Strain-induced phase diagram of the $S = \frac{3}{2}$ Kitaev material CrSiTe$_3$

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The interplay among anisotropic magnetic terms, such as the bond-dependent Kitaev interactions and single-ion anisotropy, plays a key role in stabilizing the finite-temperature ferromagnetism in the two-dimensional compound CrSiTe$_3$. While the Heisenberg interaction is predominant in this material, a recent work shows that it is rather sensitive to the compressive strain, leading to a variety of phases, possibly including a sought-after Kitaev quantum spin liquid [C. Xu, et. al., Phys. Rev. Lett. 124, 087205 (2020)]. To further understand these states, we establish the quantum phase diagram of a related bond-directional spin-3/2 model by the density-matrix renormalization group method. As the Heisenberg coupling varies from ferromagnetic to antiferromagnetic, three magnetically ordered phases, i.e., a ferromagnetic phase, a 120° phase and an antiferromagnetic phase, appear consecutively. All the phases are separated by first-order phase transitions, as revealed by the kinks in the ground-state energy and the jumps in the magnetic order parameters. However, no positive evidence of the quantum spin liquid state is found and possible reasons are discussed briefly.

I. INTRODUCTION

The bond-directional interactions have emerged as a focus of research in hunting for nontrivial quantum states ever since Kitaev proposed his famous spin-1/2 model on the honeycomb lattice in 2006, later dubbed Kitaev honeycomb model [1]. As a rare exactly solvable model which supports non-Abelian statistics and Majorana fermion excitations, the Kitaev model is believed to be a milestone in the area of quantum spin liquids (QSLs) [2–4]. However, experimental realization of such interactions is highly nontrivial due to their bond-dependent Ising nature. Later on, Jackeli and Khaliullin provided a feasible routine to realize these interactions by the collaboration of spin-orbit coupling and electron correlations [5]. Of particular interest are the 5d$^5$ iridates $A_2$IrO$_3$($A$ = Na, Li) [6] and $\alpha$-RuCl$_3$ [7–9], which are recognized as effective $j = 1/2$ Mott insulators with essential Kitaev interactions. These works soon ignited prosperous experimental and theoretical studies on such Kitaev materials (see [10–15] and references therein).

On the other hand, the non-Kitaev interactions are usually nonnegligible in candidate materials. For example, the Heisenberg ($J$) interaction is assumed to be large in Na$_2$IrO$_3$ [6], while the symmetric off-diagonal $\Gamma$ term [16], together with the distortion-induced $\Gamma'$ term [17], is essential in $\alpha$-RuCl$_3$. This leads to a prototypical JKT$\Gamma'$ model, which serves as a versatile playground to explore exotic phases and collective phenomena. These include a multi-node QSL in the vicinity of the Kitaev limit [18], a gapless QSL proposed in the honeycomb $\Gamma$ model [19–21], and a chiral spin ordering favored by a tiny $\Gamma'$ interaction in the dominated $\Gamma$ region [22].

While the spin-1/2 iridates and $\alpha$-RuCl$_3$ have been extensively studied, recent efforts are also extended to 3d$^7$ [23–26] and f$^1$ [27–29] electron states. In these materials holding large effective spins, bond-directional Ising interactions have also been proposed [30, 31]. Among them $X_3$Ni$_2$SbO$_6$(X = Li, Na) [32, 33] are candidates of $S = 1$ Kitaev materials, while Cr$_x$Te$_3$($X$ = Si, Ge) and CrI$_3$ are $S = 3/2$ ones [34–38]. In these high-spin materials, a single-ion anisotropy (SIA) coming from the partially unquenched orbital moment of the magnetic ion should also be taken into account [34, 35, 38, 39].

Recently, density functional theory calculations on CrSiTe$_3$ show that the Heisenberg interaction can be tuned significantly while the other magnetic terms are nearly unchanged under compressive strain [34, 35, 38]. Interestingly, a possible Kitaev QSL state, which is inferred from the double-peak specific heat anomaly and the associated thermal entropy plateau, is proposed to sit between a ferromagnetic (FM) phase and an antiferromagnetic (AFM) phase [35]. Their argument in mind is that the Kitaev QSL is known to display two peaks in the specific heat [40, 41], which are the reminiscence of the itinerant Majorana fermions and vison excitations [42]. Nevertheless, the proposed QSL state is far from mature for the following reasons. In the Kitaev model, intensity of the low-temperature peak in the specific heat is usually comparable to or is even larger than the high-temperature peak [40, 41, 43]. However, it is reported in Ref. [35] that the low-temperature peak is rather weak in CrSiTe$_3$. In addition, occurrence of the double-peak specific heat does not necessarily imply a QSL ground state, and several magnetically ordered states are demonstrated to possess such a specific heat anomaly in the extended Kitaev-Heisenberg model [44]. These observations motivate us to revisit the QSL state in the relevant parameter region.

In this work, we investigate the model proposed to describe CrSiTe$_3$ by the density-matrix renormalization group (DMRG) method [45–47]. As we tune the Heisen-
interactions as well as the SIA, are essential to stabilize the ferromagnetism at finite temperature. A possible minimal model Hamiltonian up to nearest-neighbor interactions is given by [31, 34, 35, 38]

\[ H = \sum_{(ij)} S_i^T \mathcal{J}_{ij} S_j + \sum_i S_i^T A S_i. \]  

The first term in Eq. (1) is known as the JKTT model. Here, \( \langle ij \rangle \) are summed over nearest neighbors, and \( \alpha (= X, Y, Z) \) represents the bond shown in Fig. 1(a). In the second term the sum runs over all sites. \( S_i = (S_i^X, S_i^Y, S_i^Z)^T \) represents the spin operators at the site \( i \). The bond-dependent interactions and SIA matrices in the \{XYZ\} coordinate system have the following form [35]

\[
\mathcal{J}^X = \begin{pmatrix}
J + K & \Gamma' & \Gamma' \\
\Gamma' & J & \Gamma \\
\Gamma' & \Gamma & J
\end{pmatrix},
\mathcal{J}^Y = \begin{pmatrix}
J & \Gamma' & \Gamma' \\
\Gamma' & J + K & \Gamma' \\
\Gamma & \Gamma' & J
\end{pmatrix},
\mathcal{J}^Z = \begin{pmatrix}
J & \Gamma' & \Gamma' \\
\Gamma & J + K & \Gamma' \\
\Gamma' & \Gamma & J + K
\end{pmatrix},
A = \frac{A_{cc}}{3} \begin{pmatrix}
1 & 1 & 1 \\
1 & 1 & 1 \\
1 & 1 & 1
\end{pmatrix}.
\]

Of important note is that in the crystallographic abc coordinate system, the SIA term could be rewritten elegantly as \( A_{cc}(S_i^c)^2 \). For \( S = 3/2 \) Kitaev materials, the mechanism for anisotropic interactions is different from the \( S = 1/2 \) case [5, 31, 34]. Therefore, the Kitaev interaction can be AFM and the off-diagonal \( \Gamma \) and \( \Gamma' \) terms could be comparable [12, 35]. We focus on the ground phase diagram of CrSiTe\(_3\) induced by compressive strain, and the parameters in (1) are referred from the supplemental material of Ref. [35]. Since \( K, \Gamma, \Gamma' \) and \( A_{cc} \) are almost independent of the strength of the strain, we take them as constants by setting \( K = 0.275 \) meV, \( \Gamma = 0.020 \) meV, \( \Gamma' = -0.083 \) meV, \( A_{cc} = 0.222 \) meV. Thus, \( J \), in unit of meV, is the only variable in the phase diagram.

The phase diagram of the model (1) induced by the strain is identified through the ground-state energy, order parameters as well as static spin structure factors (SSSFs), which are readily available in our DMRG calculation. In the simulations, we...
use different clusters to confirm that our numerical results are reliable. One should be careful whether the clusters match the magnetic orders. As shown in Fig. 1(b) and Fig. 1(c), 24-site $C_6$ cluster with periodic boundary condition and 48-site YC cylinder ($6 \times 4 \times 2$) are mainly considered in our simulations. In the calculations, the number of states we keep ranges from 1200 to 1600, and at least 12 sweeps are implemented in each calculation to ensure the convergence of DMRG results. After the calculations are converged, the utmost magnitude of truncation error in each sweep is $10^{-5}$.

**III. QUANTUM PHASE DIAGRAM**

Our DMRG results are summarized in Fig. 2. In panel (a), we plot the ground-state energy per site $E_g/N$ as a function of $J$ on the 24-site $C_6$ cluster (Fig. 1(b)) and the 48-site YC cylinder (Fig. 1(c)). Three phases are clearly detected which are separated by kinks in the energy curve. A kink in the energy curve indicates the level crossing and thus represents a first-order phase transition. Moreover, the positions of these kinks agree well on these two clusters. It is interesting to notice that the exact diagonalization (ED) on a small cluster with 12 sites [35] already provides an excellent estimate of the phase boundaries. Such a weak finite-size effect is the typical feature of first-order phase transitions. These three phases turn out to have FM, $120^\circ$ and AFM orders. One may notice that the energy of the $C_6$ cluster is lower than that of the YC cluster. This is because in the $C_6$ cluster PBC is used and more bonds are connected which contribute negative energy to $E_g$.

To gain more information on these phases, we calculate the SSSF which is defined as

$$S^{\alpha\alpha}(Q) = \frac{1}{N^2} \sum_{ij} \langle \hat{S}^\alpha_i \hat{S}^\alpha_j \rangle e^{iQ \cdot (r_i - r_j)}$$

where $N$ is the number of sites and $i, j$ run over all the sites. $\langle \cdots \rangle$ is the expectation in the ground state. The order parameter at the momenta $Q$ is then given by $M = \sqrt{\sum_{\alpha} S^{\alpha\alpha}(Q)}$. In the magnetic ordered phase, it is finite at the characteristic momentum in the thermodynamic limit. Therefore, we can tell the magnetic orders at the characteristic momenta determined by the peaks in the SSSF. In panel (b), we show the typical contour plots of the SSSF $\sum_{\alpha} S^{\alpha\alpha}(Q)$ in the three different phases. To keep the symmetry of the Brillouin zone, we use the data from the $C_6$ cluster. The data from the cylinder give the same conclusion except that the symmetry of the pattern is lower. Hence, the characteristic momenta in these three phases are $Q_{\Gamma} = (0, 0)$, $Q_K = (2\pi/3, 2\pi/(3\sqrt{3}))$, and $Q_{\Gamma'} = (2\pi/3, 2\sqrt{3}\pi/3)$, respectively, as well as others associated by the symmetry transformation of the Brillouin zone. In panel (c), we show the order parameters at the characteristic momenta as a function of $J$. The order parameters at the characteristic momenta in other phases are also shown for comparison. This figure tells us that the three phases from left to right are an FM phase, a $120^\circ$ phase and an AFM phase. At all the phase transition points, the discontinuity in the order parameters is clearly visible, indicating a first-order transition. This is consistent with the conclusions drawn from the ground-state energy.

To check the tendency of the $120^\circ$ magnetic ordering
against the system size, we calculate the order parameter of the $120^\circ$ phase on four clusters. Here, the Heisenberg interaction $J$ is taken as $-0.10$, which is close to the strength of strain $-2.34\%$ [35]. As can be seen from Fig. 3, the order parameter is rather robust and the magnetization is around one-half of the spin value $3/2$, albeit with a decreasing trend as the system size increases. The linear and quadratic fittings suggest that the magnetization is approximately $0.67(2)$ in the thermodynamic limit. It is worthy to note that the magnetization is obvious larger than the well-recognized $120^\circ$ order in the $\text{spin-1/2}$ triangular-lattice Heisenberg antiferromagnet [48], demonstrating the robustness of the $120^\circ$ order in the present honeycomb model.

IV. CLASSICAL PHASE DIAGRAMS

Large spins usually indicate weak quantum fluctuation. An $S = 3/2$ spin is considered to be near the boundary between the classical and the quantum world [49]. On the other hand, quantum fluctuation can be enhanced by anisotropic magnetic terms. So to what extent does an $S = 3/2$ spin model behave classically in frustrated systems? We will try to answer this question by a direct comparison between the quantum phase diagram and its classical counterpart. For this purpose, in this section we will establish the classical phase diagram by the parallel-tempering Monte Carlo method [50–52]. The simulations are performed on a $12 \times 12 \times 2$ rhombic cluster. The spin configurations are updated by the heat bath algorithm.

![Figure 4](image)

Figure 4. (a) The blue open circles are the ground energy obtained by the Monte Carlo. There are four phases in the phase diagram, i.e., an FM phase, a zigzag phase, a $120^\circ$ phase and an AFM phase from left to right. The transition points are $J \approx -0.32063$, $-0.26074$, $0.020$. The red lines are analytical results of the ground-state energy. (b) The SSSFs in the corresponding phases.

In the classical limit, each spin is regarded as an $O(3)$ vector in real space. We use the Monte Carlo method to find the ground state. The corresponding ground-state energy and spin configurations are then readily available. In Fig. 4 (a), we plot the classical ground-state energy per spin $E_{gg}/(NS^2)$ as a function of $J$. Interestingly, it is easy to see that the ground-state energy is a piece-wise function of $J$ and four phases can be determined in the phase diagram. The spin configurations in each phase directly tell us the characteristic features, which are an FM phase, a zigzag (ZZ) phase, a $120^\circ$ phase and an AFM phase, respectively, from left to right. Moreover, the expressions of the classical energy can be given analytically

\[
E_{\text{FM}} = \frac{S^2}{2}[3J + K - \Gamma - 2\Gamma' + P(\Gamma, \Gamma', A_{cc})\Theta(P(\Gamma, \Gamma', A_{cc}))],
\]

\[
E_{\text{ZZ}} = \frac{S^2}{4}(2J - \Gamma + 2\Gamma' + 2A_{cc}) + S^2Q(K, \Gamma, \Gamma', A_{cc}),
\]

\[
E_{120^\circ} = \frac{S^2}{2}(-K - 2\Gamma + 2\Gamma'),
\]

\[
E_{\text{AFM}} = \frac{S^2}{2}(-3J - K + \Gamma + 2\Gamma'),
\]

where $\Theta(x)$ is a step function and $P(\Gamma, \Gamma', A_{cc})$ and $Q(K, \Gamma, \Gamma', A_{cc})$ have the following forms

\[
P = 3\Gamma + 6\Gamma' + 2A_{cc},
\]

\[
Q = -\frac{1}{12}\sqrt{32(K - \Gamma + \Gamma')^2 + (6A_{cc} + 2K + 7\Gamma + 2\Gamma')^2}.
\]

Here, the occurrence of the step function $\Theta(x)$ in the FM phase is because that its classical moment direction could either lie in the $ab$ plane (when $x > 0$) or point along the $c$ direction (when $x < 0$). In Fig. 4(a), we also plot these analytical results for comparison, which are marked by red lines. The overlap of these data demonstrates the reliability of our numerical results. Just as in the quantum case, the phases can also be identified by the SSSF, which are plotted in Fig. 4(b). The SSSF in the FM phase, the $120^\circ$ phase and the AFM phase show a close resemblance to the quantum cases in Fig. 2(b). In particular, the heights of characteristic peaks are close in their value, indicating that the Eq. (1) in the quantum case is close to its classical limit.

![Figure 5](image)

Figure 5. Classical ground-state energy is plotted as a function of $J$ for $A_{cc} = 0.000, 0.100, 0.150, 0.180$ and $0.222$. We can see that when $A_{cc} \leq 0.150$, zigzag phase disappears.

From the Fig. 2 and Fig. 4, we can see that the transition point from the $120^\circ$ phase to the AFM phase agrees well in both the quantum and classical cases, which are $J = 0.022(2)$ and $J = 0.020$, respectively. However, an unexpected zigzag phase emerges in the classical phase diagram, which sits between the FM phase and $120^\circ$ phase. We have checked that when $A_{cc}$ is smaller than $0.150$, the zigzag phase disappears as shown in Fig. 5. This is not surprising because the classical spin corresponds to $S \rightarrow \infty$ and in the $S = 3/2$ case the
quantum fluctuation can not be completely neglected and thus is expected to shift the phase boundary.

V. FINITE-SIZE EFFECT IN 12-SITE CLUSTERS

Generally speaking, the QSL is known as a nonmagnetic state with fractionalized excitations, which could be manifested by topological entanglement entropy [53, 54], continuum excitation spectrum, and thermodynamic signatures [55]. In the Kitaev QSL, the spins fractionalize into localized and itinerant Majorana fermions, resulting in a double-peak specific heat anomaly [40, 41]. Nevertheless, the double-peak structure of specific heat found in the previous work on a 12-site cluster can not simply be interpreted as a signature of spin fractionalization [35]. Instead, it may originate from the artificial frustration due to the incompatible cluster with respect to the underlying magnetic ordering.

We have illustrated in Sec. III that the intermediate region is a 120° magnetically state rather than the QSL. For comparison, we fix $J = -0.10$ again where the ground state locates deep in the 120° phase. To be compatible with the six-sublattice 120° ordering, the number of lattice sites in a cluster should be a multiple of six. Even though such condition is satisfied, the cluster shape should be chosen carefully so as to match the magnetic order. As stated in Ref. [35], the 12A cluster in Fig. 6(a) is likely used in the specific heat calculation. Unfortunately, this geometry does not coordinate the 120° order in the vertical boundary, which inevitably introduces the artificial frustration and ruins the magnetic ordering significantly in the small cluster. To confirm this, we have performed the DMRG calculations on the 12A cluster. The total ground-state energy is -4.835103988, and the corresponding SSSF is shown in the first panel of Fig. 6(b). There is not a well-defined peak in the Brillouin zone, in accord with the proposed QSL inferred from the specific heat.

By contrast, a slightly modification of the geometry will remove the mismatch of the spins at boundaries, see 12B cluster in Fig. 6(a). Its SSSF in the second panel of Fig. 6(b) exhibits a well-defined peak at $Q_K$ point, in line with the desirable 120° order. We have also considered two other 12-site clusters (termed 12C and 12D) which match the 120° ordering, and their SSSFs with peaks at $Q_K$ point are shown in the last row in Fig. 6(b). Notably, All the clusters that coordinate the 120° spin textures (i.e., 12B, 12C, and 12D) share a same total energy of -5.535684265. (b) SSSF on the four clusters, which are 12A (top left), 12B (top right), 12C (bottom left) and 12D (bottom right).

VI. SUMMARY AND OUTLOOK

In this work, we study an $S = 3/2$ JKT$^\prime$+SIA model proposed to describe CrSiTe$_3$ by using the DMRG method. Upon increasing the strain, the Heisenberg ($J$) interaction varies significantly while the remaining ones almost keep unchanged. We thus map out the $J$-dependent phase diagram as a mimicry of the strain process. When $J$ varies from negative to positive, three magnetically ordered phases are found, including an FM phase, a 120° phase and an AFM phase. All the phase transitions are of the first order, characterized by the kinks in the ground-state energy and discontinuities in the order parameters. Experientially, the quantum fluctuation is expected to be strongly suppressed in the models with a large spin of $S = 3/2$, and the physics should bear resemblance to its classical counterpart [36]. We have strengthened this conclusion by performing the classical Monte Carlo simulations on the same model, with the exception that a zigzag phase is found between the FM phase and the 120° phase. The zigzag phase is sensitive to the SIA, and its territory shrinks gradually and it disappears eventually as SIA decreases.

It was argued in a previous work [35] that there is a possibly QSL, which is inferred from the double-peak structure of specific heat and the related plateau of thermal entropy on a 12-site cluster. Curiously, the proposed QSL region hap-
pens to coincide with the 120° phase identified in our work. In fact, the double-peak specific heat anomaly solely could not legitimate the Kitaev QSL. To begin with, the low-temperature peak found in Ref. [35] is rather weak when compared to the high-temperature analogy, which is in contrast to the current wisdom that the magnitudes of the two peaks are comparable in the Kitaev QSL [40, 41]. In addition, it is unclear if such a low-temperature peak will still persist with the increasing of the system size. Even though the double-peak structure indeed exists in the ground state, it is still not enough to authenticate a QSL as magnetically ordered states could also exhibit such an abnormal phenomenon [44]. In this regard, it is imperative to check if the ground state is disordered or not. Our large-scale DMRG calculation, however, suggests that the ground state is a magnetically ordered 120° state with a large order parameter.

Although our work implies that QSL is unfavorable in CrSiTe₃, three magnetic ordered phases are found under the strain. These ordered states may persist to finite temperature because the Mermin-Wagner theorem does not forbid such orders in the model (1). In this sense, this material offers a playground to study these magnetic orders and phase transitions in low dimensions[56]. Recent experiment and \textit{ab initio} calculations support Cr-based ferromagnetic van der Waals materials are \( S = 3/2 \) Kitaev materials [34–36, 38], the model here just takes the nearest-neighbor interactions into account, the other possible interactions are not considered. Due to the complexity of real material, the neutron spectra of CrI₃ can be fitted by different models [37]. In CrSiTe₃, we can not exclude other possible terms such as the next-nearest neighbors in the Hamiltonian. Moreover, the model Hamiltonian (1) and corresponding parameters remain to be verified by experiments. We expect our results can be verified in future experiments and be helpful to understand the magnetic behavior in two-dimensional ferromagnetism in high-spin Kitaev materials.

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[1] A. Kitaev, Anyons in an exactly solved model and beyond, \textit{Ann. Phys.} \textbf{321}, 2 (2006).
[2] F. W. Anderson, Resonating valence bonds: A new kind of insulator?, \textit{Mater. Res. Bull.} \textbf{8}, 153 (1973).
[3] L. Balents, Spin liquids in frustrated magnets, \textit{Nature (London)} \textbf{464}, 199 (2010).
[4] J. Wen, S.-L. Yu, S. Li, W. Yu, and Jian-Xin Li, Experimental identification of quantum spin liquids, \textit{npj Quantum Mater.} \textbf{4}, 12 (2019).
[5] G. Jackeli and G. Khaliullin, Mott insulators in the strong spin-orbit coupling limit: From Heisenberg to a quantum compass-sand Kitaev models, \textit{Phys. Rev. Lett.} \textbf{102}, 017205 (2009).
[6] J. Chaloupka, G. Jackeli, and G. Khaliullin, Kitaev-Heisenberg model on a honeycomb lattice: Possible exotic phases in iridium oxides A1IrO₃, \textit{Phys. Rev. Lett.} \textbf{105}, 027204 (2010).
[7] 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, \( \alpha \)-RuCl₃: A spin-orbit assisted mott insulator on a honeycomb lattice, \textit{Phys. Rev. B} \textbf{90}, 041112 (2014).
[8] H.-S. Kim, V. S. V., A. Catuneanu, and H.-Y. Kee, Kitaev magnetism in honeycomb \( \alpha \)-RuCl₃ with intermediate spin-orbit coupling, \textit{Phys. Rev. B} \textbf{91}, 241110 (2015).
[9] 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, S. Bhattacharjee, D. L. Kovrizhin, R. Moessner, D. A. Tennant, D. G. Mandrus, and S. E. Nagler, Proximate Kitaev quantum spin liquid behaviour in a honeycomb magnet, \textit{Nat. Mater.} \textbf{15}, 733 (2016).
[10] Wei Wang, Zhao-Yang Dong, Shun-Li Yu, and Jian-Xin Li, Theoretical investigation of magnetic dynamics in \( \alpha - \text{RuCl}_3 \), \textit{Phys. Rev. B} \textbf{96}, 115103 (2017).
[11] S. M. Winter, K. Riedl, P. A. Maksimov, A. L. Chernyshev, A. Honecker, and R. Valentì, Breakdown of magnons in a strongly spin-orbital coupled magnet, \textit{Nat. Commun.} \textbf{8}, 1152 (2017).
[12] P. A. Maksimov and A. L. Chernyshev, Rethinking \( \alpha - \text{RuCl}_3 \), \textit{Phys. Rev. Research} \textbf{2}, 033011 (2020).
[13] H. Li, H.-K. Zhang, J. Wang, H.-Q. Wu, Y. Gao, D.-W. Qu, Z.-X. Liu, S.-S. Gong, and W. Li, Identification of Magnetic Interactions and High-field Quantum Spin Liquid in \( \alpha - \text{RuCl}_3 \), \textit{Nat. Commun.} \textbf{12}, 4007 (2021).
[14] M. Hermanns, I. Kimchi, and J. Knolle, Physics of the Kitaev model: Fractionalization, dynamic correlations, and material connections, \textit{Annu. Rev. Condens. Matter Phys.} \textbf{9}, 17 (2018).
[15] H. Takagi, T. Takayama, G. Jackeli, G. Khaliullin, and S. E. Nagler, Concept and realization of Kitaev quantum spin liquids, \textit{Nat. Rev. Phys.} \textbf{1}, 264 (2019).
[16] J. G. Rau, E. K.-H. Lee, and H.-Y. Kee, Generic spin model for the honeycomb iridates beyond the Kitaev limit, \textit{Phys. Rev. Lett.} \textbf{112}, 077204 (2014).
[17] J. G. Rau and H.-Y. Kee, Trigonal distortion in the honeycomb iridates: Proximity of zigzag and spiral phases in \( \text{Na}_2\text{IrO}_3 \), arXiv:1408.4811 (2014).
[18] J. Wang, B. Normand, and Z.-X. Liu, One Proximate Kitaev Spin Liquid in the \( K'-J-\Gamma \) Model on the Honeycomb Lattice, \textit{Phys. Rev. Lett.} \textbf{123}, 197201 (2019).
[19] Q. Luo, J. Zhao, H.-Y. Kee and X. Wang, Gapless quantum spin liquid in a honeycomb \( \Gamma \) magnet, \textit{npj Quantum Mater.} \textbf{6}, 57 (2021).
[20] A. Catuneanu, Y. Yamaji, G. Wachtel, Y.-B. Kim, and H.-Y. Kee, Path to stable quantum spin liquids in spin-orbit coupled correlated materials, \textit{npj Quantum Mater.} \textbf{3}, 23 (2018).
[21] M. Gohlke, G. Wachtel, Y. Yamaji, F. Pollmann, and Y. B. Kim, Spin quantum liquid signatures in Kitaev-like frustrated magnets, \textit{Phys. Rev. B} \textbf{97}, 075126 (2018).
[22] Q. Luo, P. P. Stavropoulos, and H.-Y. Kee, Spontaneous Chiral-Spin Ordering in Spin-Orbit Coupled Honeycomb Magnets, arXiv:2010.11233 (2020).
[23] R. Sano, Y. Kato, and Y. Motome, Kitaev-Heisenberg hamilt-
nian for high-spin $d^7$ mott insulators, Phys. Rev. B 97, 014408 (2018).

[24] H. Liu and G. Khaliullin, Pseudospin exchange interactions in $d^7$ cobalt compounds: Possible realization of the Kitaev model, Phys. Rev. B 97, 014407 (2018).

[25] H. K. Vivanco, B. A. Trump, C. M. Brown, and T. M. McQueen, Competing antiferromagnetic-ferromagnetic states in a $d^7$ Kitaev honeycomb magnet, Phys. Rev. B 102, 224411 (2020).

[26] M. Songvilay, J. Robert, S. Petit, J. A. Rodriguez-Rivera, W. D. Ratcliff, F. Damay, V. Balédent, M. Jiménez-Ruiz, P. Lejay, E. Pachoud, A. Hadj-Azzem, V. Simonet, and C. Stock, Kitaev interactions in the Co honeycomb antiferromagnets $Na_3Co_2SbO_6$ and $Na_2Co_2TeO_6$, Phys. Rev. B 102, 224429 (2020).

[27] S.-H. Jang, R. Sano, Y. Kato, and Y. Motome, Antiferromagnetic Kitaev interaction in $f$-electron based honeycomb magnets, Phys. Rev. B 99, 241106 (2019).

[28] S.-H. Jang, R. Sano, Y. Kato, and Y. Motome, Computational design of $f$-electron Kitaev magnets: Honeycomb and hyper-honeycomb compounds $A_2PrO_3$ ($A = $ alkali metals), Phys. Rev. Materials 4, 104420 (2020).

[29] Y. Motome, R. Sano, S. Jang, Y. Sugita, and Y. Kato, Material designs of Kitaev spin liquids beyond the Jackeli-Khalilullin mechanism, J. Phys. Condens. Matter 32, 404001 (2020).

[30] P. P. Stavropoulos, D. Pereira, and H.-Y. Kee, Microscopic mechanism for a higher-spin Kitaev model, Phys. Rev. Lett. 123, 057203 (2019).

[31] P. P. Stavropoulos, X. Liu, and H.-Y. Kee, Magnetic anisotropy in spin-3/2 with heavy ligand in honeycomb mott insulators: Application to CrI$_3$, Phys. Rev. Research 3, 013216 (2021).

[32] E. A. Zvereva, M. I. Stratan, Y. A. Ovchenkov, V. B. Nalbandyan, J.-Y. Lin, E. L. Vavilova, M. F. Iakovlev, M. Abdel-Hafez, A. V. Silhanek, X.-J. Chen, A. Stroppa, S. Picozzi, H. O. Jeschke, R. Valentí, and A. N. Vasiliev, Zigzag antiferromagnetic quantum ground state in monolinic honeycomb lattice antimonates $A_3Ni_2SbO_6$ ($A = $ Li, Na), Phys. Rev. B 92, 144401 (2015).

[33] A. I. Kurbakov, A. N. Korshunov, S. Y. Podcheezertsev, A. L. Malyshev, M. A. Evstigneeva, F. Damay, J. Park, C. Koo, R. Klingeler, E. A. Zvereva, and V. B. Nalbandyan, Zigzag spin structure in layered honeycomb $Li_3Ni_2SbO_6$: A combined diffraction and antiferromagnetic resonance study, Phys. Rev. B 96, 024417 (2017).

[34] C. Xu, J. Feng, H. Xiang, and L. Bellaiche, Interplay between Kitaev interaction and single ion anisotropy in ferromagnetic CrI$_3$ and CrGeTe$_3$ monolayers, npj Comput. Mater. 4, 57 (2018).

[35] 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. 125, 087205 (2020).

[36] I. Lee, F. G. Utermohlen, D. Weber, K. Hwang, C. Zhang, J. van Tol, J. E. Goldberger, N. Trivedi, and P. C. Hammel, Fundamental spin interactions underlying the magnetic anisotropy in the kitaev ferromagnet CrI$_3$, Phys. Rev. Lett. 124, 017201 (2020).

[37] L. Chen, J.-H. Chung, T. Chen, C. Duan, A. Schneidewind, I. Radyelitskyi, D. J. Voneshen, R. A. Ewings, M. B. Stone, A. I. Kolesnikov, B. Winn, S. Chi, R. A. Mole, D. H. Yu, B. Gao, and P. Dai, Magnetic anisotropy in ferromagnetic CrI$_3$, Phys. Rev. B 101, 134418 (2020).

[38] C. Bacaksiz, D. Šabani, R. M. Menezes, and M. V. Milošević, Distinctive magnetic properties of CrI$_3$ and CrBr$_3$ monolayers caused by spin-orbit coupling, Phys. Rev. B 103, 125418 (2021).

[39] J. L. Lado and J. Fernández-Rossier, On the origin of magnetic anisotropy in two dimensional CrI$_3$, 2D Mater. 4, 035002 (2017).

[40] J. Nasu, M. Udagawa, and Y. Motome, Thermal fractionalization of quantum spins in a Kitaev model: Temperature-linear specific heat and coherent transport of Majorana fermions, Phys. Rev. B 92, 115122 (2015).

[41] 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).

[42] J. Knolle, D. L. Kovrizhin, J. T. Chalker, and R. Moessner, Dynamics of a Two-Dimensional Quantum Spin Liquid: Signatures of Emergent Majorana Fermions and Fluxes, Phys. Rev. Lett. 112, 207203 (2014).

[43] Q. Luo, S. Hu, and H.-Y. Kee, Unusual excitations and double-pole specific heat in a bond-alternating spin-1 K' chain, arXiv:2104.10725 (2021) [Phys. Rev. Research (to be published)].

[44] Y. Yamaji, T. Suzuki, T. Yamada, S.-i. Suga, N. Kawashima, and M. Imada, Clues and criteria for designing Kitaev spin liquid revealed by thermal and spin excitations of honeycomb iridates $Na_2IrO_3$, Phys. Rev. B 93, 174425 (2016).

[45] S. R. White, Density matrix formulation for quantum renormalization groups, Phys. Rev. Lett. 69, 2863 (1992).

[46] J. Peschel, X. Q. Wang, M. Kaulke, and K. Hallberg, Density-matrix renormalization (Springer, Berlin, 1999).

[47] U. Schollwöck, The density-matrix renormalization group, Rev. Mod. Phys. 77, 259 (2005).

[48] Qian Li, Hong Li, Rize Zhao, Hong GANG Luo, Z. Y. Xie, Magnitization of the spin-1/2 Heisenberg antiferromagnet on the triangular lattice, arXiv:2009.03765 (2020).

[49] M. Songvilay, S. Petit, F. Damay, G. Roux, N. Qureshi, H. C. Walker, J. A. Rodriguez-Rivera, B. Gao, S.-W. Cheong, and C. Stock, From one to two-magnon excitations in the $S = 3/2$ magnet $\beta$-C$_3$Cr$_2$O$_4$, Phys. Rev. Lett. 126, 017201 (2021).

[50] K. Hukushima and K. Nemoto, Exchange Monte Carlo method and application to spin glass simulations, J. Phys. Soc. Jpn. 65, 1604 (1996).

[51] A. Mitsutake and Y. Okamoto, Replica-exchange simulated tempering method for simulations of frustrated systems, Chem. Phys. Lett. 332, 131 (2000).

[52] L. Janssen, E. C. Andrade, and M. Vojta, Honeycomb-Lattice Heisenberg-Kitaev Model in a Magnetic Field: Spin Canting, Metamagnetism, and Vortex Crystals, Phys. Rev. Lett. 117, 277202 (2016).

[53] A. Kitaev and J. Preskill, Topological Entanglement Entropy, Phys. Rev. Lett. 96, 110404 (2006).

[54] Michael Levin and Xiao-Gang Wen, Detecting Topological Order in a Ground State Wave Function, Phys. Rev. Lett. 96, 110405 (2006).

[55] Seung-Hwan Do, Sang-Youn Park, Junki Yoshitake, Joji Nasu, Yukitoshi Motome, Yong Seung Kwon, D. T. Adroja, D. J. Voneshen, Kyoo Kim, T.-H. Jang, J.-H. Park, Kwang-Yong Choi, and Sungdae Ji, Majorana fermions in the Kitaev quantum spin system $alpha$-RuCl$_3$, Nat. Phys. 13, 1079 (2017).

[56] Zefang Li, Dong-Hong Xu, Xue Li, Hai-Jun Liao, Xuekui Xi, Yi-Cong Yu, Wenhong Wang, Abnormal critical fluctuations revealed by magnetic resonance in the two-dimensional ferromagnetic insulators, arXiv:2101.02440.