Exotic Cooper pairing in multi-orbital models of Sr$_2$RuO$_4$

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(Dated: May 14, 2019)

The unconventional superconductivity in Sr$_2$RuO$_4$ continues to defy a unified interpretation. In this paper, we focus on some novel aspects of its superconducting pairing by exploiting the orbital degree of freedom in this material. The multi-orbital nature, combined with the symmetry of the orbitals involved, leads to a plethora of exotic Cooper pairings not accessible in single-orbital systems. In the presence of finite spin-orbit coupling (SOC), spin-singlet and spin-triplet pairings are entangled, thus the spin susceptibility is generically expected to be suppressed below $T_c$. Essential physics is illustrated first using a two-orbital model with $d_{xz}$- and $d_{yz}$-orbitals. We classify the gap functions according to the underlying lattice symmetries, analyze the effective theories of a few representative pairings, and make connections to Sr$_2$RuO$_4$ in the course. For completeness, the classification is generalized to the three-orbital model involving the $d_{xy}$-orbital as well. The orbital-basis approach distinguishes from the itinerant-band description for Sr$_2$RuO$_4$, and hence offers an alternative perspective to investigate its enigmatic superconducting state.

I. INTRODUCTION

Superconductivity in Sr$_2$RuO$_4$ was discovered a quarter of a century ago. Widely hailed as an archetypal unconventional superconductor, no consensus is yet available regarding its pairing symmetry. Indications of spin-triplet pairing with spontaneous time-reversal symmetry breaking (TRSB) were reported in a series of earlier measurements. Taken together, they point to a chiral $p$-wave order with $d$-vector $(k_x + ik_y)\hat{z}$, which may exhibit nontrivial topology and host exotic excitations such as Majorana zero modes. Such a pairing is also supported by a number of other measurements. However, this interpretation also stands at odds with a variety of signatures not easily reconcilable with this chiral $p$-wave pairing, including the indications of nodal excitations, the absence of spontaneous surface current, and the anomalous behavior under in-plane magnetic fields and in-plane uniaxial strains. The out-of-plane $d$-vector orientation is further challenged by a recent net magnetic fields. Thus far, we still lack a pairing state that is able to coherently interpret all of the key experiments. It is hence sensible to both examine the existing theories and assumptions, and to search for alternative superconducting pairings that may ultimately bring a unified understanding.

Sr$_2$RuO$_4$ is a prominent material whose Fermi surface is considered relevant to Cooper pairing. The resultant superconductivity, in one way or another, is driven by spin or charge fluctuations reminiscent of the celebrated Kohn-Luttinger mechanism. The gap classification in the corresponding band basis is relatively straightforward. In the presence of finite SOC, spins are no longer good quantum numbers. Nonetheless, an effective pseudospin basis can be adopted, thanks to the conservation of the Kramers degeneracy in the Bloch bands. An alternative approach is the orbital-basis description. In this case Cooper pairs are formed by electrons with well-defined orbital characters. Although a corresponding full-fledged symmetry classification is lacking, many existing studies on the phenomenology of the superconducting Sr$_2$RuO$_4$ are constructed on the multi-orbital basis (e.g. some recent studies in Refs. 42–46). When transformed into band basis, the state typically allows for interband pairing, which is crucial for the appearance of the intrinsic anomalous Hall effect (which leads to Kerr rotation) below $T_c$ in a multiband chiral $p$-wave superconductor.

There are without doubt marked distinction between the band- and orbital-basis approaches. As we shall see in the present study, the latter exhibits a rich variety of exotic superconducting pairings. We illustrate this using a toy two-body model.

![FIG. 1: Top view of the $xz$, $yz$, and $xy$-orbitals on a 2D square lattice.](image-url)
II. SINGLE-PARTICLE HAMILTONIAN AND GAP CLASSIFICATION

To make connection with Sr$_2$RuO$_4$, we take a two-orbital model with $d_{xz}$ and $d_{yz}$ orbitals residing on the each site of a square lattice (see Fig. 1 for illustration). The model contains no sublattice degree of freedom. In two spatial dimensions (2d), the model also applies to systems of $p_x$ and $p_y$ orbitals. It is instructive to first construct a continuum model Hamiltonian that respects both time-reversal and the $D_{4h}$ point group symmetries. In the spinor basis $(c_{xz}^\dagger, c_{x'y}, c_{y'z}^\dagger, c_{zz})^T$,

$$H_{0k} = t(k_x^2 + k_y^2) - \mu + t'(k_x^2 - k_y^2)\sigma_z + \eta'k_xk_y\sigma_x + \eta\sigma_y \otimes s_z,$$

where $\sigma_i$ and $s_i$ with $i = x, y, z$ are the Pauli matrices operating on the respective orbital and spin degrees of freedom, $(t, t', \eta')$ designate the kinetic energy and $\eta$ the onsite spin-orbit coupling (SOC). This Hamiltonian is manifestly invariant under time-reversal, $\mathcal{T} = \sigma_0 \otimes is_y K$, where $K$ denotes complex conjugation. It is also consistent with the tight-binding construction in previous studies.

To see how Eq. (1) respects $D_{4h}$, it is important to recognize that the point group operations must act jointly on spatial, spin and orbital degrees of freedom. This involves varying the phase (gauge) of the orbital wavefunctions under certain operations, due to the peculiar symmetry properties of the two orbitals. For example, a $C_4$ rotation, in addition to rotating momentum and spin, also exchanges the label of the two orbitals and induces a $\pi$ phase change on one of them, e.g. $(d_{xz}, d_{yz}) \rightarrow (d_{yz}, -d_{xz})$. As a consequence, the bilinear $\sigma$-operators, which are formally $c_{m,s}^\dagger \sigma_m^{m'c_{n,s'}} (m = xz, yz)$, transform according to irreps of $D_{4h}$ in the following fashion:

$$c_0, \sigma_x, \sigma_z, \quad \text{and} \quad \sigma_y \quad \text{as} \quad A_{1g}, B_{2g}, B_{1g} \quad \text{and} \quad A_{2g},$$

respectively. A Hamiltonian invariant under all $D_{4h}$ operations can then be conveniently constructed by ‘multiplying’ the $\sigma$-operators by their respective basis functions, as in Eq (1). Note that, amongst the terms in the Hamiltonian, $s_z$ transforms as $A_{2g}$. Further, since the orbital wavefunction of the $t_{2g}$-electrons are even under inversion, the only effect of inversion is to invert the electron momentum. This differs from the model with $p_x$ and $p_y$-orbitals, where inversion also changes the sign of the fermion creation and annihilation operators (the bilinear operators are however unaffected by this).

III. SINGLET-TRIPLET-MIXED EVEN PARITY $A_{1g}$-PAIRING

As one can see in Table III, there are multiple one-dimensional irreps which contain more than one symmetry equivalent components and permit mixtures of spin-triplet and spin-singlet pairings. We take an example a $A_{1g}$ gap function,

$$\hat{\Delta}_k = \psi_1 \hat{\Delta}_{1k} + \psi_2 \hat{\Delta}_{2k} = (\psi_1 i\sigma_y \otimes z \cdot s + \psi_2 i) s_y, \quad (2)$$

Table I: Representative basis functions of the superconducting pairing in the two-orbital model in the two dimensional $E_{0}$ and $E_{u}$ irreps of the $D_{4h}$ point group. Here $\sigma_i$ and $s_i$ operate in the orbital and spin space, respectively. The vectors $x, y$ and $z$ denote the direction of the $d$-vector of spin-triplet and odd-parity spin-singlet pairings. We analyze the phenomenology of these states and discuss their possible relation to Sr$_2$RuO$_4$. In particular, we show explicitly the influence of SOC on mixing spin-singlet and spin-triplet pairings in various superconducting channel. For completeness, we also perform a gap classification for the three-orbital model that takes into account the $d_{xy}$-orbital as well.

| irrep. | basis function |
|-------|----------------|
| $E_{0}$ | $(i\sigma_y \otimes x \cdot s, \quad i\sigma_y \otimes y \cdot s)$ |
| $E_{u}$ | $(i\sigma_y \otimes k_{x(y)} x \cdot s, \quad i\sigma_y \otimes k_{x(y)} y \cdot s)$ |

Tabled II and III list some of the representative superconducting basis functions in different irreps of the $D_{4h}$ group. We see that most of the individual irreps contain multiple symmetry-equivalent basis functions – a prominent feature not present in single-orbital systems. Note that, the spatial parity is a good quantum number in this system, and basis functions even and odd in $k$ will not mix, because the inversion operation only acts to invert the momentum $k$ while leaves orbital and spin degrees of freedom unchanged. This is quite different from systems with sublattice degree of freedom, such as a honeycomb lattice, where the gap functions may comprise components even and odd in momentum (overall inversion symmetry is nonetheless retained).
The triplet and singlet components correspond to inter- and intra-orbital pairings, respectively. In general, the two do not necessarily coexist in the absence of SOC – when spins are good quantum numbers. To understand how SOC induces mixed pairings, we perform a standard free energy expansion, \( f = \Delta^t \Delta/V + T \sum_{k,w} \text{Tr}[G(iw_n, k)\tilde{G}(iw_n, k)\Delta^t]/(2i) \) where \( G(iw_n, k) = (iw_n - H_n(k))^{-1} \) and \( \tilde{G}(iw_n, k) = (iw_n + H_{\sigma_{-k}})^{-1} \) are the electron and hole components of the Gorkov Green’s function. The singlet and triplet pairings are coupled at quadratic order,

\[
J_{12} = i\lambda_{12}(\psi_1^* \psi_2 - \psi_2^* \psi_1),
\]

with \( \lambda_{12} \propto \eta \) a real constant. The complex phase is a consequence of the particular structure of the SOC in Eq. \( \lfloor 1 \rfloor \). A similar conclusion was reached in Ref \( \lfloor 41 \rfloor \). Therefore, SOC not only mixes but also selects a particular relative phase between the two components, e.g. \( \theta_2 - \theta_1 = \pi/2 \) if \( \lambda_{12} > 0 \). The relative phase can be absorbed into the basis function. Thus a more compact form of Eq. \( \lfloor 2 \rfloor \) reads:

\[
\Delta_k \propto [\sigma_y \otimes (z \cdot s) + c] i s_y, \]

where \( \epsilon \) is a real constant determined by the details of the microscopic model. Notice there exists no ground state degeneracy, and such a pairing is time-reversal invariant (TRI), i.e. it satisfies \( T\Delta_k = -\Delta_k \). On the contrary, the pairings with relative phases of 0 and \( \pi \) between \( \psi_1 \) and \( \psi_2 \) are degenerate and violate time reversal symmetry. It is also worth stressing that, the spin susceptibility of such a mixed singlet-triplet pairing is expected to drop below \( T_c \), although typically not as drastic as that of a pure singlet state.

In like manner, the remaining two components of \( A_{1g} \) given in Table \( \lfloor 3 \rfloor \) \( \Delta_{3k} \equiv k_x k_y \sigma_x \otimes i s_y \) and \( \Delta_{4k} = (k_x^2 - k_y^2) \sigma_z \otimes i s_y \), also couple quadratically to the first two components, besides a coupling of the similar order between themselves. In full, the free energy up to the quadratic order reads,

\[
f_{2nd} = \sum_{j=1}^{4} \alpha_j |\psi_j|^2 + i \sum_{j=2}^{4} (\lambda_{1j} \psi_1^* \psi_j - c.c.) + (\lambda_{23} \psi_2^* \psi_3 + \lambda_{24} \psi_2^* \psi_4 + \lambda_{34} \psi_3^* \psi_4 + c.c.).
\]

All of the \( \alpha_j \) and \( \lambda_{1j} \)-coefficients are real. Like \( \lambda_{12} \), the other two coefficients that couple triplet and singlet pairings, \( \lambda_{13} \) and \( \lambda_{14} \), depend both on SOC. By contrast, the remaining coefficients, \( \lambda_{23}, \lambda_{24} \) and \( \lambda_{34} \), do not rely on SOC. Instead, these three couplings are induced by the \( \sigma_x \) and/or \( \sigma_z \) terms in Eq. \( \lfloor 1 \rfloor \) with \( \lambda_{23} \propto t''/t, \lambda_{24} \propto \bar{t}/t \) and \( \lambda_{34} \propto \bar{t}''/t^2 \). The sign of \( \alpha_3 \) determines whether an intrinsic Cooper instability exists for the corresponding pairing component. The most negative \( \alpha_3 \) typically signifies the most dominant component. A component that lacks Cooper instability \( (\alpha_3 > 0) \) may still be induced due to the effective proximity effects through the finite couplings in the following sense. The free energy can be minimized by taking the lowest-energy eigenvalues of the coupling matrix, with the basis defined by \( \tilde{\psi} = (\psi_1, \psi_2, \psi_3, \psi_4)^T \):

\[
f_{2nd} = \tilde{\psi}^\dagger \left[ \begin{array}{cccc}
\alpha_1 & i\lambda_{12} & i\lambda_{13} & i\lambda_{14} \\
-i\lambda_{12} & \alpha_2 & \lambda_{23} & \lambda_{24} \\
-i\lambda_{13} & -\lambda_{23} & \alpha_3 & \lambda_{34} \\
-i\lambda_{14} & -\lambda_{24} & -\lambda_{34} & \alpha_4
\end{array} \right] \tilde{\psi}. \]

In the single most favorable eigenstate, \( \psi_1 \) should acquire a relative phase of \( \pi/2 \) or \(-\pi/2 \) with respect to the remaining components. A general \( A_{1g} \) gap function, with all of the four components emerging simultaneously, is then given by,

\[
\Delta_k \propto \epsilon_1 \Delta_{1k} + i \epsilon_2 \Delta_{2k} + i \epsilon_3 \Delta_{3k} + i \epsilon_4 \Delta_{4k},
\]

where \( (\epsilon_1, i\epsilon_2, i\epsilon_3, i\epsilon_4) \) constitutes the lowest-energy eigenvector of the coupling matrix in Eq. \( \lfloor 5 \rfloor \). In reality, one or certain subset of the \( \epsilon_i \)'s may dominate, while the rest are induced. For example, since \( \Delta_{3k} \) and \( \Delta_{4k} \) both describe intra-orbital pairing and since orbital-mixing is secondary to the intra-orbital hoppings in \( \text{Sr}_{2}\text{RuO}_4 \), \( \epsilon_2 \) and \( \epsilon_4 \) could be much larger than the others.

### IV. SPIN-TRIPLET EVEN-PARITY \( E_g \)-PAIRING

In single-orbital models, the ordinary \( E_g \) pairing is even-parity and spin-singlet in nature, and it must involve out-of-plane pairing, taking the form of \( k_z (k_x, k_y) \). However, in the present two-orbital model, the simplest \( E_g \) pairing taken from Table \( \lfloor 4 \rfloor \) is a spin-triplet given by,

\[
\Delta_k = (\psi_x \cdot i\sigma_y \otimes x \cdot s + \psi_y \cdot i\sigma_y \otimes y \cdot s) i s_y,
\]

where the two order parameters \( \psi_x \) and \( \psi_y \) form a two-dimensional irreps. In essence, the two components each describes a spin-triplet inter-orbital \( s \)-wave pairing. A Ginzburg-Landau free energy can be constructed on symmetry basis or through a straightforward expansion, which leads to

\[
f = k_1 \left( |\partial_x \psi_x|^2 + |\partial_y \psi_x|^2 \right) + k_2 \left( |\partial_x \psi_y|^2 + |\partial_y \psi_y|^2 \right) + \alpha \left( |\psi_x|^2 + |\psi_y|^2 \right)^2 + \beta \left( |\psi_x|^4 + |\psi_y|^4 \right) + \beta_{xy} |\psi_x|^2 |\psi_y|^2 + \beta' \left( |\psi_x^s \psi_x|^2 + |\psi_y^s \psi_y|^2 \right) + \cdots,
\]

where ‘\( \cdots \)’ denotes higher order terms. Note that because \( \psi_{x,y} \) are both under inversion, cross-gradient terms such as \( \partial_x \psi_x^s \partial_y \psi_y \) are disallowed. Likewise, \( \partial_x \psi_y^s \partial_y \psi_x \), \( \partial_y \psi_y^s \partial_y \psi_x \) and their complex conjugates are forbidden, as \( \psi_x \) and \( \psi_y \) exhibit opposite mirror eigenvalues about the \( xz \) (and \( yz \)) planes. Dependent on the sign of \( \beta', \) two types of superconducting phases are possible, one preserving and the other breaking time reversal symmetry. When \( \beta' > 0, \) the two components preferentially develop a relative phase of \( \pm \pi/2, \) leading to a TRSB pairing; whereas a relative phase of \( 0 \) or \( \pi \) is favored if \( \beta' < 0, \) which corresponds to a time-reversal invariant (TRI) state.

A TRSB pairing may support spontaneous current at the surface or around defects. Within Ginzburg-Landau theory, it is been well understood that the forbidden gradient terms
mentioned above would have been crucial for the existence of spontaneous 
chiral d-wave pairing with $\Delta_k \sim (k_x + ik_y)k_z$, the present 
TRSB $E_g$ pairing (when appears alone) has the salient fea-
ture that it is free of surface current. On the other hand, the 
system may exhibit superconducting domain walls separating 
regions of distinct TRSB pairings, and the neighboring cor-
ners of such domain walls carry opposite fractional quantum 
fluxes analogous to the scenario in a coupled anisotropic XY-
model. The resultant internal field distribution could be 
detected in $\mu$SR measurements. Notably, fractional vortices 
could still emerge even when the pairing is TRSB.

V. SINGLET-TRIPLET-MIXED ODD-PARITY $E_u$-PAIRING

We write down in Table II four of the simplest basis func-
tions belonging to the $E_u$ irrep. Among them, the first com-
ponent is the only singlet pairing, and the third one is re-
related to the proposal of $p$-wave instability on the quasi-1D 
component is the only singlet pairing, and the third one is re-
related to the proposal of $p$-wave instability on the quasi-1D 

$$f_{2nd} = \sum_{i=1}^{4} \sum_{\mu=x,y} \alpha_{1i} |\psi_{1\mu}|^2 + \sum_{\mu \neq \nu} [\lambda_{23} \bar{\psi}_{2\mu}^{*} \psi_{3\mu} + \lambda_{24} \bar{\psi}_{2\mu}^{*} \psi_{4\mu} + \lambda_{34} \bar{\psi}_{3\mu}^{*} \psi_{4\mu} + c.c.] + \sum_{\mu \neq \nu} \i [\psi_{1\mu}^* (\lambda_{12} \bar{\psi}_{2\mu} + \bar{\psi}_{3\mu} + \lambda_{14} \psi_{4\mu}) - c.c.]$$ (9)

where all $\lambda_{ij}$ are real quantities. In particular, $\lambda_{12}, \lambda_{13}, \lambda_{14} \propto \eta$, demonstrating once again that SOC couples the singlet to 
the triplet pairings. The couplings between the triplet pairings 
(i.e. $\lambda_{23}, \lambda_{24}, \lambda_{34}$) does not require finite SOC but other ingre-
dients such as the inter-orbital hybridization $\epsilon'$. The entangle-
ment of $\psi_3$ with the others deserves a special note: each of the 
individual component $\psi_{1\mu}$ only couples to an orthogonal 
component from $\psi_2, \psi_3$ and $\psi_4$. So the order parameters come 
conveniently in two groups, $\psi_a = (\psi_{1x}, \psi_{2y}, \psi_{3y}, \psi_{4y})^T$ and 
\(\hat{\psi}_b\) with appropriate exchange of $x$ and $y$ indices. By analogy 
with the situation in the $A_{1g}$ channel, the ground state 
must have the relative phases between the components in 
$\psi_{a(b)}$ locked in a single most favorable configuration. De-
noting the two corresponding order parameters $\psi_{a(b)}$, the 
$E_u$ gap function is more properly expressed in an alternative 
two-component form: $\Delta_k = \Delta_{ak} + \Delta_{bk}$, with,

$$\Delta_{ak} = \psi_a \left( \epsilon_1 \Delta_{1kx} + i\epsilon_2 \Delta_{2ky} + i\epsilon_3 \Delta_{3kx} + i\epsilon_4 \Delta_{4ky} \right), \tag{10}$$
$$\Delta_{bk} = \psi_b \left( \epsilon_1 \Delta_{1ky} + i\epsilon_2 \Delta_{2kx} + i\epsilon_3 \Delta_{3kx} + i\epsilon_4 \Delta_{4kx} \right), \tag{11}$$

where $\epsilon_1, \ldots, \epsilon_4$ are real constants. This leads to the following free energy in powers of $\psi_a$ and $\psi_b$,

$$f = k_1 \left( |\partial_x \psi_a|^2 + |\partial_y \psi_b|^2 \right) + k_2 \left( |\partial_x \psi_b|^2 + |\partial_y \psi_a|^2 \right) + k_3 \left( \partial_x \psi_a \partial_y \psi_b + c.c. \right) + k_4 \left( \psi_a \partial_y \psi_b + c.c. \right) + \alpha \left( |\psi_a|^2 + |\psi_b|^2 \right) + \beta \left( |\psi_a|^4 + |\psi_b|^4 \right) + \beta_{ab}|\psi_a|^2|\psi_b|^2 + \beta' \left( |\psi_a|^2 |\psi_b|^2 \right) + \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \right) \tag{12}$$

Compared to the effective theory in Eq. 8, the cross-gradient terms with coefficients $k_3$ and $k_4$ are present, and they could generate finite spontaneous current if the pairing breaks time-
reversal symmetry.

VI. THREE-ORBITAL MODEL

In extending to a full three-orbital model, the Gall-Mann 
matrices $(T_i, i = 1, \ldots, 8)$ turn out to be convenient devices. 
We define $T_{11} = (T_0 + \sqrt{3} T_3)/2$ and $T_{33} = (T_0 - \sqrt{3} T_3)/4$, 
where $T_0$ is a $3 \times 3$ identity matrix. Using the orbital spinor 
basis $(c^{|m\uparrow\rangle}, c^{|m\downarrow\rangle})$ in the order $m = x, y, z, xy, yz, xz$, up to quadratic order in $k$ and with on-site SOC, the Hamiltonian reads,

$$H_{0k} = \left[ t(k_x^2 + k_y^2) - \mu \right] T_{11} + \left[ \epsilon' (k_x^2 - k_y^2) \right] T_3 + l' k_x k_y T_1 + [l' (k_x^2 + k_y^2) - \mu_{xy}] T_{33} + \eta (T_2 \otimes s_x + T_3 \otimes s_x + T_7 \otimes s_y), \tag{13}$$

where the $l'$ term and $\mu_{xy}$ denote the kinetic energy and chem-
ical potential of the $d_{xy}$ orbital. Note that $T_{1,2,3}$ are equivalent 
to $\sigma_{x,y,z}$, and $T_{11}$ to $\sigma_0$. Hence they inherit the transformation properties of the $\sigma_{\mu}$-operators. $T_{1,5}$ and $T_{6,7}$, on the other 
hand, transform respectively as the $B_{3g}$ and $B_{2g}$ irreps of the 
$D_{2h}$ group. However, the SOC term (the last term), having 
an appropriate linear superposition of $T_3$ and $T_7$, respects $D_{4h}$.

Without further elaboration, the gap functions, especially those not involving inter-orbital pairings with the $d_{xy}$ orbital, 
can be classified rather straightforwardly following the pre-
ceding analyses involving two orbitals. Inter-orbital pairings 
involve $d_{xy}$ which are associated with pairing operators $T_{3,4,7}$. 
As can be checked, $(T_4, T_6) \text{ and } (T_5, T_7)$ transform as $E_g$ 
($E_u$) irreps under $D_{4h}$. As a consequence, any such pairing must contain both $T_3$ and $T_6$ (or $T_5$ and $T_7$) in the gap 
function. This is demonstrated in Table III for the $E_g$ and $E_u$ 
pairings, as well as in Table IV for the one-dimensional ir-
reps. As an interesting note, two recent microscopic multi-
orbital calculations both found noticeable, or even domi-
nant, interorbital $E_u$ pairing involving the $d_{xy}$-orbital in some 
regimes of the interaction parameter space.

VII. SUMMARY AND DISCUSSIONS

With an eye on the yet-unresolved myth of the supercon-
ducting Sr$_2$RuO$_4$, we explored the possibilities made avail-
able by its multi-orbital degree of freedom. The supercon-
ducting pairings are classified on the basis of the Ru $t_{2g}$, 4$d$
TABLE II: Representative superconducting basis functions of the inter-orbital pairing involving the \( d_{xy} \)-orbital in the two dimensional \( E_y \) and \( E_u \) irreps. Here \( T_1 \) and \( s \) operate respectively in the orbital and spin space, as explained in the text.

| irrep. | basis function |
|-------|----------------|
| \( E_y \) | \( (T_4, s) \) |
| \( [ k_{x(y)}, T_4 \cdot k_{y(x)}^2 T_6 ] \) |
| \( (iT_5 \cdot z \cdot s; iT_6 \cdot z \cdot s) \) |
| \([iT_5 \cdot k_{x(y)} z \cdot s; iT_7 \cdot k_{y(x)}^2 z \cdot s] \) |
| \( (T_1 \otimes k_x x \cdot s; T_6 \otimes k_y y \cdot s) \) |
| \( (T_4 \otimes k_x y \cdot s; T_6 \otimes k_y x \cdot s) \) |
| \( (T_4 \otimes k_y y \cdot s; T_6 \otimes k_x x \cdot s) \) |
| \( (T_4 \otimes k_x x \cdot s; T_6 \otimes k_y y \cdot s) \) |

orbitals according to the underlying crystal point group symmetries. This leads to multiple exotic superconducting pairings not accessible in single-orbital or itinerant-electron models. In some cases, the phenomenology of the orbital-basis description could differ considerably from that of an itinerant-band description. We discussed some of their salient aspects and made connections to \( \text{Sr}_2\text{RuO}_4 \) in due course. As a special note, as spin-singlet and spin-triplet pairings are in general mixed due to the finite SOC in this material, its spin susceptibility is expected to drop below the superconducting transition.

Our main purpose is not to rule out or identify any pairing for \( \text{Sr}_2\text{RuO}_4 \), but rather to provide a new perspective to further explore the enigmatic superconductivity in this material. Hence we have restricted, for simplicity, to in-plane pairings in our symmetry classification of the multi-orbital superconductivity. Including out-of-plane couplings, i.e. extending the model to three spatial dimensions (3d), brings about numerous additional possibilities. In fact, even within the conventional band description, some novel forms of pairings may arise due to a 3d spin-orbital entanglement in the electronic structure. In particular, the \( E_u \) pairing is recently shown to be inherently three-dimensional\(^{23,26}\), containing both in-plane and out-of-plane pairings. This is unlike what has been typically assumed for quasi-2d models. More intriguingly, a 3d nematic \( E_u \) pairing, which can be realized if the out-of-plane pairing is sizable, was argued to explain a number of outstanding puzzles, such as the absence of surface current and the anomalous response to in-plane uniaxial strains\(^{61,62}\). Notably, since the d-vector of a 3d \( E_u \) pairing has both in-plane and out-of-plane components, a drop in the NMR Knight shift is expected for generic in-plane magnetic field orientations\(^{25}\). Additionally, models containing out-of-plane pairings have appeared in several other contexts\(^{63,65}\).

**Note added** - As this manuscript was being prepared for submission, a preprint appeared on arXiv\(^{66}\) with a similar idea to exploit the multi-orbital nature of the superconductivity in \( \text{Sr}_2\text{RuO}_4 \).

**Acknowledgments**

We would like to thank Fu-Chun Zhang and Qiang-Hua Wang for valuable discussions. This work is supported in part by the NSFC under grants No. 11825404 (W.H. and H.Y.), No. 11774306 and No. 11704106 (Y.Z.), the National Key Research and Development Program of China under grant No.2016YFA0300202 (Y.Z.) and No. 2016YFA0301001 (H.Y.), the Strategic Priority Research Program of Chinese Academy of Sciences under Grant No. XDB28000000 (Y.Z. and H.Y.), a startup grant at SUSTech and the C.N. Yang Junior Fellowship at Tsinghua University (W.H.).

**Appendix A: 1D representations of the gap functions**

Table II is a supplement to Table I in the text.

Table IV is a supplement to Table III in the text.

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TABLE III: Representative superconducting basis functions of the two-orbital model in the 1D irreps of the $D_{4h}$ point group.

| irrep. | basis function |
|-------|----------------|
| $A_{1g}$ | $i\sigma y \otimes z \cdot s$, $1$, $k_x k_y \sigma_z$, $(k_x^2 - k_y^2)\sigma_z$ |
| $A_{2g}$ | $k_x k_y \sigma_z$, $(k_x^2 - k_y^2)\sigma_z$ |
| $B_{1g}$ | $\sigma_z$, $i\sigma y \otimes (k_x^2 - k_y^2)z \cdot s$ |
| $B_{2g}$ | $\sigma_z$, $i\sigma y \otimes k_x k_y z \cdot s$ |
| $A_{1u}$ | $\frac{a_{\mu \nu}}{2} \otimes k_x x \cdot s + \frac{a_{\mu \nu} + s}{2} \otimes k_y y \cdot s$, $\sigma_z \otimes (k_x y + k_y x) \cdot s$ |
| $A_{2u}$ | $\frac{a_{\mu \nu}}{2} \otimes k_y y \cdot s - \frac{a_{\mu \nu} + s}{2} \otimes k_x x \cdot s$, $\sigma_z \otimes (k_y x - k_x y) \cdot s$ |
| $B_{1u}$ | $\frac{a_{\mu \nu}}{2} \otimes k_x x \cdot s + \frac{a_{\mu \nu} + s}{2} \otimes k_y y \cdot s$, $\sigma_z \otimes (k_x y - k_y x) \cdot s$ |
| $B_{2u}$ | $\frac{a_{\mu \nu}}{2} \otimes k_y y \cdot s + \frac{a_{\mu \nu} + s}{2} \otimes k_x x \cdot s$, $\sigma_z \otimes (k_x y + k_y x) \cdot s$ |

TABLE IV: Inter-orbital pairing superconducting basis functions involving the $d_{x^2-y^2}$-orbital in the 1D irreps of the three-orbital model. Here both $T_1$ and $T_2$ are components of the Gell-Mann matrices.

| irrep. | basis function |
|-------|----------------|
| $A_{1g}$ | $T_5 \otimes x \cdot s - T_7 \otimes y \cdot s$ |
| $A_{2g}$ | $T_5 \otimes y \cdot s + T_7 \otimes x \cdot s$ |
| $B_{1g}$ | $T_5 \otimes x \cdot s + T_7 \otimes y \cdot s$ |
| $B_{2g}$ | $T_5 \otimes y \cdot s - T_7 \otimes x \cdot s$ |

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