

Topological and Trivial Valence-Bond Orders in Higher-Spin Kitaev Models

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We investigate the quantum phases of higher-spin Kitaev models using tensor network methods. Our results reveal distinct bond-ordered phases for spin-1, spin- $\frac{3}{2}$, and spin-2 models. In all cases, we find translational symmetry breaking with unit cells being tripled by forming valence-bond orders. However, these three phases are distinct, forming plaquette order, topological dimer order, and non-topological dimer order, respectively. Our findings are based on a cross-validation between variational two-dimensional tensor network calculations: an unrestricted exploration of symmetry-broken states versus the detection of symmetry breaking from cat-state behavior in symmetry-restricted states. The origin of this different orders can also be understood from a theoretical analysis. Our work sheds light upon the interplay between topological and symmetry-breaking orders as well as their detection via tensor networks.

For quantum spin lattice systems, valence-bond solids (VBS) break translational symmetry by forming valence-bond orders: some bonds spontaneously become stronger with lower energies than others. A quantum spin liquid (QSL) was originally defined by the absence of valence-bond orders and magnetic orders [1]. The competition between QSL and VBS is determined by whether tunneling and resonance processes remain dominant in the thermodynamic limit. Historically, this competition echoes concepts from chemistry (e.g., Kekulé bond resonance) and solid-state physics [2, 3]. After the initial proposal of QSLs, revolutionary concepts such as emergent gauge fields [4], anyons [5], topological orders [6], and long-range entanglement [7], were introduced to characterize general QSLs. These characterizations reshape the definition of a QSL and motivate a renewed examination of the interplay between valence-bond order and QSL concepts.

The Kitaev-type spin-spin interaction [8, 9] involves non-trivial energetics of quantum spin orientation and dimer resonance, providing a platform to study the interplay of valence-bond order, gauge theory, and topological order. For comparison, the antiferromagnetic Heisenberg interaction ($\mathbf{J}\mathbf{S}_i \cdot \mathbf{S}_j$, $J > 0$) has the singlet pairing as its lowest energy state, thereby giving rise to the well-known dimer picture and motivating dimer models based on dimer resonance [10–12]. This picture can capture various conventional VBS, e.g., [13–18] which lack QSL characters. In contrast, the Kitaev type of spin-spin interaction per bond is Ising-like (e.g. $K_x S_i^x S_j^x$) with two degenerate spin orientations with lowest energy. Therefore, the possible valence-bond orders of Kitaev models are not captured by minimal dimer models. The spin- $\frac{1}{2}$ Kitaev honeycomb model [8], an exactly solvable QSL with a $\mathbb{Z}_2$ gauge field, lacks valence-bond order. Likewise, the higher-spin Kitaev models also exhibit local $\mathbb{Z}_2$ fluxes as conserved quantities [19], which preclude any magnetic order. Analytical constructions for emergent fermions and braiding [19–22] have been found for half-integer spin S , but not for integer spin. Semiclassical analyses near the large-spin limit suggest the possibility of valence-bond order with a tripled unit cell [19, 23, 24]. Furthermore, the possibility of a spontaneous

valence-bond order coexisting with topological order has been proposed [24].

Significant computational efforts [20, 21, 25–32] have already been devoted to the spin-1 and spin- $\frac{3}{2}$ Kitaev honeycomb model. Such reference points at or near the pure Kitaev limit are relevant for material realizations [33–35]. However, consensus is still lacking on whether the isotropic spin-1 model is gapless or gapped [20, 21, 25, 27, 32], as well as on the existence of bond orders [21, 28, 32] and topological order. For larger spin up to $S = 2$, the results reported in Refs. 30–32 are not in agreement with the tripled-unit cell obtained in semiclassical results. Accurate numerics of both the spin-1 and the spin- $\frac{3}{2}$ model by exact diagonalization and density-matrix renormalization group report large finite-size effects. This is not surprising because the semiclassical argument for the bond order [19] requires at least a 6×6 unit cell and a lattice with 72 sites, which is beyond the scope of these methods. Two-dimensional tensor network (TN) methods, specifically infinite projected entangled-pair states (iPEPS) [36], are not limited by such finite-size effects. Hence, iPEPS results for the higher-spin Kitaev models, obtained using improved optimization algorithms and more systematic data analysis that go beyond earlier studies based on imaginary-time evolution [21, 28, 32], are highly desirable.

In this Letter, we address the question of the existence and nature of valence-bond orders for Kitaev models with moderately higher spin $S \in (1, \frac{3}{2}, 2)$. We reveal that for these higher-spin models, translational symmetry is broken with unit cells being tripled by forming valence-bond orders. However, three distinct phases are realized for the different spin values, namely non-topological plaquette order (spin 1), topological dimer order (spin $\frac{3}{2}$), and non-topological dimer order (spin 2). These phases are illustrated in Fig. 1 (b-d). These findings are obtained with high-performance iPEPS simulations, and complemented with an analytical argument showing that the presence and absence of topological orders for half-integer and integer values of S are a consequence of the properties of the conserved flux operators.

In line with the on-going development of variational iPEPS

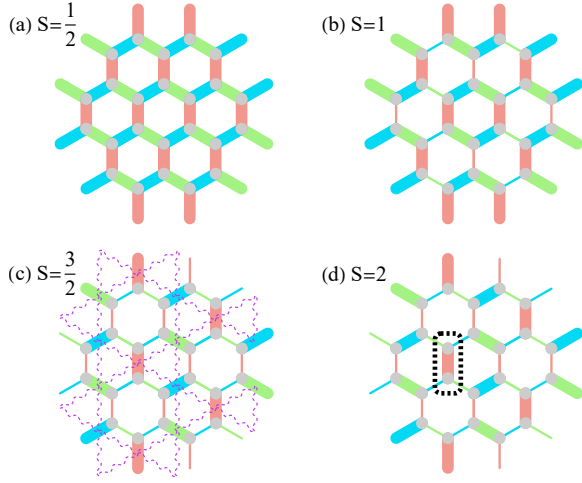


FIG. 1. Ground-state bond-energy configurations in the Kitaev model for different spin values. Different colors represent spins along the x -, y -, and z -axes, while line thickness indicates the relative bond-energy strength. (a) Spin- $\frac{1}{2}$: No bond order, exhibiting uniform bond energies. The ground state is a gapless Kitaev spin liquid. (b) Spin-1: Plaquequette order. Each site is connected to one weak and two strong bonds. (c) Spin- $\frac{3}{2}$: Topological dimer order. Each site connects to one strong and two weak bonds. Purple dashed lines mark entanglement between strong bonds, forming a kagome lattice. (d) Spin-2: trivial dimer order. Each site connects to one weak and two strong bonds, where the strong bonds form short-range entanglement. A limiting case is a configuration where one spin in a bond (e.g., within the black dashed box) only entangles with the other spin: $\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle)$, where the up and down arrows represent the largest and smallest S_z components, respectively.

techniques [37–39], we employ gradient descent optimizations, integrating further algorithmic improvements to achieve significantly higher precision with iPEPS at large bond dimensions. For the spin- $\frac{1}{2}$ Kitaev model [40–44], the transition from traditional imaginary-time evolution to modern gradient-descent schemes has pushed the ground-state energy precision to $\sim 10^{-7}$. Building on this progress, we use gradient-optimized iPEPS with enlarged unit cells to determine the translational symmetry breaking order of higher-spin Kitaev models. Further evidence is acquired by contrasting these results with symmetry-restricted iPEPS results, where we propose a novel methodology to detect cat-state behavior associated with the translational symmetry breaking. We furthermore characterize topological order through the PEPS transfer-matrix spectrum [45]. Unlike several previous works [21, 28, 45, 46], we do not impose a $\mathbb{Z}_2$ virtual symmetry on the iPEPS but find that it emerges spontaneously [47, 48] in the regime where topological order is expected. This is the case for spin- $\frac{3}{2}$. For the spin-1 model [20, 21], our methodology avoids the problem of a cat state hiding the $\mathbb{Z}_2$ condensation.

We start with a brief discussion of the Kitaev honeycomb

Hamiltonian [8] generalized to arbitrary spin:

$$H = \sum_{\langle i,j \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma \quad (1)$$

where γ denotes the x, y, z bonds and $\langle i, j \rangle_\gamma$ denotes the nearest-neighbor pair on the γ bond. The bonds of the honeycomb lattice are divided into x, y, z sets, colored darkblue, yellow, and red in Fig. 1. We focus on the isotropic case $K_x = K_y = K_z = K = -1$. The sign of K can be flipped by a four-unit-cell local π -rotations of spins [49]. Our conclusion will equally apply to the $K = 1$ models because such rotations do not alter the physical quantities characterizing the quantum phases we find. For each unit honeycomb plaquette, there is a locally conserved quantity $W_p = e^{i\pi(S_1^x + S_2^y + S_3^z + S_4^x + S_5^y + S_6^z)}$ [8, 19]. Due to Elitzur’s well-known no-go theorem [50], these local flux-symmetries cannot be broken in the ground state. The global symmetry of π – spin rotations around x -, y -, and z - can be composed from subsets of W_p operators, thereby also precluding magnetic order in the ground state(s).

To obtain the ground states of the spin-1, $\frac{3}{2}$, and 2 Kitaev models, we perform gradient-descent optimization of iPEPS. The iPEPS are a family of variational wavefunctions in the thermodynamic limit, defined by a repeated unit cell of tensors, as depicted in Fig. 2(a). The number of variational parameters scales as D^3 , with D the bond dimension of the virtual bonds connecting the site tensors to their nearest neighbors [Fig. 2(a)]. To directly identify translational symmetry breaking, we explore iPEPS with different enlarged unit cells and determine the tripled structure [Fig. 2(b)] as the optimal choice. Physical observables of iPEPS such as bond energies are computed by controlled approximate contractions. We developed the variational uniform matrix product state (VUMPS) [43, 51–53] method to directly work with iPEPS on the honeycomb lattice with a general unit cell. The VUMPS method aims to solve the fixed-point environments of the iPEPS as top, bottom, up and down boundary MPS. Parallel to the general unit cell calculations, we also use a symmetry-restricted iPEPS that is invariant under the full lattice C_{3v} and translational symmetry—contracted using the C_{3v} symmetric QR corner transfer matrix renormalization group (CTMRG) method [42, 44, 54, 55]—in order to provide a cross-check and to facilitate connecting to previous works. Both VUMPS and QR-CTMRG methods are characterized by the environment bond dimension χ , with increasing χ yielding higher precision. Energy gradients for the optimization are computed by automatic differentiation (AD) [39, 43, 44] of VUMPS and QR-CTMRG, respectively.

We optimize iPEPS with different bond dimensions D to approximate the ground state for the spin-1, $\frac{3}{2}$ and 2 models and summarize the results in Fig. 3. In the left panels, we plot the average energy per site obtained with both the unrestricted PEPS with tripled unit cell, as well as with the C_{3v} -symmetric and translational invariant PEPS. For comparison, we also include the—to our knowledge—best results for the energy per site currently available in the literature. We are able to consistently achieve lower variational energies with the symmetry-

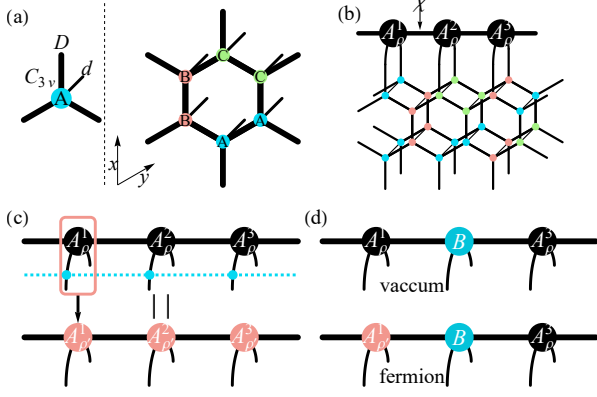


FIG. 2. iPEPS ansatz and MPS boundary solutions in VUMPS contraction. (a) Left: C_{3v} -symmetric tensor for single-tensor iPEPS, the tensor on other sublattice is identical as a function of physical leg (dimension d) and virtual bonds x, y and z (dimension D). Right: A tripled-unit-cell structure with three independent tensors (ABC). (b) Boundary MPS as the fixed-point solutions of the iPEPS transfer matrix (appearing in the norm), where the physical legs of bra and ket have been contracted. The MPS is defined by the contraction of local tensors A_p^i along the virtual legs with dimension χ . (c) For $\mathbb{Z}_2$ topologically ordered states, the iPEPS transfer matrix exhibits a doubly degenerate fixed point structure, represented by boundary MPS with tensors A_p and $A_{p'}$ and associated with breaking of an emerging virtual $\mathbb{Z}_2$ symmetry. (d) The transfer matrix spectrum then contains trivial excitations, targeted by the ansatz in the upper panel, as well as domain wall excitations between the two fixed points, targeted by the lower ansatz.

unrestricted iPEPS with tripled unit cell. These results exhibit, to good approximation, only two distinct values of the bond energies. The bond ratio is thus defined as the magnitude of the stronger bond energy divided by that of the weaker one, as obtained from the tripled-unit-cell data, and is plotted in the right panels of Fig. 3.

The deviation of the bond ratio from 1 indicates the existence of valence-bond solid orders. The spin- $\frac{3}{2}$ and spin-2 states both exhibit the tripled-unit-cell dimer bond-energy configuration [Fig. 1(c)(d)] expected in the semiclassical limit, which features isolated strong bonds, i.e. dimers. We will show below that only the spin- $\frac{3}{2}$ ground state exhibits topological order. For spin- $\frac{3}{2}$ and 2, Ref. [32] obtained anisotropic bond ordered states that do not break translational symmetry using iPEPS, optimized with imaginary-time evolution using simple update (TN-SU). Our results show that the tripled-unit-cell states are energetically more favored. For the spin-1 model, our results exhibit a different bond energy configuration resulting in plaquette order: connected strong bonds form a unit honeycomb plaquette on the lattice [Fig. 1(b)]. Our spin-1 energy density is lower than previous results [20, 27], including a DMRG calculation for an infinite cylinder geometry with 4-unit vector width [27] and an iPEPS calculation using imaginary time evolution with simple update that preserves the translational, C_{3v} , and flux conservation symmetries (STN-SU) [28]. The latter result only differs

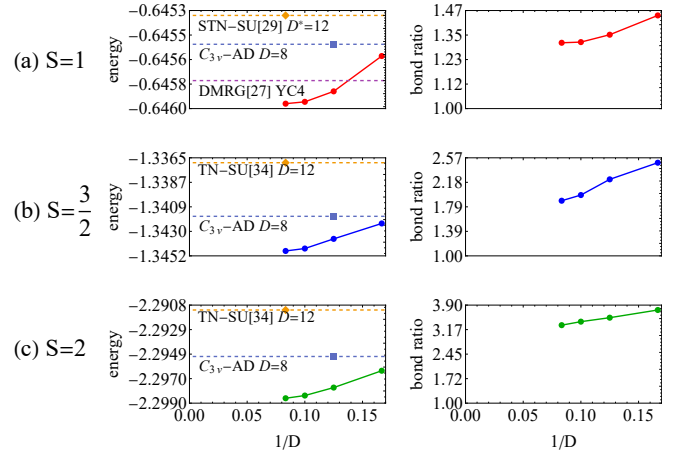


FIG. 3. Average ground-state energy per site (left) and bond ratio (right) for higher-spin Kitaev models for (a) spin 1, (b) spin $\frac{3}{2}$, (c) spin 2. Our results are plotted as points on solid lines for unrestricted tripled-unit-cell iPEPS (see Fig. 2.), with bond dimension D up to 12, and as a square box for the C_{3v} -symmetric PEPS with $D = 8$. The contraction dimension χ for the environment is ramped to be large enough for convergence (up to 512). Previous (symmetric) iPEPS results obtained using simple-update [(S)TN-SU] [28, 32] as well as DMRG results [27] are also indicated. The bond ratio is defined as the ratio between strong and weak bond energies $|\langle S_i^\gamma S_j^\gamma \rangle|_{\max} / |\langle S_i^\gamma S_j^\gamma \rangle|_{\min}$.

by ~ 0.001 with our gradient-optimized symmetry-restricted calculation (C_{3v} -AD), thus raising the question whether both results are capturing the same phase, which may furthermore be compatible with the symmetry breaking that is explicitly present in our symmetry-unrestricted results.

Indeed, while the symmetry-restricted iPEPS results (STN-SU [20] and our own C_{3v} -AD results) cannot explicitly break the translational symmetry due to the single-tensor unit cell, it is possible that they encode a cat-state superposition of bond-ordered states. Following standard practices [56], we determine whether the bond-bond correlation function exhibits long-range order. As shown in Fig. 6, the data indicates that the tripled-unit-cell oscillation may survive in the long-range limit. The behavior is different from that of the spin- $\frac{1}{2}$ case, where an algebraic decay with exponent 4 has been determined [44, 57, 58]. Detecting cat state superpositions from correlation functions is standard practice in many other numerical methods. However, using the iPEPS transfer matrix, we can probe this behavior even more explicitly. The momentum-resolved transfer-matrix spectra, parameterized by the transverse momentum $k_{\parallel}$ as

$$\lambda_n(k_{\parallel}) = \exp[-\epsilon_n(k_{\parallel}) + i\kappa_n(k_{\parallel})], \quad (2)$$

directly encodes both the decay rate and oscillatory behavior of static two-point correlation functions [45, 59, 60], where the angular frequency $\kappa(k)$ encodes the longitudinal momentum $k_{\perp}$ and $\epsilon(k)$ can be interpreted as an inverse correlation length, but also serves as a proxy for the physical excitation energies in the system. We plot the spectra $\epsilon(k)$ of

the symmetry-restricted iPEPS transfer matrix for the spin-1 model in Fig. 4(a). A gap closing tendency at K and K' indicates either a slowly decaying oscillation or long-range order [61], compatible with the symmetry broken state obtained in the tripled unit cell. To relate this gap closing to a specific order, we can use an enlarged unit cell environment to contract the C_{3v} -AD iPEPS, after it has been optimized in a translation invariant environment. We then find that the plaquette and dimer orders appear (Fig. 1), consistent with our enlarged unit cell iPEPS results. This analysis thus provides more detailed information than the bond-bond correlation function that we calculated to distinguish these two orders. Further details are provided in the End Matter.

The transfer matrix also encodes the presence or absence of topological orders. Hereto, we first determine the number of dominant eigenvalues, i.e. distinct VUMPS boundary MPS solutions. Starting from random initializations, we find the two-fold degeneracy characteristic of $\mathbb{Z}_2$ topological order for the spin- $\frac{3}{2}$ model, whereas unique boundary states are obtained for the spin-1 and spin-2 cases. This is consistent with our theoretical analysis, discussed later, which explains the odd-even $2S$ discrepancy for the presence of topological order. The occurrence of different boundary states can be directly related to the existence of 1-form (i.e. string-like) symmetries in topologically ordered states. Absorbing a virtual string in the boundary environment must give rise to another degenerate boundary solution, in order to support deconfined anyonic excitations [45], as depicted in Fig. 2(c-d).

With the boundary solutions at hand, we now also plot the transfer matrix spectra of $\epsilon(k)$ for unrestricted iPEPS with tripled unit cell in Fig. 4(b-d). Gapped spectra for $\epsilon(k)$ are observed for the spin-1, $\frac{3}{2}$ and 2 models, indicating the existence of physical excitation gaps. As we find two degenerate boundary MPS for the spin- $\frac{3}{2}$ case, a domain wall structure can be used to construct a transfer matrix that encodes the string correlation [see Fig. 2(d)], which corresponds to the two-point correlation of emergent fermions.

Now we provide a theoretical analysis to explain the absence and presence of topological order in the case of integer and half-integer spin, respectively. The idea is to examine the flux operators. For dimer orders, one can construct a prototype of short-ranged entangled states by maximally entangling the spins in a dimer: $\frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle)$ for $K = -1$ and $\frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$ for $K = 1$. A straightforward calculation shows that a product state of such dimers is an eigenstate of the flux operators for integer spins but not for half-integer spins. This applies for all dimer coverings. The non-topological decoupled plaquette state can also be constructed. Our calculation shows the terms involving a plaquette for spin 1 have a unique ground state. The above constructions show that for integer spins there is a non-topological state with the same broken symmetries, indicating a possible adiabatic connection between the optimized iPEPS and these prototypical zero-correlation-length states.

Furthermore, we consider an effective theory for strong

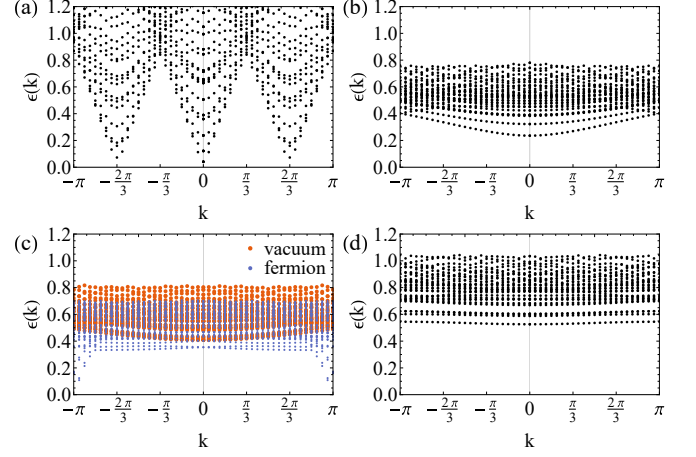


FIG. 4. Spectra of inverse correlation lengths [see Eq. (2)] obtained from the transfer matrix for the single-tensor unit cell C_{3v} -restricted iPEPS for the spin-1 model (a), the tripled-unit-cell iPEPS for spin-1 (b), spin- $\frac{3}{2}$ (c) and spin-2 (d). The transverse momentum along the Brillouin zone corresponds to a cut in the Brillouin zone (a), or in the three-fold reduced Brillouin zone (b-d), where $\frac{\pm 2\pi}{3}$ corresponds to (reduced) K and K' points. Only for spin- $\frac{3}{2}$ (b) did we find two degenerate boundary MPS and do we thus plot both a trivial and a domain wall spectrum, using the ansatz in Fig 2(d).

tripled-unit-cell dimer orders. As mentioned before, the Kitaev type of spin-spin interaction per bond is Ising-like (e.g. $K_x S_i^x S_j^x$) with two degenerate spin orientations. An effective theory can be constructed by assigning a spin variable σ^z with a basis of two low-energy states of the bond. The construction gives an effective spin 1/2 kagome lattice [Fig. 1(c)] model [24]. To reveal the discrepancy between the two classes, we find that it is enough to consider the restriction put to possible effective terms from the flux conservation. For half-integer spin cases, one can construct a Hamiltonian with W_p symmetry:

$$H_{\text{eff}}^{\text{top}} = \sum_{i,j,n \in \bigcirc} J_e \sigma_i^x \sigma_j^x \sigma_k^x \sigma_l^x \sigma_m^x \sigma_n^x + \sum_{a,b,c \in \triangle} J_m \sigma_a^z \sigma_b^z \sigma_c^z, \quad (3)$$

where $i-n$ and $a-c$ label the sites of honeycomb and triangular plaquette in the kagome lattice, respectively. The first and second terms are effective W_p operators commuting with each other. This is the toric code model on the kagome lattice realizing $\mathbb{Z}_2$ topological order, which has been obtained before from semiclassical analysis [24]. For the integer S case, we instead obtain a Hamiltonian:

$$H_{\text{eff}}^{\text{tri}} = \sum_i h \sigma_i^x, \quad (4)$$

where i labels the lattice sites. Eq. (4) commutes with the effective W_p operators $W_{p,i} = I$ for honeycomb and $W_{p,j} = \sigma_{j,a}^x \sigma_{j,b}^x \sigma_{j,c}^x$ for triangle. This stabilizes the trivial state of isolated dimers.

The mechanism for valence-bond order formation beyond the semiclassical picture deserves discussion. Previous theoretical investigations of spin- $\frac{3}{2}$ Kitaev models have developed

an interacting pseudo-fermionic theory [30, 31] for spin- $\frac{3}{2}$. It has been found at the mean-field level, by assuming translational symmetry, that fermions form nearly flat bands with Majorana cones. We find that the spin bond order is translated into interaction-induced pseudo-fermion bond order, while the $\mathbb{Z}_2$ gauge structure is intact. For the spin-1 Kitaev model, as there is no obvious emergent fermion and braiding [20, 21], previous works proposed candidate symmetric phases with a picture of anyonless $\mathbb{Z}_2$ spin liquids [19–22, 62]. Our work shows that spin-1 Kitaev model forms trivial plaquette order. Since the order is moderate, certain additional interactions may help suppress the plaquette order to stabilize a possible nearby symmetric state.

In conclusion, we have demonstrated that the higher-spin Kitaev models provide a platform for an intriguing interplay among valence bond orders and spin-liquid concepts. Considering additional terms relevant for Kitaev materials may lead to even richer phenomena, such as interplay between bond and magnetic order and between anisotropy and translational symmetry breaking. Furthermore, our work provides an improved tensor network workflow for future numerical investigations of symmetry breaking and topological order in frustrated magnets.

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VUMPS algorithms for native honeycomb lattice

To compute gradients and physical observables, we employ two contraction schemes depending on the iPEPS unit cell. For the translationally invariant and C_{3v} -symmetric iPEPS, we use the efficient QR-CTMRG method [44], while for enlarged unit cells without we adopt the VUMPS algorithm. Both approaches offer a computational advantage over the original CTMRG in obtaining fixed-point environments. Although randomized SVD (rSVD) [42] can reduce the cost of CTMRG to a similar level, QR-CTMRG and VUMPS remain faster in practice. Our QR-CTMRG implementation is adapted to the honeycomb lattice and exploits its C_{3v} symmetry for further speedup. As QR-CTMRG is not yet generalized to enlarged unit cells, VUMPS is used in those cases, as well as for cross-validation and analysis of C_{3v} -symmetric states with larger environment cells, as discussed in the main text. Details of the VUMPS can be found in Ref. 43, and our additional general performance improvements will be reported elsewhere.

We focus on the efficient contraction of native honeycomb iPEPS using the VUMPS algorithm. In contraction, the bracket network surrounding a lattice site is approximated by environment tensors (denoted as T in Fig. 5). The primary computational cost in VUMPS arises from applying the linear transfer matrix map. The depicted contraction scheme scales as $O(\chi^3 D^4 + \chi^2 D^5 d)$; C_{3v} CTMRG exhibits the same scaling when using a similar method. A key advantage of contracting the honeycomb lattice natively is computational efficiency. A common alternative is to merge two sites to form a square lattice, but this is effectively a more costly contraction order. The merged square iPEPS tensor (with four virtual bonds) has a higher contraction cost of $O(\chi^3 D^4 + \chi^2 D^6 d^2)$. Assuming the typical empirical relation $\chi = O(D^2)$, both methods ultimately scale as $O(D^{10})$. However, working directly with the honeycomb lattice reduces the prefactor, offering a practical speedup.

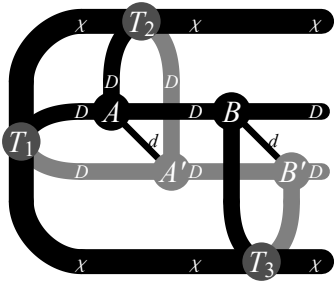


FIG. 5. The linear transfer matrix map: the optimal contraction order is $T_1 \rightarrow T_2 \rightarrow A \rightarrow A' \rightarrow T_3 \rightarrow B \rightarrow B'$, and the corresponding computational costs are $O(\chi^3 D^4) \rightarrow O(\chi^2 D^5 d) \rightarrow O(\chi^2 D^5 d) \rightarrow O(\chi^3 D^4) \rightarrow O(\chi^2 D^5 d) \rightarrow O(\chi^2 D^5 d)$.

Inferring cat states from symmetry-restricted iPEPS

In the main text, we demonstrate that signatures of symmetry breaking can be inferred from symmetry-restricted iPEPS via cat-state behavior. Here we provide further details and supporting data.

Bond-bond correlation functions. In the thermodynamic limit, a cat state associated with the expected tripled-unit-cell symmetry-breaking pattern corresponds to a superposition of three degenerate states, yielding long-range order in specific connected correlation functions. For valence-bond order, the relevant quantity is the bond-bond correlation function

$$C_{D_z, D_z}(r_{i,j}) = \langle S_i^z S_{i'}^z S_j^z S_{j'}^z \rangle - \langle S_i^z S_{i'}^z \rangle \langle S_j^z S_{j'}^z \rangle, \quad (5)$$

where i, i' and j, j' denote nearest-neighbor pairs and $r_{i,j}$ is the distance between sites i and j . Based on our optimized C_{3v} iPEPS, correlations can be reliably inferred over finite ranges, which extend as D increases. Since long-range order is defined only in the infinite-distance limit, its emergence must be inferred from the trend with increasing D . Both tripled-unit-cell dimer and plaquette orders in the bond-bond correlation function manifest as oscillations with period three, taking values $(2c, -c, -c)$, where c is a constant. In variational iPEPS, correlation functions are generally more challenging to compute accurately than ground-state energies, with precision estimated from results at different bond dimensions D . Figure 6(a) presents our spin-1 results. For $D = 9$, the energy (nearest-neighbor correlation) error is on the order of 10^{-4} . The enlarged-unit-cell iPEPS yields a correlation length of roughly three lattice constants for the symmetry-broken state, suggesting that correlation data up to ten sites, accurate to about 10^{-3} , can reliably indicate the ordering tendency. The expected oscillation is clearly observed, although the plateau shows a slight decay, which we attribute to numerical artifacts. Similar finite-accuracy effects have been reported in large-scale PEPS studies of other spin models [64]. As a consistency check, the decay is slower than a $1/r$ form, excluding an algebraic QSL scenario.

Transfer matrix spectrum. Long-range order in correlation functions is formally associated with exact degeneracy of the dominant eigenvalues in the boundary MPS transfer-matrix spectrum [61]. In practice, iPEPS optimized at finite bond dimension D need not exhibit exact degeneracy, but our results show a clear convergence toward it with increasing D . This behavior corresponds to the suppression of the apparent artificial decay in correlation functions. A static connected two-point correlation function, including Eq. (5), can be expressed as

$$f(r) = \sum_{i>0} f_i e^{-\epsilon_i r}, \quad \epsilon_i = -\ln(\lambda_i/\lambda_0), \quad (6)$$

where λ_i are the eigenvalues of the transfer matrix determined by the MPS bond dimension χ for a given iPEPS bond dimension D . Accurate estimation of the spectrum requires a simultaneous increase of both χ and D . The approach of the

inverse dominant correlation length $\xi^{-1} = \ln(|\lambda_1/\lambda_0|) \rightarrow 0$ signals that the connected correlation function ceases to decay exponentially. As shown in Figs. 6 (b,c), we perform finite-correlation-length scaling following Refs. [44, 60]. For spin 1, the extrapolated ξ ($\chi \rightarrow \infty$, $\delta \rightarrow 0$) appears to converge for fixed D , yet remains relatively large even at moderate D . This behavior is consistent with Refs. 21 and 28, which achieve comparable energy accuracy to our $D = 5, 6$ data. We further find that ξ tends to increase with D , suggesting an approach toward degeneracy consistent with cat-state behavior. To contrast this with a known algebraic QSL, we compare to the $S = \frac{1}{2}$ case: for $S = \frac{1}{2}$, data with $D \geq 6$ collapse, whereas for $S = 1$ no such collapse occurs up to $D = 9$. Moreover, the slope decreases with increasing D for both even and odd D , indicating that, at fixed δ , the gap between the leading and first excited transfer-matrix eigenvalues closes more rapidly as D grows.

Spontaneous symmetry breaking by environments. As a third approach, contracting the optimized symmetry-restricted iPEPS with an unrestricted environment directly demonstrates symmetry breaking. Explicitly enlarging the unit cells of the environments to 2×6 in combination with replicated PEPS tensors from the single unit cell, the calculated bond energies become nonuniform due to environmental symmetry breaking, as shown in Fig. 7. This is a direct consequence of strong oscillatory bond-bond correlations. This method can directly reveal plaquette order and dimer order. When the degeneracy of transfer matrix dominant eigenvalues is not exact, the order exists only for finite χ .

Determine topological order from boundary MPS

As shown in the main text, the action of string operators relating topological sectors leads to a two-fold degeneracy in the boundary MPS of the iPEPS. Our approach employs unstructured site tensors and searches for distinct boundary MPS of the optimized iPEPS [47]. To explore different solutions, we use random initializations in the approximate boundary solver. Different boundary solutions yield energy densities that agree up to 10^{-9} relative precision. Distinct solutions are identified by computing the inner-product density between six consecutive MPS tensors, with overlaps within 10^{-6} regarded as identical. Unlike most previous works that impose $\mathbb{Z}_2$ virtual symmetry to realize topological degeneracy through virtual symmetry breaking, our method avoids such constraints. This is important because in such symmetric constructions, even trivial phases can exhibit artificial four-fold boundary degeneracy—interpreted as condensations that break all $\mathbb{Z}_2 \times \mathbb{Z}_2$ symmetries of the iPEPS bra-ket structure [45]—which doubles the required bond dimension D and increases computational cost. The relationship between degeneracy and symmetry breaking becomes subtle when condensation coincides with translational-symmetry breaking under restricted translational invariance. Following Ref. 21, we apply projectors [28, 41] to construct $\mathbb{Z}_2$ -iPEPS from our C_{3v} data. Random initialization then reveals a degeneracy exceed-

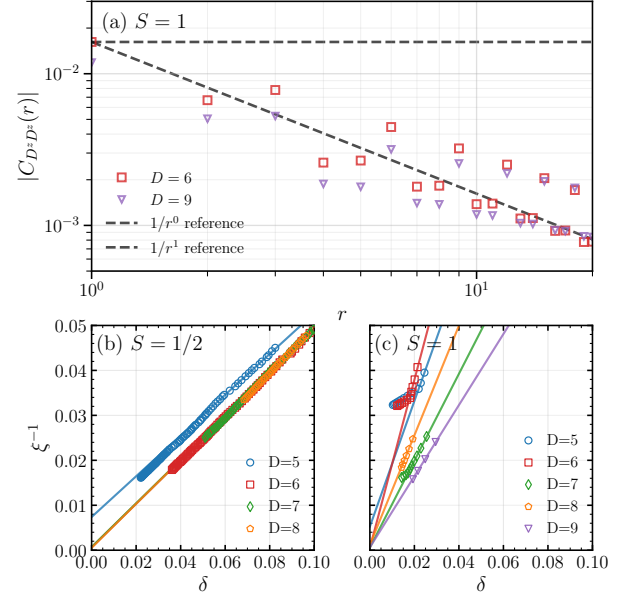


FIG. 6. (a) Bond-bond correlation function of symmetry restricted iPEPS for $S = 1$. Horizontal dashed lines are plateau behavior expected for oscillatory long-range orders. Another reference dashed line indicates $1/r$ algebraic decay. More accurate data (larger D) shows that the three unit-cell oscillation has no obvious decaying tendency as algebraic QSL. (b-c) Finite correlation length scaling for $S = \frac{1}{2}$ (b) and $S = 1$ (c), where $\delta = \ln(|\lambda_1/\lambda_3|)$ with λ_i the i -th-largest magnitude eigenvalue of transfer matrices, and $\xi^{-1} = -\ln(|\lambda_1/\lambda_0|)$ is the inverse dominant correlation length.

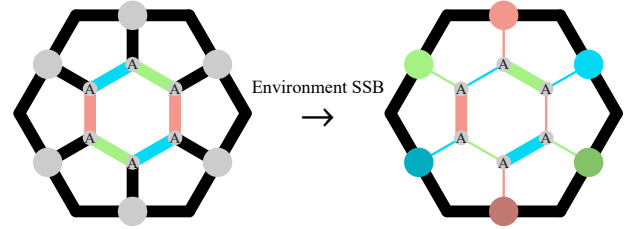


FIG. 7. Spontaneous symmetry breaking of environment for contracting uniform C_{3v} iPEPS tensor once the uniform restriction of environment is lifted.

ing two, with overlaps around 0.4, indicating cat states of virtual symmetry, which is consistent with the absence of topological order in the $S = 1$ phase.

More details of iPEPS and contraction ansatz

Our calculations and all the referenced previous iPEPS work [20, 21, 32] of higher-spin Kitaev models are based on honeycomb iPEPS, where the tensor on each site has three virtual legs, with the dimension set all to be D . As the network is native to the lattice geometry, it brings no artifact for studying the lattice symmetry breaking of our interests. To investigate translational symmetry breaking beyond the scope of Ref. [20, 21, 32], we extend the translational symmetry of iPEPS from the minimal two-site unit to enlarged cases.

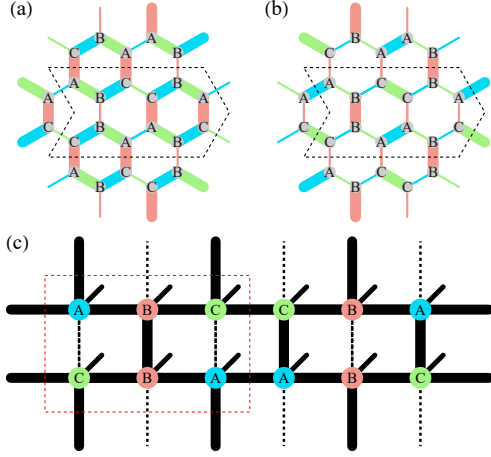


FIG. 8. iPEPS ansatz with large unit cell. (a)Bond pattern for spin-1; (b)Bond pattern for spin- $\frac{3}{2}$ and spin-2; (c) the 2×6 unit cell with brickwall shape. The dashed black lines are the bond missing as compared to the square lattice, while the dashed red box are put around the six sets of boundary MPS environments.

This process begins with the construction of a unit cell. The configuration presented in Fig. 1 of the main text has a unit cell with 6 sites. For VUMPS contraction, it is better to illustrate the assignment of the environment tensors in a further enlarged 2×6 unit cell (see Fig. 8). We draw the network alternatively by embedding the honeycomb lattice in a square lattice. The honeycomb iPEPS network remains unchanged

because it is defined by the unchanged connection topology. The variational parameters are contained in the independent tensors A , B , and C , optimized by gradient descent. We set site tensors expected with the same bond energy x, y, z distribution to be the same, a generalization of the single-tensor-for-two-sublattice restriction in [41, 44]. We verify the validity by lifting the restriction. However, we cannot fully reduce the number of environments in the same spirit, as a converging contraction cannot be achieved with the restrictions we tried. The working way is using six sets of boundary MPS environments, surrounding the six tensors enclosed in the red dashed square in Fig. 8(c) respectively. The environments for the parts not enclosed is obtained from translation.

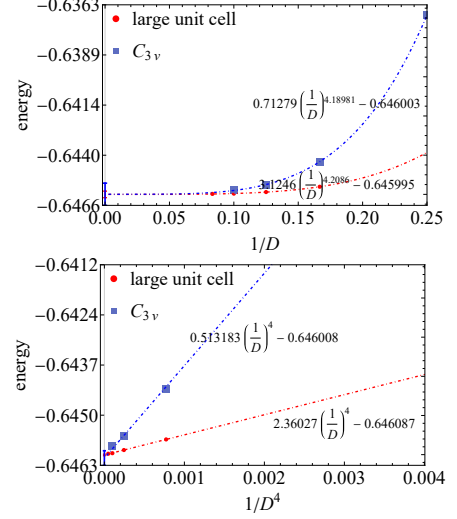


FIG. 9. Extrapolation of the energy with $1/D^\alpha$ scaling for both our symmetry-breaking iPEPS (large unit cell) and symmetry restricted iPEPS results.

Energy extrapolation

We adopt a $1/D^\alpha$ energy extrapolation for both our symmetry-breaking iPEPS (large unit cell) and symmetry restricted iPEPS results. The extrapolated results (Fig. 9) show better agreement.