Spin-orbital liquids in non-Kramers magnet on Kagome lattice

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Localised magnetic moments with crystal-field doublet or pseudo-spin 1/2 may arise in correlated insulators with even number of electrons and strong spin-orbit coupling. Such a non-Kramers pseudo-spin 1/2 is the consequence of crystalline symmetries as opposed to the Kramers doublet arising from time-reversal invariance, and is necessarily a composite of spin and orbital degrees of freedom. We investigate possible spin-orbital liquids with fermionic spinons for such non-Kramers pseudo-spin 1/2 systems on the Kagome lattice. Using the projective symmetry group analysis, we find ten new phases that are not allowed in the corresponding Kramers systems. These new phases are allowed due to unusual action of the time reversal operation on non-Kramers pseudo-spins. We compute the spin-spin dynamic structure factor that shows characteristic features of these non-Kramers spin-orbital liquids arising from their unusual coupling to neutrons, which is therefore relevant for neutron scattering experiments. We also point out possible anomalous broadening of Raman scattering intensity that may serve as a signature experimental feature for gapless non-Kramers spin-orbital liquids.

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I. INTRODUCTION

The low energy magnetic degrees of freedom of a Mott insulator, in the presence of strong spin-orbit coupling, are described by states with entangled spin and orbital wave functions. In certain crystalline materials, for ions with even numbers of electrons, a low energy spin-orbit entangled "pseudo-spin"-1/2 may emerge, which is not protected by time-reversal symmetry (Kramers degeneracy) but rather by the crystal symmetries. Various phases of such non-Kramers pseudo-spin systems on geometrically frustrated lattices, particularly various quantum paramagnetic phases, are of much recent theoretical and experimental interest in the context of a number of rare earth materials including frustrated pyrochlores and heavy fermion systems.

In this paper, we explore novel spin-orbital liquids that may emerge in these systems due to the unusual transformation of the non-Kramers pseudo-spins under the time reversal transformation. Contrary to Kramers spin-1/2, where the spins transform as \( S \rightarrow -S \) under time reversal, here only one component of the pseudo-spin operators changes sign under time reversal: \( \{ \sigma^1, \sigma^2, \sigma^3 \} \rightarrow \{ \sigma^1, \sigma^2, -\sigma^3 \} \). Because, due to the nature of the wave-function content, the \( \sigma^3 \) component of the pseudo-spin carries a dipolar magnetic moment while the other two components carry quadrupolar moments of the underlying electrons. Hence the time reversal operator for the non-Kramers pseudo-spins is given by \( T = \sigma^1 K \) (where \( K \) is the complex conjugation operator), which allows for new spin-orbital liquid phases. Since the magnetic degrees of freedom are composed out of wave functions with entangled spin and orbital components, we refer to the above quantum paramagnetic states as spin-orbital liquids, rather than spin liquids.

Since the degeneracy of the non-Kramers doublet is protected by crystal symmetries, the transformation properties of the pseudo-spin under various lattice symmetries intimately depend on the content of the wave-functions that make up the doublet. To this end, we focus our attention on the example of Praseodymium ions (Pr³⁺) in a local \( D_{3d} \) environment, which is a well known non-Kramers ion that occurs in a number of materials with interesting properties. Such an environment typically occurs in Praseodymium pyrochlores given by the generic formulae \( \text{Pr}_2\text{TM}_2\text{O}_7 \), where \( \text{TM}(=\text{Zr}, \text{Sn}, \text{Hf}, \text{or} \text{Ir}) \) is a transition metal. In these compounds, the Pr³⁺ ions host a pair of 4f electrons which form a \( J = 4 \) ground state manifold with \( S = 1 \) and \( L = 5 \), as expected due to Hund’s rules. In terms of this local environment we have a nine fold degeneracy of the electronic states. This degeneracy is broken by the crystalline electric field. The oxygen and TM ions form a \( D_{3d} \) local symmetry environment around the Pr³⁺ ions, splitting the nine fold degeneracy. A standard analysis of the symmetries of this system (see Appendix A) shows that the \( J = 4 \) manifold splits into three doublets and three singlets \( (\Gamma_1 = 3E_g + 2A_{1g} + A_{2g}) \) out of which one of the doublets is found to have the lowest energy, usually well separated from the other crystal field states. This doublet (details in Appendix A), formed out of a linear combination of the \( J_z = \pm 4 \) with \( J_z = \pm 1 \) and \( J_z = \pm 2 \) states, is given by

\[
|\pm\rangle = \alpha|m = \pm 4\rangle \pm \beta|m = \pm 1\rangle - \gamma|m = \mp 2\rangle.
\]

The non-Kramers nature of this doublet is evident from the nature of the “spin” raising and lowering operators within the doublet manifold; the projection of the angular momentum raising and lowering operators to the space of doublets is zero \((\mathcal{P}J^\pm|\sigma\rangle = 0 \) where \( \mathcal{P} \) projects into the doublet manifold\( )\). However, the projection of the \( J^z \) operator to this manifold is non-zero, and describes the \( z \) component of the pseudo-spin \( (\sigma^3) \). In addition, there is a non-trivial projection of the quadrupole operators \( \{ J^\pm, J^z \} \) in this manifold. These have off-diagonal matrix elements, and are identified with the pseudo-spin raising and lowering operators \( (\sigma^\pm = \sigma^1 \pm i\sigma^2) \).

In a pyrochlore lattice the local \( D_{3d} \) axes point to the centre of the tetrahedra. On looking at the pyrochlore lattice along...
the [111] direction, it is found to be made out of alternate layers of Kagome and triangular lattices. For each Kagome layer (shown in Fig. 1) the local $D_{3d}$ axes make an angle of $\cos^{-1}(\sqrt{2}/3)$ with the plane of the Kagome layer. We imagine replacing the Pr$^{3+}$ ions from the triangular lattice layer with non-magnetic ions so as to obtain decoupled Kagome layers with Pr$^{3+}$ ions on the sites. The resulting structure is obtained in the same spirit as the now well-known Kagome compound Herbertsmithite was envisioned. As long as the local crystal field has $D_{3d}$ symmetry, the doublet remains well defined. A suitable candidate non-magnetic ion may be iso-

valent but non-magnetic La$^{3+}$. Notice that the most extended orbitals in both cases are the fifth shell orbitals and the crystal field at each Pr$^{3+}$ site is mainly determined by the surrounding oxygens and the transition metal element. Hence, we expect that the splitting of the non-Kramers doublet due to the above substitution would be very small and the doublet will remain well defined. In this work we shall consider such a Kagome lattice layer and analyze possible Z$_2$ spin-orbital liquids, with gapped or gapless fermionic spinons.

The rest of the paper is organized as follows. In Sec. II, we begin with a discussion of the symmetries of the non-Kramers system on a Kagome lattice and write down the most general pseudo-spin model with pseudo-spin exchange interactions up to second nearest neighbours. In Sec. III formulate the projective symmetry group (PSG) analysis for singlet and triplet decouplings. Using this we demonstrate that the non-Kramers transformation of our pseudo-spin degrees of freedom under time reversal leads to a set of ten spin-orbital liquids which cannot be realized in the Kramers case. In Sec. IV we derive the dynamic spin-spin structure factor for a representa-

tive spin liquid for the case of both Kramers and non-Kramers doublets, demonstrating that experimentally measurable properties of these two types of spin-orbital liquids differ qualita-

tively. Finally, in Sec. V, we discuss our results, and propose an experimental test which can detect a non-Kramers spin liquid for the case of both Kramers and non-Kramers doublets, demonstrating that experimentally measurable properties of these two types of spin-orbital liquids differ qualita-

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freedom, and the remaining operators act as

\[ T = \sigma_1 K, \quad \sigma = \sigma_1, \quad S_0 = \begin{bmatrix} \bar{\omega} & 0 \\ 0 & \omega \end{bmatrix}, \tag{3} \]

where \( K \) refers to complex conjugation. The above expressions can be derived by examining the effect of these operators on the wave-function describing the doublet (Eq. 1).

We can now write down the most generic pseudo-spin Hamiltonian allowed by the above lattice symmetries that is bilinear in pseudo-spin operators. The form of the time-reversal symmetry restricts our attention to those products which are formed by a pair of \( \sigma^3 \) operators or those which mix the pseudo-spin raising and lowering operators. Any term which mixes \( \sigma^3 \) and \( \sigma^\pm \) changes sign under the symmetry, and can thus be excluded. Under the \( C_3 \) transformation about a site, the terms \( C_3 : \sigma^3_+ \sigma^3_+ \rightarrow \sigma^3_{C_3(r)} \sigma^3_{C_3(r')} \) and \( C_3 : \sigma^+_+ \sigma^-_+ \rightarrow \sigma^+_{C_3(r)} \sigma^-_{C_3(r')} \). However, the term \( \sigma^+_+ \sigma^-_+ \) (and its Hermitian conjugate) gain additional phase factors when transformed; under the \( C_3 \) symmetry transformation, this term becomes \( C_3 : \sigma^+_+ \sigma^-_+ \rightarrow \omega^3 \sigma^+_+ \sigma^-_+ \) and \( C_3 : \sigma^+_+ \sigma^-_+ \rightarrow \omega \sigma^+_+ \sigma^-_+ \). In addition, under the \( \sigma \) symmetry, this term transforms as \( \sigma^+_+ \sigma^-_+ \rightarrow \sigma^+_+ \sigma^-_+ \). Thus the Hamiltonian with spin-spin exchange interactions up to next-nearest neighbour is given by

\[
H_{c,f} = J_{nn} \sum_{(r,r')} \left[ \sigma^+_r \sigma^+_\tau \sigma^-_\tau \sigma^-_r + 2(\delta \sigma^+_r \sigma^-_r + h.c.) \right] \\
+ 2q(\epsilon \sigma^+_r \sigma^-_\tau \sigma^+_\tau \sigma^-_r + h.c.) \\
+ J_{nnn} \sum_{\langle (r,r') \rangle} \left[ \sigma^3_\tau \sigma^3_\tau \sigma^3_r \sigma^3_r + 2(\delta \sigma^+_r \sigma^-_r + h.c.) \right] \\
+ 2q' \left( \epsilon \sigma^+_r \sigma^-_\tau \sigma^+_\tau \sigma^-_r + h.c. \right), \tag{4} \]

where \( \phi \) and \( \phi' \) take values 0, 1 and 2 depending on the bonds on which they are defined (Fig. 2(b)).

### III. SPINON REPRESENTATION OF THE PSEUDO-SPINS AND PSG ANALYSIS

Having written down the pseudo-spin Hamiltonian, we now discuss the possible spin-orbital liquid phases. We do this in two stages in the following sub-sections.

#### A. Slave fermion representation and spinon decoupling

In order to understand these phases, we will use the fermionic slave-particle decomposition of the pseudo-spin operators. At this point, we note that the pseudo-spins satisfy \( S = 1/2 \) representations of a “SU(2)” algebra among their generators (not to be confused with the regular spin rotation symmetry). We represent the pseudo-spin degrees of freedom in terms of a fermion bilinear. This is very similar to usual slave fermion construction for spin liquids.\(^{13,14}\) We take

\[
\sigma^\alpha_j = \frac{1}{2} f_j^\dagger \rho^\alpha \xi_{\alpha j} f^\beta, \tag{5} \]

where \( \alpha, \beta = \uparrow, \downarrow \) is defined along the local z axis and \( f_j^\dagger \) (\( f^\dagger \)) is an \( S = 1/2 \) fermionic creation (annihilation) operator. Following standard nomenclature, we refer to the \( f(f^\dagger) \) as the spinon annihilation (creation) operator, and note that these satisfy standard fermionic anti-commutation relations. The above spinon representation, along with the single occupancy constraint

\[
f_{i\uparrow} f^\dagger_{i\uparrow} + f_{i\downarrow} f^\dagger_{i\downarrow} = 1, \tag{6} \]

form a faithful representation of the pseudo-spin-1/2 Hilbert space. The above representation of the pseudo-spins, when used in Eq. 4, leads to a quartic spinon Hamiltonian. Following standard procedure,\(^{13,14}\) this is then decomposed using auxiliary fields into a quadratic spinon Hamiltonian (after writing down the corresponding Euclidean action). The mean field description of the phases is then characterized by the possible saddle point values of the auxiliary fields. There are eight such auxiliary fields per bond, corresponding to

\[
\chi_{ij} = \langle f_{i\alpha} f_{j\alpha} \rangle; \quad \eta_{ij} = \langle f_{i\alpha} [i\tau^2]_{\alpha\beta} f_{j\beta} \rangle; \tag{7a} \]

\[
E_{ij}^\alpha = \langle f_{i\alpha} [\tau^a]_{\alpha\beta} f_{j\beta} \rangle; \quad D_{ij}^\alpha = \langle f_{i\alpha} [i\tau^2 \tau^a]_{\alpha\beta} f_{j\beta} \rangle; \tag{7b} \]

where \( \tau^a \) (\( a = 1, 2, 3 \)) are the Pauli matrices. While Eq. 7a represents the usual singlet spinon hopping (particle-hole) and pairing (particle-particle) channels, Eq. 7b represents the corresponding triplet decoupling channels. Since the Hamiltonian (Eq. 4) does not have pseudo-spin rotation symmetry, both the singlet and the triplet decouplings are necessary.\(^{16,17}\)

From this decoupling, we obtain a mean-field Hamiltonian which is quadratic in the spinon operators. We write this compactly in the following form\(^{17}\) (subject to the constraint Eq.6)

\[
H_0 = \sum_{ij} J_{ij} f_{i\uparrow}^\dagger U_{ij} f_{j\uparrow}, \tag{8} \]

\[
f_{i\uparrow}^\dagger = \left[ f_{i\uparrow}^\dagger f_{i\downarrow} f_{i\downarrow}^\dagger - f_{i\uparrow} \right], \tag{9} \]

\[
U_{ij} = \xi_{ij} \sigma_{ij} \Gamma_{ij}, \tag{10} \]

\[
\Sigma_{ij}^\alpha = \rho^\alpha \otimes I; \quad \Gamma_{ij} = I \otimes \tau^a, \tag{11} \]

where \( \rho^\alpha \) are the Identity (for \( \alpha = 0 \)) and Pauli matrices (\( \alpha = 1, 2, 3 \)) acting on pseudo-spin degrees of freedom, and \( \tau^a \) represents the same in the gauge space. We immediately note that

\[
[\Sigma_{ij}^\alpha, \Gamma_{ij}^\beta] = 0 \quad \forall \alpha, \beta. \tag{12} \]

The requirement that our \( H_0 \) be Hermitian restricts the coefficients \( \xi_{ij} \) to satisfy

\[
\xi_{ij}^{00}, \xi_{ij}^{ab} \in \mathbb{C}; \quad \xi_{ij}^{0a}, \xi_{ij}^{ab} \in \mathbb{R}. \tag{13} \]

for \( a, b \in \{1, 2, 3\} \). The relations between \( \xi_{ij}s \) and \( \{\chi_{ij}, \eta_{ij}, E_{ij}, D_{ij}\} \) are given in Appendix C.\(^{17}\) As a straightforward extension of the SU(2) gauge theory formulation for
spin liquids,\textsuperscript{13,18} we find that $H_0$ is invariant under the gauge transformation

\[ \tilde{f}_j \rightarrow W_j \tilde{f}_j, \]

\[ U_{ij} \rightarrow W_i U_{ij} W_j^\dagger, \]

where the $W_i$ matrices are SU(2) matrices of the form $W_i = e^{i\vec{a}_i \cdot \vec{f}_i} (\vec{G} \equiv (\Gamma^1, \Gamma^2, \Gamma^3))$. Noting that the physical pseudo-spin operators are given by

\[ \tilde{\sigma}_i = \frac{1}{4} \tilde{f}_i \tilde{\Sigma}_i \tilde{f}_i^\dagger, \]

Eq. 12 shows that the spin operators, as expected, are gauge invariant. It is useful to define the "$\Sigma$-components" of the $U_{ij}$ matrices as follows:

\[ U_{ij} = V_{ij}^\alpha \Sigma^\alpha, \]

where

\[ V_{ij}^\alpha = \xi_{ij}^{\alpha \beta} \Gamma^{\beta} = \begin{bmatrix} \mathcal{J}_{ij}^\alpha & 0 \\ 0 & \mathcal{J}_{ij}^\alpha \end{bmatrix}, \]

and

\[ \mathcal{J}_{ij}^\alpha = \begin{bmatrix} \xi_{ij}^{\alpha 0} + \xi_{ij}^{\alpha 1} & \xi_{ij}^{\alpha 1} - i \xi_{ij}^{\alpha 2} \\ \xi_{ij}^{\alpha 1} + i \xi_{ij}^{\alpha 2} & \xi_{ij}^{\alpha 0} - i \xi_{ij}^{\alpha 1} \end{bmatrix}. \]

Under global spin rotations the fermions transform as

\[ \tilde{f}_j \rightarrow V \tilde{f}_j, \]

where $V$ is an SU(2) matrix of the form $V = e^{i\vec{S} \cdot \vec{b}} (\vec{S} \equiv \{\Sigma^1, \Sigma^2, \Sigma^3\})$. So while $V_{ij}^{\alpha}$ (the singlet hopping and pairing) is invariant under spin rotation, $\{V_{ij}^1, V_{ij}^2, V_{ij}^3\}$ transforms as a vector as expected since they represent triplet hopping and pairing amplitudes.

\section*{B. PSG Classification}

We now classify the non-Kramers spin-orbital liquids based on projective representation similar to that of the conventional quantum spin liquids.\textsuperscript{13} Each spin-orbital liquid ground state of the quadratic Hamiltonian (Eq 11) is characterized by the mean field parameters (eight on each bond, $\chi, \eta, E^1, E^2, E^3, D^1, D^2, D^3$, or equivalently $U_{ij}$). However, due to the gauge redundancy of the spinon parametrization (as shown in Eq. 15), a general mean-field ansatz need not be invariant under the symmetry transformations on their own but may be transformed to a gauge equivalent form without breaking the symmetry. Therefore, we must consider its transformation properties under a projective representation of the symmetry group.\textsuperscript{13} For this, we need to know the various projective representations of the lattice symmetries of the Hamiltonian (Eq. 4) in order to classify different spin-orbital liquid states.

Operationally, we need to find different possible sets of gauge transformations \{GG\} which act in combination with the symmetry transformations \{SG\} such that the mean-field ansatz $U_{ij}$ is invariant under such a combined transformation. In the case of spin rotation invariant spin-liquids (where only the singlet channels $\chi$ and $\eta$ are present), the above statement is equivalent to demanding the following invariance:

\[ U_{ij} = [G_S S] U_{ij} [G_S S]^\dagger = G_S(i) U_{ij} S(i, j) G_S^\dagger(j), \]

where $S \in SG$ is a symmetry transformation and $G_S \in GG$ is the corresponding gauge transformation. The different possible $\{G_S[S \in SG]\}$ give the possible algebraic PSGs that can characterize the different spin-orbital liquid phases. To obtain the different PSGs, we start with various lattice symmetries of the Hamiltonian. The action of various lattice transformations\textsuperscript{15} is given by

\[ T_1 : (x, y, s) \rightarrow (x + 1, y, s); \]
\[ T_2 : (x, y, s) \rightarrow (x, y + 1, s); \]
\[ \sigma : (x, y, u) \rightarrow (y, x, u); \]
\[ (x, y, v) \rightarrow (y, x, v); \]
\[ (x, y, w) \rightarrow (y, x, v); \]
\[ S_6 : (x, y, u) \rightarrow (-y - 1, x, y + 1, v); \]
\[ (x, y, v) \rightarrow (-y - 1, x, y, w); \]
\[ (x, y, w) \rightarrow (-y - 1, x, y, u); \]

where $(x, y)$ denotes the lattice coordinates and $s \in \{u, v, w\}$ denotes the sub-lattice index (see figure 2).

In terms of the symmetries of the Kagome lattice, these operators obey the following conditions

\[ T^2 = \sigma^2 = (S_6)^6 = e, \]
\[ g^{-1} T^{-1} g T = e \quad \forall g \in SG, \]
\[ T_1^{-1} T_2^{-1} T_1^{-1} T_2^{-1} T_1 = e, \]
\[ \sigma^{-1} T_1^{-1} \sigma T_2 = e, \]
\[ \sigma^{-1} T_2^{-1} \sigma T_1 = e, \]
\[ S_6^{-1} T_2^{-1} S_6 T_1 = e, \]
\[ S_6^{-1} T_2^{-1} T_1 S_6 T_2 = e, \]
\[ \sigma^{-1} S_6 \sigma S_6 = e. \]

In addition, these commutation relations are valid in terms of the operations on the pseudo-spin degrees of freedom, as can be verified from Eq. 3.

In addition to the conditions in Eq. 23, the Hamiltonian is trivially invariant under the identity transformation. The invariant gauge group (IGG) of an ansatz is defined as the set of all pure gauge transformations $G_I$ such that $G_I : U_{ij} \rightarrow U_{ij}$. The nature of such pure gauge transformations immediately dictates the nature of the low energy fluctuations about the mean field state. If these fluctuations do not destabilize the mean-field state, we get stable spin liquid phases whose low energy properties are controlled by the IGG. Accordingly, spin liquids obtained within projective classification are primarily labelled by their IGGs and we have $Z_2, U(1)$ and $SU(2)$ spin liquids corresponding to IGGs of $Z_2, U(1)$ and
$SU(2)$ respectively. In this work we concentrate on the set of $Z_2$ "spin liquids" (spin-orbital liquids with a $Z_2$ IGG).

We now focus on the PSG classification. As shown in Eq. 2, in the present case, the pseudo-spins transform non-trivially under different lattice symmetry transformations. Due to the presence of the triplet decoupling channels the non-Kramers doublet transforms non-trivially under lattice symmetries (Eq. 3). Thus, the invariance condition on the $U_{ij}$s is not given by Eq. 21, but by a more general condition

$$U_{ij} = [G_S]U_{ij} [G_S]^\dagger = G_S(i)\phi_S [U_{S(i)S(j)}] G_S^\dagger(j).$$

(24)

Here

$$\phi_S [U_{S(i)S(j)}] = D_S U_{S(i)S(j)} D_S^\dagger,$$

(25)

and $D_S$ generates the pseudo-spin rotation associated with the symmetry transformation ($S$) on the doublet. The matrices $D_S$ have the form

$$D_S = \frac{1}{2} \Sigma^0 - \frac{i \sqrt{2}}{2} \Sigma^3,$$

(26)

$$D_T = i \Sigma^1, \quad D_{T_1} = D_{T_2} = \Sigma^0.$$  

(27)

Under these constraints, we must determine the relations between the gauge transformation matrices $G_S(i)$ for our set of ansatz. The additional spin transformation (Eq. 25) does not affect the structure of the gauge transformations, as the gauge and spin portions of our ansatz are naturally separate (Eq. 12). In particular, we can choose to define our gauge transformations such that

$$G_S : U_{ij} \rightarrow G_S : \xi_{ij}^{\alpha\beta} \Sigma^\alpha \Gamma^\beta \rightarrow \xi_{ij}^{\alpha\beta} \Sigma^\alpha G_S^\dagger(i) \Gamma^\beta G_S(j),$$

(28)

$$S : U_{ij} \rightarrow S : \xi_{ij}^{\alpha\beta} \Sigma^\alpha \Gamma^\beta \rightarrow \xi_{S(i)S(j)}^{\alpha\beta} D_S \Sigma^\alpha D_S^\dagger \Gamma^\beta,$$

(29)

where we have used the notation $G_S : U_{ij} \equiv G_S^\dagger(i)U_{ij}G_S(j)$ and so forth. As a result, we can build on the general construction of Lu et al.\textsuperscript{15} to derive the form of the gauge transformation matrices. The details are given in Appendix B.

A major difference arises when examining the set of algebraic PSGs for $Z_2$ spin liquids found on the Kagome lattice due to the difference between the structure of the time reversal symmetry operation on the Kramers and non-Kramers pseudo-spin-1/2s. In the present case, we find there are 30 invariant PSGs leading to thirty possible spin-orbital liquids. This is in contrast with the Kramers case analysed by Lu et al.,\textsuperscript{15} where ten of the algebraic PSGs cannot be realized as invariant PSGs, as all bonds in these ansatz are predicted to vanish identically due to the form of the time reversal operator, and hence there are only twenty possible spin liquids. However, with the inclusion of spin triplet terms and the non-Kramers form of our time reversal operator, these ansatz are now realizable as invariant PSGs as well. The time reversal operator, as defined in Appendix B, acts as

$$T : \xi_{ij}^{\alpha\beta} \Sigma^\alpha \Gamma^\beta \rightarrow \xi_{ij}^{\alpha\beta} \Sigma^\alpha \Gamma^\beta,$$

(30)

where $\xi^{\alpha\beta} = \xi^{\alpha\beta}$ if $\alpha \in \{1, 2\}$ and $\xi^{\alpha\beta} = -\xi^{\alpha\beta}$ if $\alpha \in \{0, 3\}$. The projective implementation of the time-reversal symmetry condition (Eq. 23) takes the form (see Appendix B)

$$[G_T(i)]^2 = \eta_T I \quad \forall i,$$

(31)

where $G_T(i)$ is the gauge transformation associated with time reversal operation and $\eta_T = \pm 1$ for a $Z_2$ IGG.

Therefore, the terms allowed by the time reversal symmetry to be non-zero are, for $\eta_T = 1$,

$$\xi^{10}, \xi^{11}, \xi^{12}, \xi^{13}, \xi^{14}, \xi^{15}, \xi^{16}, \xi^{17}, \xi^{18}, \xi^{19}, \xi^{20}, \xi^{21}, \xi^{22}, \xi^{23}$$

(32)

and for $\eta_T = -1$, with the choice $G_T(i) = i \Gamma^1$ (see appendix B),

$$\xi^{02}, \xi^{03}, \xi^{10}, \xi^{11}, \xi^{12}, \xi^{13}, \xi^{14}, \xi^{15}, \xi^{16}, \xi^{17}, \xi^{18}, \xi^{19}, \xi^{20}, \xi^{21}, \xi^{22}, \xi^{23}, \xi^{32}, \xi^{33}.$$  

(33)

This contrasts with the case of Kramers doublets, in which no terms are allowed for $\eta_T = -1$, and for $\eta_T = -1$ the allowed terms are

$$\xi^{02}, \xi^{03}, \xi^{10}, \xi^{11}, \xi^{12}, \xi^{13}, \xi^{14}, \xi^{15}, \xi^{16}, \xi^{17}, \xi^{18}, \xi^{19}, \xi^{20}, \xi^{21}, \xi^{22}, \xi^{23}, \xi^{32}, \xi^{33}.$$  

(34)

Further restrictions on the allowed terms on each link arise from the form of the gauge transformations defined for the symmetry transformations. All nearest neighbour bonds can then be generated from $U_{ij}$ defined on a single bond, by performing appropriate symmetry operations.

Using the methods outlined in earlier works (Ref. 13, 15) we find the minimum set of parameters required to stabilize $Z_2$ spin-orbital liquids. We take into consideration up to second neighbour hopping and pairing amplitudes (both singlet and triplet channels). The results are listed in Table I.

The spin-orbital liquids listed from 21–30 are not allowed in the case of Kramers doublets and, as pointed out before, their existence is solely due to the unusual action of the time-reversal symmetry operator on the non-Kramers spins. Hence these ten spin-orbital liquids are qualitatively new phases that may appear in these systems. Of these ten phases, only two (labelled as 21 and 22 in Table I) require next nearest neighbour amplitudes to obtain a $Z_2$ spin-orbital liquid. For the other eight, nearest neighbour amplitudes are already sufficient to stabilize a $Z_2$ spin-orbital liquid.

It is interesting to note (see below) that bond-pseudo-spin-nematic order (Eq. 35 and Eq. 36) can signal spontaneous time-reversal symmetry breaking. Generally, since the triplet decouplings are present, the bond nematic order parameter for the pseudo-spins\textsuperscript{21,22}

$$Q_{ij}^{\alpha\beta} = \langle S_i^\alpha S_j^\beta + S_j^\beta S_i^\alpha \rangle /2 - \delta^{\alpha\beta}(S_i \cdot S_j) /3,$$

(35)

as well as vector chirality order

$$\vec{J}_{ij} = \langle \vec{S}_i \times \vec{S}_j \rangle,$$

(36)

are non zero. Since the underlying Hamiltonian Eq. 4) generally does not have pseudo-spin rotation symmetry, the above non-zero expectation values do not spontaneously break any
pseudospin rotation symmetry. However, because of the unusual transformation property of the non-Kramers pseudospins under time reversal, the operators corresponding to $Q_{ij}^{13}$, $Q_{ij}^{23}$, $\mathcal{J}_{ij}^{1}$, $\mathcal{J}_{ij}^{2}$ are odd under time reversal, a symmetry of the pseudospin Hamiltonian. Hence if any of the above operators gain a non-zero expectation value in the ground state, then the corresponding spin-orbital liquid breaks time reversal symmetry. While this can occur in principle, we check explicitly (see Appendix C) that in all the spin-orbital liquids discussed above, the expectation values of these operators are identically zero. This provides a non-trivial consistency check on our PSG calculations.

We now briefly discuss the effect of the fluctuations about the mean-field states. In the absence of pairing channels (both singlet and triplet) the gauge group is $U(1)$. In this case, the fluctuations of the gauge field about the mean field (Eq. 15) are related to the scalar pseudospin chirality $\tilde{S}_1 \cdot \tilde{S}_2 \times \tilde{S}_3$, where the three sites form a triangle. Such fluctuations are gapless in a $U(1)$ spin liquid. It is interesting to note that the scalar spin-chirality is odd under time-reversal symmetry and it has been proposed that such fluctuations can be detected in neutron scattering experiments in presence of spin rotation symmetry breaking. In the present case, however, due to the presence of spinon pairing, the gauge group is broken down to $Z_2$ and the above gauge fluctuations are rendered gapped through Anderson-Higgs mechanism.

In addition to the above gauge fluctuations, because of the triplet decouplings which break pseudospin rotational symmetry, there are bond quadrupolar fluctuations of the pseudospins $Q_{ij}^{\alpha \beta}$ (Eq. 35), as well as vector chirality fluctuations $\mathcal{J}_{ij}$ (Eq. 36) on the bonds. These nematic and vector chirality fluctuations are gapped because the underlying pseudospin Hamiltonian (Eq. 4) breaks pseudospin-rotation symmetry. However, we note that because of the unusual transformation of the non-Kramers pseudospins under time reversal (only the $z$-component of pseudospins being odd under time reversal), $Q_{ij}^{13}$, $Q_{ij}^{23}$, $\mathcal{J}_{ij}^{1}$ and $\mathcal{J}_{ij}^{2}$ are odd under time reversal. Hence, while their mean field expectation values are zero (see above), the fluctuations of these quantities can in principle linearly couple to the neutrons in addition to the $z$-component of the pseudospins.

Having identified the possible $Z_2$ spin-orbital liquids, we can now study typical dynamic structure factors for these spin-orbital liquids. In the next section we examine the typical spinon band structure for different spin-orbital liquids obtained above and find their dynamic spin structure factor.

### Table I: Symmetry allowed terms:

| No. | $\Lambda$ | n.n. | n.n.n. | $Z_2$ |
|-----|-----------|------|--------|-------|
| 1-2 | $\Gamma^2$, $\Gamma^3$ | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 3-4 | 0 | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 5-6 | $\Gamma^3$ | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{10}_1 \xi^{21}_1, \xi^{12}_1 \xi^{20}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 7-8 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n.n. |
| 9-10 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 11-12 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n.n. |
| 13-14 | $\Gamma^3$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 15-16 | $\Gamma^3$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 17-18 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 19-20 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 21-22 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n.n. |
| 23-24 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 25-26 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 27-28 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
| 29-30 | 0 | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | $\xi^{11}_1 \xi^{20}_1, \xi^{10}_1 \xi^{21}_1, \xi^{03}_1 \xi^{32}_1, \xi^{33}_1$ | n.n. |
IV. DYNAMIC SPIN STRUCTURE FACTOR

We compute the dynamic spin structure factor

$$S(q, \omega) = \int \frac{dt}{2\pi} e^{i\omega t} \sum_{ij} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \sum_{a=1,2,3} \langle \sigma_i^a(t) \sigma_j^a(0) \rangle,$$

for an example ansatz of our spin liquid candidates, in order to demonstrate the qualitative differences between the Kramers and non-Kramers spin-orbital liquids. In the above equation, the pseudo-spin variables are defined in a global basis (with the z-axis perpendicular to the Kagome plane). In computing the structure factor for the non-Kramers example, we include only the $\sigma^z$ components of the pseudo-spin operator in the local basis, since only the $z$-components carry magnetic dipole moment (see discussion before). Hence, only this component couples linearly to neutrons in a neutron scattering experiment.

Eq. 37 fails to be periodic in the first Brillouin zone of the Kagome lattice, as the term $r_i - r_j$ in eq. 37 is a half-integer multiple of the primitive lattice vectors when the sublattices of sites i and j are not equal. As such, we examine the structure factor in the extended brillouin zone, which consists of those momenta of length up to double that of those in the first brillouin zone. We plot the structure factor along the cut $\Gamma \rightarrow M' \rightarrow K' \rightarrow \Gamma$, where $M' = 2M$ and $K' = 2K$. We examine the structure factors for two ansatz of spin liquid #17 which has both Kramers and non-Kramers analogues.

As expected, we find that the structure factor has greater weight in the case of a Kramers spin liquid. This is partially due to the fact that the moment of the scattering particle couples with all components of the spin, rather than simply the $z$-component. In addition, we note that the presence of terms allowed in the non-Kramers spin-orbital liquid induces the formation of a gap, which is absent for the Kramers case with up to second nearest neighbour singlet and triplet terms in this particular spin-orbital liquid. Qualitative and quantitative differences such as these, which can be observed in these structure factors between Kramers and non-Kramers spin-orbital liquids, provides one possible distinguishing experimental signature of these states. We shall not pursue this in detail in the present work.

V. DISCUSSION AND POSSIBLE EXPERIMENTAL SIGNATURE OF NON-KRAMERS SPIN-ORBITAL LIQUIDS

In this work, we have outlined the possible $Z_2$ spin-orbital liquids, with gapped or gapless fermionic spinons, that can be obtained in a system of non-Kramers pseudo-spin-1/2s on a Kagome lattice of Pr$^{+3}$ ions. We find a total of thirty, 10 more than in the case of corresponding Kramers system, allowed within PSG analysis in presence of time reversal symmetry. The larger number of spin-orbital liquids is a result of the difference in the action of the time-reversal operator, when realized projectively. We note that the spin-spin dynamic structure factor can bear important signatures of a non-Kramers spin-orbital liquid when compared to their Kramers counterparts. Our analysis of the number of invariant PSGs leading to possibly different spin-orbital liquids that may be realizable in other lattice geometries will form interesting future directions.

We now briefly discuss an experiment that can play an important role in determining non-Kramers spin-orbital liquids. Since the non-Kramers doublets are protected by crystalline symmetries, lattice strains can linearly couple to the pseudo-spinons. As we discussed, the transverse ($x$ and $y$) components of the pseudo-spins $\{\sigma^x, \sigma^y\}$ carry quadrupolar moments and hence are even under the time reversal transformation. Further, they transform under an $E_g$ irreducible representation of the local $D_{3d}$ crystal field. Hence any lattice strain which has this symmetry can linearly couple to the above two transverse components. It turns out that in the crystal type that we are concerned, there is indeed such a mode related to the distortion of the oxygen octahedra. Symmetry considerations show that the linear coupling is of the form $E_{g1}\sigma^1 + E_{g2}\sigma^2$ ($\{E_{g1}, E_{g2}\}$ being the two components of the distortion in the local basis). The above mode is Raman active. For a spin-liquid, we expect that as the temperature is lowered, the spinons become more prominent as deconfined quasiparticles. So the Raman active phonon can efficiently decay into the spinons due to the above coupling channel. If the spin liquid is gapless, this will lead to anomalous broaden-
ing of the above Raman mode as the temperature is lowered, which, if observed, can be an experimental signature of the non-Kramers spin-orbital liquid. The above coupling is forbidden in Kramers doublets by time-reversal symmetry and hence no such anomalous broadening is expected.

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Appendix A: Crystal Field Effects

In this appendix, we explore the breaking of the \( J = 4 \) spin degeneracy by the crystalline electric field. The oxygen and TM ions form a \( D_{3d} \) local symmetry environment around the \( Pr^{3+} \) ions, splitting the ground state degeneracy of the electrons. This symmetry group contains 6 classes of elements: \( E, 2C_3, 3C_2, i, 2S_6, \) and \( 3\sigma_d \), where the \( C_3 \) are rotations by \( 2\pi/3 \) about the local z axis, the \( C_2 \) are rotations by \( \pi \) about axis perpendicular to the local z axis, \( i \) is inversion, \( S_6 \) is a rotation by \( 4\pi/3 \) combined with inversion and \( \sigma_d \) is a reflection about the plane connecting one corner and the opposing plane, running through the \( Pr \) molecule about which this is measured (or, equivalently, a rotation about the x axis combined with inversion). For our \( J=4 \) manifold, these have characters given by

\[
\chi^{(4)}(E) = 2 \ast 4 + 1 = 9 = \chi^{(4)}(i) (A1)
\]

\[
\chi^{(4)}(C_3) = \chi^{(4)}(\frac{2\pi}{3}) = \frac{\sin(3\pi)}{\sin(\pi/3)} = 0 = \chi^{(4)}(S_6) (A2)
\]

\[
\chi^{(4)}(\sigma_d) = \chi^{(4)}(\pi) = \frac{\sin(9\pi/2)}{\sin(\pi/2)} = 1 = \chi^{(4)}(C_2') (A3)
\]

where the latter equalities are given by the fact that our \( J=4 \) manifold is inversion symmetric. Thus, decomposing this in terms of \( D_{3d} \) irreps, our \( J=4 \) manifold splits into a sum of doublet and singlet manifolds as

\[
\Gamma_{\ell=4} = 3E_g + 2A_{1g} + A_{2g}. (A4)
\]

To examine this further, we need to consider the matrix elements of the crystal field potential between the states of different angular momenta. We know that this potential must be invariant under all group operations of \( D_{3d} \), so we can examine the transformation properties of individual matrix elements, \( \langle m|V|m'\rangle \). Under the \( C_3 \) operation, these states of fixed \( m \) transform as

\[
C_3|m\rangle = e^{\frac{2\pi i}{3}}|m\rangle = \omega^m|m\rangle \quad (\omega = e^{\frac{2\pi i}{3}}) (A5)
\]

and thus the matrix elements transform as

\[
C_3 : \langle m|V|m'\rangle \rightarrow \langle m|(C_3)^{-1}VC_3|m'\rangle = \omega^{m'-m}\langle m|V|m'\rangle. (A6)
\]

By requiring that this matrix be invariant under this transformation, we can see that this potential only contains matrix elements for mixing of states which have the \( z \)-component of angular momentum which differ by 3. Thus, our eigenstates are mixtures of the \( |m = 4\rangle, |m = 1\rangle, \) and \( |m = -2\rangle \) states, of the \( |m = 3\rangle, |m = 0\rangle, \) and \( |m = -3\rangle \) states, and of the \( |m = -4\rangle, |m = -1\rangle, \) and \( |m = 2\rangle \) states.

In addition to this, we have the transformation properties

\[
T|m\rangle = (-1)^m|m\rangle \quad (A7)
\]

and

\[
\sigma|m\rangle = (-1)^m|m\rangle \quad (A8)
\]

(where the operators for time reversal and reflection are bolded for future clarity). Inversion acts trivially on these states, as we have total angular momentum even. Thus our time-reversal and lattice reflection (about one axis) symmetries give us doublet states of eigenstates \( \alpha|m = 4\rangle + \beta|m = 1\rangle - \gamma|m = -2\rangle \) and \( \alpha|m = -4\rangle - \beta|m = -1\rangle - \gamma|m = 2\rangle \) (with \( \alpha, \beta, \gamma \in \mathbb{R} \) in order to respect the time reversal symmetry) for the three eigenstates of \( V \) in these sectors. The eigenstates of the \( |m = 3\rangle, |m = 0\rangle, \) and \( |m = -3\rangle \) portion of \( V \) must therefore split into three singlet states, by our representation theory argument \( A4 \). Due to the expected strong Ising term in our potential, we expect the eigenstate with maximal \( J \) to be the ground state, meaning that to analyze the properties of this ground state we are interested in a single doublet state, one with large \( \alpha \) close to one). We will restrict ourselves to this manifold from this point forward, and define the two states in this doublet as

\[
|+\rangle = \alpha|m = 4\rangle + \beta|m = 1\rangle - \gamma|m = -2\rangle \quad (A9)
\]

\[
|-\rangle = \alpha|m = -4\rangle - \beta|m = -1\rangle - \gamma|m = 2\rangle. \quad (A10)
\]

We shall also refer to states of angular momentum \( |m = n\rangle \) as \( |n\rangle \) for simplicity of notation.

Appendix B: Gauge transformations

We begin by describing the action of time reversal on our ansatz. The operation is antunitary, and must be combined with a spin transformation \( \sigma^1 \) in the case of non-Kramers doublets. As a result, the operation acts as \( T : \xi_{ij}^{\alpha\beta}\Sigma^\alpha\Sigma^\beta \rightarrow \xi_{ij}^{\alpha\beta}\Sigma^1\Sigma^{\alpha\Sigma^1\Sigma^1\Gamma^\beta} \). However, we can simplify this considerably by performing a gauge transformation in addition to the above transformation, which yields the same transformation on any physical variables. The gauge transformation we perform is \( \tilde{\xi}^{\alpha\beta} \), which changes the form of the time reversal operation to \( T : \xi_{ij}^{\alpha\beta}\Sigma^\alpha\Sigma^\beta \rightarrow \xi_{ij}^{\alpha\beta}\Sigma^1\Sigma^{\alpha\Sigma^1\Sigma^1\Gamma^\beta} = \xi_{ij}^{\alpha\beta}\Sigma^1\Sigma^1\Sigma^1\Gamma^\beta \), where \( \tilde{\xi}^{\alpha\beta} \equiv \xi^{\alpha\beta} \) if \( \alpha \in \{1, 2\} \) and \( \tilde{\xi}^{\alpha\beta} = -\xi^{\alpha\beta} \) if \( \alpha \in \{0, 3\} \).

On the Kagome lattice, the allowed form of the gauge transformations has been determined by Yuan-Ming Lu et al.\(^{15}\). For completeness, we will reproduce that calculation, valid also
for our spin triplet ansatz. Here, the relations between the

gauge transformation matrices,

\begin{align}
[G_T(i)]^2 &= \eta T I, \\
G_\sigma(\sigma(i))G_\sigma(i) &= \eta \sigma I, \\
G^\dagger_{T T}(i)G^\dagger_T(i)G_T(i)G_T^{-1}(i) &= \eta T T I, \\
G^\dagger_T(i)G^\dagger_{TT}(i)G_T(i)G_T^{-1}(i) &= \eta T T I, \\
G^\dagger_\sigma(i)G^\dagger_\sigma(i)G_\sigma(i)G_\sigma^{-1}(i) &= \eta \sigma T I, \\
G^\dagger_\sigma(i)G^\dagger_\sigma(i)G_\sigma(i)G_\sigma^{-1}(i) &= \eta \sigma T I, \\
G^\dagger_S(i)G^\dagger_S(i)G_S(i)G_S^{-1}(i) &= \eta S T I, \\
G^\dagger_S(i)G^\dagger_S(i)G_S(i)G_S^{-1}(i) &= \eta S T I,
\end{align}

are valid for our case as well, due to the decoupling of spin and
gauge portions of our ansatz. In the above, the relations are
valid for all lattice sites \( i = (x, y, s) \). I is the 4x4 identity
matrix, and the \( G_T \) matrices are gauge transformation
matrices generated by exponentiation of the \( T \) matrices. The \( \eta \)'s
are \( \pm 1 \), the choice of which characterize different spin liquid
states. In deriving this form of the commutation relations, we
have included a gauge transformation \( i^2 \) in our definition of
the time reversal operator, as this simplifies the effect of
the operator on the mean field ansatz.

We turn next to the calculation of the gauge transformations.
We look first at the gauge transformations associated
with the translations. We can perform a site dependent
gauge transformation \( W(i) \), under which the gauge transformations
associated with the translational symmetries transform as

\begin{align}
G_{T_1}(i) &\rightarrow W(i)G_{T_1}(i)W^\dagger(i - \hat{x}), \\
G_{T_2}(i) &\rightarrow W(i)G_{T_2}(i)W^\dagger(i - \hat{y}).
\end{align}

As such, we can choose a gauge transformation \( W(i) \) to
simplify the form of \( G_{T_1} \) and \( G_{T_2} \). Using such a transformation,
along with condition B7, we can restrict the form of these
gauge transformations to be

\begin{align}
G_{T_1}(i) &= \eta_{T_1}^y I, \\
G_{T_2}(i) &= I.
\end{align}

To preserve this choice, we can now only perform gauge transformations which are equivalent on all lattice positions
\((W(x, y, s) = W(s))\) or transformations which change the
shown matrices by an IGG transformation.

Next, we look at adding the reflection symmetry \( \sigma \). Given
our formulae for \( G_{T_1} \) and \( G_{T_2} \), along with the relations be-

 tween the gauge transformations, we have that

\begin{align}
G_{T_1}(T_2^{-1}(i))G_\sigma(i)\eta_{T_2}^y &= \eta_{T_1}^r I, \\
G_{T_1}(T_1^{-1}(i))G_\sigma(i)\eta_{T_2}^y &= \eta_{T_2}^r I.
\end{align}

Defining \( G_\sigma(0,0,s) = g_\sigma(s) \), we have, by repeated application
of the above,

\begin{align}
G_\sigma(0, y, s) &= \eta_{T_1}^y g_\sigma(s) \\
G_\sigma(x, y, s) &= \eta_{T_1}^y \eta_{T_2}^y g_\sigma(s).
\end{align}

Next, using

\begin{align}
G_\sigma(\sigma(i))G_\sigma(i) &= \eta \sigma I
\end{align}

we find that

\begin{align}
\eta_T I &= G_\sigma(y, x, \sigma(s))G_\sigma(x, y, s) \\
&= (\eta_{T_1} \eta_{T_2})^{x+y} g_\sigma(s) \eta \sigma(s).
\end{align}

Since this is true for all \( x \) and \( y \), \( \eta_{T_1} \eta_{T_2} = 1 \) and thus
\( \eta_{T_1} \eta_{T_2} = \eta_{T_1} \eta_{T_2} \) and \( g_\sigma(s) \eta \sigma(s) = \eta \sigma I \), where \( \sigma(u) = u, \sigma(v) = w \) and \( \sigma(w) = v \). Our final form for the gauge transformation is

\begin{align}
G_\sigma(x, y, s) &= \eta_{T_1}^y \eta_{T_2}^y g_\sigma(s).
\end{align}

Next we look at adding the \( S_6 \) symmetry to our calculation.
We can do an IGG transformation, taking \( G_{T_1}(T_1(i)) \)
to \( \eta S_6 T_2 G_{T_1}(T_1(i)) \), with the net effect being that \( \eta S_6 T_2 \) becomes one (previous calculations are unaffected). We now have that

\begin{align}
G_{S_6}(T_2^{-1} T_1(i)) &= \eta S_6 \eta_{T_1}^r I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I, \\
G_{S_6}(T_2^{-1} T_1(i))G_{S_6}(i)\eta_{T_1}^{y-1} &= \eta S_6 T_1 I.
\end{align}

Defining \( G_{S_6}(0,0,s) = g_{S_6}(s) \), we find that

\begin{align}
G_{S_6}(n, -n, s) &= \eta_{S_6}^{n(n-1)/2} g_{S_6}(s), \\
G_{S_6}(x, y, s) &= \eta_{S_6}^{x(x-1)/2+y+y} \eta_{S_6 T_1} g_{S_6}(s), \\
G_{S_6}(x, y, s) &= \eta_{S_6}^{x(x-1)/2+y+y} \eta_{S_6 T_1} g_{S_6}(s).
\end{align}

Using the commutation relation between the \( \sigma \) and \( S_6 \)
gauge transformations, we find that

\begin{align}
\eta \sigma S_6 I &= \eta_{T_1}^y \eta_{S_6 T_1} g_{S_6}(v) g_{S_6}(v) \eta \sigma(u) g_{S_6}(u), \\
\eta \sigma S_6 I &= \eta_{T_1}^y \eta_{S_6 T_1} g_{S_6}(v) g_{S_6}(v) \eta \sigma(u) g_{S_6}(u).
\end{align}

Giving us that \( \eta \sigma_{T_1} \eta_{S_6 T_1} = 1 \) and
\( g_{S_6}(u) g_{S_6}(w) g_{S_6}(v) g_{S_6}(v) \eta \sigma(u) = \eta \sigma_{S_6} \eta \sigma I \). A similar calculation on a different sublattice gives us

\begin{align}
\eta \sigma S_6 I &= \eta_{T_1}^y \eta_{S_6 T_1} g_{S_6}(w) g_{S_6}(w) \eta \sigma(u) g_{S_6}(u), \\
\eta \sigma S_6 I &= \eta_{T_1}^y \eta_{S_6 T_1} g_{S_6}(w) g_{S_6}(w) \eta \sigma(u) g_{S_6}(u).
\end{align}
TABLE II: We list the solutions of Eq. B43 - B54, along with a set of gauge transformations which realize these solutions.

| No. | $\eta_T$ | $\eta_{sT}$ | $\eta_{sT}^0$ | $\eta_S$ | $\eta_S^0$ | $\eta_{f_1}$ | $g_\sigma(u)$ | $g_\sigma(v)$ | $g_\sigma(w)$ | $g_{S_0}(u)$ | $g_{S_0}(v)$ | $g_{S_0}(w)$ |
|-----|----------|-------------|--------------|---------|------------|-------------|--------------|--------------|--------------|--------------|--------------|--------------|
| 1,2 | -1       | 1           | 1            | 1       | $\mp$      | 1           | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   |
| 3,4 | -1       | 1           | 1            | 1       | -1         | $\mp$      | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $-\Gamma^0$  | $i\Gamma^0$  | $i\Gamma^0$  |
| 5,6 | -1       | 1           | -1           | -1      | $\mp$      | 1           | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $i\Gamma^0$  | $i\Gamma^0$  | $i\Gamma^0$  |
| 7,8 | -1       | 1           | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $i\Gamma^0$  | $i\Gamma^0$  | $i\Gamma^0$  |
| 9,10 | -1      | -1          | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 11,12 | -1   | -1          | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 13,14 | -1 | -1           | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 15,16 | -1 | -1           | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 17,18 | -1 | -1           | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 19,20 | -1 | -1           | -1           | -1      | $\mp$      | 1           | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 21,22 | 1       | 1           | 1            | 1       | $\mp$      | 1           | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   |
| 23,24 | 1       | 1           | 1            | 1       | -1         | $\mp$      | $\Gamma^0$   | $\Gamma^0$   | $\Gamma^0$   | $-\Gamma^0$  | $i\Gamma^0$  | $i\Gamma^0$  |
| 25,26 | 1       | 1           | 1            | 1       | -1         | $\mp$      | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 27,28 | 1       | 1           | 1            | 1       | -1         | $\mp$      | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |
| 29,30 | 1       | 1           | 1            | 1       | -1         | $\mp$      | $i\Gamma^0$  | $-\Gamma^0$  | $\Gamma^0$   | $-i\Gamma^0$ | $i\Gamma^0$  | $i\Gamma^0$  |

Because this is true for all x and y, and $f_1(i)$ is not equal to $f_2(i)$, the $G_T(i)$ is $i \hat{a} \cdot \vec{k}$, we perform a gauge transformation $W$ on $G_T(i)$ such that $W^\dagger G_T(i) W = i \vec{k}^3$ as this is the same on all sites, it does not affect our gauge fixing for the translation gauge transformations. Collecting the necessary results for further use,

\[
G_{T_1}(x, y, s) = \eta_{T_1}^y I \tag{B43}
\]

\[
G_{T_2}(x, y, s) = I \tag{B44}
\]

\[
G_{\sigma}(x, y, s) = \eta_{\sigma}^y g_\sigma(s) \tag{B45}
\]

\[
G_{S_0}(x, y, s) = \eta_{S_0}^{xy+(x+y)x/2} g_{S_0}(s) \quad s = u, v \tag{B46}
\]

\[
G_T(s) = I = g_T(s) \quad \eta_T = 1 \tag{B48}
\]

\[
G_T(s) = i\Gamma^1 = g_T(s) \quad \eta_T = 1 \tag{B49}
\]

\[
g_\sigma(\sigma(s)) g_s(s) = \eta_s I \tag{B50}
\]

\[
(g_{S_0}(u) g_{S_0}(u) g_{S_0}(v) g_{S_0}(w)) = (g_\sigma(v) g_{S_0}(w))^2 = \eta_{sT} \eta_s I \tag{B51}
\]

\[
(g_{S_0}(u) g_{S_0}(v) g_{S_0}(u))^2 = \eta_{s} \eta_{sT} I \tag{B52}
\]

\[
g_\sigma(\sigma(s)) g_s(s) = \eta_s I \tag{B53}
\]

\[
(g_{S_0}(u) g_{S_0}(v) g_{S_0}(u)) = \eta_{s} \eta_{sT} g_{S_0}(s) \tag{B54}
\]

We also have the gauge freedom left to perform a gauge rotation arbitrarily at all positions for $\eta_T = 1$ or an arbitrary gauge rotation about the x axis for $\eta_T = -1$.

The solution to the above equations is derived in detail by Lu et al.\textsuperscript{15} and as such we simply list the results in table II. The basic method of obtaining these solutions is as follows:
for each choice of $Z_2$ parameter set, we determine whether there is a choice of gauge matrices \{ $g$s \} which satisfy the equations B43 - B54. In order to do so, we determine the allowed forms of the $g$s matrices from the equations, then use the gauge freedom on each site to fix the form of these. Of particular not is the fact that in the consistency equations for the $g$ matrices, the terms $\eta_{12}$ and $\eta_{86}$ only appear multiplied together, meaning that for any choice of the gauge matrices $g$s we can choose $\eta_{12} = \pm 1$, which fixes the form of $\eta_{86}$.

Appendix C: Relation among the mean-field parameters

The relation among the different singlet and triplet parameters in terms of $\xi_{ij}$ is given by

\[
\chi_{ij} = \xi_{ij}^{00} + \xi_{ij}^{03}, \quad \eta_{ij} = -\xi_{ij}^{01} + i\xi_{ij}^{02},
\]

\[
E_{ij}^{1} = \xi_{ij}^{10} + \xi_{ij}^{13}, \quad E_{ij}^{2} = \xi_{ij}^{20} + \xi_{ij}^{23}, \quad E_{ij}^{3} = \xi_{ij}^{30} + \xi_{ij}^{33},
\]

\[
D_{ij}^{1} = -\xi_{ij}^{11} + i\xi_{ij}^{12}, \quad D_{ij}^{2} = -\xi_{ij}^{21} + i\xi_{ij}^{22}, \quad D_{ij}^{3} = -\xi_{ij}^{31} + i\xi_{ij}^{32}. \quad (C1)
\]

Using these, we can derive the form of the bond nematic order parameter and vector chirality order parameters, which are given in terms of the mean field parameters\(^{21}\) as

\[
Q_{ij}^{\mu\nu} = -\frac{1}{2} \left( E_{ij}^{\mu} E_{ij}^{\nu*} - \frac{1}{3} \delta_{\mu\nu} |E_{ij}|^2 \right) + h.c.,
\]

\[
J_{ij}^{\lambda} = \frac{1}{2} \left( \chi_{ij} E_{ij}^{\lambda*} - \chi_{ij}^{*} E_{ij}^{\lambda} \right),
\]

\[
\frac{J_{ij}^{\lambda}}{2} \left( \eta_{ij} D_{ij}^{\lambda*} - \eta_{ij}^{*} D_{ij}^{\lambda} \right). \quad (C2)
\]

where our definition of $\eta_{ij}$ differs by a factor of $(-1)$ from that of the cited work. We rewrite this in terms of our variables, finding

\[
Q_{ij}^{\mu\nu} = -\frac{1}{2} \xi_{ij}^{\mu 0} \xi_{ij}^{\nu 0} + \sum_a \xi_{ij}^{\mu a} \xi_{ij}^{\nu a} + \frac{1}{3} \sum_b \left( (\xi_{ij}^{0b})^2 - \sum_a (\xi_{ij}^{ba})^2 \right) + h.c.,
\]

\[
J_{ij}^{\lambda} = 2i (\xi_{ij}^{00} \lambda_0 - \sum_a \xi_{ij}^{0a} \lambda_a). \quad (C3)
\]

In particular, we find that $J_{ij}^{1}$, $J_{ij}^{2}$, $Q_{ij}^{13}$ and $Q_{ij}^{23}$ must be zero for all non-Kramers spin liquids, as the terms allowed by symmetry in Eq. 32 and 33 do not allow non-zero values for these order parameters.

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