Motivated by the recent discovery of high temperature antiferromagnet SrRu$_2$O$_6$ [1, 2] and its potential to be the parent of a new superconductor upon doping, we construct a minimal $t_{2g}$-orbital model on a honeycomb lattice to simulate its low energy band structure. Local Coulomb interaction is taken into account through both random phase approximation and mean field theory. Experimentally observed antiferromagnetic order is obtained in both approximations. In addition, our theory predicts that the magnetic moments on three $t_{2g}$-orbitals are non-collinear as a result of the strong spin-orbit coupling of Ru atoms.

I. INTRODUCTION

Magnetism and superconductivity are closely related to each other, as a common thread in several families of unconventional superconductors. [3] Singlet Cooper pairing is expected to be mediated by antiferromagnetic (AF) fluctuations near the AF phase boundary like in most cuprates [4], iron-based [5], and heavy fermion [6] superconductors. While triplet Cooper pairing is widely believed to be triggered by ferromagnetic fluctuations near the AF phase boundary like in most cuprates [4]. In parallel, SrRu$_2$ lattice revealed Wigner crystallization [14] and anomalous quantum Hall effect [15, 16]. In parallel, SrRu$_2$O$_6$ provides us a natural realization of the $t_{2g}$ d-orbital on the honeycomb lattice.

In this paper, we first derive an effective $t_{2g}$-orbital tight-binding Hamiltonian as a minimal model for SrRu$_2$O$_6$. We then consider the correlation effect through both random phase approximation (RPA) and mean field theory. We obtain the experimentally observed AF order and estimate the Neel moment and transition temperature within the mean field theory. Furthermore, we find the orbital-resolved AF moments on three $t_{2g}$-orbitals are non-collinear as a result of the strong spin-orbit coupling (SOC) on Ru atoms. Our minimal model provides the basis for further theoretical studies, and the magnetic structure we uncovered would trigger further experimental interests in this new member of the ruthenate family.

II. $t_{2g}$-ORBITAL TIGHT-BINDING

HAMILTONIAN

In SrRu$_2$O$_6$, the low energy bands mainly come from the $t_{2g}$-orbitals of Ru atoms as reported by first-principle calculations. [2, 12] The Ru atoms form a honeycomb lattice in each RuO$_6$ layer. Therefore, to mimic the system by a minimal model, we only consider the $t_{2g}$-orbital model on a honeycomb lattice: application to antiferromagnet SrRu$_2$O$_6$

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electrons on a honeycomb lattice as shown in Fig. 1. The coordinate is set up with the origin at Ru-site and three axes point to three oxygen atoms above the Ru-plane in the undistorted RuO$_6$ octahedron. In this coordinate, the c-axis perpendicular to Ru-plane is along (1,1,1)-direction, and the $d_{xy}$, $d_{xz}$ and $d_{yz}$ orbitals forms an exact $t_2g$ multiplet, as schematically represented in Fig. 1. The blue/yellow/red lobes lie within/above/below the Ru-plane. Although in SrRu$_2$O$_6$, the RuO$_6$ octahedron is slightly twisted around and stretched along the c-axis, the $t_{2g}$-orbital degeneracy is protected by the $C_{3v}$ symmetry. As a result, the coordinate established in the undistorted octahedron will be used in this work.

To construct a tight-binding Hamiltonian, we keep nearest neighbor hopping $t_{1,4}$, next nearest neighbor hopping $t_{5,8}$, and the third-nearest hopping $t_{1'4'}$. All these hopping elements are schematically shown in Fig. 1. We notice that $t_5 \neq t_5'$ and $t_8 \neq t_8'$ due to the distortion of the RuO$_6$ octahedra. Furthermore, we add intra- and inter-orbital on-site energies $V_0$ and $V_0'$, respectively. In a pure two dimensional model, $V_0'$ should be exactly zero due to the orthogonality of different orbitals. But here we are looking for an effective two dimensional model in the $k_z = 0$ plane and thus the inter-layer hopping could lead to an effective on-site inter-orbital mixing $V_0'$. In addition, we add an SOC term $H_{SOC} = -\lambda \sum_{\mu} \psi_{i}^\dagger L_{\mu} \otimes \sigma_{\mu} \psi_{i}$, where $\psi_{i} = \{d_{i,xy}, d_{i,xz}, d_{i,yz}, d_{i,xy}, d_{i,xz}, d_{i,yz}, \}$, $\sigma_{\mu} = \{\sigma_{x}, \sigma_{y}, \sigma_{z} \}$ are three Pauli’s matrices, and $L_{\mu} = [L_{x}, L_{y}, L_{z}]$ are rank-3 angular momentum matrices acting on orbital space, with all nonzero elements given by

$$
L_{x,12} = -L_{x,21} = -i, \quad L_{y,13} = -L_{y,31} = -i,
L_{z,23} = -L_{z,32} = i.
$$

(1)

With the above ingredients and for a given inter-layer momentum $k_z = 0$, we arrive at an effective two-dimensional tight-binding Hamiltonian $H = \sum_{k_{\perp}} \psi_{i,\kappa}^\dagger H_{k} \psi_{i}$ in the basis $\psi_{i}^\dagger = [\psi_{i}^{A\kappa}, \psi_{i}^{B\kappa}, \psi_{i}^{A\kappa'], \psi_{i}^{B\kappa} ]$ where $\psi_{i}^{A,B}(d_{i,xy}, d_{i,xz}, d_{i,yz}, d_{i,xy}, d_{i,xz}, d_{i,yz}, )$ for $s = A, B$ on the two sublattices. The matrix $H_{k}$ is explicitly written as

$$
H_{k} = \begin{bmatrix}
T_{AA}(k) + V - \lambda L_z & T_{AB}(k) & T_{BB}(k) + V - \lambda L_z & -\lambda L_x + i\lambda L_y \\
-\lambda L_x - i\lambda L_y & 0 & -\lambda L_x - i\lambda L_y & -\lambda L_x + i\lambda L_y \\
0 & T_{AB}(k)^\dagger & T_{BB}(k) + V + \lambda L_z & 0 \\
-\lambda L_x + i\lambda L_y & 0 & T_{AB}(k) & T_{BB}(k) + V + \lambda L_z
\end{bmatrix},
$$

(2)

in which

$$
T_{AB}(k) = \begin{bmatrix}
t_1 & t_4 & t_4 \\
t_4 & t_2 & t_3 \\
t_4 & t_3 & t_2
\end{bmatrix} e^{ik a_1} + \begin{bmatrix}
t_2 & t_3 & t_4 \\
t_3 & t_1 & t_4 \\
t_3 & t_4 & t_1
\end{bmatrix} e^{ik a_2} + \begin{bmatrix}
t_2 & t_4 & t_3 \\
t_4 & t_1 & t_3 \\
t_4 & t_3 & t_1
\end{bmatrix} e^{ik a_3},
$$

$$
T_{AA}(k) = \begin{bmatrix}
t_7 & t_8 & t_7 \\
t_8 & t_6 & t_6 \\
t_8 & t_6 & t_6
\end{bmatrix} e^{ik (a_2-a_3)} + \begin{bmatrix}
t_6 & t_5 & t_5 \\
t_5 & t_7 & t_7 \\
t_5 & t_7 & t_7
\end{bmatrix} e^{ik (a_3-a_1)} + \begin{bmatrix}
t_6 & t_5 & t_5 \\
t_5 & t_7 & t_7 \\
t_5 & t_7 & t_7
\end{bmatrix} e^{ik (a_1-a_2)} + h.c.,
$$

$$
T_{BB}(k) = \begin{bmatrix}
t_7 & t_8 & t_7 \\
t_8 & t_6 & t_6 \\
t_8 & t_6 & t_6
\end{bmatrix} e^{-ik (a_2-a_3)} + \begin{bmatrix}
t_6 & t_5 & t_5 \\
t_5 & t_7 & t_7 \\
t_5 & t_7 & t_7
\end{bmatrix} e^{-ik (a_3-a_1)} + \begin{bmatrix}
t_6 & t_5 & t_5 \\
t_5 & t_7 & t_7 \\
t_5 & t_7 & t_7
\end{bmatrix} e^{-ik (a_1-a_2)} + h.c.,
$$

(3)

and

$$
V = \begin{bmatrix}
V_0 & V_0' & V_0' \\
V_0' & V_0 & V_0' \\
V_0' & V_0' & V_0
\end{bmatrix},
$$

(4)

where $(a_1, a_2, a_3)$ are three displacements from an A-site to its nearest neighbour B-sites. All the model parameters are determined by fitting the first-principle band structure [12]: (in unit of eV) $t_1 = 0.16$, $t_2 = -0.01$, $t_3 = 0.30$, $t_4 = -0.02$, $t_5 = -0.10$, $t_6 = 0.01$, $t_7 = -0.02$, $t_8 = 0.02$, $t_9 = -0.04$, $t_{10} = 0.01$, $t_{11} = 0.01$, $t_{12} = -0.01$, $V_0 = -0.09$, $V_0' = -0.07$, $\lambda = 0.16$. In particular, we obtain the SOC strength $\lambda = 0.16$eV, which is in agreement with the literature [17]. Based on these parameters, the band structure of our minimal model is plotted in Fig. 2.

In the paramagnetic state SrRu$_2$O$_6$ is a band insulator, as seen in our band structure and the first-principle
result [2, 12]. However, a strong Hund’s coupling would bind up the electrons to form a spin-3/2 state. Due to the bipartite lattice an AF order with moment 3µB/Ru is expected. Such an AF order has already been observed by Neutron scattering experiment in SrRu2O6 [2] but the observed moment is only 1.3µB/Ru, much smaller than 3µB. This indicates the inadequacy of a naive local moment picture. Instead, the itinerant property of electrons and SOC may play important roles. In the following, we will investigate the effect of correlation and SOC on the AF order in the itinerant picture of the t2g-orbital model.

III. INTERACTION AND ANTIFERROMAGNETISM

We adopt general multi-orbital local Coulomb interactions as follows,

\[ H_I = \sum_{I} \left[ U \sum_a n_{ia \uparrow} n_{ia \downarrow} + V \sum_{a>b} n_{ia} n_{ib} + J \sum_{a>b, \sigma \sigma'} a_{ia \sigma}^\dagger b_{ia \sigma} b_{ib \sigma'} a_{ib \sigma'} \right], \]

(5)

where \( n_{ia} = \sum_{\sigma} n_{ia \sigma} = \sum_{\sigma} a_{ia \sigma}^\dagger a_{ia \sigma} \). \( U \) is the Hubbard interaction, \( V \) is the inter-orbital charge interaction, \( J \) is the Hund’s coupling and \( J' \) is the pair hopping term. These four interactions satisfies the relation \( J' = J \) and \( U = V + 2J \). [18] Among these four terms, only \( U \) and \( J \) are responsible for magnetic channel instabilities. [19] So in the following discussions, we will only retain the \( U \) and \( J \) terms.

Since the non-interacting model is a band insulator, the bare susceptibility only depends on momentum very weakly. So we perform an RPA level calculation instead, since RPA will pick out relevant channels and strongly enhance their susceptibilities.

The interactions (\( U \) and \( J \) terms) are first written in magnetic channels: \(-S_I \hat{\Gamma}(I; J) S_{IJ}, \) where \( I = (a \mu n) \) and \( J = (b \nu m) \) with the orbital index \( (a/b), \) spin-direction index \( (\mu/\nu), \) and sublattice index \( (n/m) \). The spin operator is \( S_{I=(a\mu n)} = \frac{1}{2} \sum_{\alpha \beta} a_{\alpha \mu n}^\dagger \sigma_{\alpha \beta}^\dagger a_{\beta \mu n} \), and the vertex function \( \hat{\Gamma}(a \mu n; b \nu m) \) represented by Fig. 3(a) is diagonal in both spin-direction and sublattice subspaces and is given by

\[ \hat{\Gamma}(a \mu n; b \nu m) = \begin{cases} 2U \delta_{\mu \nu} \delta_{nm}, & a = b \\ 2J \delta_{\mu \nu} \delta_{nm}, & a \neq b \end{cases}. \]

(6)

The RPA is a bubble summation as represented in Fig. 3(b). After solving the iterate equation we obtain the magnetic susceptibility matrix as

\[ \hat{\chi}(q, i\nu_n) = \left[ 1 - \hat{\Gamma}(q, i\nu_n) \right]^{-1} \hat{\chi}^{(0)}(q, i\nu_n). \]

(7)

\( \hat{\chi}^{(0)}(q, i\nu_n) \) is the bare susceptibility whose matrix element is defined as

\[ \chi_{I,J}^{(0)}(q, i\nu_n) = \int_0^{1/T} \langle S_I(-q, \tau) S_J(q, 0) \rangle e^{i\nu_n \tau} d\tau. \]

(8)

Here, we have used Matsubara frequency with \( T \) the temperature.

We use the interaction parameters \( U = 1.35eV \) and \( J = 0.14eV \), being half of the values obtained by first principle calculation [2], since RPA is known to overestimate the magnetic instability. We plot the leading (largest) eigenvalue \( \Lambda \) of the hermitian susceptibility matrix \( \hat{\chi}(q, 0) \) as a function of \( q \) in Fig. 4. The peak at \( q = 0 \) implies a magnetic instability that is periodic across the unit cell. The corresponding leading eigenvector decides the form factor of the magnetic order, namely the magnetic structure within a unit cell. This is shown in Fig. 5(a). We find the total moment (of the three orbitals) is along the c-axis and changes sign from one to
the other sublattice within the unit cell. This is exactly the AF order seen in the neutron scattering experiment. More interestingly, the moments on the three orbitals are non-collinear about the c-axis. This is a prediction of the present work.

Next, we employ mean field theory to quantitatively investigate the AF moment and the transition temperature. The interactions are decoupled in magnetic channels:

\[ U_{\pi\alpha\pi\alpha'} \rightarrow \frac{-2U}{3} \langle S_{\pi\alpha}\rangle \cdot S_{\pi\alpha} \]

\[ \rightarrow -4U \frac{S_{\pi\alpha}}{3} \cdot S_{\pi\alpha} + \frac{2U}{3} \langle S_{\pi\alpha}\rangle \cdot \langle S_{\pi\alpha}\rangle, \tag{9} \]

and

\[ J \sum_{\sigma\sigma'} a_{\sigma\alpha}^\dagger a_{\sigma\alpha'}^\dagger a_{\sigma\alpha} \rightarrow -2JS_{\pi\alpha} \cdot S_{\pi\beta} \]

\[ \rightarrow -2\langle S_{\pi\alpha}\rangle \cdot S_{\pi\beta} - 2JS_{\pi\alpha} \cdot \langle S_{\pi\beta}\rangle + 2J \langle S_{\pi\alpha}\rangle \cdot \langle S_{\pi\beta}\rangle. \tag{10} \]

Then we obtain the mean field Hamiltonian

\[ H_{MF} = H_0 - \frac{4U}{3} \sum_{\pi\pi'} \langle S_{\pi\alpha}\rangle \cdot S_{\pi\alpha} \]

\[ -2J \sum_{\pi\pi',\alpha\neq\beta} \langle S_{\pi\alpha}\rangle \cdot S_{\pi\beta}, \tag{11} \]

where \( H_0 \) is the non-interacting part, which is the same as Eq. 2 but written in real space. The order parameters \( \langle S_{\pi\alpha}\rangle = \frac{1}{2} \sum_{\alpha\beta} \langle a_{\alpha\beta}^\dagger a_{\alpha\beta} \rangle \) are then numerically solved iteratively until convergence is achieved.

Our mean field result confirms the non-collinear AF configuration [Fig. 5(a)] revealed by RPA in the normal state. We have performed unrestricted mean field calculations starting from random initial spin configurations. No translation symmetry is assumed in advance. However, the results all converge to the same non-collinear AF configuration up to a shift of the sublattice. The magnetic unit cell is always found to be equal to the lattice unit cell, which is in agreement with the unique peak \( q = 0 \) in the leading eigenvalue of the RPA magnetic susceptibility in the momentum space as shown in Fig. 4. The non-collinear AF configuration is the result of the strong SOC on Ru atoms. The values of the total moment \( M \) and the deviation angle \( \theta \) vs temperature within the mean field approximation. (a) The AF configuration is schematically shown within a unit cell, with three orbital resolved moment represented by three different colors. The total moment is along c-axis [(1,1,1)-direction]. (b) The total AF moment \( M \) and the deviation angle \( \theta \) vs temperature within the mean field approximation.
can be observed in more delicate experiments like orbital-selective nuclear magnetic resonance\cite{20, 21} through the anisotropic hyperfine interaction \cite{22} or orbital-resolved angle-resolved photoemission spectroscopy\cite{23} through the photon polarization selection rule \cite{24}.

**IV. SUMMARY AND FUTURE WORKS**

In summary, we have constructed a $t_{2g}$-orbital model on a honeycomb lattice. Local Coulomb interaction was investigated in both RPA and mean field theory. Experimentally observed Neel order is obtained. Furthermore, our theory predicts that the magnetic moments on three orbitals are non-collinear as a result of the strong spin-orbit coupling of Ru atoms. This particular kind of orbital-resolved AF order is expected to be observed in future experiments.

For future works, possible superconductivity in this compound after doping or under pressure is an interesting direction. Our $t_{2g}$-orbital model can be used as a minimal model to study possible superconductivity upon doping. The AF fluctuation may induce singlet Cooper pairing between the nearest neighbours. However, due to the strong SOC, triplet pairing may coexist with the singlet pairing. This part of work is being in progress.

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