

# Tangle of spin double helices in the honeycomb Kitaev- $\Gamma$ model

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We investigate the ground-state nature of the honeycomb Kitaev- $\Gamma$  model in the material-relevant parameter regime through a combination of analytics and classical and quantum simulations. We find the classical model is imprinted with a tangle of highly structured spin double helices. This helix tangle consists of 18 inequivalent helices and features modulation of multiple rotation axes, an anisotropy in spacial periodicities indicative of spontaneous symmetry breaking, and a  $\text{sgn}(\Gamma)$ -determined chirality pattern. Infinite projected entangled pair state simulations with clusters of up to 36 sites identify hallmarks of this unprecedented many-body order in the quantum spin-1/2 model. Our findings provide a fresh perspective of the Kitaev- $\Gamma$  model and enrich the physics of the Kitaev magnetism.

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## I. INTRODUCTION

The honeycomb Kitaev- $\Gamma$  Hamiltonian is a paradigmatic model for the physics of two-dimensional Kitaev magnets [1–7]. The Kitaev exchange leads to an exactly solvable spin liquid [8] and can be realized in  $d$ -electron transition-metal compounds with edge-shared geometry [1]. However, symmetries of real materials permit a generic existence of a Heisenberg and an off-diagonal  $\Gamma$  term [3–5], which may dramatically modify the desired spin-liquid ground state. In particular, in the prime candidate material  $\alpha$ - $\text{RuCl}_3$  [9–18], the  $\Gamma$  exchange is comparable with the Kitaev one [19–25]. Theoretical studies further suggest the competition between  $\Gamma$  and Kitaev interactions can induce novel exotic phases beyond a single spin liquid [24–41], while the Heisenberg term stabilizes regular magnetic orders [2]. Nonetheless, state-of-the-art numerical methods, including exact diagonalization [26,39], tensor network [27–30], variational Monte Carlo [31], and functional renormalization group [32] techniques, have yielded highly diverse results for the spin-1/2 Kitaev- $\Gamma$  model, leaving the quantum phase diagram obscure. Consensus exists for a Kitaev spin liquid (KSL) at finite  $\Gamma$ , but even its extent is debated.

In this article we conduct large-scale classical and quantum simulations to unravel the ground-state nature of the honeycomb Kitaev- $\Gamma$  model in the most puzzling yet material-relevant regimes. We demonstrate that the classical ground state imprints a tangle of emergent *spin double helices* (Fig. 1).

It exhibits several prominent characteristics, including modulation of multiple helical axes, anisotropic periodicities, and a  $\text{sgn}(\Gamma)$ -determined chirality patterns, as summarized in the *Ansatz* Eq. (5). The emergence of such a sophisticated helix tangle poses fundamental challenges to quantum algorithms and is the source of noncoherent observations in the literature. Nevertheless, our infinite projected entangled pair states (iPEPS) calculations identify signatures of this unprecedented many-body order, showing that it can survive quantum fluctuations in the spin-1/2 case.

## II. MODEL AND SYMMETRY

The Kitaev- $\Gamma$  model on a honeycomb lattice comprises two bond-dependent terms: A directional Ising-type interaction and a symmetric off-diagonal  $\Gamma$  interaction:

$$H = \sum_{\langle ij \rangle_\gamma} K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha). \quad (1)$$

Here,  $\gamma$  labels the three different types of bonds, and  $\alpha, \beta, \gamma \in \{x, y, z\}$  are mutually exclusive. For example, the local Hamiltonian on a  $z$  bond reads  $H_z = K S_i^z S_j^z + \Gamma (S_i^x S_j^y + S_i^y S_j^x)$ . For simplicity we assume the coupling strengths on the three bonds are uniform. Nevertheless, one expects that a small bond anisotropy (e.g.,  $K_z \neq K_{x,y}$ ) does not alter the nature of the underlying phases as in the pure Kitaev model [8].

The Hamiltonian Eq. (1) has a *hidden global* symmetry that intertwines. This is seen by rewriting Eq. (1) as  $H = \sum_{\langle ij \rangle_\gamma} \mathbf{S}_i \cdot \hat{J}_{ij}^\gamma \mathbf{S}_j$ , with three exchange matrices:

$$\hat{J}_{ij}^x = \begin{bmatrix} K & & \\ & \Gamma & \\ & & \Gamma \end{bmatrix}, \quad \hat{J}_{ij}^y = \begin{bmatrix} & K & \\ & & \Gamma \\ \Gamma & & \end{bmatrix}, \quad \hat{J}_{ij}^z = \begin{bmatrix} & & \Gamma \\ \Gamma & & \\ & K & \end{bmatrix}. \quad (2)$$

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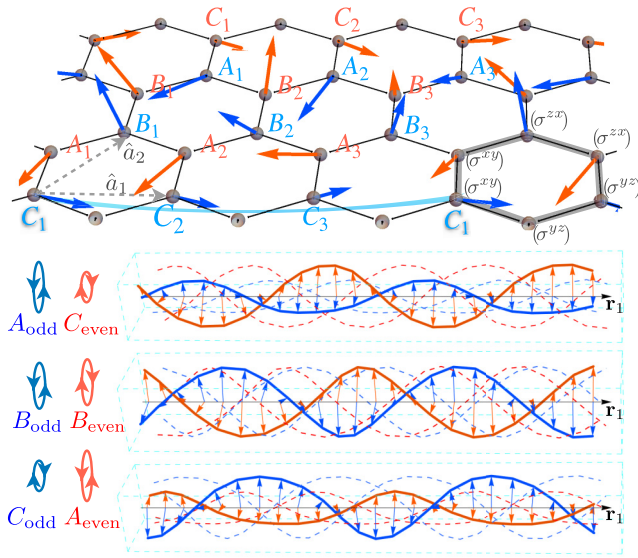


FIG. 1. A classical ground state of the honeycomb Kitaev- $\Gamma$  model at  $\Gamma = -K$  on a  $L = 72$  lattice. Upper panel: Structure of the  $3 \times 3$  supercell determined from the helical axes (longitudinal components) of spins.  $A, B, C$  label three independent directions of the helical axes, and  $j = 1, 2, 3$  distinguish different orientations due to the hidden symmetry. Blue and red colors mark the odd and even honeycomb sublattices and the helices' chirality. A supercell specifies 18 inequivalent helices as spins form a helix *only if* they belong to the same sublattice, e.g., the linked blue  $C_1$  spins. The gray hexagon marks a unit cell of the six-site hidden symmetry transformation. Lower panel: Spiral (transverse) components of the 18 helices in their respective sublattice basis. Each helix consists of  $\frac{1}{3}$  spins due to the  $3 \times 3$  supercell structure. The helical pitches are *spontaneously anisotropic* and are  $\frac{1}{6}$  and  $\frac{1}{3}$  supercells in size along the directions of the two lattice vectors  $\hat{a}_{1,2}$ . Cycles on the side reflect the strength and staggered chirality of the corresponding helix. The helix ensemble can be viewed as nine pairs of double helices.

One can verify that  $\hat{J}_{ij}^\gamma$ , and hence the local environment of spins, is invariant under the transformations

$$\sigma_i^{\alpha\beta} \hat{J}_{ij}^\gamma \sigma_j^{\alpha\beta} = \hat{J}_{ij}^\gamma, \quad (3)$$

$$\sigma_i^{\alpha\gamma} \hat{J}_{ij}^\gamma \sigma_j^{\alpha\gamma} = \hat{J}_{ij}^\gamma, \quad (4)$$

where  $\sigma_i^{\alpha\beta}$  denotes spin reflections transforming a spin as  $\sigma_i^{\alpha\beta} S_i^\gamma = -S_i^\gamma$ . These transformations alternate the three spin reflections in accordance with the index rule of Eqs. (3) and (4), until covering the entire lattice (see the Supplemental Material, SM [42], for an example). In contrast to usual global symmetries, such as the time reversal and homogeneous spin rotations, this hidden symmetry can modify correlations in the system and make two *distinct* orders degenerate.

### III. SELECTED PARAMETER REGION

The main open problems in constructing the phase diagram of the spin-1/2 Kitaev- $\Gamma$  model can be viewed from three fronts. First, although mounting numerical evidence suggests an extended KSL regime under a small  $\Gamma$ , different algorithms find different extents of the regime [28–32]. Furthermore,

in the large  $\Gamma$  limits, the fate of classical  $\Gamma$  spin liquids [37,43,44] is unsettled and prone to simulation techniques [45,46]. Away from these two limits, a diversity of candidate ground states have been suggested, including non-Kitaev spin liquids, quantum paramagnets, incommensurate or spiral orders, and various magnetic states [27–32]. As these open problems cannot be resolved in a single work, we focus our efforts on the regime where both interactions are sizable and competing, which is of the highest relevance for real materials. For convenience we parametrize the two interactions using an angle parameter  $\theta$  as  $K = \sin \theta$ ,  $\Gamma = \cos \theta$ . We consider the frustrated region with a ferromagnetic  $K$  and an antiferromagnetic  $\Gamma$  ( $1.5\pi < \theta < 2\pi$ ) as realized in the  $d$ -electron Ir- and Ru-based compounds [23,25,47]. Through a mapping  $\theta \rightarrow \theta + \pi$  and simultaneously  $\mathbf{S}_{2i} \rightarrow \mathbf{S}_{2i}$ ,  $\mathbf{S}_{2i+1} \rightarrow -\mathbf{S}_{2i+1}$  [48], the results also enlighten the physics at  $0.5\pi < \theta < \pi$ , where  $2i$  and  $2i+1$  denote the even and odd honeycomb sublattices, respectively. Other parameter regions with  $\theta \in (0, 0.5\pi)$  and  $(\pi, 1.5\pi)$  are unfrustrated and develop strong  $120^\circ$ -type magnetic orders [26,37].

### IV. TANGLE OF SPIN HELICES

We first discuss the classical ground states in the selected parameter regime, which is crucial for understanding the quantum ground states. The growing interest in the classical Kitaev- $\Gamma$  model is also rewarded with rich physics [35–41]. In particular, two recent works [35,39] based on analysis of small systems reported various large-unit-cell states, including two degenerate 6- and 18-site structures. By examining large systems at temperature  $T = 10^{-3}\sqrt{K^2 + \Gamma^2}$ , these two states were further mapped to a frustrated phase spanning over  $1.58\pi \lesssim \theta < 2\pi$  [37]. This phase can be understood by the competition between two classical spin liquids, and its magnetization exhibits an intrinsic undersaturation indicating the lack of perfect translationally invariant order [37].

We now reveal that the missing magnitude encodes the essential nature of the classical ground state, which only manifests at very large lattices and cold temperatures. We utilize parallel tempering Monte Carlo methods to reach temperatures down to  $T = 10^{-5}\sqrt{K^2 + \Gamma^2}$  for systems of linear size up to  $L = 72, 90, 108$ , under periodic boundary conditions. We further cool the system to  $T \rightarrow 0$  by eliminating remaining thermal noise; see the SM [42] for details of simulations.

To understand the structure of the ordering, we first discuss its longitudinal components, which lead to a  $3 \times 3$  supercell containing 18 sublattices as depicted in Fig. 1. The corresponding magnetic moments  $\mathbf{m}_\mu^\parallel$  are obtained by averaging spins over all supercells as  $\mathbf{m}_\mu^\parallel = \frac{1}{N_{\text{cell}}} \sum_{\text{cells}} \mathbf{S}_\mu$ , where  $\mu$  distinguishes the 18 sublattices. These magnetic moments are the source of the stable  $\frac{2}{3}\mathbf{M}$  magnetic Bragg peaks reported in the literature [35,37] and also act as the rotation axes of the highly structured spin helices.

There are two degenerate magnetization patterns due to the hidden symmetry. The example in the upper panel of Fig. 1 shows a relatively simpler pattern whose helical axes fall into three different orientations, labeled with italic letters  $A, B, C$ . In this case, all  $A_j$  spins have identical longitudinal components  $\mathbf{S}_{A_j}^\parallel = \mathbf{m}_{A_j}^\parallel$ ; similarly for  $B_j$  and  $C_j$  spins. Its

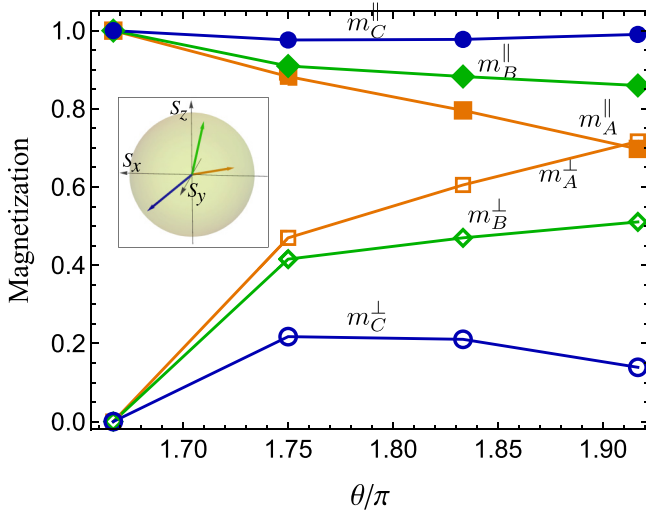


FIG. 2. Magnetizations of the classical spin-helix tangle with comparable Kitaev and  $\Gamma$  interaction,  $K = \sin \theta$ ,  $\Gamma = \cos \theta$ . The longitudinal and spiral components of the order are measured by  $m_{A,B,C}^{\parallel}$  and  $m_{A,B,C}^{\perp}$ , respectively, leading to saturated total magnetizations  $(m_{A,B,C}^{\parallel})^2 + (m_{A,B,C}^{\perp})^2 = 1$ . The spiral component at  $\theta \approx 1.67\pi$  is small but nonvanishing (see Fig. 3). The inset exemplifies the orientations of  $\mathbf{m}_A^{\parallel}$ ,  $\mathbf{m}_B^{\parallel}$ , and  $\mathbf{m}_C^{\parallel}$  (distinguished by colors) at  $\theta = 1.75\pi$ .

degenerate state is obtained by applying the transformation Eqs. (3) and (4) as indicated by the gray hexagon, leading to nine distinct rotation axes distinguished by the numerical subscripts  $j = 1, 2, 3$ . Since the hidden symmetry is exact, one can always infer all helical axes from three independent moments  $\mathbf{m}_{A,B,C}^{\parallel}$ . These magnetic moments are noncollinear and noncoplanar and vary with  $K/\Gamma$  in both magnitude and orientation. Figure 2 measures their magnitudes  $m_{A,B,C}^{\parallel}$  in the helical phase and shows an explicit example of their orientations. A procedure to semianalytically estimate their directions is provided in the SM [42].

It is important to note that the supercell is only defined for the longitudinal order. The full ordering does not have a translational symmetry due to its incommensurate transverse components, as illustrated in the lower panel of Fig. 1. Moreover, the 18 helices specified by 18 sublattices are *inequivalent*, despite that their rotation axes may have the same orientations. Namely, there is a  $3 \times 3$  modulation in the helical axes, and spins form a helix *only if* they belong to the same sublattice instead of with their neighbors.

To describe the spiral components, it is convenient to work in sublattice coordinates where  $\mathbf{m}_{\mu}^{\parallel}$  is rotated to  $\tilde{\mathbf{m}}_{\mu}^{\parallel} = (0, 0, m_{\mu}^{\parallel})$  for each sublattice. Then the spiral component of a spin can be formulated by an *Ansatz*:

$$\tilde{\mathbf{S}}_{\mu,\mathbf{r}}^{\perp} = m_{\mu}^{\perp} \left( \cos \frac{6\pi}{L} (2r_1 + r_2), \text{sgn}(-\Gamma)^{\eta} \sin \frac{6\pi}{L} (2r_1 + r_2), 0 \right). \quad (5)$$

Here,  $m_{\mu}^{\perp} = \frac{1}{N_{\text{cell}}} \sum_{\text{cells}} |\tilde{\mathbf{S}}_{\mu}^{\perp}|$  defines a spiral magnetization, which fulfils  $(m_{\mu}^{\parallel})^2 + (m_{\mu}^{\perp})^2 = 1$  as measured in Fig. 2;  $\mathbf{r} = (r_1, r_2)$  with  $r_1, r_2 = 0, 1, \dots, \frac{L}{3} - 1$  labels the  $3 \times 3$

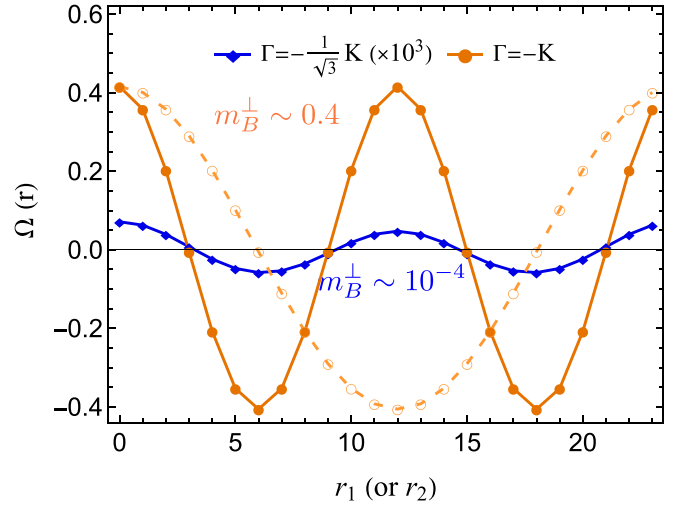


FIG. 3. The spiral correlation function  $\Omega_{\mu}(\mathbf{r})$  for B helices at  $\Gamma = -K/\sqrt{3}$  (blue; rescaled by  $10^3$ ) and  $-K$  (orange) ( $\theta = 1.67\pi, 1.75\pi$ ). The oscillation and the amplitude reflect the periodicity of the helices and strength of the spiral magnetization  $m_B^{\perp}$ , respectively. Filled and open symbols are measured along spontaneously chosen lattice  $\mathbf{r}_1$  and  $\mathbf{r}_2$  directions, with pitch sizes of  $\frac{L}{6}$  and  $\frac{L}{3}$  supercells, respectively. A  $L = 72$  lattice is considered, and  $r_1, r_2 = 0, 1, \dots, \frac{L}{3} - 1$ .

supercells, and  $\eta = 0, 1$  distinguishes the even and odd honeycomb sublattices.

One notices that the relative chirality of the spin helices is determined by the sign of  $\Gamma$ . Hence, in the parameter region  $\Gamma > 0$  and  $K < 0$ , spin helices living on the odd and even honeycomb sublattices (blue and red sites in Fig. 1) can be viewed as nine pairs of double helices with opposite chirality. We also verify that the other frustrated region with  $\Gamma < 0$  and  $K > 0$  has a uniform chirality pattern.

Another highly unusual feature of the helical phase is that it displays a *spontaneously* anisotropy in spatial periodicity. The helical pitch sizes are  $\frac{L}{6}$  and  $\frac{L}{3}$  supercells along the directions of the two lattice vectors, respectively. Namely, a helix completes two periods along a spontaneously chosen  $\mathbf{r}_1$  direction but only one period along the other direction, despite the same amplitude.

Such a periodicity anisotropy has been encoded in the *Ansatz* Eq. (5) and can be shown explicitly by measuring a spiral correlation function:

$$\Omega_{\mu}(\mathbf{r}) = \frac{1}{m_{\mu}^{\perp}} (\mathbf{S}_{\mu,\mathbf{r}_0} \cdot \mathbf{S}_{\mu,\mathbf{r}_0+\mathbf{r}} - |\mathbf{m}_{\mu}^{\parallel}|^2), \quad (6)$$

where the distance  $\mathbf{r}$  is measured in units of supercells.  $\Omega_{\mu}(\mathbf{r})$  shall develop a cosine curve reflecting the periodicity of the underlying spin helix, and its amplitude indicates strength of the spiral magnetization  $m_{\mu}^{\perp}$ . In Fig. 3 we compare the behaviors of  $\Omega_{\mu}(\mathbf{r})$  for two B helices along spontaneously chosen  $\mathbf{r}_1$  and  $\mathbf{r}_2$  at  $\Gamma = -K$  (orange curves) for a  $L = 72$  lattice, by which the spontaneous anisotropy is confirmed.

We stress that the helical order is *genuinely incommensurate*. Thus, in any finite-size simulations the observed pitch sizes  $\frac{L}{6}$  and  $\frac{L}{3}$  will grow linearly with  $L$ ; see the SM [42].

In Fig. 3 we also measure  $\Omega_\mu(\mathbf{r})$  for a  $B$  helix at  $\Gamma = -K/\sqrt{3}$  (blue curve). Despite an extremely small  $m_B^\perp \sim 10^{-4}$ , the expected cosine oscillation remains developed. This means that the helicity is an imprinted feature of the entire frustrated phase  $1.58\pi \lesssim \theta < 2\pi$ , while it may be easily overlooked at weaker  $\Gamma$  values.

## V. PROXIMATE QUANTUM GROUND STATES

The identification of the classical ground state provides a reference for understanding the quantum model. Indeed, strong magnetic orders established in the classical limit often can persist in the quantum spin-1/2 case with a reduced ordering moment and shifted phase boundary. Examples can be found in both Kitaev magnets [26,38,49] and highly frustrated triangular antiferromagnets [50,51]. Our full characterization of the classical helix angle is especially valuable in the current problem, as its unbiased simulations are far beyond the reach of any state-of-the-art quantum algorithm.

In particular, before  $\Gamma$  gets too strong, and hence the helicity, we hope iPEPS may capture the longitudinal magnetizations consistently. We consider three tensor network *Ansätze* built from 4-, 16-, and 36-site clusters. These clusters cover potential competing orders such as ferromagnetic, Néel,  $\sqrt{3} \times \sqrt{3}$ , stripy, and zigzag type orders which are commonly found in Kitaev magnets [26,38,52]. The 36-site cluster can further fit two supercells of the classical helical axes. We scan the parameter space through a simple update scheme [53,54] up to a large bond dimension  $D = 8$ . Typically over 600 initializations are simulated at each  $\theta$  value. Physical quantities are measured using a CTMRG method (corner transfer matrix renormalization group method) [55,56] with environmental bond dimensions  $\chi > D^2$ . For comparison, we have also examined a full update scheme [49,57] but find the improvements are limited for the symmetry-broken states. See the SM for simulation details [42].

As we show in Fig. 4, within the parameter regime  $1.6\pi < \theta < 1.9\pi$ , the 36-site cluster systematically leads to the lowest energies whose variations in  $\chi$  are significantly smaller than the energy distinctions between different clusters. This confirms the convergence of our simulations and excludes competing magnetic orders. Remarkably, the quantum magnetic moments  $\mathbf{m}_\mu$  display a very similar sublattice structure as the classical longitudinal magnetizations  $\mathbf{m}_\mu^\parallel$  [42]. Their strengths reduce from the classical values  $0.8 \lesssim |\mathbf{m}_{A,B,C}^\parallel| \lesssim 1$  to  $0.3S \lesssim |\mathbf{m}_{A,B,C}| \lesssim 0.5S$  but remain sizable to distinguish from paramagnetic states. Orientations of the quantum magnetizations  $\mathbf{m}_\mu$  also nearly coincide with that of the classical longitudinal moments  $\mathbf{m}_\mu^\parallel$ , as compared in Fig. 5.

The resemblance between the quantum and classical longitudinal magnetic moments indicates the persistence of the classical helical phase. Although the spiral components are intractable with existing quantum algorithms, at a weaker  $\Gamma$  value  $\theta \approx 1.67\pi$ , their strength remains negligible in the classical state (Fig. 3) and affects the energy in the order of  $|m_\mu^\perp|^2 \sim 10^{-8}$ . Thus, one may expect the iPEPS *Ansätze* to remain legitimate at such  $\Gamma$  values. Moreover, given we do not observe signals of a phase transition at immediately stronger  $\Gamma$  in both the quantum and classical cases, the classical order

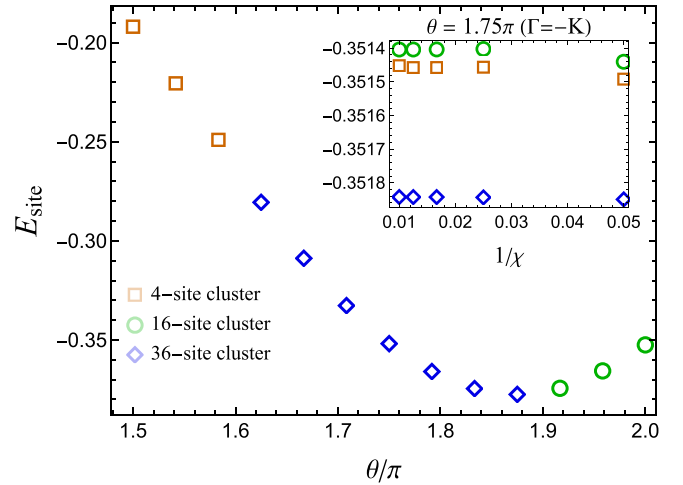


FIG. 4. (Proximate) Ground-state energies computed with iPEPS. In the regime with comparable Kitaev and  $\Gamma$  interactions ( $1.6\pi < \theta < 1.9\pi$ ), the 36-site cluster systematically gives the lowest energies and develops a magnetic pattern similar to classical longitudinal magnetizations. At larger  $\Gamma$  ( $\theta > 1.9\pi$ ), the 16-site and 4-site clusters take over and capture a zigzag-type order. Small magnetizations are obtained near the Kitaev corner ( $\theta < 1.6\pi$ ) where the 4-site cluster is preferred. The inset shows the energy convergence with the environmental bond dimension  $\chi$ .

can be anticipated to survive quantum fluctuations at least for a finite extent of moderate  $\Gamma$  values, which is also the experimentally interested parameter regime.

In the large- $\Gamma$  regime ( $\theta > 1.9\pi$ ), the 16-site and 4-site clusters take over with indistinguishable energies, but both show a clear zigzag-type magnetization [42]. Nevertheless, here the iPEPS *Ansätze* likely fail due to growing helicity. The observed zigzag order may reflect a numerical artifact or a consequence of a possible quantum order-by-disorder at the  $\Gamma$  point [45,46].

Near the Kitaev corner ( $\theta < 1.6\pi$ ) we obtain small magnetizations, in line with the literature [29–31].

## VI. SUMMARY AND DISCUSSION

Understanding the physics of the honeycomb Kitaev- $\Gamma$  model is crucial for both interpreting experimental

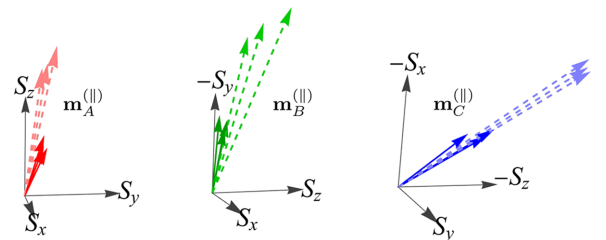


FIG. 5. Comparison between quantum (solid arrows) and classical (dashed arrows) sublattice magnetizations at  $\theta = 1.67\pi, 1.75\pi, 1.83\pi$  ( $\Gamma = -K/\sqrt{3}, -K, -\sqrt{3}K$ ). Orientations of the quantum magnetic moments are very close to the classical longitudinal magnetizations, while the magnitudes reduce to  $0.3S \lesssim |\mathbf{m}_{A,B,C}| \lesssim 0.5S$  from the classical values  $0.8 \lesssim |\mathbf{m}_{A,B,C}^\parallel| \lesssim 1$ . Axes are rotated for visualization.

observations and exploring novel phases in Kitaev magnets. In this work we have investigated its ground-state properties through a combination of comprehensive classical and quantum simulations. We discovered a tangle of highly structured spin double helices imprinted in the classical ground states at material-relevant  $\Gamma$  values. This helix tangle distinguished itself from typical spiral magnets [58–61] by a number of prominent features, including an intricate modulation of helical axes, a spontaneous periodicity anisotropy, and a well regulated chirality pattern. The complete characterization of the classical ground state becomes particularly valuable given fundamental limitations in state-of-the-art quantum numerical algorithms. Converged iPEPS calculations reproduced magnetic moments resembling the classical longitudinal magnetization and signal persistence of the helix tangle against quantum fluctuations. The emergence of unconventional helicity may leave fingerprints to dynamical and transport behaviors, such as spectrum broadening

and splitting, long-living currents, and anomalous diffusions [62–64].

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