = e^{O/N} (\partial O / \partial u) / N$. Therefore,

$$\begin{aligned} \frac{\partial}{\partial u} e^O &= \sum_{n=1}^N (e^{O/N})^{(n-1)} \frac{\partial e^{O/N}}{\partial u} (e^{O/N})^{(N-n)} \\ &\simeq \frac{1}{N} \sum_{n=1}^N e^{(n/N)O} \frac{\partial O}{\partial u} e^{(1-n/N)O}. \end{aligned} \quad (\text{E4})$$

Taking the limit $N \rightarrow \infty$, we arrive at Eq. (E3). With this, we can express the susceptibility as

$$\begin{aligned} \chi &= \frac{\beta}{Z(\beta)} \int_0^1 \text{Tr} [S_{i_0}^z e^{-\lambda \beta H} S_{\text{tot}}^z e^{-(1-\lambda)\beta H}] d\lambda - \beta \langle S_{i_0}^z \rangle_\beta \langle S_{\text{tot}}^z \rangle_\beta \\ &= \frac{1}{Z(\beta)} \int_0^\beta \text{Tr} [S_{i_0}^z e^{-\tau H} S_{\text{tot}}^z e^{(\tau-\beta)H}] d\tau - \beta \langle S_{i_0}^z \rangle_\beta \langle S_{\text{tot}}^z \rangle_\beta \end{aligned}$$

$$\begin{aligned} &= \frac{1}{Z(\beta)} \int_0^\beta \text{Tr} [e^{\tau H} S_{i_0}^z e^{-\tau H} S_{\text{tot}}^z e^{-\beta H}] d\tau - \beta \langle S_{i_0}^z \rangle_\beta \langle S_{\text{tot}}^z \rangle_\beta \\ &= \sum_j \int_0^\beta \langle S_{i_0}^z(\tau) S_j^z \rangle_\beta d\tau - \beta \langle S_{i_0}^z \rangle_\beta \langle S_{\text{tot}}^z \rangle_\beta, \end{aligned} \quad (\text{E5})$$

where in the second line we have made the variable substitution $\tau \equiv \lambda\beta$ and in the penultimate line we perform a cyclic permutation within the trace. For the sake of simplicity (and without loss of generality), we assume $M = \langle S_{i_0}^z \rangle_\beta = 0$ in the discussion of fractional liquid, and thus arrive at the Kubo formula (3).

If the total spin S_{tot}^z commutes with the Hamiltonian H , e.g., in the Heisenberg model, the two factors $e^{\tau H}$ and $e^{-\tau H}$ cancel each other and the Kubo formula reduces to the conventional susceptibility expression in terms of equal-time spin correlators. Here in the Kitaev model, although S_{tot}^z is not a good quantum number, $e^{\tau H}$ and $e^{-\tau H}$ nevertheless approximately cancel in the fractional liquid regime (see the discussion in Sec. IV) and Eq. (5) also holds, which in turn gives rise to a Curie behavior with a modified Curie constant $C' > 1/4$.

FIG. 14. (a) Uniform susceptibility χ of the AF Kitaev model under various fields h . The black dashed line indicates the high- T Curie-Weiss fitting, and at intermediate temperature no emergent Curie behavior is found, which is also evident as shown in the $1/\chi$ plot (inset). Since the AF and FM Kitaev models are exactly equivalent under zero fields, here the two temperature scales T_L and T_H coincide with those in Fig. 5(a). (b) Staggered susceptibility χ_s of AF Kitaev model exhibiting a universal Curie behavior in the intermediate- T regime (see also $1/\chi_s$ in the inset). In fact, the χ_s data of the AF Kitaev model are in quantitative agreement with the uniform susceptibility χ of the FM Kitaev model.

FIG. 15. Inverse susceptibility $1/\chi$ of (a) the Kitaev-Heisenberg and (b) Kitaev- Γ models with various J or Γ interactions. The linear fittings are shown in both the high- T (the red dashed lines) and intermediate- T regimes, with the same fitting parameters as those in Figs. 6(g) and 6(h).

However, as the temperature further decreases to the KSL regime, the dynamical correlation decays exponentially as the gauge fluxes freeze at low temperature below the finite flux excitation gap, which can suppress the universal Curie-law susceptibility at intermediate temperature.

APPENDIX F: UNIFORM AND STAGGERED MAGNETIC SUSCEPTIBILITIES IN THE ANTIFERROMAGNETIC KITAEV MODEL

For the isotropic Kitaev model with the AF Kitaev interaction $K_{x,y,z} = 1$, we also compute the uniform magnetic susceptibility χ by applying small fields that coupled to the S^z component, i.e., along the $[001]$ direction. In sharp contrast to the FM Kitaev model results in Fig. 5, the uniform susceptibilities χ in Fig. 14(a) do not exhibit a universal Curie behavior in the intermediate- T regime. The susceptibility curves there are nondiverging convex curves and cannot be fitted by the Curie-Weiss formula. Our cylindrical results of AF Kitaev susceptibility in Fig. 14(a) are again in excellent agreement with previous QMC results [29].

The underlying reason for the absence of emergent Curie behaviors under uniform fields is that the nearest spin pairs in the AF Kitaev model are also correlated along the d_z bond, but in an antiparallel manner. Therefore, the change of total spin that relates to uniform susceptibility corresponds to flipping one of the two spins, which has to overcome a finite amount of AF binding energy along the d_z bond and does not reflect the fluctuating fluxes. In fact, as shown in Fig. 14(b), the staggered susceptibility χ_S under a staggered magnetic field h_S (which has alternating sign on A and B sublattices of the Kitaev honeycomb) exhibits a universal Curie behavior at intermediate temperature.

FIG. 16. Landscape of specific-heat data for the (a) Kitaev-Heisenberg and (b) Kitaev- Γ models. The corresponding gauge fluxes W_p are contour plotted in (c) and (d) for the two models.

APPENDIX G: THERMODYNAMICS OF THE KITAEV-HEISENBERG AND KITAEV- Γ MODELS

In this Appendix we provide additional thermodynamic results of the Kitaev-Heisenberg and Kitaev- Γ models. In Fig. 15 we plot the inverse susceptibility data $1/\chi$ vs T , under the Heisenberg [Fig. 15(a)] and Γ [Fig. 15(b)] interactions. The emergent Curie-Weiss susceptibility scaling is evident at intermediate temperature in Fig. 15, complementing the χ vs T fittings in Figs. 6(g) and 6(h).

In Fig. 16 we show contour plots of the specific heat C_V and gauge flux W_p results of both models. From Figs. 16(a) and 6(b) we see that the C_V curves are always double peaked, which defines two temperature scales T_H and T_L that bound the fractional liquid regime. The corresponding flux expectations W_p are plotted in Figs. 16(c) and 16(d), where the asymptotic KSL regimes with nonzero W_p can be clearly identified.

For the Kitaev-Heisenberg model, the low- T scale T_L determined from the lower peak position of C_V exhibits a clear dip at $J_c \simeq 0.12$ in Fig. 16(a) and for $J \lesssim J_c$ W_p quickly establishes itself at around T_L as shown in Fig. 16(c). The scenario is altered for $J \gtrsim J_c$, where a different low- T scale T'_L emerges, which represents the temperature scale between the SAF phase and Kitaev fractional liquid. In contrast, for the Kitaev- Γ model in Fig. 16(b), the low- T scale T_L remains stable for various Γ values, suggesting that the low- T KSL regime is very robust against Γ interactions. This is also supported by the flux results in Fig. 16(d), where W_p remains nonzero at low temperature at least up to $\Gamma = 0.1$.

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