Magnetic field induced spin liquids in $S = 1$ Kitaev honeycomb modelZheng Zhu^{1,*}, Zheng-Yu Weng² and D. N. Sheng³¹Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA²Institute for Advanced Study, Tsinghua University, Beijing 100084, China³Department of Physics and Astronomy, California State University, Northridge, California 91330, USA

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We investigate the ground state properties of the spin $S = 1$ Kitaev honeycomb model under a magnetic field based on the density matrix renormalization group (DMRG) calculation. With the time-reversal symmetry breaking due to the magnetic field, a gapped Kitaev spin liquid is identified for both ferromagnetic (FM) and antiferromagnetic (AFM) Kitaev couplings. The topological nature of such a Kitaev spin liquid is manifested by a nearly quantized Wilson loop, degeneracy in the entanglement spectra, and the existence of edge modes. While the FM Kitaev spin liquid is destroyed by a weaker magnetic field H_*^{FM} , the AFM one demonstrates a robustness up to an order of magnitude larger critical field H_*^{AFM} . Moreover, an intermediate nonmagnetic phase appears only for the AFM case at larger fields, $H_*^{\text{AFM}} < H < H_{**}^{\text{AFM}}$, before the transition to a high-field polarized paramagnet. The stability of the Kitaev spin liquid against the Heisenberg interactions is also examined. Our findings may further inspire the investigation of recently proposed $S = 1$ Kitaev materials.

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Introduction. The search for exotic states of matter such as highly entangled quantum spin liquids (QSLs) has attracted considerable attention in the modern condensed matter field [1–4]. Among various proposed QSLs, the Kitaev model [5] and its candidate materials [6–10] are of particular interest.

The Kitaev model on a tricoordinated lattice with $S = 1/2$ local degree of freedom is exactly solvable by mapping it into a model of Majorana fermions coupled to $\mathbb{Z}_2$ gauge fields [5]. The ground state harbors both a gapless QSL for a parameter region around equal coupling strengths and a gapped QSL for more anisotropic couplings. Interestingly, the former becomes a non-Abelian topological spin liquid upon time-reversal symmetry breaking [5], for example, by applying a magnetic field [5], adding a three-spin chirality term [11], or decorating the honeycomb lattice [12]. The realization of Kitaev physics in real materials has been proposed in transition metal oxides with strong spin-orbit coupling (SOC) and $S = 1/2$ local moments, in which the edge-sharing oxygen octahedra result in bond-dependent Ising interactions [6,7]. By now, various materials including A_2IrO_3 ($\text{A} = \text{Na}, \text{Li}$) [13,14], $\alpha\text{-RuCl}_3$ [15], and $\text{H}_3\text{LiIr}_2\text{O}_6$ [16] have been discovered to be proximate to the Kitaev spin liquid (KSL) [17–29].

Remarkably, the recent observation of half-integer quantized thermal Hall conductivity in $\alpha\text{-RuCl}_3$ under a magnetic field offers smoking-gun evidence of the fractionalized topological state [29], leading to substantial activities in examining the magnetic field induced QSLs in the Kitaev models and ma-

terials [30–51]. Although numerous efforts have been devoted to the $S = 1/2$ case, the magnetic field induced phases in the $S = 1$ Kitaev model and the corresponding experiments are of equal interest and importance. Different from $S = 1/2$, the nature of the ground state for the pure $S = 1$ Kitaev model is still controversial [52–57]. Recently, a mechanism for realizing the $S = 1$ Kitaev interactions by considering both SOC and strong Hund's couplings in transition metal oxides $\text{A}_3\text{Ni}_2\text{XO}_6$ ($\text{A} = \text{Li}, \text{Na}$ and $\text{X} = \text{Bi}, \text{Sb}$) has been proposed [58], providing a platform to explore the $S = 1$ Kitaev physics. The honeycomb layer lies in a plane perpendicular to the [111] direction and the dominant Kitaev coupling is antiferromagnetic [58]. It is then natural to ask if the perpendicular magnetic field can induce new QSLs in such real materials, which is distinct from the pure $S = 1$ Kitaev model [52–57] due to the lack of time-reversal symmetry [5]. Here, both the possible field induced phases and their instabilities are equally important to the ones in the $S = 1/2$ case [30–51]. Therefore, establishing the quantum phase diagram is the very essence of understanding various important questions and providing valuable information for the experiments. Motivated by the above, in this Rapid Communication we study the magnetic field induced phases in the $S = 1$ Kitaev honeycomb model by density matrix renormalization group (DMRG).

Model, method, and key results. We consider the $S = 1$ Kitaev honeycomb model with a perpendicular magnetic field $\mathbf{H}$ along the [111] direction. The Hamiltonian is

$$H = \sum_{\langle i,j \rangle} K_\gamma S_i^\gamma S_j^\gamma - \sum_i \mathbf{H} \cdot \mathbf{S}_i + J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (1)$$

Here, $\gamma = x, y, z$ represent the three nearest-neighbor links $\langle i, j \rangle$ of the honeycomb lattice, and S_i^γ denotes the effective spin $S = 1$ degrees of freedom sitting on each site and interacting via Kitaev coupling K_γ . The second term is the Zeeman

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term with a uniform field, and the third term represents the Heisenberg interactions with coupling J . We consider isotropic coupling $K_\gamma \equiv K$ and set $|K|$ as the unit of energy, where $K = +1$ ($K = -1$) corresponds to the antiferromagnetic (ferromagnetic) [AFM (FM)] model. Below we will identify the magnetic field induced phases at $J = 0$ and examine the stability of KSL against Heisenberg perturbations.

We use DMRG to study the Hamiltonian (1) on a honeycomb lattice spanned by length vectors $\mathbf{L}_x = L_x \mathbf{e}_x$ and $\mathbf{L}_y = L_y \mathbf{e}_y$, where $\mathbf{e}_x = (1, 0)$ and $\mathbf{e}_y = (1/2, \sqrt{3}/2)$ are two primitive vectors. As each unit cell contains two sites, the total number of sites $N = L_x \times L_y \times 2$. In the present calculation, we choose both the cylinder and torus geometries and keep up to 1600–2800 states for good convergence (depending on the nature of states and system sizes). We keep the truncation error to be smaller than 10^{-6} in most cases.

We shall identify a gapped KSL phase for both AFM and FM Kitaev couplings in weak magnetic fields. Interestingly, while the FM gapped KSL is fragile against the magnetic field, the AFM gapped KSL is extremely robust up to an order of magnitude larger critical field. We also find the topological nature of this gapped KSL, including the $\mathbb{Z}_2$ gauge structure, the edge modes, and a robust multiple degeneracy in the entanglement spectra on ladder systems. Moreover, we discover an intermediate phase (IP) only in the AFM model before a second transition to the high-field polarized paramagnet. The phase diagram is depicted in Fig. 1(a). Finally, we show that field induced KSLs eventually give way to magnetic orders under sufficient Heisenberg perturbations.

Phase diagram. We begin by establishing the phase diagram of the model Hamiltonian (1) [see Fig. 1(a)]. We obtain the ground state for a given magnetic field, and the phase boundaries are determined by consistent evidence from the measurement results of magnetization M , magnetic susceptibility χ , and the Wilson loop W_y .

Figures 1(b) and 1(c) show the magnetization M and magnetic susceptibility $\chi = \partial M / \partial H$ for systems with both toroidal and cylindrical boundary conditions. At the FM Kitaev coupling side (see the left-hand side in Fig. 1), we find a strong response of the system to the magnetic field, and the single kink in magnetization M [see Fig. 1(b)] and single peak of χ [see Fig. 1(c)] demonstrate a single phase transition to the high-field partially polarized paramagnet at $H_*^{\text{FM}} \sim 0.01$, which smoothly connects to the fully polarized phase in the infinite-field limit. In contrast, in the case with AFM Kitaev coupling (see the right-hand side in Fig. 1), the system exhibits a weak response to an external field with a critical field $H_*^{\text{AFM}} \sim 0.24$, which is larger than H_*^{FM} by more than an order of magnitude [see Fig. 1(c)]. Moreover, the two-peak structure in the magnetic susceptibility χ suggests that there is an intermediate phase at $H_*^{\text{AFM}} < H < H_{**}^{\text{AFM}}$ before a second transition to a partial polarized paramagnet beyond H_{**}^{AFM} .

We also note that the ground state of the $S = 1$ Kitaev model at zero field can be characterized by a conserved $\mathbb{Z}_2$ quantum number determined by the Wilson loop operators $\hat{W}_l$, which is generalized from the $S = 1/2$ model and can be defined by $\hat{W}_l = \prod_{i=1}^{L_l} \exp[i\pi S_i^{\alpha_i}]$ in every closed loop on the lattice, where α_i represents the normal direction of site i [52]. For example, $\hat{W}_y$ denotes the Wilson loop operator with

FIG. 1. Phase diagram of the $S = 1$ Kitaev honeycomb model subject to a [111] magnetic field H . (a) shows the distinct phases in the FM (left side) and AFM (right side) Kitaev model as a function of magnetic field strength, including the gapped Kitaev spin liquid (KSL) phase, the intermediate phase (IP), and the partial polarized paramagnet. The zero-field limit (blue circle) corresponds to the pure Kitaev model. The phase boundaries are probed by (b) the magnetization M and (c) the magnetic susceptibility χ for systems on a torus with 4×3 unit cells (sphere), and on a cylinder with 8×4 (circle). The peaks in χ suggest the phase transitions. Note the stark difference between the AFM and FM models, $H_*^{\text{AFM}} \gg H_*^{\text{FM}}$. (c) shows the Wilson loop W_y (illustrated by the green loop in the inset) as a function of H , and the nearly quantized W_y implies the approximate $\mathbb{Z}_2$ gauge structure in the KSL phase.

the loop winding once around the cylinder which only covers $\gamma = x, z$ links [see the inset in Fig. 1(d)]. It is straightforward to verify that these Wilson loop operators commute with each other and also commute with the Hamiltonian of the pure Kitaev model. Since $\hat{W}_l$ squares to identity $\hat{W}_l^2 = 1$, the eigenvalues of $\hat{W}_l$ are $W_l = \pm 1$ at $H = 0$, which is associated with flux excitations such as in the spin-1/2 Kitaev model. At $H \neq 0$, the Wilson loop $W_l = \langle \hat{W}_l \rangle$ measured at the ground state is shown in Fig. 1(d), where W_y takes the exact quantized value at $H = 0$. After the time-reversal symmetry breaking by the magnetic field, W_y takes nearly quantized values for a finite range of magnetic fields at $H < H_*^{\text{AFM}}$ ($H < H_*^{\text{FM}}$) for AFM (FM) Kitaev coupling. This indicates the $\mathbb{Z}_2$ gauge structure in the pure $S = 1$ Kitaev model remains a good description of the perturbed Kitaev model away from the zero-field limit. Here, we should note $W_y > 0$ for $L_y = 3$ and $W_y < 0$ for $L_y = 4$, while its absolute value is nearly quantized. This indicates distinct topological sectors can be accessed with changing L_y . Beyond H_*^{FM} or H_*^{AFM} , W_y tends to be negligibly small, particularly for wider systems, implying the absence of the $\mathbb{Z}_2$ gauge structure. The sudden drop in W_y characterizes the phase transition at H_*^{AFM} and H_*^{FM} [59].

FIG. 2. Magnetic field induced Kitaev spin liquid in the AFM/FM Kitaev models under the [111] magnetic field. (a) shows the decay of spin correlations along the zigzag chain (the yellow dots in the inset) in semilogarithmic scale. The gapped nature of KSL can also be confirmed from the von Neumann entanglement entropy S_{vN} in (b), which is independent of the cut position L_A (the cuts are parallel to $\mathbf{e}_y$). The corresponding entanglement spectra at $L_A = L_x/2$ is shown in (c), which shows four quasidegenerate states separated from higher spectra by a finite entanglement gap Δ_ξ . In (d), the magnetization distribution $\langle \mathbf{S} \cdot \mathbf{e}_H \rangle$ [$\mathbf{e}_H = (1, 1, 1)$] along the zigzag chain explicitly exhibits the edge modes. The inset of (d) shows the uniform distribution of the Wilson loop W_y along the cylinder.

The KSL phase. Below we show that the phase in Fig. 1(a) at $0 < H < H_*^{\text{AFM}}$ in the AFM model or $0 < H < H_*^{\text{FM}}$ in the FM model is a gapped KSL phase with a topological nature. We first examine the spin-spin correlations $S_{ij} \equiv \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - \langle \mathbf{S}_i \rangle \cdot \langle \mathbf{S}_j \rangle$ and the von Neumann entanglement entropy S_{vN} to show the gapped nature, and then we probe its topological properties from the Wilson loop, the entanglement spectra, and the edge modes.

In the Kitaev limit (i.e., $H = 0$), we confirm that the spin-spin correlations vanish beyond the nearest-neighbor links [52] such as the $S = 1/2$ pure Kitaev model [5]. Beyond the Kitaev limit, as shown in Fig. 2(a) for both AFM and FM Kitaev couplings, the spin-spin correlations decay exponentially along the zigzag chains of the cylinders [see the inset of Fig. 2(a)], indicating a nonmagnetic ground state with a finite spin gap. The gapped nature can be further confirmed by the von Neumann entanglement entropy $S_{vN} = -\text{Tr}(\rho_A \ln \rho_A)$, where ρ_A is the reduced density matrix of part A for the bipartition of the system into A and B, with ρ_A obtained by tracing out the degrees of freedom of the B part. Here, we consider the cut parallel to $\mathbf{e}_y$ and measure S_{vN} for each cut with subsystem length L_A . As shown in Fig. 2(b), we find that S_{vN} are independent on the positions of each cut and display flat behavior for cylinders with different widths, suggesting zero central charge or the existence of a finite energy gap in the bulk.

In addition, we also examine the entanglement spectra for the cut parallel to $\mathbf{e}_y$ at $L_A = L_x/2$, as shown in Fig. 2(c), with a finite entanglement gap Δ_ξ separating the ground state manifold from the higher spectra. On cylinders of width $L_y = 4$, the entanglement spectra displays fourfold (quasi)degeneracy [see Fig. 2(c)], indicating the existence of boundary zero modes and their topological nature. The structure of the entanglement spectra is similar to the topological KSL in the spin $S = 1/2$ Kitaev model [30]. The topological nature is also exhibited by the Wilson loop operator $\hat{W}_y$, which takes a nearly quantized mean value in the KSL phase [see Fig. 1(c)]. Moreover, the distribution of W_y along the cylinders is uniform with the nearly quantized value [see the inset of Fig. 2(d)]. All of these facts imply the $\mathbb{Z}_2$ gauge structure remains a good description of the gapped KSL phase. To explicitly show the presence of edge modes, we compute the local on-site magnetization distributions $\langle \mathbf{S} \cdot \mathbf{e}_H \rangle$ [$\mathbf{e}_H = (1, 1, 1)$] along the zigzag chain of the cylinders, as shown in Fig. 2(d), where the

spins near the edge are much easier to be polarized than the bulk, indeed implying gapless edge modes.

From the above analysis, the nearly quantized Wilson loop operator mean value, the fourfold degeneracy in the entanglement spectra, and the existence of edge modes all manifest the topological nature of the gapped KSL phase. Different from the FM model, the susceptibility χ in Fig. 1(b) suggests the AFM model further hosts an intermediate phase sandwiched between the KSL and partially polarized paramagnet phase at $H_*^{\text{AFM}} < H < H_{**}^{\text{AFM}}$. The intermediate phase is distinct from the KSL phase by the vanishingly small Wilson loop, together with the edge mode. As shown in Fig. 3(a) and its inset, S_{ij} decays exponentially while S_{vN} is flat. However, the local magnetization $\langle \mathbf{S} \cdot \mathbf{e}_H \rangle$ shows strong oscillation in real space, which is similar to the intermediate spin-liquid phase in the $S = 1/2$ case. Due to the nonuniform pattern of magnetization, much larger system sizes are demanded to resolve its nature, which is beyond the scope of this work.

Instability of the KSL. We have found that the KSL in the AFM Kitaev model is much more robust against increasing magnetic field than the one in the FM model (i.e., $H_*^{\text{AFM}} \gg H_*^{\text{FM}}$), though at zero field these two models are equivalent [7]. For finite magnetic fields, the indications of such a significant difference can be gained from the low-field magnitude of susceptibility χ [see Fig. 1(c)].

FIG. 3. Intermediate phase (IP) and paramagnet phase in the AFM Kitaev model. (a) shows the decay of spin correlations along the zigzag chain in semilogarithmic scale for the $L_y = 3, L_x = 12$ cylinders, where we compared the intermediate phase with the paramagnets. The inset of (a) exhibits the flat behavior of the entanglement entropy S_{vN} as a function of the cut position L_A . (b) shows the spatial distribution of $\langle \mathbf{S} \cdot \mathbf{e}_H \rangle$ [$\mathbf{e}_H = (1, 1, 1)$] along the zigzag chain.

FIG. 4. Instability of the KSL. (a) and (b) show the instability of the (a) AFM KSL and (b) FM KSL against AFM/FM Heisenberg perturbations. The colored shadows correspond to the KSL depicted in Fig. 1. The ground state energy per site (orange color) and the entanglement entropy for the cuts bisecting the torus (olive color) show that the AFM/FM KSLs survive in a narrow range of Heisenberg coupling strength and eventually give way to magnetic orders. Here, we consider a torus geometry with 4×3 unit cells. The natures of these magnetic orders are identified by the static spin structure factor (the insets) at $|J/K| = 0.1$. The black dots represent the accessible momenta in the Brillouin zone for finite-sized cylinders with $L_y = 4, L_x = 6$.

Furthermore, in the $S = 1$ Kitaev materials, the superexchange processes would also give rise to the Heisenberg interactions between nearest-neighbor sites [58]. Thus we study the instability of the KSL against the Heisenberg interactions in the vicinity of the pure Kitaev model. Here, we consider both AFM and FM Heisenberg coupling J . The phase transitions are characterized by the singular behavior of entanglement entropy and the kinks in the ground state energy density. As illustrated in the Figs. 4(a) and 4(b), the magnetic field induced gapped KSLs can survive in a finite range of the Heisenberg interactions before they finally give way to the magnetic ordered phases. These magnetic ordered phases are identified by the peaks in the static spin structure factor, as shown in the insets of Fig. 4. Here, the magnetic ordered phases at $H > 0$ are the same as the ones identified in the Kitaev-Heisenberg model with the same ratio J/K but $H = 0$ [56,58]. In other words, if the system is tuned proximate to the Kitaev phase at a finite J , the spin liquids we have discovered can be still induced by applying a magnetic field along the [111] direction.

Discussions and summary. We investigate the magnetic field induced phases in the $S = 1$ Kitaev honeycomb model,

which is distinct from the studies on the pure $S = 1$ Kitaev model in Refs. [52–57] because the magnetic field breaks the time-reversal symmetry and gives rise to new spin liquids. The FM and AFM models are no longer equivalent beyond the zero-field limit either. The gapped topological KSL identified in this work is indeed different from the gapless KSL proposed in the pure Kitaev model [52–56].

We identify a field induced gapped KSL phase with a topological nature for both the AFM and FM Kitaev couplings, including the $\mathbb{Z}_2$ gauge structure, the edge modes, and degenerate entanglement spectra on the cylinders. Interestingly, such a KSL phase is much more stable against increasing magnetic field for the AFM Kitaev coupling than the FM one, which resembles the $S = 1/2$ case [30]. However, there are differences between the $S = 1$ and $S = 1/2$ cases such as the degeneracy in the entanglement spectrum. Moreover, the interaction type of $S = 1/2$ Kitaev materials is proposed to be FM [9,10] while the $S = 1$ Kitaev materials harbor AFM Kitaev interactions [58].

A theoretical understanding of the $S = 1$ Kitaev model in a magnetic field is still a challenge partially due to the lack of an exact solution even in the zero-field limit, and thus it will be an excellent task to pursue in the future. Our findings may also stimulate investigations on the effects of more complicated interactions or anisotropies, as well as the theories of the intermediate phase. Recently, the $S = 1$ and high-spin Kitaev materials [58,60] have been proposed to host AFM Kitaev couplings, so then it is promising to realize stable topological spin liquids based on our findings. The signatures such as the quantized thermal Hall conductivity and fractionalized excitations in inelastic neutron scattering can be directly probed.

Note added. Recently, we became aware of Ref. [57], where the authors constructed a tensor network ground state wave function for a pure $S = 1$ Kitaev model. The magnetic field effect was also examined in Ref. [57] via the magnetizations, which are consistent with our results in Figs. 1(b) and 1(c). In our work, we identify the magnetic field induced AFM/FM KSL from different aspects of a systematical characterization based on the unbiased DMRG method. We should also note that the largest field for the AFM model in Ref. [57] is up to $H \approx 0.3/\sqrt{3} < H_*^{\text{AFM}}$, which lies within the AFM KSL phase identified in our work. In addition, we became aware of Refs. [61–63], who also reported on investigations of a $S = 1$ Kitaev model in a magnetic field.

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[1] P. A. Lee, An end to the drought of quantum spin liquids, *Science* **321**, 1306 (2008).

[2] L. Savary and L. Balents, Quantum spin liquids: A review, *Rep. Prog. Phys.* **80**, 016502 (2017), and references therein;

- L. Balents, Spin liquids in frustrated magnets, *Nature (London)* **464**, 199 (2010), and references therein.
- [3] Y. Zhou, K. Kanoda, and T.-K. Ng, Quantum spin liquid states, *Rev. Mod. Phys.* **89**, 025003 (2017), and references therein.
- [4] J. Knolle and R. Moessner, A field guide to spin liquids, *Annu. Rev. Condens. Matter Phys.* **10**, 451 (2019), and references therein.
- [5] A. Kitaev, Anyons in an exactly solved model and beyond, *Ann. Phys.* **321**, 2 (2006).
- [6] G. Jackeli and G. Khaliullin, Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models, *Phys. Rev. Lett.* **102**, 017205 (2009).
- [7] J. Chaloupka, G. Jackeli, and G. Khaliullin, Kitaev-Heisenberg Model on a Honeycomb Lattice: Possible Exotic Phases in Iridium Oxides $A_2\text{IrO}_3$, *Phys. Rev. Lett.* **105**, 027204 (2010).
- [8] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Spin-orbit physics giving rise to novel phases in correlated systems: Iridates and related materials, *Annu. Rev. Condens. Matter Phys.* **7**, 195 (2016), and references therein.
- [9] S. Trebst, Kitaev materials, [arXiv:1701.07056](https://arxiv.org/abs/1701.07056), and references therein.
- [10] M. Hermanns, I. Kimchi, and J. Knolle, Physics of the Kitaev model: Fractionalization, dynamical correlations, and material connections, *Annu. Rev. Condens. Matter Phys.* **9**, 17 (2018), and references therein.
- [11] D.-H. Lee, G.-M. Zhang, and T. Xiang, Edge Solitons of Topological Insulators and Fractionalized Quasiparticles in Two Dimensions, *Phys. Rev. Lett.* **99**, 196805 (2007).
- [12] H. Yao and S. A. Kivelson, Exact Chiral Spin Liquid with Non-Abelian Anyons, *Phys. Rev. Lett.* **99**, 247203 (2007).
- [13] S. K. Choi, R. Coldea, A. N. Kolmogorov, T. Lancaster, I. I. Mazin, S. J. Blundell, P. G. Radaelli, Yogesh Singh, P. Gegenwart, K. R. Choi, S.-W. Cheong, P. J. Baker, C. Stock, and J. Taylor, Spin Waves and Revised Crystal Structure of Honeycomb Iridate Na_2IrO_3 , *Phys. Rev. Lett.* **108**, 127204 (2012).
- [14] Y. Singh and P. Gegenwart, Antiferromagnetic Mott insulating state in single crystals of the honeycomb lattice material Na_2IrO_3 , *Phys. Rev. B* **82**, 064412 (2010); Y. Singh, S. Manni, J. Reuther, T. Berlijn, R. Thomale, W. Ku, S. Trebst, and P. Gegenwart, Relevance of the Heisenberg-Kitaev Model for the Honeycomb Lattice Iridates $A_2\text{IrO}_3$, *Phys. Rev. Lett.* **108**, 127203 (2012).
- [15] K. W. Plumb, J. P. Clancy, L. J. Sandilands, V. V. Shankar, Y. F. Hu, K. S. Burch, H.-Y. Kee, and Y.-J. Kim, $\alpha\text{-RuCl}_3$: A spin-orbit assisted Mott insulator on a honeycomb lattice, *Phys. Rev. B* **90**, 041112 (2014); L. J. Sandilands, Y. Tian, K. W. Plumb, Y.-J. Kim, and K. S. Burch, Scattering Continuum and Possible Fractionalized Excitations in $\alpha\text{-RuCl}_3$, *Phys. Rev. Lett.* **114**, 147201 (2015); H.-S. Kim, V. V. Shankar, A. Catuneanu, and H.-Y. Kee, Kitaev magnetism in honeycomb RuCl_3 with intermediate spin-orbit coupling, *Phys. Rev. B* **91**, 241110(R) (2015).
- [16] S. Bette, T. Takayama, K. Kitagawa, R. Takano, H. Takagi, and R. E. Dinnebier, Solution of the heavily stacking faulted crystal structure of the honeycomb iridate $\text{H}_3\text{LiIr}_2\text{O}_6$, *Dalton Trans.* **46**, 15216 (2017).
- [17] H.-C. Jiang, Z.g.-C. Gu, X.-L. Qi, and S. Trebst, Possible proximity of the Mott insulating iridate Na_2IrO_3 to a topological phase: Phase diagram of the Heisenberg-Kitaev model in a magnetic field, *Phys. Rev. B* **83**, 245104 (2011).
- [18] I. Kimchi and A. Vishwanath, Kitaev-Heisenberg models for iridates on the triangular, hyperkagome, kagome, fcc, and pyrochlore lattices, *Phys. Rev. B* **89**, 014414 (2014).
- [19] V. M. Katukuri, S. Nishimoto, V. Yushankhai, A. Stoyanova, H. Kandpal, S. Choi, R. Coldea, I. Rousochatzakis, L. Hozoi, and J. van den Brink, Kitaev interactions between $j = 1/2$ moments in honeycomb Na_2IrO_3 are large and ferromagnetic: Insights from *ab initio* quantum chemistry calculations, *New J. Phys.* **16**, 013056 (2014).
- [20] J. Osorio Iregui, P. Corboz, and M. Troyer, Probing the stability of the spin-liquid phases in the Kitaev-Heisenberg model using tensor network algorithms, *Phys. Rev. B* **90**, 195102 (2014).
- [21] A. Banerjee, C. A. Bridges, J.-Q. Yan, A. A. Aczel, L. Li, M. B. Stone, G. E. Granroth, M. D. Lumsden, Y. Yiu, J. Knolle, D. L. Kovrizhin, S. Bhattacharjee, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, Proximate Kitaev quantum spin liquid behaviour in a honeycomb magnet, *Nat. Mater.* **15**, 733 (2016); A. Banerjee, J. Yan, J. Knolle, C. A. Bridges, M. B. Stone, M. D. Lumsden, D. G. Mandrus, D. A. Tennant, R. Moessner, and S. E. Nagler, Neutron scattering in the proximate quantum spin liquid $\alpha\text{-RuCl}_3$, *Science* **356**, 1055 (2017); A. Banerjee, P. Lampen-Kelley, J. Knolle, C. Balz, A. A. Aczel, B. Winn, Y. Liu, D. Pajerowski, J.-Q. Yan, C. A. Bridges, A. T. Savici, B. C. Chakoumakos, M. D. Lumsden, D. A. Tennant, R. Moessner, D. G. Mandrus, and S. E. Nagler, Excitations in the field-induced quantum spin liquid state of $\alpha\text{-RuCl}_3$, *npj Quantum Mater.* **3**, 8 (2018).
- [22] X.-Y. Song, Y.-Z. You, and L. Balents, Low-Energy Spin Dynamics of the Honeycomb Spin Liquid Beyond the Kitaev Limit, *Phys. Rev. Lett.* **117**, 037209 (2016).
- [23] M. Gohlke, R. Verresen, R. Moessner, and F. Pollmann, Dynamics of the Kitaev-Heisenberg Model, *Phys. Rev. Lett.* **119**, 157203 (2017); M. Gohlke, G. Wachtel, Y. Yamaji, F. Pollmann, and Y. B. Kim, Quantum spin liquid signatures in Kitaev-like frustrated magnets, *Phys. Rev. B* **97**, 075126 (2018).
- [24] Z. Wang, S. Reschke, D. Hüvonen, S.-H. Do, K.-Y. Choi, M. Gensch, U. Nage, T. Rööm, and A. Loidl, Magnetic Excitations and Continuum of a Possibly Field-Induced Quantum Spin Liquid in $\alpha\text{-RuCl}_3$, *Phys. Rev. Lett.* **119**, 227202 (2017).
- [25] S. M. Winter, K. Riedl, D. Kaib, R. Coldea, and R. Valenti, Probing $\alpha\text{-RuCl}_3$ Beyond Magnetic Order: Effects of Temperature and Magnetic Field, *Phys. Rev. Lett.* **120**, 077203 (2018).
- [26] L. Janssen, E. C. Andrade, and M. Vojta, Magnetization processes of zigzag states on the honeycomb lattice: Identifying spin models for $\alpha\text{-RuCl}_3$ and Na_2IrO_3 , *Phys. Rev. B* **96**, 064430 (2017); Honeycomb-Lattice Heisenberg-Kitaev Model in a Magnetic Field: Spin Canting, Metamagnetism, and Vortex Crystals, *Phys. Rev. Lett.* **117**, 277202 (2016).
- [27] A. N. Ponomaryov, E. Schulze, J. Wosnitzer, P. Lampen-Kelley, A. Banerjee, J.-Q. Yan, C. A. Bridges, D. G. Mandrus, S. E. Nagler, A. K. Kolezhuk, and S. A. Zvyagin, Unconventional spin dynamics in the honeycomb-lattice material $\alpha\text{-RuCl}_3$: High-field electron spin resonance studies, *Phys. Rev. B* **96**, 241107 (2017).
- [28] A. Little, Liang Wu, P. Lampen-Kelley, A. Banerjee, S. Patankar, D. Rees, C. A. Bridges, J.-Q. Yan, D. Mandrus, S. E. Nagler, and J. Orenstein, Antiferromagnetic Resonance

- and Terahertz Continuum in α -RuCl₃, *Phys. Rev. Lett.* **119**, 227201 (2017).
- [29] Y. Kasahara, T. Ohnishi, Y. Mizukami, O. Tanaka, S. Ma, K. Sugii, N. Kurita, H. Tanaka, J. Nasu, Y. Motome, T. Shibauchi, and Y. Matsuda, Majorana quantization and half-integer thermal quantum Hall effect in a Kitaev spin liquid, *Nature (London)* **559**, 227 (2018).
- [30] Z. Zhu, I. Kimchi, D. N. Sheng, and L. Fu, Robust non-Abelian spin liquid and a possible intermediate phase in the antiferromagnetic Kitaev model with magnetic field, *Phys. Rev. B* **97**, 241110(R) (2018).
- [31] M. Gohlke, R. Moessner, and F. Pollmann, Dynamical and topological properties of the Kitaev model in a [111] magnetic field, *Phys. Rev. B* **98**, 014418 (2018).
- [32] J. Nasu, Y. Kato, Y. Kamiya, and Y. Motome, Successive Majorana topological transitions driven by a magnetic field in the Kitaev model, *Phys. Rev. B* **98**, 060416(R) (2018); J. Nasu and Y. Motome, Nonequilibrium Majorana dynamics by quenching a magnetic field in Kitaev spin liquids, *Phys. Rev. Research* **1**, 033007 (2019); Y. Motome and J. Nasu, Hunting Majorana fermions in Kitaev magnets, *J. Phys. Soc. Jpn.* **89**, 012002 (2020).
- [33] S. Liang, M.-H. Jiang, W. Chen, J.-X. Li, and Q.-H. Wang, Intermediate gapless phase and topological phase transition of the Kitaev model in a uniform magnetic field, *Phys. Rev. B* **98**, 054433 (2018).
- [34] Z.-X. Liu and B. Normand, Dirac and Chiral Quantum Spin Liquids on the Honeycomb Lattice in a Magnetic Field, *Phys. Rev. Lett.* **120**, 187201 (2018).
- [35] C. Hickey and S. Trebst, Emergence of a field-driven U(1) spin liquid in the Kitaev honeycomb model, *Nat. Commun.* **10**, 530 (2019).
- [36] D. C. Ronquillo, A. Vengal, and N. Trivedi, Signatures of magnetic-field-driven quantum phase transitions in the entanglement entropy and spin dynamics of the Kitaev honeycomb model, *Phys. Rev. B* **99**, 140413 (2019); N. D. Patel and N. Trivedi, Magnetic field-induced intermediate quantum spin liquid with a spinon Fermi surface, *Proc. Natl. Acad. Sci. USA* **116**, 12199 (2019); S. Pradhan, N. D. Patel, and N. Trivedi, Two-magnon bound states in the Kitaev model in a [111] field, *Phys. Rev. B* **101**, 180401 (2020).
- [37] J. S. Gordon, A. Catuneanu, E. S. Sørensen, and H.-Y. Kee, Theory of the field-revealed Kitaev spin liquid, *Nat. Commun.* **10**, 2470 (2019).
- [38] H.-C. Jiang, C.-Y. Wang, B. Huang, and Y.-M. Lu, Field induced quantum spin liquid with spinon Fermi surfaces in the Kitaev model, *arXiv:1809.08247*.
- [39] Y.-F. Jiang, T. P. Devereaux, and H.-C. Jiang, Field-induced quantum spin liquid in the Kitaev-Heisenberg model and its relation to α -RuCl₃, *Phys. Rev. B* **100**, 165123 (2019).
- [40] J.-Y. Chen, L. Vanderstraeten, S. Capponi, and D. Poilblanc, Non-Abelian chiral spin liquid in a quantum antiferromagnet revealed by an iPEPS study, *Phys. Rev. B* **98**, 184409 (2018).
- [41] D. Takikawa and S. Fujimoto, Impact of off-diagonal exchange interactions on the Kitaev spin liquid state of α -RuCl₃, *Phys. Rev. B* **99**, 224409 (2019).
- [42] L. Janssen and M. Vojta, Heisenberg-Kitaev physics in magnetic fields, *J. Phys.: Condens. Matter* **31**, 423002 (2019).
- [43] W. M. H. Natori, R. Moessner, and J. Knolle, Orbital magnetic field effects in Mott insulators with strong spin-orbit coupling, *Phys. Rev. B* **100**, 144403 (2019).
- [44] D. A. S. Kaib, S. M. Winter, and R. Valenti, Kitaev honeycomb models in magnetic fields: Dynamical response and hidden symmetries, *Phys. Rev. B* **100**, 144445 (2019).
- [45] Y. Sugita, Y. Kato, and Y. Motome, Antiferromagnetic Kitaev interactions in polar spin-orbit Mott insulators, *Phys. Rev. B* **101**, 100410 (2020).
- [46] K. Ido and T. Misawa, Correlation effects on the magnetization process of the Kitaev model, *Phys. Rev. B* **101**, 045121 (2020).
- [47] S.-H. Jang, R. Sano, Y. Kato, and Y. Motome, Antiferromagnetic Kitaev interaction in *f*-electron based honeycomb magnets, *Phys. Rev. B* **99**, 241106(R) (2019); Computational design of *f*-electron Kitaev magnets: honeycomb and hyperhoneycomb compounds A₂PrO₃ (A = alkali metals), *arXiv:1912.03422*.
- [48] Y. D. Li and G. Chen, Non-spin-ice pyrochlore U(1) quantum spin liquid: Manifesting mixed symmetry enrichments, *Phys. Rev. Research* **2**, 013334 (2020).
- [49] J. Wen, S.-L. Yu, S. Li, W. Yu, and J.-X. Li, Experimental identification of quantum spin liquids, *npj Quantum Mater.* **4**, 12 (2019).
- [50] Y. H. Gao, C. Hickey, T. Xiang, S. Trebst, and G. Chen, Thermal Hall signatures of non-Kitaev spin liquids in honeycomb Kitaev materials, *Phys. Rev. Res.* **1**, 013014 (2019).
- [51] H.-Y. Lee, R. Kaneko, L. E. Chern, T. Okubo, Y. Yamaji, N. Kawashima, and Y. B. Kim, Magnetic-field induced quantum phases in tensor network study of Kitaev magnets, *Nat. Commun.* **11**, 1639 (2020).
- [52] G. Baskaran, D. Sen, and R. Shankar, Spin-*S* Kitaev model: Classical ground states, order from disorder, and exact correlation functions, *Phys. Rev. B* **78**, 115116 (2008); G. Baskaran, S. Mandal, and R. Shankar, Exact Results for Spin Dynamics and Fractionalization in the Kitaev Model, *Phys. Rev. Lett.* **98**, 247201 (2007).
- [53] T. Minakawa, J. Nasu, and A. Koga, Quantum and classical behavior of spin-*S* Kitaev models in the anisotropic limit, *Phys. Rev. B* **99**, 104408 (2019).
- [54] A. Koga, H. Tomishige, and J. Nasu, Ground-state and thermodynamic properties of an *S* = 1 Kitaev model, *J. Phys. Soc. Jpn.* **87**, 063703 (2018).
- [55] J. Oitmaa, A. Koga, and R. R. P. Singh, Incipient and well-developed entropy plateaus in spin-*S* Kitaev models, *Phys. Rev. B* **98**, 214404 (2018).
- [56] X.-Y. Dong and D. N. Sheng, Spin-1 Kitaev-Heisenberg model on a two-dimensional honeycomb lattice, *arXiv:1911.12854*.
- [57] H.-Y. Lee, N. Kawashima, and Y. B. Kim, Tensor network wavefunction of *S* = 1 Kitaev spin liquids, *arXiv:1911.07714*.
- [58] P. P. Stavropoulos, D. Pereira, and H.-Y. Kee, Microscopic Mechanism for a Higher-Spin Kitaev Model, *Phys. Rev. Lett.* **123**, 037203 (2019).
- [59] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevResearch.2.022047> for more discussions on the phase transition.
- [60] C. Xu, J. Feng, M. Kawamura, Y. Yamaji, Y. Nahas, S. Prokhorenko, Y. Qi, H. Xiang, and L. Bellaiche, Possible Kitaev Quantum Spin Liquid State in 2D Materials with *S* = 3/2,

- [Phys. Rev. Lett.](#) **124**, 087205 (2020); C. Xu, J. Feng, H. Xiang *et al.*, Interplay between Kitaev interaction and single ion anisotropy in ferromagnetic CrI_3 and CrGeTe_3 monolayers, [npj Comput. Mater.](#) **4**, 57 (2018).
- [61] I. Khait, P. P. Stavropoulos, H.-Y. Kee, and Y. B. Kim, Characterizing spin-one Kitaev quantum spin liquids, [arXiv:2001.06000](#).
- [62] C. Hickey, C. Berke, P. P. Stavropoulos, H.-Y. Kee, and S. Trebst, Field-driven gapless spin liquid in the spin-1 Kitaev honeycomb model, [arXiv:2001.07699](#).
- [63] A. Koga, T. Minakawa, Y. Murakami, and J. Nasu, Spin transport in the quantum spin liquid state in the $S = 1$ Kitaev model: Role of the fractionalized quasiparticles, [J. Phys. Soc. Jpn.](#) **89**, 033701 (2020).