

Supplemental Material for “Chiral spin-orbital liquids with nodal lines”

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1. Symmetry of the spin liquid ansatz

Time reversal $T = K e^{-i\pi J_{\text{tot}}^y} = K \prod_j (i\Gamma_j^{13})$ acts on pseudospins and pseudo-orbitals as

$$T^{-1} \mathbf{s}_j T = \mathbf{s}_j, \quad T^{-1} \tau_j^{x,z} T = \tau_j^{x,z}, \quad T^{-1} \tau_j^y T = -\tau_j^y. \quad (1)$$

In the Majorana fermion representation for \mathbf{s} and $\boldsymbol{\tau}$ this transformation can be implemented by $T = K \prod_j \theta_j^1 \theta_j^3$. This is equivalent to complex conjugation combined with the operation $\theta_j^1 \rightarrow -\theta_j^1$ and $\theta_j^3 \rightarrow -\theta_j^3$. Thus, if we focus on the decoupled flavors $\gamma \in \{\eta^a, \theta^2\}$, we can take time reversal to be represented simply by complex conjugation.

Let $\gamma_{j\ell}$ denote a Majorana fermion on site j belonging to sublattice $\ell = \text{A,B,C,D}$. The operators $\gamma_{\mathbf{k}\ell}$ in momentum space are defined by

$$\gamma_{j\ell} = \sqrt{\frac{8}{N}} \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} [\gamma_{\mathbf{k}\ell} e^{i\mathbf{k}\cdot\mathbf{R}_j} + \gamma_{\mathbf{k}\ell}^\dagger e^{-i\mathbf{k}\cdot\mathbf{R}_j}], \quad (2)$$

$$\gamma_{\mathbf{k}\ell} = \sqrt{\frac{2}{N}} \sum_{j \in \ell} \gamma_{j\ell} e^{-i\mathbf{k}\cdot\mathbf{R}_j}, \quad (3)$$

and $\gamma_{\mathbf{k}\ell}$ are normalized such that $\{\gamma_{\mathbf{k}\ell}, \gamma_{\mathbf{k}'\ell'}^\dagger\} = \delta_{\mathbf{k}\mathbf{k}'} \delta_{\ell\ell'}$. For each flavor of Majorana fermion we combine the four sublattice modes into a single “spinor” $\boldsymbol{\gamma}_{\mathbf{k}} = (\gamma_{\mathbf{k}\text{A}}, \gamma_{\mathbf{k}\text{B}}, \gamma_{\mathbf{k}\text{C}}, \gamma_{\mathbf{k}\text{D}})^t$.

In momentum space, time reversal takes $\mathbf{k} \rightarrow -\mathbf{k}$. Up to a hopping amplitude (determined by self-consistent equations, see next section), the mean-field Hamiltonian for a decoupled flavor is of the form $\tilde{H}_{\text{MF}} = \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} \boldsymbol{\gamma}_{\mathbf{k}}^\dagger \mathcal{H}(\mathbf{k}) \boldsymbol{\gamma}_{\mathbf{k}}$ with

$$\mathcal{H}(\mathbf{k}) = i \begin{pmatrix} 0 & f(k_x, k_y) & f(k_y, k_z) & f(k_x, k_z) \\ -f(k_x, k_y) & 0 & -f(k_x, k_z) & f(k_y, k_z) \\ -f(k_y, k_z) & f(k_x, k_z) & 0 & -f(k_x, k_y) \\ -f(k_x, k_z) & -f(k_y, k_z) & f(k_x, k_y) & 0 \end{pmatrix}, \quad (4)$$

where $f(k_a, k_b) = 4 \cos(k_a/2) \cos(k_b/2)$. Notice the factor of i . It follows that

$$T^{-1} \mathcal{H}(\mathbf{k}) T = \mathcal{H}^*(-\mathbf{k}) = -\mathcal{H}(\mathbf{k}). \quad (5)$$

We define inversion P as the reflection by the mirror plane that exchanges A and C sublattices (plane perpendicular to $\boldsymbol{\delta}_{yz} = (0, \frac{1}{2}, \frac{1}{2})$). In momentum space, $P : k_x \rightarrow k_x, k_y \rightarrow -k_z, k_z \rightarrow -k_y$. In addition, we have the action in the internal (sublattice) space given by the matrix (with determinant -1)

$$P = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (6)$$

We also define the Z_2 gauge transformation that changes the sign of fermions on the B sublattice:

$$G_P = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (7)$$

It is easy to check that inversion anticommutes with the mean-field Hamiltonian:

$$(PG_P)^{-1}\mathcal{H}(P\mathbf{k})PG_P = -\mathcal{H}(\mathbf{k}). \quad (8)$$

It follows that the combined transformation PTG_P is a symmetry of the Hamiltonian:

$$(PTG_P)^{-1}\mathcal{H}(P\mathbf{k})PTG_P = \mathcal{H}(\mathbf{k}). \quad (9)$$

The C_3 rotation about a (111) axis that leaves an A site invariant is represented by

$$C_3 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix}, \quad (10)$$

and the rotation in momentum space takes $R_3 : k_x \rightarrow k_z, k_y \rightarrow k_x, k_z \rightarrow k_y$. In this case a gauge transformation is not required; we obtain immediately that

$$C_3^{-1}\mathcal{H}(R_3\mathbf{k})C_3 = \mathcal{H}(\mathbf{k}). \quad (11)$$

The C_2 rotation along the z axis that exchanges $A \leftrightarrow B, C \leftrightarrow D$ is represented by

$$C_2 = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}, \quad (12)$$

and in momentum space $R_2 : k_x \rightarrow -k_x, k_y \rightarrow -k_y$. We need to combine the C_2 rotation with the gauge transformation

$$G_2 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \quad (13)$$

We then have

$$(C_2G_2)^{-1}\mathcal{H}(R_2\mathbf{k})(C_2G_2) = \mathcal{H}(\mathbf{k}). \quad (14)$$

Translation by $\boldsymbol{\delta}_{xy} = (\frac{1}{2}, \frac{1}{2}, 0)$, which we denote by T_{xy} , has the same effect of exchanging sublattices as the above C_2 rotation. Thus, conjugation by $T_{xy}G_2$, together with $\mathbf{R}_j \rightarrow \mathbf{R}_j + \boldsymbol{\delta}_{xy}$ in real space, is also a symmetry of the Hamiltonian (and likewise for the equivalent translations in yz and xz planes).

Now consider a C_4 rotation along the z axis going through an A site, which exchanges C and D sublattices. This can be represented in sublattice space by

$$\hat{C}_4 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{pmatrix}. \quad (15)$$

In momentum space, $R_4 : k_x \rightarrow k_y, k_y \rightarrow -k_x$. Combining with the gauge transformation:

$$\hat{G}_4 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (16)$$

we find

$$(C_4G_4)^{-1}\mathcal{H}(R_4\mathbf{k})(C_4G_4) = -\mathcal{H}(\mathbf{k}). \quad (17)$$

Thus, like P and T , the C_4 rotation inverts the chirality of the ansatz. It is then easy to see that $C_4 G_4 T$ is a symmetry of the Hamiltonian.

The C_4 rotation can be used to construct a symmetry transformation that accounts for the twofold degeneracy of the Majorana fermion bands.

It is also interesting to consider the C_4 rotation that exchanges A and B sublattices, given by

$$\hat{C}'_4 = \begin{pmatrix} 0 & -1 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (18)$$

If we define this to be a rotation around z axis in the opposite direction than the one in Eq. (15), the transformation in momentum space is $R'_4 = (R_4)^{-1}$, i.e., $R'_4 : k_x \rightarrow -k_y, k_y \rightarrow k_x$. It is easy to check that the composition $M = \hat{C}_4 \hat{C}'_4$ commutes with the Hamiltonian, $M^{-1} \mathcal{H}(\mathbf{k}) M = \mathcal{H}(\mathbf{k})$, and obeys $M^2 = -\mathbb{1}$. Thus, we can block diagonalize $\mathcal{H}(\mathbf{k})$ by sectors labeled by the eigenvalue $\pm i$ of the matrix M :

$$U_M^\dagger \mathcal{H}(\mathbf{k}) U_M = \begin{pmatrix} \tilde{\boldsymbol{\sigma}} \cdot \mathbf{h}_{\mathbf{k}} & 0 \\ 0 & -\tilde{\boldsymbol{\sigma}} \cdot \mathbf{h}_{\mathbf{k}} \end{pmatrix}, \quad (19)$$

where $\tilde{\boldsymbol{\sigma}} \equiv (-\sigma^z, \sigma^y, \sigma^x)$ and U_M is the unitary matrix that diagonalizes M . It is then clear that the spectrum of $\mathcal{H}(\mathbf{k})$ is twofold degenerate with eigenvalues $\pm |\mathbf{h}_{\mathbf{k}}|$. Two degenerate states can be distinguished by the eigenvalue ± 1 of the Hermitean matrix iM (which is analogous to the chirality of Weyl fermions in the massless Dirac equation).

In summary, the chiral spin-orbital liquid ansatz lowers the symmetry of the Hamiltonian from $O_h \times Z_2$ (where Z_2 is time reversal) to O_h (where the new group contains combinations of broken point group symmetries with the broken time reversal).

2. Berry connection

The nodal lines can be characterized as topological defects of a Berry connection in reciprocal space. In our case, the Berry connection has to be non-Abelian due to the double degeneracy of the bands. Away from the nodal lines, we define the $SU(2)$ connection

$$A_{mn}^a(\mathbf{k}) = i \langle \psi_m(\mathbf{k}) | \partial_{k_a} \psi_n(\mathbf{k}) \rangle, \quad (20)$$

where $|\psi_m(\mathbf{k})\rangle$ and $|\psi_n(\mathbf{k})\rangle$, $m, n \in \{1, 2\}$, are degenerate eigenstates of $\mathcal{H}(\mathbf{k})$ (say with energy $\varepsilon_+(\mathbf{k})$) chosen so as to obey $\langle \psi_m(\mathbf{k}) | \psi_n(\mathbf{k}) \rangle = \delta_{mn}$ and to diagonalize $A^z(\mathbf{k})$. The generalized Berry phase is the Wilson loop

$$U = \mathcal{P} \exp[-i \oint dk_a A^a(\mathbf{k})], \quad (21)$$

where \mathcal{P} denotes path ordering. The calculation of U is simplified if we consider a path around the line node parametrized by $\mathbf{k} \approx (\pi + \epsilon \cos \alpha, \pi + \epsilon \sin \alpha, k_z)$, with $\alpha \in [-\pi, \pi)$. For infinitesimal radius $\epsilon \ll 1$, we obtain

$$A^x = -\frac{1}{2\epsilon} \sin \alpha \sigma^y + \mathcal{O}(\epsilon^0), \quad A^y = \frac{1}{2\epsilon} \cos \alpha \sigma^y + \mathcal{O}(\epsilon^0), \quad (22)$$

which is precisely the singular ϵ dependence of a vortex line. We then find $U = -\mathbb{1}$, equivalent to a π Berry phase.

3. Solving the mean-field Hamiltonian

In this section we outline the steps required to diagonalize the mean-field Hamiltonian and calculate the ground state energy.

Using the mode expansion Eq. (2), we can rewrite the various hopping terms for Majorana fermions in terms of operators in reciprocal space. For instance,

$$i \sum_{\langle j,l \rangle \in XY} \gamma_{jA} \gamma_{lB} = 2i \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} h_{\mathbf{k}}^1 (\gamma_{\mathbf{k}A}^\dagger \gamma_{\mathbf{k}B} - \gamma_{\mathbf{k}B}^\dagger \gamma_{\mathbf{k}A}), \quad (23)$$

where $h_{\mathbf{k}}^1 = 4 \cos(k_x/2) \cos(k_y/2)$ is the first component of $\mathbf{h}_{\mathbf{k}}$. The mean-field Hamiltonian becomes

$$\begin{aligned} \tilde{H}_{\text{MF}} = & \frac{J}{18} \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} \left[(2u + \bar{w}) \sum_{a=1}^3 (\eta_{\mathbf{k}}^a)^\dagger \mathcal{H}_1(\mathbf{k}) \eta_{\mathbf{k}}^a - w (\theta_{\mathbf{k}}^2)^\dagger \mathcal{H}_1(\mathbf{k}) \theta_{\mathbf{k}}^2 + (\Theta_{\mathbf{k}})^\dagger \mathcal{H}_2(\mathbf{k}) \Theta_{\mathbf{k}} \right] \\ & - \frac{NJ}{6} + \frac{J}{36} \sum_{\langle i,j \rangle \in \alpha} (3u_{ij}^2 + 3\bar{w}_{ij}^\alpha u_{ij} - w_{ij}^\alpha v_{ij}), \end{aligned} \quad (24)$$

where $\mathcal{H}_1(\mathbf{k})$ is the 4×4 matrix in Eq. (4), $\Theta_{\mathbf{k}} = (\theta_{\mathbf{k}A}^1, \dots, \theta_{\mathbf{k}D}^3)^t$ is an eight-component spinor that combines θ^1 and θ^3 fermions and $\mathcal{H}_2(\mathbf{k})$ is an 8×8 matrix to be specified below.

First consider the fermions $\zeta \in \{\eta^a, \theta^2\}$, whose spectrum is determined by $\mathcal{H}_1(\mathbf{k})$. Let $U_{\mathbf{k}}$ be the unitary transformation that diagonalizes $\mathcal{H}_1(\mathbf{k})$:

$$U_{\mathbf{k}}^\dagger \mathcal{H}_1(\mathbf{k}) U_{\mathbf{k}} = \Lambda_1(\mathbf{k}), \quad (25)$$

where $\Lambda_1(\mathbf{k}) = \text{diag}\{-|\mathbf{h}_{\mathbf{k}}|, -|\mathbf{h}_{\mathbf{k}}|, |\mathbf{h}_{\mathbf{k}}|, |\mathbf{h}_{\mathbf{k}}|\}$ is a diagonal matrix. The operators that annihilate fermions in eigenstates of $\mathcal{H}_1(\mathbf{k})$ are

$$\tilde{\gamma}_{\mathbf{k}\lambda} = \sum_{\ell=A,\dots,D} (U^\dagger)_{\lambda\ell} \gamma_{\mathbf{k}\ell}, \quad (26)$$

$$\gamma_{\mathbf{k}\ell} = \sum_{\lambda=1}^4 (U)_{\ell\lambda} \tilde{\gamma}_{\mathbf{k}\lambda}, \quad (27)$$

where $\lambda = 1, \dots, 4$ is the band index. The mean-field ground state $|\text{GS}\rangle$ is the state in which all single-fermion states with negative energy are occupied. This leads to the self-consistent equation for expectation values, e.g.

$$\begin{aligned} \langle \gamma_{j,A} \gamma_{j+\delta_{xy},B} \rangle &= \frac{8}{N} \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} [e^{i\mathbf{k}\cdot\delta_{xy}} \langle \gamma_{\mathbf{k}A}^\dagger \gamma_{\mathbf{k}B} \rangle + e^{-i\mathbf{k}\cdot\delta_{xy}} \langle \gamma_{\mathbf{k}A} \gamma_{\mathbf{k}B}^\dagger \rangle] \\ &= i \text{Im} \left[\frac{16}{N} \sum_{\mathbf{k} \in \frac{1}{2}\text{BZ}} \sum_{\lambda} (U^\dagger)_{\lambda A} U_{B\lambda} e^{i\mathbf{k}\cdot\delta_{xy}} \langle \tilde{\gamma}_{\mathbf{k}\lambda}^\dagger \tilde{\gamma}_{\mathbf{k}\lambda} \rangle \right] \\ &= \frac{i}{2\pi^3} \text{Im} \left\{ \sum_{\lambda (\epsilon_\lambda < 0)} \int_{\frac{1}{2}\text{BZ}} d^3k (U^\dagger)_{\lambda A} U_{B\lambda} \right\}, \end{aligned} \quad (28)$$

where in the last line the sum is over bands with negative energy and we took the thermodynamic limit to replace $\sum_{\mathbf{k}} \rightarrow \frac{N}{32\pi^3} \int d^3k$ (corresponding to $N/4$ states in the Brillouin zone of the cubic sublattice).

Since $\mathcal{H}_1(\mathbf{k})$ determines the spectrum of η^a and θ^2 fermions, the self-consistent equations for $u_{ij} = -i \langle \eta_i^a \eta_j^a \rangle$ and $v_{ij} = -i \langle \theta_i^2 \theta_j^2 \rangle$ are the same up to an overall minus sign, depending on the relative sign of the hopping amplitudes $2u + \bar{w}$ and $-w$ in Eq. (24). We then have the constraint $|u| = |v|$, but must analyze two possibilities, namely $u = v$ and $u = -v$. Without loss of generality (by choosing one of the two degenerate ground states with opposite chiralities), we can set $u > 0$. Numerical evaluation of the integral in Eq. (28) then yields $u \approx 0.258$.

The relation between u and v determines the 8×8 matrix for $\Theta_{\mathbf{k}}$. For $v = u$, we obtain

$$\mathcal{H}_2(\mathbf{k}) = u \mathbf{h}_{\mathbf{k}} \cdot \boldsymbol{\Sigma}', \quad (29)$$

where $\boldsymbol{\Sigma}' = (2K^z \otimes \sigma^y, -2K^y \otimes \mathbb{1}, -2K^x \otimes \sigma^y)$, with

$$K^x = \frac{1}{2} \begin{pmatrix} 0 & 2 & 0 & \sqrt{3} \\ 2 & 0 & \sqrt{3} & 0 \\ 0 & \sqrt{3} & 0 & 0 \\ \sqrt{3} & 0 & 0 & 0 \end{pmatrix}, \quad K^y = \frac{i}{2} \begin{pmatrix} 0 & -2 & 0 & \sqrt{3} \\ 2 & 0 & -\sqrt{3} & 0 \\ 0 & \sqrt{3} & 0 & 0 \\ -\sqrt{3} & 0 & 0 & 0 \end{pmatrix}, \quad K^z = \frac{1}{2} \begin{pmatrix} 1 & & & \\ & -1 & & \\ & & -3 & \\ & & & 3 \end{pmatrix}. \quad (30)$$

The components of the matrix vector \mathbf{K} satisfy the $\text{SU}(2)$ algebra. We then obtain the spectrum of $\mathcal{H}_2(\mathbf{k})$ and use it to solve the self-consistent equations for $w_{ij} = -i \langle \theta_i^\alpha \theta_j^\alpha \rangle$ and $\bar{w}_{ij} = -i \langle \bar{\theta}_i^\alpha \bar{\theta}_j^\alpha \rangle$ analogous to Eq. (28). In this case

of $u = v$ we find a self-consistent solution with $w \approx -0.081$ and $\bar{w} \approx 0.317$. Having fixed the order parameters, we obtain the mean-field ground states energy $E_{\text{MF}}(v = u) = \langle \bar{H}_{\text{MF}} \rangle \approx -0.244NJ$.

For $v = -u$ we obtain

$$H_{\mathbf{k}}^{(2)} = u \mathbf{h}_{\mathbf{k}} \cdot \boldsymbol{\Sigma}'', \quad (31)$$

where $\boldsymbol{\Sigma}'' = ((2 - \sigma^{xy}) \otimes \Sigma^1, (2 - \sigma^{yz}) \otimes \Sigma^2, (2 - \sigma^{xz}) \otimes \Sigma^3)$, with $\boldsymbol{\Sigma}$ the matrix vector defined in the main text, and the 2×2 matrices σ^α given by $\sigma^{xy} = \sigma^z$, $\sigma^{yz(xz)} = \frac{1}{2}(-\sigma^z \pm \sqrt{3}\sigma^x)$. In this case we find a self-consistent solution with $w \approx 0.161$ and $\bar{w} \approx 0.318$. The ground state energy is $E_{\text{MF}}(v = -u) \approx -0.248NJ$, slightly lower than the result for $u = v$. This is the value quoted in the main text. We note that the small difference between the two energies may change beyond the mean-field level. However, we have verified that both solutions give rise to a spectrum with nodal lines along MR directions, qualitatively similar to the spectrum for η^a and θ^2 fermions. Therefore, the properties derived from the low-energy density of states $\rho(\epsilon) \propto \sqrt{\epsilon}$ are generic.

4. Variational Monte Carlo

To check the viability of the proposed chiral spin-orbital liquid beyond the mean-field level, we now enforce the local constraint exactly by considering a Gutzwiller projection of the mean-field wave function [1] by means of a variational Monte Carlo calculation [2].

We begin by rewriting the Majorana fermions in terms of three Dirac fermions, closely following the representation used in Ref. [3]

$$c_i^\dagger = \frac{1}{2} (\eta_i^2 + i\theta_i^2), \quad d_i^\dagger = \frac{1}{2} (\eta_i^1 + i\eta_i^3), \quad f_i^\dagger = \frac{1}{2} (\theta_i^1 + i\theta_i^3). \quad (32)$$

In terms of this representation, the local constraint is now translated into the fact that a given site may either have no Dirac fermions, a state denoted by $|0\rangle$, or two Dirac fermions, in which case there are three possible states at each site defined as

$$|x_i\rangle = c_i^\dagger d_i^\dagger |0\rangle, \quad |y_i\rangle = d_i^\dagger f_i^\dagger |0\rangle, \quad |z_i\rangle = f_i^\dagger c_i^\dagger |0\rangle. \quad (33)$$

Given a real space configuration specified by the locations of the doubly occupied sites, $X \equiv \{x_i\}, Y \equiv \{y_i\}, Z \equiv \{z_i\}$, the wave function assigns an amplitude $\Psi(\{x_i\}, \{y_j\}, \{z_m\})$ to it. Notice that the locations of the $|0\rangle$ states are automatically specified. We point out that the local constraint only fixes the parity of the number of fermions but not the number itself. Moreover, our Hamiltonian contains terms which not only create/annihilate two particles, e.g. $|x_i\rangle \leftrightarrow |0\rangle$, but also terms which preserve the total number of fermions while changing the number of each of the three individual fermionic flavors, e.g. $|x_i\rangle \leftrightarrow |y_i\rangle$. Although it is possible to write down a projected wave functions with varying particle number [4], we refrain from doing so in this work for the sake of computational simplicity. Instead, we consider a restrictive form for the ground state wave function: each one of the four states is equally distributed over $N/4$ distinct sites, with $\{x_i\} = \{x_1, x_2, \dots, x_{N/4}\}$, etc.

The mean-field Hamiltonian in the main text may be rewritten, in terms of the three Dirac fermions in Eq. (32), as

$$\begin{aligned} \mathcal{H}_{\text{MF}} = & -\frac{NJ}{6} + \frac{J}{36} \sum_{\langle jl \rangle \in \alpha} \left[(3u_{jl}^2 + 3\bar{w}_{jl}^\alpha u_{jl} - w_{jl}^\alpha v_{jl}) + i(4u_{jl} + 2\bar{w}_{jl}^\alpha) d_j^\dagger d_l \right. \\ & \left. + i(2u_{jl} + \bar{w}_{jl}^\alpha - w_{jl}^\alpha) c_j^\dagger c_l + i(2u_{jl} + \bar{w}_{jl}^\alpha + w_{jl}^\alpha) c_j^\dagger c_l^\dagger + i(3u_{jl} - v_{jl}) f_j^\dagger f_l + \text{h.c.} \right] \\ & - \frac{J}{36} \sum_{\langle jl \rangle \in XY} \left[i(3u_{jl} + v_{jl}) f_j^\dagger f_l^\dagger + \text{h.c.} \right] + \frac{J}{36} \sum_{\langle jl \rangle \in YZ} \left[i(3u_{jl} e^{i\pi/3} + v_{jl} e^{-2i\pi/3}) f_j^\dagger f_l^\dagger + \text{h.c.} \right] \\ & + \frac{J}{36} \sum_{\langle jl \rangle \in XZ} \left[i(3u_{jl} e^{i\pi/3} - v_{jl} e^{-2i\pi/3}) f_j^\dagger f_l^\dagger + \text{h.c.} \right]. \end{aligned} \quad (34)$$

The three fermion flavors are decoupled and we may thus write the mean-field wave function as a product of three Slater determinants. Thus, after the Gutzwiller projection, we obtain

$$\Psi(\{x_i\}, \{y_j\}, \{z_m\}) = \Phi_d(\{x_i\}, \{y_j\}) \cdot \Phi_f(\{y_j\}, \{z_m\}) \cdot \Phi_c(\{z_m\}, \{x_i\}). \quad (35)$$

The d -fermion sector of the Hamiltonian in Eq. (34) corresponds to free fermions and thus their mean-field ground state is obtained by filling up the states with negative energy. For the c and f -fermions we have a BCS-like Hamiltonian instead and their ground state is given by the vacuum of their respective Bogoliubov quasiparticles [1]. The different status of the d fermion is expected from symmetry: the hidden global SU(2) symmetry of the original Hamiltonian implies the global U(1) symmetry corresponding to the conservation of the total number of Dirac fermions defined by a combination of η Majorana fermions. On the other hand, there is no continuous symmetry associated with θ Majorana fermions; as a result, the total number of c and f fermions is not conserved. The need to work with BCS-type wave functions in our case should be contrasted with the case of SU(4) symmetric models [3], where the SU(4) symmetry implies the conservation of the numbers of all three flavors of Dirac fermions.

After constructing the mean-field wave function, we then implemented a variational Monte Carlo calculation of the Gutzwiller-projected ground state energy $E_{\text{gs}}^{\text{VMC}}$. We started by generating an initial state in which we populate $N/4$ randomly chosen sites with the x -state ($\{x_i\}$), then $N/4$ of the remaining sites with the y -state ($\{y_i\}$), and finally $N/4$ of the further remaining sites with the z -state ($\{z_i\}$). Our Monte Carlo moves consist in exchanging random pairs of sites containing distinct states. We allow for moves involving widely separated sites — and which would not be connected by the Hamiltonian — because this improves the sampling over the space of configurations. We accept or reject these moves according to the usual Metropolis algorithm. After N such exchange attempts, we are said to have performed one Monte Carlo sweep and after every sweep we compute $E_{\text{gs}}^{\text{VMC}}$. N_{warm} sweeps are performed before measurements of physical quantities for “thermalization”. Averages are then performed over N_{sweep} sweeps. We typically considered $N_{\text{warm}} = N_{\text{sweep}} \sim 10^4$. The results were obtained for lattices of size $N = 4L^3$ with $L = 4, 6, \text{ and } 8$. We find that the change in the ground state energy with N is smaller than the Monte Carlo error bars for the system sizes considered here. Thus, we quote the results for $L = 8$ as the converged ones.

We computed the ground state energy for the two sets of mean-field parameters quoted in this supplemental material. For $u = v = 0.258$, $\bar{w} = 0.317$, and $w = -0.081$ we obtain $E_{\text{gs}}^{\text{VMC}} = -0.39(1) NJ$. As for $u = -v = 0.258$, $\bar{w} = 0.318$, and $w = 0.161$ we obtain $E_{\text{gs}}^{\text{VMC}} = -0.40(1) NJ$. Clearly, the Gutzwiller projection decreases significantly the mean-field energy down to values which are already comparable to that obtained, for instance, for a valence-bond covering of the lattice ($E_{\text{VBS}} = -0.417NJ$) [5], thus showing that the proposed chiral spin-orbital liquid is a competitive ground state candidate.

In light of this favorable energy of our proposed ansatz, we conclude by pointing out two important restrictions in our variational Monte Carlo calculation that, once lifted, should further decrease the value of the ground state energy $E_{\text{gs}}^{\text{VMC}}$:

1. For the quoted values of $E_{\text{gs}}^{\text{VMC}}$, we considered the optimal values for the mean-field amplitudes v , \bar{w} and w obtained *before* the Gutzwiller projection, i.e., at the mean-field level;
2. The restrictive form of the considered wave function neglects variations both in the populations of the fermionic flavors and in the total number of fermions.

We stress that these restrictions were important for this first calculation beyond mean field due to the complexity of the chiral spin-orbital liquid ansatz considered here. We leave a more detailed investigation, together with a more precise estimate for the variational energy of our spin liquid ansatz, for future work.

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