Hidden $SU(2)$ Symmetries, the Symmetry Hierarchy and the Emergent Eight-Fold Way in Spin-1 Quantum Magnets

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The largest allowed symmetry in a spin-1 quantum system is an $SU(3)$ symmetry rather than the $SO(3)$ spin rotation. In this Letter, we reveal some $SU(2)$ symmetries as subgroups of $SU(3)$ that, to the best of our knowledge, have not previously been recognized. Then, we construct $SU(2)$ symmetric Hamiltonians and explore the ground-state phase diagram in accordance with the $SU(3) \supset SU(2) \times U(1)$ symmetry hierarchy. It is natural to treat the eight generators of the $SU(3)$ symmetry on an equal footing; this approach is called the eight-fold way. We find that the spin spectral functions and spin quadrupole spectral functions share the same structure, provided that the elementary excitations are flavor waves at low energies, which serves as a clue to the eight-fold way. An emergent $S = 1/2$ gapless quantum spin liquid is found to coexist with spin nematic order in one of the ground states.

The symmetry principle plays a fundamental role with respect to the laws of nature. It provides an infrastructure and coherence for summarizing physical laws that are independent of any specific dynamics. Noether’s theorem says that every continuous symmetry of the action of a physical system is associated with a corresponding conservation law. The standard paradigm for describing phase transitions and critical phenomena is Landau’s theorem, which says that every continuous symmetry of the action of a physical system is associated with a corresponding order parameter. According to the relevant symmetries, a hierarchy of order parameters and is described by all terms that are allowed according to the relevant symmetries. A hierarchy of symmetries is also widely used in particle physics to understand the dynamics of elementary particles.

Meanwhile, spin-1 quantum magnets are of great interest in physics. One famous example is the Haldane phase in one-dimensional (1D) spin-1 chains, in which fractional spin-1/2 end states are protected by the spin rotational symmetry in a phenomenon called symmetry-protected topological order. Spin-1 systems are also able to host spin nematic orders in dimensions of $D > 1$; such orders are characterized by long-range spin quadrupolar correlations, and the possibility for fractional spinon excitations to coexist with spin nematic orders has also been proposed. Such quantum magnets are widely encountered in various materials, especially transition metal compounds, in which a local $S = 1$ magnetic moment can be formed in a cation via Hund’s coupling; examples include $3d^8$ Ni$^{2+}$ and $3d^9$ Fe$^{2+}$. In this Letter, we shall reveal several hidden $SU(2)$ symmetries in spin-1 quantum magnets in addition to spin rotational symmetry, and we will study spin-1 quantum systems with the help of the symmetry hierarchy.

Models and symmetries. For a spin-1 quantum magnet, there are three local states, namely, $|S^z = \pm 1\rangle$ and $|S^z = 0\rangle$, and eight independent local Hermitian operators: three spin vector operators, $S^x$, $S^y$, and $S^z$, and five spin quadrupolar operators, $Q^{x^2-y^2} = (S^x)^2 - (S^y)^2$, $Q^{(x^2+y^2)-z^2} = \frac{1}{\sqrt{3}} [2(S^x)^2 - (S^y)^2]$, $Q^{xy} = S^x S^y + S^y S^x$, $Q^{yz} = S^y S^z + S^z S^y$, and $Q^{zx} = S^z S^x + S^x S^z$. To illustrate the symmetry hierarchy, we consider a generic two-body interacting Hamiltonian as follows:

$$
H = \sum_{\langle i,j \rangle} \left( \sum_{\alpha \beta} J_{\alpha \beta} S_i^\alpha S_j^\beta + \sum_{\mu \nu} J_{\mu \nu} Q_i^\mu Q_j^\nu + \sum_{\alpha \mu} I_{\alpha \mu} S_i^\alpha Q_j^\mu \right),
$$

where $\langle i,j \rangle$ is a pair of nearest neighboring sites; $\alpha$ and $\beta$ denote $x$, $y$, and $z$; and $\mu$ and $\nu$ denote $x^2 - y^2$, $3z^2 - r^2$, $xy$, $yz$, and $zx$. The $SO(3)$ spin rotational symmetry is achieved when $I_{\alpha \mu} = 0$, $J_{\alpha \beta} = \delta_{\alpha \beta} J_1$, and $J_{\mu \nu} = \delta_{\mu \nu} J_2$. Furthermore, $H$ will be $SU(3)$ symmetric when $J_1 = J_2$, and the $SU(3)$ group is generated by eight operators $\{S, Q\}$. The $SO(3)$ model is well studied: a phase diagram consisting of a ferromagnetic phase, a dimerized phase, Haldane phases and a critical phase has been constructed in one dimension [10-13], and the $SO(3)$ model can host spin nematic ground states in dimensions of $D > 3$ [14,21].

The model defined in Eq. (1) has typically been studied in accordance with the $SU(3) \supset SO(3) \cdots$ symmetry hierarchy. Nevertheless, there are other $SU(2)$ subgroups belonging to the $SU(3)$ group, and this fact implies the existence of a slice of $SU(2)$ symmetries in addition to the $SO(3)$ spin rotation in spin-1 quantum magnets of which, to the best of our knowledge, the research community is not aware. This situation inspires us to search for Hamiltonians that respect these hidden symmetries; for this purpose, a new symmetry hierarchy, $SU(3) \supset SU(2) \times U(1) \cdots$, will be adopted to reveal novel states with various low-energy excitations. To describe these states, it is natural to treat all operators $\{S, Q\}$ on
Hidden SU(2) symmetries. It turns out that there are three hidden SU(2) symmetries, which are generated as follows: (1) $SU(2)_\alpha : \{Q^{xx}, S_y \}, \frac{1}{2} Q^{x^2-y^2} + \frac{\sqrt{2}}{Q^{3z^2-r^2}}\}$, (2) $SU(2)_\beta : \{Q^{yy}, S_x, \frac{1}{2} Q^{z^2-y^2} - \frac{\sqrt{2}}{2} Q^{3z^2-r^2}\}$, and (3) $SU(2)_\gamma : \{Q^{xy}, S_z, Q^{x^2-y^2}\}$ (see the Supplementary Materials). Each set of these generators consists of one component of the spin vector $S$ and two components of the spin quadrupole $Q$. Note that these three sets of generators are related to each other by the following cycle: $S^x \rightarrow S^y \rightarrow S^z$. In the remaining part of this Letter, we shall focus on $SU(2)_\gamma$; $SU(2)_\alpha$ and $SU(2)_\beta$ can then be obtained in accordance with this cycle. For the $SU(2)_\gamma$ symmetry, $S^z$ generates spin rotations along the $z$-axis, and the other two generators, $Q^{xy}$ and $Q^{x^2-y^2}$, correspond to two-magnon processes, as can be seen from $Q^{x^2-y^2} = \frac{1}{2} \left( (S^+)^2 + (S^-)^2 \right)$ and $Q^{xy} = \frac{1}{2} \left( (S^+)^2 - (S^-)^2 \right)$, where $S^z = S^\pm \pm i S^y$. Let us define $J^x = \frac{1}{2} S^x$ and $J^z = \frac{1}{2} (Q^{x^2-y^2} \pm i Q^{xy}) = \frac{1}{2} (S^\pm)^2$. It is easy to verify that $\{J^x, J^z\}$ satisfy the $SU(2)$ Lie algebra. Therefore, the spontaneous breaking of the $SU(2)_\gamma$ symmetry along the $S^z$ direction will give rise to two-magnon low-energy excitations, while spontaneous symmetry breaking along the $Q^{xy}$ and $Q^{x^2-y^2}$ directions will give rise to an admixture of one- and two-magnon excitations, which will tend to restore the $SU(2)_\gamma$ symmetry.

The underlying $SU(3)$ structure and the hidden $SU(2)$ symmetries will be more transparent in the Cartesian representation of the spin states: $|x\rangle = i(|1\rangle - |\bar{1}\rangle) / \sqrt{2}$, $|y\rangle = (|1\rangle + |\bar{1}\rangle) / \sqrt{2}$, and $|z\rangle = - |0\rangle$. Then, a spin state can be written as $|d\rangle = d^x|x\rangle + d^y|y\rangle + d^z|z\rangle$, where $d = (d^x, d^y, d^z)$ is a complex vector and the normalization condition is given by $|d|^2 = 1$. The expectation values for $\{S, Q\}$ can be expressed in terms of $d$ as follows: $(S^\alpha) = -i \epsilon_{\alpha\beta\gamma} d^\beta d^\gamma$, $(Q^{\alpha\beta}) = -(d^\alpha d^\beta + d^\beta d^\alpha)$, $(Q^{x^2-y^2}) = |d^x|^2 - |d^y|^2$ and $(Q^{3z^2-r^2}) = \frac{1}{\sqrt{3}} (2 |d|^2 - |d^y|^2 - |d^z|^2)$, where $d^\alpha$ is the complex conjugate of $d^\alpha$ and $\epsilon_{\alpha\beta\gamma}$ is a three-rank antisymmetric tensor. Thus, a spin-1 quantum system can be described by the following path integral:

$$Z = \int \mathcal{D}[d, \bar{d}] \delta(|d|^2 - 1) e^{-\int_0^\beta d\tau \{\mathcal{H}, d, \bar{d}, \partial_\tau d, -\bar{d}\}},$$

where the Hamiltonian $\mathcal{H}$ is given by Eq. [1] with $\{S, Q\}$ replaced with their expectation values. Now, it is clear that all of the special unitary transformations of $d$ give rise to the $SU(3)$ group and that the special unitary transformations of any two components of $d$ lead to either $SU(2)_\alpha$, $SU(2)_\beta$ or $SU(2)_\gamma$.

$SU(2)_\gamma$-symmetric Hamiltonians. Now, we are in a position to construct Hamiltonians in accordance with the $SU(2)_\gamma$ symmetry. A generic spin-1 Hamiltonian can be written in terms of $\{S, Q\}$ in a bilinear form as shown in Eq. [1]. Using group theory, one is able to obtain all

| Hamiltonian | $T$ | $T'$ | Global | Local |
|-------------|-----|-----|--------|-------|
| $H_1$ | Yes | Yes | SU(2) x U(1) | U(1) |
| $H_2$ | Yes | Yes | SU(2) x U(1) | U(2) |
| $H_3$ | Yes | Yes | SU(2) x U(1) | U(2) |
| $H_4$ | Yes | No | SU(2) | |
| $H_5$ | No | No | SU(2) x U(1) | |
| $H_6$ | No | No | SU(2) | |
$a_{m}(j)$, to each site $j$ on the $n^{th}$ sublattice, where $\alpha = x, y, z$ refers to the local spin states. For example, $n = 1$ for a uniform state, while $n = 1, 2$ for a bipartite-lattice ordered state. The operators $\{S, Q\}$ can be written bilinearly in terms of the Schwinger bosons, and the physical Hilbert space can be restored by imposing a single-occupancy condition (see the Supplementary Materials). Third, without loss of generality, we let the Schwinger bosons condense at $a_{n\tilde{z}}$ to obtain ordered states, where $a_{n\tilde{z}}$ and the other two orthogonal components, $a_{n\tilde{g}}$ and $a_{n\tilde{z}}$, are related to $(a_{nx}, a_{ny}, a_{nz})$ by an $SU(3)$ rotation $\Omega$, as follows: $(a_{n\tilde{z}}, a_{n\tilde{g}}, a_{n\tilde{z}}) = \Omega_n(a_{nx}, a_{ny}, a_{nz})$. Such an $\Omega_n$ is determined by the mean-field vector $\mathbf{d}$ and enables us to attribute the condensate to $a_{n\tilde{z}}$ alone, while treating $a_{n\tilde{g}}$ and $a_{n\tilde{z}}$ as small fractions. Then, the low-energy Hamiltonian can be bilinearized by the Holstein-Primakoff transformation: $a_{n\tilde{z}}^\dagger(j) = a_{n\tilde{z}}(j) = \sqrt{M - a_{n\tilde{g}}^2(j)} a_{n\tilde{g}}(j) - a_{n\tilde{z}}^2(j) a_{n\tilde{z}}(j)$, where we will ultimately take $M = 1$ for the single-occupancy case. Expansion in $1/M$ and Bogoliubov transformation will give rise to a diagonalized Hamiltonian in $k$-space (see the Supplementary Materials): $\mathcal{H} = \sum_{m,k} \omega_m(k) b_m^\dagger(k) b_m(k) + \mathcal{C}$, where $\omega_m(k)$ is the energy dispersion of the $m$-th flavor-wave branch, $b_m(k)$ is a bosonic Bogoliubov quasiparticle, and $\mathcal{C}$ is a constant. For a uniform state, $m = 1, 2$, while for a bipartite-lattice ordered state, $m = 1, 2, 3, 4$. As long as the vector $\mathbf{d}$ is given by the mean-field theory, we will be able to obtain $\omega_m(k)$ and $b_m(k)$ simultaneously.

$SU(2) \times U(1) \times T \times I$-symmetric model. In particular, we are interested in Hamiltonians with the time-reversal symmetry $T$ and the spatial inversion symmetry $I$, which can be parameterized in terms of three real numbers $K_1$, $K_2$, and $K_3$ as follows:

$$\mathcal{H} = K_1 \mathcal{H}_1 + K_2 \mathcal{H}_2 + K_3 \mathcal{H}_3.$$  \hspace{1cm} (4)

Note that the model given in Eq. (4) respects the $SU(2)_c \times U(1)$ symmetry rather than the $SU(2)$ symmetry. For simplicity, we shall consider bipartite lattices only, including a 1D chain, a square lattice and a cubic lattice.

To explore the ground-state phase diagram, we set $K_1^2 + K_2^2 + K_3^2 = 1$, such that the parameter space is a sphere. Top and bottom views (along the $K_3$ axis) of this sphere are displayed in Fig. 1, where the mean-field phase diagram is presented. There are six ordered phases, FQ1, FQ2, FQ3, AFQ1, AFQ2, and AFQ3. Here, FQ refers to a ferro-quadrupolar state, and AFQ refers to an antiferro-quadrupolar state (or, to be precise, a state with a staggered quadrupolar order). When $K_{1(2,3)}$ is negative and predominates, the ground states are FQ states, while when $K_{1(2,3)}$ is positive and predominates, the ground states are AFQ states. The solid lines in the phase diagram represent first-order transitions, while the dashed lines represent continuous transitions. The $SU(3)$ symmetry will be achieved at two points where $K_1 = K_2 = K_3$. Both $SU(3)$ points are tricritical points.

The one with $K_{1,2,3} < 0$ corresponds to three phases, FQ1, FQ2 and FQ3, while the other one, with $K_{1,2,3} > 0$, corresponds to AFQ1, AFQ2 and AFQ3. The mean-field ground states and low-energy flavor-wave excitations for these six phases are summarized in Table I. Notably, dipolar and quadrupolar orders may coexist in a ground state in the FQ1, FQ2, AFQ1 and AFQ2 phases, while only a quadrupolar order exists in the FQ3 and AFQ3 phases.

The low-energy excitations can be understood in the framework of the symmetry hierarchy as follows. (1) The spontaneous symmetry breaking is distinct in the different phases: (a) $SU(2)$ is broken in FQ1 (AFQ1), but $U(1)$ is not (i.e., $SU(2) \times U(1) \rightarrow U(1)$); (b) both $SU(2)$ and $U(1)$ are broken in FQ2 (AFQ2) (i.e., $SU(2) \times SU(2) \rightarrow SU(2)$); and (c) neither $SU(2)$ nor $U(1)$ is broken in FQ3 (AFQ3). (2) For FQ1, the $\omega_{1FQ1}$ mode is gapless, while the other mode, $\omega_{2FQ1}$, is gapful. Since $SU(2)$ is broken, the gapless Goldstone mode $\omega_{1FQ1}$ tends to recover the symmetry. However, $U(1)$ is unbroken, so $\omega_{2FQ1}$ is not required to be gapless as well. The gapless $\omega_{1FQ1}$ mode corresponds to two-magnon excitations, while the gapful $\omega_{2FQ1}$ mode corresponds to one-magnon excitations (see the Supplementary Materials). (3) For FQ2, there are two gapless Goldstone modes, $\omega_{1FQ2}$ and $\omega_{2FQ2}$, because both $SU(2)$ and $U(1)$ are broken. The $\omega_{1FQ2}$ mode is an admixture of one- and two-magnon excitations, while the $\omega_{2FQ2}$ mode consists of one-magnon excitations only. (4) For FQ3, there are two gapful modes, $\omega_{1FQ3} = \omega_{2FQ3}$, which are related to each other through the $SU(2)$ symmetry. Both of them correspond to one-magnon excitations. (5) The AFQ1, AFQ2 and AFQ3 phases can be analyzed similarly.

**Spectral functions.** We find that inelastic neutron scattering and resonant inelastic X-ray scattering (RIXS), which measure the spin spectral function $S(\mathbf{q}, \omega)$ and the spin quadrupole spectral function $Q(\mathbf{q}, \omega)$, respectively, can be used to detect flavor waves and distinguish the various FQ and AFQ phases. With the help of flavor-
TABLE II. Summary of the $SU(2) \times U(1) \times T \times I$-symmetric model defined in Eq. [4]. The parameters $\vartheta$ and $\tilde{\vartheta}$ are given by $\sin^2 \vartheta = \frac{2|K_2| + K_1 + K_3}{4|K_2| + K_1 + K_3}$ and $\sin^2 \tilde{\vartheta} = \frac{2K_2 + K_1 + K_3}{4|K_2| + K_1 + K_3}$, respectively. $\mathcal{R}(\chi, \theta, \phi, \varphi) = \text{diag}(e^{\frac{i}{2} \pi \sigma^z \varphi}, e^{\frac{i}{2} \pi \sigma^z \varphi}, e^{\frac{i}{2} \pi \sigma^z \varphi}, e^{i \pi \sigma^z \varphi})$ is an $SU(2) \times U(1)$ rotation. $A_K, B_K, C_K, D_K$ and $\gamma(k)$ are defined as follows: $A_K = \frac{2|K_2| - K_1 - K_3 + 2K_1 K_3}{4|K_2| + K_1 + K_3}$, $B_K = \frac{K_1 + 3K_3 + 2K_1(K_1 + K_3)}{4|K_2| + K_1 + 3K_3}$, $C_K = (3 + K_1/K_3)/2$, $D_K = 2K_2/K_3$, and $\gamma(k) = Z^{-1} \sum e^{ik\delta}$, where $Z = 2D$ is the coordination number and $\delta$ is a nearest-neighbor displacement. "1" refers to one-magnon excitations, "2" refers to two-magnon excitations, and "1+2" refers to an admixture of one- and two-magnon excitations.

|      | $d$ vector(s) | Flavor-wave dispersion | Gap       | Magnon |
|------|---------------|------------------------|-----------|--------|
| FQ1  | $d = \mathcal{R}(\chi, \theta, 0, 0)$ | $\omega^{FQ1}_1(k) = 2Z[K_1[1 - \gamma(k)]]$ | gapless   | 2      |
|      |               | $\omega^{FQ1}_2(k) = Z[|K_1| - K_3] + 2\bar{2}K_1\gamma(k)$ | gapful    | 1      |
| FQ2  | $d = \mathcal{R}(\chi, \theta, \phi, 0, 0)$ | $\omega^{FQ2}_1(k) = 2Z[K_2A_K(1 - \gamma(k))]$ | gapless   | 1      |
|      |               | $\omega^{FQ2}_2(k) = 2Z[K_2[1 - |\gamma(k)|][1 + B_K\gamma(k)]]$ | gapful    | 1      |
| FQ3  | $d = \mathcal{R}(0, 0, 0)$ | $\omega^{FQ3}_1(k) = \omega^{FQ3}_2(k) = 2Z[K_3 + K_2\gamma(k)]$ | gapful    | 1      |
| AFQ1 | $d_1 = \mathcal{R}(\chi, \theta, 0, 0, 0)$ | $\omega^{AFQ1}_1(k) = \omega^{AFQ1}_2(k) = 2ZK_1[1 - \gamma(2k)]$ | gapless   | 2      |
|      | $d_2 = \mathcal{R}(\chi, \theta, 0, 0, 1)$ | $\omega^{AFQ1}_3(k) = \omega^{AFQ1}_4(k) = (Z - K_3 - K_3)$ | gapless   | 1      |
| AFQ2 | $d_1 = \mathcal{R}(\chi, \theta, 0, 0, 1)$ | $\omega^{AFQ2}_1(k) = 2ZK_2A_K[1 - \gamma(k)]$ | gapless   | 1      |
|      | $d_2 = \mathcal{R}(\chi, \theta, 0, -0, 0)$ | $\omega^{AFQ2}_2(k) = 2ZK_2[1 + |\gamma(k)|][1 + B_K\gamma(k)]$ | gapless   | 1      |
| AFQ3 | $d_1 = \mathcal{R}(0, 0, 0, 0, 0)$ | $\omega^{AFQ3}_1(k) = 0 + O(k^3)$ | gapless   | 2      |
|      | $d_2 = \mathcal{R}(\chi, \theta, 0, 0, 0)$ | $\omega^{AFQ3}_2(k) = 2Z[K_3\sqrt{C_k^2 - D_k^2Z^2}(k)$ | gapless   | 1      |
|      |               | $+ k_{1-k}Z]$ | gapless   | 1      |

Wave theory, these spectral functions can be evaluated for each FQ or AFQ state; these functions are distinct in different phases but do not qualitatively change within a single phase. Moreover, $S(q, \omega)$ and $Q(q, \omega)$ share the same structure as long as the elementary excitations are flavor waves, as demonstrated in Fig. [3]. This similarity provides evidence for the underlying $SU(3)$ structure and serves as a clue to the eight-fold way. The details of these spectral functions for all FQ and AFQ phases can be found in the Supplementary Materials.

Emergent gapless spin liquid. In the mean-field theory, the AFQ3 ground states are locally degenerate inside an energy gap. This degeneracy arises from the unperturbative Hamiltonian $K_3H_3$ and will be lifted by a finite $K_1$ and $K_2$. To address this case and go beyond the mean-field theory, we consider perturbations of up to the third order in the limit $K_3 \gg |K_1(2)|$. As an example, consider a square lattice; the spins have a quadratic order on one of the two sublattices, and we have the following effective Hamiltonian on the other sublattice (see the Supplementary Materials):

$$
\mathcal{H}_{eff} = J_1 \sum_{\langle ij \rangle_1} \mathcal{P}(S_i^x S_j^x + Q_i^x Q_j^x + Q_i^y Q_j^y - y_i Q_j^x y_j) \mathcal{P} + J_2 \sum_{\langle ij \rangle_2} \mathcal{P}(S_i^x S_j^y + Q_i^x Q_j^y + Q_i^y Q_j^x - y_i Q_j^x y_j) \mathcal{P},
$$

where $\mathcal{P}$ projects a state into the subspace spanned by the local basis $\{|x_i|, |y_i|\}, \langle ij \rangle_1(2)$ denotes a pair of (next) nearest neighboring sites on the sublattice, $J_1 = 2K_1K_2^2/K_3^2$ and $J_2 = K_1K_2^2/K_3^2$. Note that this is an effective $S = 1/2$ $J_1$-$J_2$ Heisenberg model constructed by $SU(2)$, generators. When $K_1 < 0$, $J_1(2) < 0$, and the ground state is of ferromagnetic order. When $K_1 > 0$, $J_1 > 2J_2 > 0$ gives rise to a gapless quantum spin liquid (QSL) ground state [27,30]. We expect the QSL state to be stable against higher-order perturbations because there is a QSL phase in the vicinity of $J_1 = 2J_2$ in the $J_1$-$J_2$ Heisenberg model.

In summary, we have revealed hidden $SU(2)$ symmetries in spin-1 quantum magnets, studied them in accordance with the $SU(3) \supset SU(2) \times U(1)$ symmetry hierarchy, demonstrated novel emergent phenomena, and found some clues to the emergent eight-fold way. These $SU(2)$ symmetries may be realized in cold atoms as well as $d^8$ and/or $d^6$ electrons with the proper specific choices of the spin-orbital couplings.

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FIG. 2. The spin spectral functions $S(q, \omega)$ and the spin quadrupole spectral functions $Q(q, \omega)$ for the FQ and AFQ phases. Here, we set $K_2 = K_3 = 0.1K_1$ for FQ1 and AFQ1, $K_1 = K_3 = 0.1K_2$ for FQ2 and AFQ2 and $K_1 = K_2 = 0.1K_3$ for FQ3 and AFQ3.

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Supplementary Materials

Appendix A: Fundamentals of SU(3) Lie algebra

The eight Gell-Mann matrices are defined as,

\[
\begin{align*}
\lambda_1 &= \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\
\lambda_2 &= \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\
\lambda_3 &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\
\lambda_4 &= \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \\
\lambda_5 &= \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, \\
\lambda_6 &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \\
\lambda_7 &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \\
\lambda_8 &= \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -2 \end{pmatrix}.
\end{align*}
\]

The generators of SU(3) Lie group are given by \( T_i = \lambda_i/2, \ i = 1, \ldots, 8. \)

In SU(3) representations, a state in an irreducible representation (IR) is labelled by \((p, q)\), corresponding to the weight vector \( \mu = p\mu^1 + q\mu^2 \), where \( \mu^1 = (1/2, \sqrt{3}/6) \) and \( \mu^2 = (1/2, -\sqrt{3}/6) \). The weights are defined by the eigenvalues of the Cartan generators \( H_1 \) and \( H_2 \), \( H, \mu = \mu_i \mu \) where \( i = 1, 2 \). Thus a state in a SU(3) IR can be written as \( |n, m, (p, q)\rangle \). Note that there may exist more than one \((p, q)\) state in IR \((n, m)\), these different \((p, q)\) states are distinguished by the subscript \( k \), which will be neglected when there is only one \((p, q)\) state.

Appendix B: SU(3) structure and Hidden SU(2) symmetries

Firstly, it is straightforward to examine the SU(3) Lie algebra relation among \( \{S, Q\} \), through the commutators \([S^\alpha, S^\beta], [S^\alpha, Q^\mu]\) and \([Q^\mu, Q^\nu]\) directly. As mentioned, besides the SU(3) algebra \( \{S^\alpha, S^\beta, S^\gamma\} \), there are other SU(2) subalgebras belonging to the SU(3) Lie algebra.

In order to find out the other SU(2) subalgebras, we consider the Cartan subalgebra \( H \), the largest commutative subalgebra, of the SU(3) Lie algebra, which can be chosen to be made of linear combinations of two commutative operators \( H_1 = T_3 \) and \( H_2 = T_8 \), where \( T_i = \lambda_i/2 \) and satisfy \( \text{Tr}(H_i H_j) = \frac{1}{2} \delta_{ij} \). An SU(2) subalgebra can be constructed as follows. Let us select an operator in the Cartan subalgebra \( H \), which serves as \( J^z \) in the SU(2) algebra. Writing \( \mathbf{H} = \{H_1, H_2\} \), we have \( J^z = |\alpha|^2 \mathbf{H} \), where \( \alpha \) is a two dimensional vector. Then the raising and lowering operators \( J^\pm \) can be obtained through \( J^\pm = |\alpha|^{-1} E_{\pm \alpha}, \) with \( \alpha \) a root vector. It is easy to verify that \( [J^+, J^-] = \pm J^z \) and \( [J^+, J^-] = 2J_z \). So that a nonzero root \( \alpha \) of SU(3) will give rise to an SU(2) subgroup.

\[
J^\pm = |\alpha|^{-1} E_{\pm \alpha}.
\]

\[
\begin{align*}
J^\pm &= |\alpha|^{-1} E_{\pm \alpha}, \quad \text{with} \ \alpha \ \text{a root vector}. \\
\text{It is easy to verify that} \ [J^+, J^-] &= \pm J^z \ \text{and} \ [J^+, J^-] = 2J_z. \\
\text{So that a nonzero root} \ \alpha \ \text{of SU(3) will give rise to an SU(2) subgroup.}
\end{align*}
\]
where \( \mathbf{d} = (d^x, d^y, d^z) \) is a complex vector, and normalization condition is given by \( |\mathbf{d}|^2 = 1 \). So that a time reversal invariant state is given by \( |\mathbf{d}|^2 = 1 \). The expectation values for \( \{\mathbf{S}, \mathbf{Q}\} \) can be expressed in terms of the \( \mathbf{d} \) vector, \( \langle \mathbf{S} \rangle = \mathbf{d}^2 \), \( \langle \mathbf{Q} \rangle = \mathbf{d}^3 \), \( \langle \mathbf{Q}^2 \rangle = \mathbf{d}^4 \), where \( \mathbf{d}^3 \) is the conjugate complex number of \( \mathbf{d}^3 \). Then the path integral for a spin \( S = 1 \) system can be written as in Eq. (2), where the Hamiltonian \( \mathcal{H} \) is given by Eq. (1), while the operators \( \mathbf{S} \) and \( \mathbf{Q} \) are replaced by their expectation values as follows,

\[
\begin{pmatrix}
S^x \\
S^y \\
S^z \\
Q^{x^2-y^2} \\
Q^{x^2-z^2} \\
Q^{y^2} \\
Q^{z^2} \\
\end{pmatrix}
= \begin{pmatrix}
+d^1\lambda_d^1 \\
-d^1\lambda_d^1 \\
+d^2\lambda_d^2 \\
-d^2\lambda_d^2 \\
+d^3\lambda_d^3 \\
-d^3\lambda_d^3 \\
+d^4\lambda_d^4 \\
-d^4\lambda_d^4
\end{pmatrix}.
\]

Now it is clear that the unitary transformation of the three dimensional complex \( \mathbf{d} \) vector (apart from a global phase factor) gives rise to the underlying \( SU(3) \) structure. Thus the \( SU(3) \) algebra of \( \{\mathbf{S}, \mathbf{Q}\} \) can be visualized from Eq. (B3). Since the complex \( \mathbf{d} \) vector transfer as a 1-rank tensor under the \( SU(3) \) rotations, one can find how \( \mathbf{S} \) and \( \mathbf{Q} \) and other physical quantities will transfer under \( SU(3) \) as well, which can be written in bilinear or biquadratic terms of \( \mathbf{d} \) and \( \mathbf{d}^* \) in the path integral.

\section*{Appendix C: \( SU(2) \gamma \) symmetric states/Hamiltonians}

In the language of group theory, the three components of \( \mathbf{d} \) belong to the 3-dimensional (3D) fundamental representation \( 3 \equiv (1, 0) \) of \( SU(3) \) group, and those of \( \mathbf{d}^* \) belong to its complex conjugate representation \( 3 \equiv (1, 0) \). So that each \( (\mathbf{d}, \mathbf{d}^*) \) bilinear term belongs to the representations \( 3 \otimes 3 = 1 \oplus 8 \), where \( 1 \equiv (0, 0) \) and \( 8 \equiv (1, 1) \). Explicitly, \( |\mathbf{d}|^2 \) belongs to the 1D IR \( (0, 0) \), and \( \langle \mathbf{S}, \mathbf{Q} \rangle \) belong to the 8D IR \( (3, 3) \). Furthermore, each \( \langle \mathbf{S}, \mathbf{Q} \rangle \) bilinear term in Eq. (1) belongs to the representations \( (1, 1) \otimes (1, 1) = (0, 0) \oplus (1, 1) \oplus (3, 0) \oplus (0, 3) \oplus (2, 2) \). Therefore, we are able to classify the terms in Eq. (1) according to group theory and find possible spin Hamiltonians respecting the hidden \( SU(2) \) symmetries.

Begin with \( \mathbf{d} \) vector and its complex conjugate \( \mathbf{d}^* \), the Cartesian coordinate representation of the three spin-1 states is isomorphic to \( SU(3) \) IR \( (1, 0) \),

\[
\begin{array}{ccc}
|\mathbf{0}, (0, 1)\rangle & \leftrightarrow & |x\rangle \leftrightarrow d^x, \\
|\mathbf{0}, (1, -1)\rangle & \leftrightarrow & |z\rangle \leftrightarrow d^z, \\
|\mathbf{0}, (0, -1)\rangle & \leftrightarrow & |y\rangle \leftrightarrow d^y,
\end{array}
\]

and its complex conjugate representation \( (0, 1) \),

\[
\begin{array}{ccc}
|\mathbf{0}, (0, 1)\rangle & \leftrightarrow & |y\rangle \leftrightarrow \tilde{d}^y, \\
|\mathbf{0}, (1, -1)\rangle & \leftrightarrow & |z\rangle \leftrightarrow -\tilde{d}^z, \\
|\mathbf{0}, (1, 0)\rangle & \leftrightarrow & |x\rangle \leftrightarrow -\tilde{d}^x,
\end{array}
\]

where \( |\mathbf{m}, n\rangle \) was defined in previous section. Then \( \langle \mathbf{S}, \mathbf{Q} \rangle \) can be obtained through \( (0, 1) \otimes (1, 0) = (0, 0) \oplus (1, 1) \), which belong to the 8D IR \( (1, 1) \),

\[
\begin{array}{ccc}
|\mathbf{1}, (1, 1)\rangle & \leftrightarrow & -i\sqrt{3}Q^{xy}, \\
|\mathbf{1}, (1, -1)\rangle & \leftrightarrow & -i\sqrt{3}Q^{yz}, \\
|\mathbf{1}, (1, 0)\rangle & \leftrightarrow & -i\sqrt{3}Q^{zx}, \\
\end{array}
\]

In what follows, we shall construct \( SU(2) \gamma \) symmetric two-body interactions in terms of bilinear forms of \( \langle \mathbf{S}, \mathbf{Q} \rangle \). As mentioned, such bilinear forms belong to the representations \( (1, 1) \otimes (1, 1) = (0, 0) \oplus (1, 1) \oplus (1, 1) \oplus (3, 0) \oplus (3, 0) \oplus (2, 2) \). Firstly we shall find out all the \( SU(2) \gamma \) symmetric states in the IR decomposition of \( (1, 1) \otimes (1, 1) \), which would be annihilated by both the raising operator \( E_\gamma \) and the lowering operator \( E_{-\gamma} \). According to the block diagram shown in FIG. 4, there exist six linear independent states in the IR decomposition. We list six linear independent self-conjugate \( SU(2) \gamma \) symmetric states as follows,

\[
\begin{array}{ccc}
\Gamma_0 & = & |\mathbf{0}, (0, 0)\rangle, \\
\Gamma_1 & = & |(1, 1), (0, 0)\rangle_S + \sqrt{3}|(1, 1), (0, 0)\rangle_A, \\
\Gamma_2 & = & |(1, 1), (0, 0)\rangle_A + \sqrt{3}|(1, 1), (0, 0)\rangle_A, \\
\Gamma_3 & = & |(3, 0), (3, -3)\rangle + |(0, 3), (3, -3)\rangle, \\
\Gamma_4 & = & i|\mathbf{0}, (1, 1)\rangle - i|\mathbf{0}, (3, 3)\rangle, \\
\Gamma_5 & = & \sqrt{3}|(2, 2), (0, 0)\rangle - |(2, 2), (0, 0)\rangle
\end{array}
\]

Note that we have already made the bilinear forms symmetrized or antisymmetrized, where \( |(1, 1), (0, 0)\rangle_S \) is a symmetric state and \( |(1, 1), (0, 0)\rangle_A \) is an antisymmetric state. All the possible \( SU(2) \gamma \) symmetric states can be written as a linear combination of \( \Gamma_n, n = 0, \ldots, 5 \) in Eq. (C4). Expanding \( \Gamma_n \) in terms of \( (1, 1) \otimes (1, 1) \) states, \( |(1, 1), (p_1, q_1)\rangle \otimes |(1, 1), (p_2, q_2)\rangle \), through \( SU(3) \) Clebsch-Gordan coefficients, and replacing abstract states \( |(1, 1), (p, q)\rangle \) by physical operators \( \langle \mathbf{S}, \mathbf{Q} \rangle \) and \( \mathbf{S}^{xy} \), we obtain all the \( SU(2) \gamma \) symmetric spin Hamiltonians.
Eq. (3) in the main text. For simplicity, putting Eq. (C3) into Eq. (C5), we obtain

$$\Gamma_{\gamma} = \frac{1}{\sqrt{8}} \left( |0, 0\rangle_i |0, 0\rangle_j + |2, 0\rangle_i |0, 0\rangle_j \right) + \frac{1}{\sqrt{8}} \left( |1, 1\rangle_i [-1, -1]_j - |[-1, 2]_i |1, -2]_j - |[2, -1]_i |[-2, 1]_j + (i \leftrightarrow j) \right)$$

$$\Gamma_{1} = \frac{1}{\sqrt{20}} \left( 2 |2, 1\rangle_i |[-2, 1]_j + 2 |[-2, 1]_i |1, -2]_j + 4 |1, 1\rangle_i [-1, -1]_j + (i \leftrightarrow j) \right)$$

$$+ \frac{1}{\sqrt{20}} \left( -2 |0, 0\rangle_i |0, 0\rangle_j + 2 |2, 0\rangle_i |0, 0\rangle_j - 2 \sqrt{3} |0, 0\rangle_i |0, 0\rangle_j - 2 \sqrt{3} |2, 0\rangle_i |0, 0\rangle_j \right),$$

$$\Gamma_{2} = \left( |2, -1\rangle_i |[-2, 1]_j - |[-2, 1\rangle_i |1, -2]_j - (i \leftrightarrow j) \right),$$

$$\Gamma_{3} = \frac{1}{\sqrt{2}} \left( |[-1, 2]_i |[-2, 1]_j + |1, -2\rangle_i |2, -1]_j - (i \leftrightarrow j) \right),$$

$$\Gamma_{4} = \frac{1}{\sqrt{2}} \left( |[-1, 2]_i |[-2, 1]_j - |[-1, 2\rangle_i |2, -1]_j - (i \leftrightarrow j) \right),$$

$$\Gamma_{5} = \frac{1}{\sqrt{40}} \left( 3 |2, -1\rangle_i |[-2, 1]_j + 3 |[-1, 2]_i |1, -2]_j + |1, 1\rangle_i [-1, -1]_j + (i \leftrightarrow j) \right)$$

$$+ \frac{1}{\sqrt{40}} \left( 7 |0, 0\rangle_i |0, 0\rangle_j + 3 |0, 0\rangle_i |0, 0\rangle_j + 2 \sqrt{3} |0, 0\rangle_i |0, 0\rangle_j + 2 \sqrt{3} |0, 0\rangle_i |0, 0\rangle_j \right),$$

where we use $|p, q\rangle_i$ to denote state $|p, q\rangle$ at site $i$ for simplicity. Putting Eq. (C3) into Eq. (C5), we obtain Eq. (3) in the main text.

### Appendix D: Flavor-wave theory

In this appendix, we provide details for the flavor-wave theory\cite{23,26} In order to study low energy excitations, we assign three flavors of Shwinger bosons $a_{\alpha}(j)$ at each
site \( j \) on the \( n^{th} \) sublattice, where \( \alpha = x, y, z \) corresponds to \( x, y, z \) spin states defined in Eq. 12. Here \( n = 1 \) for the uniform states, while \( n = 1, 2 \) for the bipartite-lattice ordered states. Thus, the operators \( (S, Q) \) can be written bilinearly in terms of Schwinger bosons,

\[
S_n^a(j) = -i \sum_{\alpha,\beta} \epsilon_{\alpha\beta} a_{n\alpha}^\dagger(j)a_{n\beta}(j), \quad \text{(D1a)}
\]

\[
Q_n^{m\nu}(j)|_{m \neq \nu} = -[a_{nm}^\dagger(j)a_{m\nu}(j) + a_{m\nu}(j)a_{nm}(j)], \quad \text{(D1b)}
\]

\[
Q_n^{x^2-y^2}(j) = -[a_{nx}(j)a_{nx}(j) - a_{ny}(j)a_{ny}(j)], \quad \text{(D1c)}
\]

\[
Q_n^{z^2-r^2}(j) = -\frac{1}{\sqrt{3}}[3a_{nz}(j)a_{nz}(j) - 1], \quad \text{(D1d)}
\]

where the single occupancy constraint

\[
\sum_\alpha a_{n\alpha}^\dagger(j)a_{n\alpha}(j) = 1 \quad \text{(D2)}
\]

is imposed.

To obtain various spin ordered states, we shall condense these Schwinger bosons at some components. Without loss of generality, the condensate components are constructed by an \( SU(3) \) rotation \( \Omega_n \) in the \( n \)-th sublattice, which is defined as follows,

\[
\begin{pmatrix}
  a_{n\bar{x}} \\
  a_{n\bar{y}} \\
  a_{n\bar{z}}
\end{pmatrix} = \Omega_n \begin{pmatrix}
  a_{nx} \\
  a_{ny} \\
  a_{nz}
\end{pmatrix}. \quad \text{(D3)}
\]

Such an \( SU(3) \) rotation \( \Omega_n \) is site-independent and determined by corresponding mean-field \( d \) vectors and enable us to attribute the condensate to \( a_{n\bar{z}} \) component only. And \( a_{n\bar{y}} \) and \( a_{n\bar{z}} \) components are thought as small fractions. Then the low energy Hamiltonian can be bilinearized by the Holstein-Primakoff transformation. Approximately, \( a_{n\bar{z}}^\dagger(j) \) and \( a_{n\bar{z}}(j) \) can be written as,

\[
a_{n\bar{z}}^\dagger(j) = a_{n\bar{z}}(j) = \sqrt{M - a_{n\bar{y}}^2(j)a_{n\bar{y}}(j) - a_{n\bar{z}}^2(j)a_{n\bar{z}}(j)}, \quad \text{(D4)}
\]

where \( M = 1 \) in present case considering the single occupancy constraint.

Then we carry out the 1/M expansion in the Holstein-Primakoff bosons \( a_{n\bar{y}} \) and \( a_{n\bar{z}} \) up to quadratic order, and perform the Fourier transformation \( a_{n\alpha}(k) = \sum_y e^{i\mathbf{k} \cdot \mathbf{r}_a(j)/\sqrt{N}} \) to obtain the Hamiltonian \( \mathcal{H} \) in the \( k \)-space, where \( \mathbf{r}_a \) is the position of the lattice site \( j \) and \( N \) is the number of magnetic unit cells. Thus the \( k \)-space Hamiltonian can be diagonalized by the bosonic Bogoliubov transformation,

\[
\mathcal{H} = \sum_{m,k} \omega_m(k)b_m^\dagger(k)b_m(k) + C, \quad \text{(D5)}
\]

where \( \omega_m(k) \) is the energy dispersion of \( m \)-th branch flavor wave, \( b_m(k) \) and \( b_m^\dagger(k) \) are bosonic Bogoliubov quasiparticle annihilation and creation operators, and the constant \( C \) does not depend on boson fields. For uniform states, say, FQ states, \( m = 1, 2 \); while for AFQ states, \( m = 1, 2, 3, 4 \). As long as the ground states of \( \mathcal{H} \) determined by \( K_1, K_2 \) and \( K_3 \) are given, we are able to obtain the dispersions \( \omega_m(k) \) simultaneously.

**FQ1 phase** For FQ1 phase, the d-vector of ground state reads \( \mathbf{d}^{\text{FQ1}} = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \), and the global rotational matrix \( \Omega^{\text{FQ1}} \) is a \( 3 \times 3 \) unit matrix. We introduce the \( SU(3) \) Schwinger bosons as

\[
\begin{pmatrix}
  a_y \\
  a_z
\end{pmatrix} = \begin{pmatrix}
  \sqrt{n_x} \\
  a_y \\
  a_z
\end{pmatrix}, \quad \text{(D6)}
\]

where

\[
\sqrt{n_x} \equiv 1 - a_y^\dagger a_y - a_z^\dagger a_z \quad \text{(D7)}
\]

Expanding spin dipolar and quadrupolar operators \( S \) and \( Q \) up to quadratic order of \( a_y \) and \( a_z \) gives rise to

\[
S^x = -i(a_y^\dagger a_z - a_z^\dagger a_y)
\]

\[
S^y = -i(a_y^\dagger a_z - a_z^\dagger a_y)
\]

\[
S^z = i(a_y^\dagger a_y)
\]

\[
Q^{xy} = -(a_y^\dagger a_y + a_z^\dagger a_z)
\]

\[
Q^{yz} = -(a_y^\dagger a_z + a_z^\dagger a_y)
\]

\[
Q^{xz} = -a_z^\dagger a_z
\]

\[
Q^{z^2-r^2} = \frac{1}{3}(3a_y^\dagger a_y - 1). \quad \text{(D8)}
\]

Put them into the Hamiltonian and keep all the terms up to quadratic order of \( a_y \) and \( a_z \), we obtain

\[
\mathcal{H}^{\text{FQ1}} = \sum_{m=1}^2 \omega_m^{\text{FQ1}}(k)b_m^\dagger(k)b_m(k), \quad \text{(D9)}
\]

where

\[
b_1(k) = a_y(k), \quad b_2(k) = a_z(k). \quad \text{(D10)}
\]

Here \( \omega_m^{\text{FQ1}}(k) \) are given in Table II in the main text. Note that all the spins condense at the \( |x\rangle \) state. So that \( b_1 = a_y^\dagger \) creates a \( |y\rangle \) state and must annihilates an \( |x\rangle \) state simultaneously to satisfy the single occupancy constraint in Eq. 12. It means that the \( \omega_1^{\text{FQ1}}(k) \) mode corresponds to a two-magnon excitation. Similarly, \( \omega_2^{\text{FQ1}}(k) \) causes an \( |x\rangle \rightarrow |z\rangle \) transition and is a one-magnon mode.

**FQ2 phase** Considering the d-vector of a FQ2 state being the form of \( d^{\text{FQ2}} = \begin{pmatrix} \cos \vartheta \\ 0 \\ -\sin \vartheta \end{pmatrix} \) and the global rotational matrix

\[
\Omega^{\text{FQ2}} = \begin{pmatrix}
  \cos \vartheta & 0 & -\sin \vartheta \\
  0 & 1 & 0 \\
  \sin \vartheta & 0 & \cos \vartheta
\end{pmatrix}, \quad \text{(D11)}
\]
we introduce rotated Schwinger bosons as follows,

\[
\begin{pmatrix}
\alpha_x \\
\alpha_y \\
\alpha_z
\end{pmatrix} = \begin{pmatrix}
\cos \vartheta \sqrt{n_x} & -\sin \vartheta a_z \\
\sin \vartheta \sqrt{n_x} + \cos \vartheta a_z
\end{pmatrix}.
\] (D12)

where \( \sin \vartheta \) is determined by the mean-field theory and given in the caption in Table II in the main text. Similarly, the operators \((S, Q)\) can be expanded to quadratic order of \(a_y\) and \(a_z\) as follows,

\[
S^x = -i \sin \vartheta (a_y^\dagger a_y - a_y a_y^\dagger) - i \cos \vartheta (a_y^\dagger a_z - a_z^\dagger a_y),
\]

\[
S^y = -i (a_z^\dagger - a_z),
\]

\[
S^z = i \cos \vartheta (a_y^\dagger - a_y) - i \sin \vartheta (a_y^\dagger a_z - a_z^\dagger a_y),
\]

\[
Q^{xy} = -\cos \vartheta (a_y^\dagger + a_y) + \sin \vartheta (a_y^\dagger a_z + a_z^\dagger a_y),
\]

\[
Q^{yz} = -\sin \vartheta (a_y^\dagger + a_y) + \sin \vartheta (a_y^\dagger a_z + a_z^\dagger a_y),
\]

\[
Q^{zx} = (\sin^2 \vartheta - \cos^2 \vartheta)(a_z + a_z^\dagger)
- 2 \sin \vartheta \cos \vartheta (1 - a_y^\dagger a_y - 2a_z^\dagger a_z),
\]

\[
Q^{x^2-y^2} = -\cos^2 \vartheta + \cos \vartheta \sin \vartheta (a_z + a_z^\dagger)
+ (1 + \cos^2 \vartheta) a_y^\dagger a_y + (2 \cos^2 \vartheta - 1)a_z^\dagger a_z,
\]

\[
Q^{x^2-z^2} = \sqrt{3} \cos^2 \vartheta (a_z^\dagger a_z - \tan^2 \vartheta (a_y^\dagger a_y + a_y a_z^\dagger))
+ \frac{1}{\sqrt{3}} (3 \sin \vartheta \cos \vartheta (a_z^\dagger + a_z) + 3 \sin^2 \vartheta - 1).
\] (D13)

Finally we obtain the diagonalized Hamiltonian

\[
\mathcal{H}^{FQ2} = \sum_k \sum_{m=1}^2 \omega_m^{FQ2}(k)b_m^\dagger(k)b_m(k),
\] (D14)

where \(\omega_{1,2}^{FQ2}(k)\) are given in Table II and the Bogoliubov transformation reads

\[
a_y(k) = b_1(k),
\]

\[
a_z(k) = \cosh(\rho^F_{k}^{Q2})b_2(k) + \sinh(\rho^F_{k}^{Q2})b_2^\dagger(-k),
\] (D15)

with

\[
\exp(\rho^F_{k}^{Q2}) = \sqrt{\frac{1 - \gamma(k)}{1 + B_K \gamma(k)}}.
\] (D16)

where \(B_K\) and \(\gamma(k)\) are given in Table II. In this case, condensate components are of \(|x\) and \(|z\) spins. Such that \(\omega_{2}^{FQ2}(k)\) mode corresponds to \(|x\rangle \leftrightarrow |z\rangle\) transition, and is a one-magnon mode, while \(\omega_{1}^{FQ2}(k)\) mode corresponds to \(|x\rangle \leftrightarrow |y\rangle\) transition, and is an admixture of one-magnon and two-magnon modes.

**FQ3 phase** Now the d-vector is \(d^{FQ3} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}\), and the global rotational matrix reads

\[
\Omega^{FQ3} = \begin{pmatrix}
0 & 0 & -1 \\
0 & 1 & 0 \\
1 & 0 & 0
\end{pmatrix}.
\] (D17)

Then the rotated Schwinger bosons becomes

\[
\begin{pmatrix}
\alpha_x \\
\alpha_y \\
\alpha_z
\end{pmatrix} = \begin{pmatrix}-a_z \\ a_y \\ a_z \sqrt{n_x}\end{pmatrix}.
\] (D18)

And the operators \(S\) and \(Q\) read

\[
S^x = -i(a_y^\dagger - a_y),
\]

\[
S^y = -i(a_z^\dagger - a_z),
\]

\[
S^z = i(a_z^\dagger a_y - h.c),
\]

\[
Q^{xy} = a_y^\dagger a_y + h.c,
\]

\[
Q^{yz} = -i(a_z^\dagger + a_y),
\]

\[
Q^{zx} = a_z^\dagger + a_z,
\]

\[
Q^{x^2-y^2} = (a_z^\dagger a_z - a_y^\dagger a_y),
\]

\[
Q^{x^2-z^2} = \frac{1}{\sqrt{3}} (3a_y^\dagger a_y + 3a_z^\dagger a_z - 2).
\] (D19)

Put them into the Hamiltonian we obtain

\[
\mathcal{H}^{FQ3} = \sum_k \sum_{m=1}^2 \omega_m^{FQ3}(k)b_m^\dagger(k)b_m(k),
\] (D20)

where \(\omega_1^{FQ3}(k)\) are given in Table II and the Bogoliubov transformation reads

\[
a_y(k) = b_1(k),\ a_z(k) = b_2(k).
\] (D21)

In this case, the condensate component is \(|z\). Such that \(\omega_1^{FQ3}(k)\) mode gives rise to \(|z\rangle \rightarrow |y\rangle\) transition and is a one-magnon mode, and \(\omega_2^{FQ3}(k)\) mode gives rise to \(|x\rangle \leftrightarrow |x, y\rangle\) transition and is a one-magnon mode too.

**AFQ1 phase** The d-vectors in sublattices 1 and 2 are of the form \(d_1^{AFQ1} = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, d_2^{AFQ1} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}\), and the corresponding global rotational matrices \(\Omega_1^{AFQ1}\) and \(\Omega_2^{AFQ1}\) read,

\[
\Omega_1^{AFQ1} = \begin{pmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{pmatrix}, \Omega_2^{AFQ1} = \begin{pmatrix}
0 & -1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 1
\end{pmatrix}.
\] (D22)

Therefore the \(SU(3)\) Schwinger bosons in the rotated representation can be written as

\[
\begin{pmatrix}
a_{1x} \\
a_{1y} \\
a_{1z}
\end{pmatrix} = \begin{pmatrix}
\sqrt{n_{1x}} \\
a_{1y} \\
a_{1z}
\end{pmatrix},
\]

\[
\begin{pmatrix}
a_{2x} \\
a_{2y} \\
a_{2z}
\end{pmatrix} = \begin{pmatrix}
0 \\
a_{2y} \\
\sqrt{n_{2x}}
\end{pmatrix},
\]

where \(\sqrt{n_{mx}} = 1 - a_{my} a_{my} - a_{mx} a_{mx}\) for \(m = 1, 2\). Expanding \((S, Q)\) to quadratic order of \(a_{my}\) and \(a_{mx}\) in
each sublattice gives rise to

\[ S_1^x = -i(a_{11}^y a_{12} - a_{12}^y a_{11}), \quad S_2^x = i(a_{12}^y a_{22} - a_{22}^y a_{12}), \]

\[ S_1^y = -i(a_{11}^y a_{12} - a_{12}^y a_{11}), \quad S_2^y = i(a_{12}^y a_{22} - a_{22}^y a_{12}), \]

\[ S_1^z = i(a_{11}^y a_{12} + a_{12}^y a_{11}), \quad S_2^z = i(a_{12}^y a_{22} + a_{22}^y a_{12}), \]

\[ Q_1^{xy} = -(a_{11}^y a_{12} + a_{12}^y a_{11}), \quad Q_2^{xy} = -(a_{12}^y a_{22} + a_{22}^y a_{12}), \]

\[ Q_1^{xz} = -(a_{11}^y a_{12} + a_{12}^y a_{11}), \quad Q_2^{xz} = (a_{12}^y a_{22} + 2a_{12}^z a_{22} z), \]

\[ Q_1^{yz} = -(1 - 2a_{11}^y a_{12} - a_{12}^y a_{12}), \quad Q_2^{yz} = (1 - 2a_{12}^y a_{22} - a_{22}^y a_{22}), \]

\[ Q_1^{zx} = \sqrt{3}(3a_{11}^y a_{12} - 1), \quad Q_2^{zx} = \sqrt{3}(3a_{12}^y a_{22} - 1). \]

(D24)

Then the mean-field Hamiltonian of AFQ1 becomes

\[ \mathcal{H}_{\text{AFQ1}} = \sum_k \sum_{m=1}^4 \omega_m^{\text{AFQ1}}(k) b_m^\dagger(k) b_m(k), \] (D25)

where \( \omega_{1,2}^{\text{AFQ1}}(k) \) are given in Table II in the main text. The Bogoliubov transformation reads

\[
\begin{align*}
a_{1z}(k) &= b_3(k), \\
a_2z(k) &= b_4(k), \\
\begin{pmatrix} a_{1y}(k) \\
a_{2y}(k) \\
\end{pmatrix} &= \begin{pmatrix} c_k^A & c_k^A \\
-c_k^A & s_k^A \\
\end{pmatrix} \begin{pmatrix} b_1(k) \\
b_2(k) \\
\end{pmatrix} \\
&\quad+ \begin{pmatrix} s_k^A \\
c_k^A \\
\end{pmatrix} \begin{pmatrix} b_2(-k) \\
b_1(-k) \\
\end{pmatrix}
\end{align*}
\] (D26)

with

\[
c_k^A \equiv \frac{1}{\sqrt{2}} \cosh(\rho_k^A), \quad s_k^A \equiv \frac{1}{\sqrt{2}} \sinh(\rho_k^A),
\]

and \( \rho_k^A \) is given as

\[
\exp(2\rho_k^A) = \sqrt{\frac{1 + \gamma(k)}{1 - \gamma(k)}}.
\]

(D28)

Similar to the case of FQ1, all the spins condense at the \(|x\rangle\) state. So that \( \omega_{1,2}^{\text{AFQ1}}(k) \) modes give rise to \(|x\rangle \rightarrow |y\rangle\) transitions and correspond to two-magnon excitations. And \( \omega_{3,4}^{\text{AFQ1}}(k) \) modes give rise to \(|x\rangle \leftrightarrow |z\rangle\) transition and are one-magnon excitations.

**AFQ2 phase** In this phase, the d-vectors in two sublattices are \( d_1^{\text{AFQ2}} = (\cos \vartheta, 0, \sin \vartheta) \) and \( d_2^{\text{AFQ2}} = (\cos \vartheta, 0, -\sin \vartheta) \), and the global rotational matrices read

\[
\begin{align*}
\Omega_1^{\text{AFQ2}} &= \begin{pmatrix} \cos \vartheta & 0 & -\sin \vartheta \\
0 & 1 & 0 \\
\sin \vartheta & 0 & \cos \vartheta \\
\end{pmatrix}, \\
\Omega_2^{\text{AFQ2}} &= \begin{pmatrix} \cos \vartheta & 0 & \sin \vartheta \\
0 & 1 & 0 \\
-\sin \vartheta & 0 & \cos \vartheta \\
\end{pmatrix}.
\end{align*}
\]

(D29)

We have \( SU(3) \) Schwinger bosons in such rotated representation as follows,

\[
\begin{pmatrix} a_{1\hat{e}} \\
 a_{1\hat{g}} \\
 a_{1\hat{f}} \\
\end{pmatrix} = \begin{pmatrix} \cos \vartheta \sqrt{\rho_{1\hat{e}}} - \sin \vartheta \rho_{1\hat{z}} \\
 a_{1\hat{g}} \\
 \sin \vartheta \sqrt{\rho_{1\hat{e}}} + \cos \vartheta \rho_{1\hat{z}} \\
\end{pmatrix},
\]

(D30)

Then the forms of \((S,Q)\) for each sublattice of AFQ2 are very similar to Eq. (D13), and here we do not list them explicitly.

The corresponding Hamiltonian becomes

\[ \mathcal{H}_{\text{AFQ2}} = \sum_k \sum_{m=1}^4 \omega_m^{\text{AFQ2}}(k) b_m^\dagger(k) b_m(k), \] (D31)

where \( \omega_{1,2}^{\text{AFQ2}} \) are given in Table II in the main text. The Bogoliubov transformation are chosen as

\[
\begin{pmatrix} a_{1y}(k) \\
a_{2y}(k) \\
\end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\
1 & -1 \\
\end{pmatrix} \begin{pmatrix} b_1(k) \\
b_2(k) \\
\end{pmatrix},
\]

and

\[
\begin{pmatrix} a_{1z}(k) \\
a_{2z}(k) \\
\end{pmatrix} = \begin{pmatrix} c_k^A & c_k^A & s_k^A & s_k^A \\
c_k^A & -c_k^A & s_k^A & -s_k^A \\
\end{pmatrix} \begin{pmatrix} b_3(k) \\
b_4(k) \\
b_1(-k) \\
b_2(-k) \\
\end{pmatrix},
\]

(D33)

where

\[
c_m^A \equiv \frac{1}{\sqrt{2}} \cosh(\rho_m^A), \quad s_m^A \equiv \frac{1}{\sqrt{2}} \sinh(\rho_m^A),
\]

with \( m = 1, 2 \) and

\[
\exp(2\rho_k^A) = \sqrt{\frac{1 + \gamma(k)}{1 - B_k \gamma(k)}}, \quad \exp(2\rho_k^A),
\]

(D35)

Similar to the case of FQ2, in this case condensate components are of \(|x\rangle \) and \(|z\rangle \) spins. So \( \omega_{1,2}^{\text{AFQ2}}(k) \) modes which correspond to \(|x\rangle \leftrightarrow |z\rangle\) transition are one-magnon modes, while \( \omega_{3,4}^{\text{AFQ2}}(k) \) modes correspond to \(|x\rangle \pm \tan \vartheta |z\rangle \leftrightarrow |y\rangle\) transitions, and are admixtures of one-magnon and two-magnon modes.

**AFQ3 phase** The AFQ3 ground states are given by the d-vectors in two sublattices as the following, \( d_1^{\text{AFQ3}} = \begin{pmatrix} 0 \\
0 \\
1 \\
0 \end{pmatrix} \), \( d_2^{\text{AFQ3}} = \begin{pmatrix} 0 \\
0 \\
0 \\
1 \end{pmatrix} \). Now the global rotational matrices \( \Omega_1^{\text{AFQ3}} \) and \( \Omega_2^{\text{AFQ3}} \) read,

\[
\begin{pmatrix} a_{1\hat{e}} \\
a_{1\hat{g}} \\
a_{1\hat{f}} \\
a_{2\hat{e}} \\
a_{2\hat{g}} \\
a_{2\hat{f}} \\
\end{pmatrix} = \begin{pmatrix} 0 & 0 & -1 & 0 \\
0 & 1 & 0 & 0 \\
1 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 \end{pmatrix}.
\]

(D36)
The corresponding Schwinger bosons in the rotated representations are
\[
\begin{pmatrix}
  a_{1\bar{x}} \\
  a_{2\bar{y}} \\
  a_{3\bar{z}}
\end{pmatrix} = \begin{pmatrix}
  -a_{1z} \\
  a_{1y} \\
  \sqrt{a_{1x}}
\end{pmatrix},
\begin{pmatrix}
  a_{2\bar{x}} \\
  a_{2\bar{y}} \\
  a_{2\bar{z}}
\end{pmatrix} = \begin{pmatrix}
  \sqrt{a_{2x}} \\
  a_{2y} \\
  a_{2z}
\end{pmatrix}.
\] (D37)

And operators \((S, Q)\) for each sublattice read
\[
S_1^y = -i(a_{1y}^\dagger - a_{1y}), \quad S_2^y = -i(a_{2y}^\dagger - a_{2y}),
\]
\[
S_1^y = i(a_{1y}^\dagger a_{1y} - a_{1y}^\dagger a_{1y}), \quad S_2^y = i(a_{2y}^\dagger a_{2y} - a_{2y}^\dagger a_{2y}),
\]
\[
Q_1^{xy} = a_{1y}^\dagger a_{1y} + a_{1y} a_{1y}^\dagger, \quad Q_2^{xy} = -(a_{2y}^\dagger a_{2y} + a_{2y} a_{2y}^\dagger),
\]
\[
Q_1^{yz} = -a_{1y}^\dagger a_{1y} + a_{1y} a_{1y}^\dagger, \quad Q_2^{yz} = -(a_{2y}^\dagger a_{2y} + a_{2y} a_{2y}^\dagger),
\]
\[
Q_1^{zx} = a_{1z}^\dagger a_{1z} + a_{1z} a_{1z}^\dagger, \quad Q_2^{zx} = -(a_{2z}^\dagger a_{2z} + a_{2z} a_{2z}^\dagger),
\]
\[
Q_1^{z^2} = -(a_{1z} a_{1z}^\dagger - a_{1z}^\dagger a_{1z}), \quad Q_2^{z^2} = -(a_{2z} a_{2z}^\dagger - a_{2z}^\dagger a_{2z}).
\] (D38)

The diagonalized Hamiltonian reads
\[
\mathcal{H}_{AFQ_3} = \sum_k \sum_{m=1}^4 \omega_{m_{AFQ_3}}(k) b_m^\dagger(k) b_m(k).
\] (D39)

where \(\omega_{1,2,3,4}\) are given in Table II and
\[
\begin{pmatrix}
  a_{1y}(k) \\
  a_{2y}(k) \\
  a_{1z}(k) \\
  a_{2z}(k)
\end{pmatrix} = \begin{pmatrix}
  \cosh(\rho_{k}^{A_1}) \sinh(\rho_{k}^{A_2}) \\
  \sinh(\rho_{k}^{A_1}) \cosh(\rho_{k}^{A_2})
\end{pmatrix}
\begin{pmatrix}
  b_3(k) \\
  b_4(k)
\end{pmatrix}.
\] (D40)

Here
\[
\exp(2\rho_{k}^{A_1}) = \frac{3K_3 + K_1 - 4K_2 \gamma(k)}{3K_3 + K_1 + 4K_2 \gamma(k)}.
\] (D41)

In this case all spins condense at the \(|x\rangle\) state. Thus \(\omega_{1,2,3,4}\) modes corresponding to \(|x\rangle \leftrightarrow |y\rangle\) transitions are two-magnon modes. While \(\omega_{3,4}\) modes corresponding to \(|x\rangle \leftrightarrow |z\rangle\) transitions are one-magnon modes.

Appendix E: Spin spectral functions

Inelastic neutron scattering measures the spin spectral function in \((q,\omega)\) space, which is defined as \(S^{\alpha\beta}(q,\omega) = \text{Im} \{ i \int_0^\infty dt e^{i\omega t} \langle [S^\alpha(q,t), S^\beta(-q,0)] \rangle \} \), where we have set \(\hbar = 1\) for simplicity. With the help of flavor-wave theory, the spin spectral function in each FQ or AFQ phase can be evaluated. At zero temperature, \(S^{\alpha\beta}(q,\omega)\) depends on the choice of ground state. However, the spectral function \(S(q,\omega) = S^{xx}(q,\omega) + S^{yy}(q,\omega) + S^{zz}(q,\omega)\) do not change qualitatively within a single phase. In this appendix, we provide details for spin spectral function, \(S(q,\omega)\), which is calculated by the linearized flavor-wave theory.

In the mean-field theory, all the degenerate ground states can be obtained from one of them by an \(SU(2) \times U(1)\) rotation of the \(d\)-vector. We parameterize a general \(SU(2)\) rotational matrix as
\[
R = r_0 \sigma_0^0 + i \sum_{n=1}^3 r_n \sigma_n,
\] (E1)

where \(\sigma_0\) are identity as well as \(\sigma_1\), \(\sigma_2\) and \(\sigma_3\) are three Pauli matrices. Here \(\hat{r} = \{r_0, r_1, r_2, r_3\}\) is a 4-dimensional real vector with \(\sum_{n=0}^3 r_n^2 = 1\). Thus, apart from a global phasefactor, two \(d\) vectors of two degenerate ground states, \(d\) and \(d'\), are related by a \(3 \times 3\) matrix as follows,
\[
d' = \begin{pmatrix}
  e^{-i\phi} & 0_{1,2} \\
  0 & 1
\end{pmatrix} d,
\] (E2)

where \(0_{1,2}(0_{1,2})\) is a \(1 \times 2 (2 \times 1)\) zero matrix. In the \(SU(3)\) Schwinger boson representation, the expression for the spin operators \(S\) depends on the parameters \(r_0, r_1, r_2\) and \(r_3\).

FQ1 phase The spin operators in the flavor-wave theory read
\[
S^x = (r_1 - i r_2) a_x + (r_1 + i r_2) a_x^\dagger,
\]
\[
S^y = -(r_3 - i r_0) a_y + (r_3 + i r_0) a_y^\dagger,
\]
\[
S^z = u a_y + u^* a_y^\dagger,
\] (E3)

where
\[
u = i (r_0 - r_3) a_y^\dagger a_y + (r_2 + i r_1) a_x^\dagger a_x^\dagger,
\] (E4)

and the quadratic boson and constant terms are omitted. Note that the constant does not contribute to any excitations thereby the spectral functions. Then spin spectral function reads
\[
S_{FQ1}(q, \omega) = 2\pi \left[ \delta(\omega - \omega_{FQ1}(q)) \right] + |u|^2 \delta(\omega - \omega_{FQ1}(q)),
\] (E5)

AFQ1 phase The spin operators for sublattice 1 are the same as Eq. (E3) but with additional sublattice subindex and the spin operators for sublattice 2 read
\[
S_1^x = (i r_0 - r_3) a_z + (-i r_0 - r_3) a_z^\dagger,
\]
\[
S_1^y = -(r_1 - i r_2) a_z + (-r_1 + i r_2) a_z^\dagger,
\]
\[
S_1^z = -u a_y - u^* a_y^\dagger.
\] (E6)

And spin spectral function is
\[
S_{AFQ1}(q, \omega) = 2\pi \delta(\omega - \omega_{AFQ1}(q))
\]
\[
+ 2\pi \left| \frac{1 - \gamma(q)}{1 + \gamma(q)} \right| |u|^2 \delta(\omega - \omega_{AFQ1}(q)),
\] (E7)
And spin spectral function reads

\[ S_{\text{AFQ3}}(q, \omega) = 2\pi F_1(\theta, \tilde{r}) \delta(\omega - \omega_{1}^{\text{AFQ3}}(q)) + 2\pi F_2(\theta, \tilde{r}) \sqrt{\frac{1 + B_K \gamma(q)}{1 - \gamma(q)}} \delta(\omega - \omega_{2}^{\text{AFQ3}}(q)), \tag{E8} \]

where \( u \) is defined in Eq. (E4) and

\[
F_1(\theta, \tilde{r}) = 1 + (|u|^2 - 1) \cos^2 \theta, \quad F_2(\theta, \tilde{r}) = 1 + \frac{4u^2 + 4r_1^2 - 3 - |u|^2}{4} \sin^2 2\theta. \tag{E10} \]

**AFQ3 phase**  The forms of spin operators for sublattice 1 are the same as Eq. (E8). And for sublattice 2 we cannot obtain spin operators by taking \( \theta \rightarrow -\theta \). So here we do not list them explicitly. The dipolar spin spectral function reads

\[
S_{\text{AFQ3}}(q, \omega) = 2\pi \sin^2 \theta \delta(\omega - \omega_{1}^{\text{AFQ3}}(q)) + 2\pi |u|^2 \cos^2 \theta \delta(\omega - \omega_{2}^{\text{AFQ3}}(q)) + 2\pi (r_0^2 + r_2^2) \sqrt{\frac{1 - B_K \gamma(q)}{1 + \gamma(q)}} \delta(\omega - \omega_{3}^{\text{AFQ3}}(q)) \]

\[ + 2\pi (r_1^2 + r_3^2) \cos^2 \theta \sqrt{\frac{1 + \gamma(q)}{1 - B_K \gamma(q)}} \delta(\omega - \omega_{3}^{\text{AFQ3}}(q)), \]

\[ + \pi \left( \frac{1 - |u|^2}{2} \right) \sin^2 2\theta \sqrt{\frac{1 - \gamma(q)}{1 + B_K \gamma(q)}} \delta(\omega - \omega_{4}^{\text{AFQ3}}(q)), \tag{E11} \]

where \( u \) is defined in Eq. (E4).

**FQ2 phase**  The spin operators are

\[ S^x = (ir_2 + r_3)a_z - \sin \theta(r_1 + ir_0)a_y + h.c., \]
\[ S^y = (-ir_0 - r_3 \cos 2\theta)a_z - \sin \theta(r_1 - ir_2)a_y + h.c., \]
\[ S^z = u \cos \theta a_y + (r_0 r_1 + r_2 r_3) \sin 2\theta a_z + h.c.. \tag{E12} \]

Note that \( S_{\text{FQ2}}(q, \omega) \) does not depend on the \( d \) vector.

**AFQ3 phase**  The forms of spin operators for sublattice 1(2) are the same as Eq. (E12)(Eq. (E13)) with additional sublattice index. Thus the dipolar spin spectral function for an AFQ3 state does not depend on the \( d \) vector as well and reads

\[ S_{\text{AFQ3}}(q, \omega) = \pi \delta(\omega - \omega_{1}^{\text{AFQ3}}(q)) + \pi \sqrt{\frac{C_K - D_K \gamma(q)}{C_K + D_K \gamma(q)}} \delta(\omega - \omega_{3}^{\text{AFQ3}}(q)) \]

\[ + \pi \sqrt{\frac{C_K - D_K \gamma(q)}{C_K + D_K \gamma(q)}} \delta(\omega - \omega_{4}^{\text{AFQ3}}(q)). \tag{E14} \]

The \( S(q, \omega) \) for all FQ and AFQ states are plotted in FIG 5.

**Appendix F: Quadrupole spectral functions**

Resonant inelastic X-ray scattering (RIXS) measures two-magnon processes, which is described by spin quadrupole spectral functions. In this appendix, we provide details for quadrupolar spectral functions, \( Q(q, \omega) = \sum_q Q^{\mu\nu}(q, \omega) \), where \( Q^{\mu\nu}(q, \omega) = \text{Im} \{ \int_0^\infty \text{d}t \ e^{i\omega t} \langle |Q^\mu(q, t) Q^{\nu\nu}(-q, 0)| \rangle \} \), and \( \mu \) and \( \nu \) denote \( xy, yz, zx, xz \) and \( x^2 - y^2 \) and \( 3z^2 - r^2 \).

**FQ1 phase**  The \( Q \) operators read

\[ Q^{xy} = v a_z + h.c., \quad Q^{yx} = (r_2 + ir_1)a_z + h.c., \]
\[ Q^{xz} = (ir_3 - r_0)a_z + h.c., \quad Q^{zx} = 2(i r_3 - r_0)(r_2 + ir_1)a_y + h.c., \tag{F1} \]

where

\[ v = (ir_0 + r_2)^2 - (r_3 - r_0)^2. \tag{F2} \]

Notice the \( r_0, r_1, r_3, r_2 \) are defined in Eq. (E1) and again the quadratic boson and constant terms are omitted. Then the quadrupolar spin spectral function reads

\[ Q_{\text{FQ1}}(q, \omega) = 2\pi \delta(\omega - \omega_{1}^{\text{FQ1}}(q)) + 2\pi \delta(\omega - \omega_{2}^{\text{FQ1}}(q)), \tag{F3} \]

where \( u \) is defined in Eq. (E4).

**AFQ1 phase**  The \( Q \) operators for sublattice 1 are the same as Eq. (F1) but with additional sublattice subindex and the \( Q \) operators for sublattice 2 read

\[ Q_2^{xy} = v a_z + h.c., \quad Q_2^{yx} = -(r_0 + ir_3)a_z + h.c., \]
\[ Q_2^{xz} = (ir_1 - r_2)a_z + h.c., \quad Q_2^{zx} = 2(ir_3 + r_0)(r_2 - ir_1)a_z + h.c., \tag{F4} \]

where \( v \) is defined in Eq. (F2). The quadrupolar spin spectral function is

\[ Q_{\text{AFQ1}}(q, \omega) = 2\pi \delta(\omega - \omega_{3}^{\text{AFQ1}}(q)) + 2\pi \delta(\omega - \omega_{4}^{\text{AFQ1}}(q)) \]

\[ + 2\pi \delta(\omega - \omega_{5}^{\text{AFQ1}}(q)) \delta(\omega - \omega_{6}^{\text{AFQ1}}(q)). \tag{F5} \]
where \( u \) is defined in Eq. \((E4)\).

**FQ2 phase** The \( Q \) operators read

\[
Q_{xy} = v \cos \vartheta a_y - (r_0 r_2 - r_1 r_3) \sin 2\vartheta a_z + h.c.,
Q_{xz} = (r_3 - r_0 \cos 2\vartheta) a_z - \sin \vartheta (ir_1 + r_2) a_y + h.c.,
Q_{yz} = (ir_1 + r_2 \cos 2\vartheta) a_z + \sin \vartheta (ir_3 - r_0) a_y + h.c.,
Q_{x^2-y^2} = (1/2 - r_1^2 - r_2^2) \sin 2\vartheta a_z
+ 2 \cos \vartheta (ir_1 + r_2)(ir_3 - r_0) a_y + h.c.,
Q_{z^2-r^2} = \sqrt{3} \sin 2\vartheta a_z/2 + h.c.,
\]

\((F6)\)

where \( v \) is defined in Eq. \((F2)\). And quadrupolar spin spectral function reads

\[
Q_{FQ2}(q, \omega) = 2\pi F_3(\vartheta, \hat{r}) \delta(\omega - \omega^{FQ2}_1(q))
+ 2\pi F_4(\vartheta, \hat{r}) \sqrt{1 + B_K \gamma(q) \over 1 - \gamma(q)} \delta(\omega - \omega^{FQ2}_2(q)),
\]

\((F7)\)

where \( u \) is defined in Eq. \((E4)\) and

\[
F_3(\vartheta, \hat{r}) = 1 + (1 - |u|^2) \cos^2 \vartheta,
F_4(\vartheta, \hat{r}) = 1 + 4r_1^2 + 4r_3^2 - 1 + |u|^2 \sin^2 2\vartheta.
\]

**AFQ2 phase** The forms of \( Q \) operators for sublattice 1 are the same as Eq. \((F6)\). And for sublattice 2 we can obtain the \( Q \) operators by taking \( \vartheta \to - \vartheta \). The quadrupolar spin spectral function reads

\[
Q_{AFQ2}(q, \omega) = 2\pi \sin^2 \vartheta \delta(\omega - \omega^{AFQ2}_1(q))
+ 2\pi (2 - |u|^2) \cos^2 \vartheta \delta(\omega - \omega^{AFQ2}_2(q))
+ 2\pi (1 + r_1^2) \sqrt{1 + B_K \gamma(q) \over 1 + \gamma(q)} \delta(\omega - \omega^{AFQ2}_3(q))
+ 2\pi (1 + r_2^2) \cos^2 2\vartheta \sqrt{1 + \gamma(q) \over 1 - B_K \gamma(q)} \delta(\omega - \omega^{AFQ2}_4(q))
+ \pi (3 + |u|^2) \sin^2 2\vartheta \sqrt{1 + B_K \gamma(q) \over 1 + B_K \gamma(q)} \delta(\omega - \omega^{AFQ2}_5(q)),
\]

\((F9)\)

where \( u \) is defined in Eq. \((E4)\).

**FQ3 phase** The \( Q \) operators are

\[
Q_{xy} = Q_{z^2-r^2} = Q_{x^2-y^2} = 0,
Q_{xz} = (ir_3 - r_0) a_z + (ir_1 + r_2) a_y + h.c.,
Q_{yz} = (ir_1 + r_2) a_z - (ir_3 - r_0) a_y + h.c.,
\]

\((F10)\)

And the quadrupolar spin spectral function \( Q_{FQ3}(q, \omega) \) is the same as Eq. \((E13)\) and does not depend on the \( d \) vector.

**AFQ3 phase** The forms of \( Q \) operators for sublattice 1(2) are the same as Eq. \((F10)\) (Eq. \((F1)\)) with additional sublattice index. Thus the quadrupolar spin spectral function \( Q_{AFQ3}(q, \omega) \) is the same as Eq. \((E14)\) and does not depend on the \( d \) vector.

The quadrupolar spectral function \( Q(q, \omega) \) for all FQ and AFQ states are plotted in Fig. \( \text{[Figure]} \). Note that \( Q(q, \omega) \) are of the same form as \( S(q, \omega) \), and difference between them is in the spectral weight.

**Appendix G: Effective Hamiltonian in the AFQ3 phase**

In the mean-field solution, the AFQ3 ground states are locally degenerate inside a bulk energy gap. This huge degeneracy arises from the unperturbative Hamiltonian \( h_1 \) defined in Eq. \((3)\), and consider their matrix elements. Firstly, \( h_1 = Q_{1z}^{\text{z-r}} Q_{2z}^{\text{z-r}} \), which is diagonal in the basis \( \{|\sigma_1 \sigma_2\}\} \), where \( \sigma_{1,2} = x, y, z \). Secondly, \( h_1 = S_1^z S_2^z + Q_1^{xy} Q_2^{xy} + Q_1^{z^2-y^2} Q_2^{z^2-y^2} \), and we have

\[
h_1|\sigma_1 \sigma_2\rangle = \begin{cases} |\sigma_1 \sigma_2\rangle, & \sigma_1 = \sigma_2 \neq z, \\ 2|\sigma_2 \sigma_1\rangle, & \sigma_1 = x(y), \sigma_2 = y(x), \\ 0, & \text{otherwise}. \end{cases}
\]

\((G1)\)

Thirdly, \( h_2 = S_1^z S_2^z + S_1^y S_2^y + Q_1^{xy} Q_2^{xy} + Q_1^{z^2-y^2} Q_2^{z^2-y^2} \), we have

\[
h_2|\sigma_1 \sigma_2\rangle = \begin{cases} 2|\sigma_2 \sigma_1\rangle, & \sigma_1 = x, y(z) \text{ and } \sigma_2 = z(x, y), \\ 0, & \text{otherwise}. \end{cases}
\]

\((G2)\)

Taking into account all the nonzero matrix elements, the leading perturbation is of the third order and the effective Hamiltonian reads

\[
H_{\text{eff}} = K_1 K_2^2 \over K_3^2 \mathcal{P} H_2 H_1 H_2 \mathcal{P}.
\]

\((G3)\)

Regarding the projector \( \mathcal{P} \), \( H_{\text{eff}} \) can be written as a \( H_1 \) type Hamiltonian defined on one of the sublattices. For instance, the effective Hamiltonian on a square lattice can be written as

\[
H_{\text{eff}} = J_1 \sum_{\langle ij \rangle} \mathcal{P} \left( S_i^z S_j^z + Q_{i}^{xy} Q_{j}^{xy} + Q_{i}^{z^2-y^2} Q_{j}^{z^2-y^2} \right) \mathcal{P}
+ J_2 \sum_{\langle ij \rangle} \mathcal{P} \left( S_i^z S_j^z + Q_{i}^{xy} Q_{j}^{xy} + Q_{i}^{z^2-y^2} Q_{j}^{z^2-y^2} \right) \mathcal{P},
\]

where \( \langle ij \rangle \) denotes a pair of nearest neighboring (next nearest neighboring) sites \( i \) and \( j \) on one of the two sublattices, \( J_1 = \frac{2K_1 K_2^2}{K_3^2} \) and \( J_2 = \frac{K_1 K_2^2}{K_3^2} \). Note that we
FIG. 5. Spin spectral function $S(q, \omega)$ for FQ and AFQ phases. Here we set the $K_2 = K_3 = K_1/10$ for FQ1 and AFQ1, $K_1 = K_3 = K_2/10$ for FQ2 and AFQ2 and $K_1 = K_2 = K_3/10$ for FQ3 and AFQ3. And we choose the ground states with $\hat{r} = (0.3, 0.4, 0.3, 0.4)/\sqrt{2}$ for FQ2 and AFQ2, and with $\hat{r} = (1, 0, 0, 0)$ for the other phases.

FIG. 6. Quadrupole spectral function $Q(q, \omega)$ for FQ and AFQ phases. Here we set the $K_2 = K_3 = K_1/10$ for FQ1 and AFQ1, $K_1 = K_3 = K_2/10$ for FQ2 and AFQ2 and $K_1 = K_2 = K_3/10$ for FQ3 and AFQ3. And we choose the ground states with $\hat{r} = (0.3, 0.4, 0.3, 0.4)/\sqrt{2}$ for FQ2 and AFQ2, and with $\hat{r} = (1, 0, 0, 0)$ for the other phases.
always have $J_2/J_1 = 1/2$. Since there are two paths contribute to $J_1$, while there is only one path contributes to $J_2$, as illustrated in FIG. 7.

When $K_1 < 0$, we have $J_1 < 0$ and $J_2 < 0$ and the effective model is a ferromagnetic spin-$1/2$ Heisenberg model on a square lattice, which gives rise to a ferromagnetic ordering state on one of the sublattices. When $K_1 > 0$, the effective model is an antiferromagnetic spin-$1/2$ Heisenberg model on a square lattice. And $J_2/J_1 = 1/2$ gives rise to a gapless quantum spin liquid ground state.

Appendix H: Local wavefunctions for ordered states

In this appendix we use local spin density to illustrate the wavefunctions for various FQ and AFQ states. If $d$ vector is real, the state is time reversal invariant, and the local spin density $|\langle \hat{S}(\hat{n})|d \rangle|^2$ is invariant under an $SO(2)$ rotation along the axis of $(|d_x|, |d_y|, |d_z|)$, which is so-called $SO(2)$ symmetric pattern. Otherwise, $|d|$ breaks the time-reversal symmetry, $|\langle \hat{S}(\hat{n})|d \rangle|^2$ will be distorted from an $SO(2)$ symmetric pattern to an $SO(2)$ non-symmetric pattern. Two examples for time reversal symmetric state and time reversal asymmetric state are given in FIG. 8.

All the six quadrupolar ordered states are illustrated on square lattice in FIG. 9, where we choose time-reversal symmetric ground states to eliminate spin dipolar orders and manifest quadrupolar orders.
Appendix I: Connection with $t$-$J$-$V$ model.

The local spin state $|z\rangle$ is $SU(2)_\gamma$ invariant and belongs to $SU(2)_\gamma$ irreducible representation (IR) $J = 0$, while $|x\rangle$ and $|y\rangle$ belong to $SU(2)_\gamma$ IR $J = \frac{1}{2}$. Therefore, $|z\rangle$ can be treated as a “hole” state, and the $SU(2) \times U(1)$ symmetric model defined in Eq. (4) can be mapped to the $t$-$J$-$V$ model, which reads

$$H = -t \sum_{j,a} P \psi_{j,a}^\dagger \psi_{j+1,a} P + h.c$$

$$+ \sum_j P (J \vec{s}_j \cdot \vec{s}_{j+1} + V n_j n_{j+1}) P,$$

where $P$ projects out states with double occupancy and electron spin $\vec{s}_j$ and $n_j$ on site $j$ are defined as

$$\vec{s}_j = \sum_{a,b} \psi_{j,a}^\dagger \vec{\sigma}_{ab} \psi_{j,b}, \quad n_j = \sum_a \psi_{j,a}^\dagger \psi_{j,a}. \quad (I2)$$

The spin-1/2 indices, $a, b = \uparrow, \downarrow$. Let $|\uparrow\rangle$ and $|\downarrow\rangle$ correspond to $|x\rangle$ and $|y\rangle$, and the “hole” state correspond to $|z\rangle$ in spin-1 system (see the definition of $|x\rangle$, $|y\rangle$, $|z\rangle$ states in main text), we can establish a mapping from the $t$-$J$-$V$ model to the $SU(2) \times U(1)$ model defined in Eq. (4) through

$$K_1 = J/4, \quad K_2 = t/2, \quad K_3 = V/3 \quad (I3)$$

Hamiltonian given in Eq. (I1) is equivalent to the $SU(3)$ symmetric model when $J = 2t$ and $V = 3t/2$. And the supersymmetric $t$-$J$-$V$ model can be realized when $K_1 = K_2 = -K_3/3$. 