Exotic Cooper pairing in multiorbital models of Sr_2RuO_4

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The unconventional superconductivity in Sr_2RuO_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 multiorbital nature, combined with the symmetry of the orbitals involved, leads to a plethora of exotic Cooper pairings not accessible in single-orbital systems. 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_2RuO_4 in the course. In particular, we show how spin-orbit coupling may entangle spin-triplet and spin-singlet pairings. 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_2RuO_4 and hence offers an alternative perspective to investigate its enigmatic superconducting state.

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I. INTRODUCTION

Superconductivity in Sr_2RuO_4 was discovered twenty-five years ago [1]. Widely hailed as an archetypal unconventional superconductor, no consensus is yet available regarding its pairing symmetry [2–9]. Indications of spin-triplet [10,11], odd-parity [12] pairing with spontaneous time-reversal symmetry breaking (TRSB) [13,14] 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 [4–6,9]. However, this interpretation stands at odds with a variety of signatures not easily reconcilable with this chiral p-wave pairing [4–6,9], including the indications of nodal excitations [15,16], the absence of spontaneous surface current [17–19], and the anomalous behavior under in-plane magnetic fields [20–22] and in-plane uniaxial strains [23–25]. The out-of-plane d-vector orientation is further challenged by a recent observation of a Knight shift drop below T_c under in-plane magnetic fields [26]. 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_2RuO_4 has three Fermi sheets derived mainly from the Ru $4d$ t_{2g} orbitals [27,28]. As superconductivity appears to emerge from a coherent Fermi liquid [29], plenty of microscopic theories take an itinerant-electron perspective, in which only intraband superconducting pairing is active although multiple bands are considered [30–38]. In this setting, only electrons near the Fermi level are 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 [39]. The gap classification in the corresponding band basis is relatively straightforward [40]. In the presence of finite SOC, spins are no longer good quantum numbers. Nonetheless, an effective pseudospin basis can be adopted [35,38], thanks to the conservation of the Kramers degeneracy in the Bloch bands. An alternative approach is the orbital-basis description. In this description, Cooper pairs are formed by electrons with well-defined orbital characters [41]. Although a corresponding full-fledged symmetry classification is lacking, many existing studies on the phenomenology of the superconducting Sr_2RuO_4 are constructed on the multiorbital 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 [14]) below T_c in a multiband chiral p-wave superconductor [45–47].

There is without a doubt a 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-orbital model with the t_{2g} d_{xz} and d_{yz} orbitals. Similarly to some of the previous studies on the multiorbital iron-based superconductors [48–52], the gap functions are classified according to the underlying lattice point group symmetry. The orbital manifold

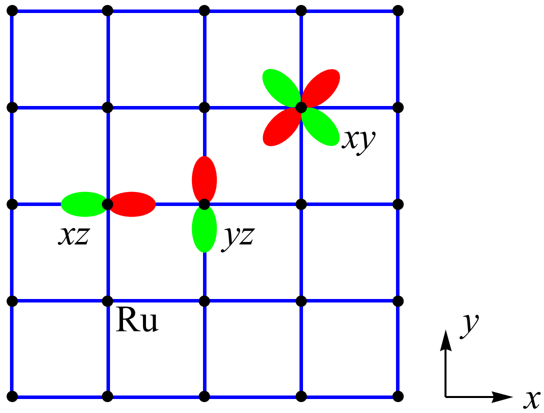

FIG. 1. Top view of the xz, yz, and xy orbitals on a 2D square lattice.

in Sr_2RuO_4 introduces numerous novel possibilities not available in single-orbital models, such as even-parity spin-triplet and odd-parity spin-singlet pairings. We analyze the phenomenology of these states and discuss their possible relation to Sr_2RuO_4 when necessary. In particular, we show explicitly the influence of SOC on mixing spin-singlet and spin-triplet pairings in various superconducting channels [53]. For completeness, we also present a gap classification for the three-orbital model that takes into account the d_{xy} orbital as well.

II. SINGLE-PARTICLE HAMILTONIAN AND GAP CLASSIFICATION

To make a connection with Sr_2RuO_4 , we take a two-orbital model with d_{xz} and d_{yz} orbitals residing on each site of a square lattice (see Fig. 1). 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 $(c_{xz\uparrow}, c_{xz\downarrow}, c_{yz\uparrow}, c_{yz\downarrow})^T$,

$$H_{0\boldsymbol{k}} = t\left(k_x^2+k_y^2\right) - \mu + \tilde{t}\left(k_x^2-k_y^2\right)\sigma_z + t'' k_x k_y \sigma_x + \eta\sigma_y \otimes s_z, \tag{1}$$

where σ_i and s_i with $i = x, y, z$ are the Pauli matrices operating on the respective orbital and spin degrees of freedom, $(t, \tilde{t}, t'')$ designate the kinetic energy, and η designates the onsite spin-orbit coupling (SOC). This Hamiltonian is manifestly invariant under time reversal, $\mathcal{T} = \sigma_0 \otimes is_y\mathcal{K}$, where $\mathcal{K}$ denotes complex conjugation. It is also consistent with the tight-binding construction in previous studies Refs. [35] and [38].

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 wave functions 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 π phase change on one of them, e.g., $(d_{xz}, d_{yz}) \to (d_{yz}, -d_{xz})$. As a consequence, the bilinear σ operators, which are formally $c^\dagger_{m,s}\sigma_i^{mn}c_{n,s'}$ ($m = xz, yz$), transform according to irreps of D_{4h} in the following

TABLE I. Representative basis functions of the superconducting pairing in the two-orbital model in various irreps of the D_{4h} point group. Here σ_i and s_i operate in the orbital and spin space, respectively. The vectors $\boldsymbol{x}$, $\boldsymbol{y}$, and $\boldsymbol{z}$ denote the direction of the d-vector of spin-triplet pairings, and the pairing gap functions are obtained by multiplying the basis function by is_y (same below). Throughout this work, we neglect out-of-plane pairing for simplicity (see Sec. VII).

Irrep	Basis function
A_{1g}	$i\sigma_y \otimes \boldsymbol{z}\cdot\boldsymbol{s}$, $\mathbf{1}$, $k_xk_y\sigma_x$, $(k_x^2-k_y^2)\sigma_z$
A_{2g}	$k_xk_y\sigma_z$, $(k_x^2-k_y^2)\sigma_x$
B_{1g}	σ_z, $i\sigma_y \otimes (k_x^2-k_y^2)\boldsymbol{z}\cdot\boldsymbol{s}$
B_{2g}	σ_x, $i\sigma_y \otimes k_xk_y\boldsymbol{z}\cdot\boldsymbol{s}$
E_g	$(i\sigma_y \otimes \boldsymbol{x}\cdot\boldsymbol{s},\ \ i\sigma_y \otimes \boldsymbol{y}\cdot\boldsymbol{s})[i\sigma_y \otimes k_{x(y)}^2\boldsymbol{x}\cdot\boldsymbol{s},\ \ i\sigma_y \otimes k_{y(x)}^2\boldsymbol{y}\cdot\boldsymbol{s}]$
A_{1u}	$\frac{\sigma_0\pm\sigma_z}{2} \otimes k_x\boldsymbol{x}\cdot\boldsymbol{s} + \frac{\sigma_0\mp\sigma_z}{2} \otimes k_y\boldsymbol{y}\cdot\boldsymbol{s}$, $\sigma_x \otimes (k_x\boldsymbol{y}+k_y\boldsymbol{x})\cdot\boldsymbol{s}$
A_{2u}	$\frac{\sigma_0\pm\sigma_z}{2} \otimes k_x\boldsymbol{y}\cdot\boldsymbol{s} - \frac{\sigma_0\mp\sigma_z}{2} \otimes k_y\boldsymbol{x}\cdot\boldsymbol{s}$, $\sigma_x \otimes (k_x\boldsymbol{x}-k_y\boldsymbol{y})\cdot\boldsymbol{s}$
B_{1u}	$\frac{\sigma_0\pm\sigma_z}{2} \otimes k_x\boldsymbol{x}\cdot\boldsymbol{s} - \frac{\sigma_0\mp\sigma_z}{2} \otimes k_y\boldsymbol{y}\cdot\boldsymbol{s}$, $\sigma_x \otimes (k_x\boldsymbol{y}-k_y\boldsymbol{x})\cdot\boldsymbol{s}$
B_{2u}	$\frac{\sigma_0\pm\sigma_z}{2} \otimes k_x\boldsymbol{y}\cdot\boldsymbol{s} + \frac{\sigma_0\mp\sigma_z}{2} \otimes k_y\boldsymbol{x}\cdot\boldsymbol{s}$, $\sigma_x \otimes (k_x\boldsymbol{x}+k_y\boldsymbol{y})\cdot\boldsymbol{s}$
	$(ik_x\sigma_y,\ \ ik_y\sigma_y)$
E_u	$(\sigma_x \otimes k_y\boldsymbol{z}\cdot\boldsymbol{s},\ \ \sigma_x \otimes k_x\boldsymbol{z}\cdot\boldsymbol{s})$
	$(\frac{\sigma_0\pm\sigma_z}{2} \otimes k_x\boldsymbol{z}\cdot\boldsymbol{s},\ \ \frac{\sigma_0\mp\sigma_z}{2} \otimes k_y\boldsymbol{z}\cdot\boldsymbol{s})$

fashion [50]: σ_0, σ_x, σ_z, and σ_y as A_{1g}, B_{2g}, B_{1g}, and A_{2g}, respectively. A Hamiltonian invariant under all D_{4h} operations is then constructed by appropriate product of the σ, s, and momentum-space basis functions, as in Eq. (1). Note that, among the terms in the Hamiltonian, s_z transforms as A_{2g}. Further, since the orbital wave function of the t_{2g} electrons are even under inversion, the only effect of inversion is to invert 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). Taken together, it can be verified that the Hamiltonian Eq. (1) respects the full D_{4h} symmetry.

The possible pairing symmetries are typically classified according to the irreducible representations (irreps) of the underlying crystalline point group. This is straightforwad in single-orbital models, as has been well documented by Sigrist and Ueda [40]. However, the presence of multiple orbitals adds a layer of complexity. The usual classifications into even-parity spin-singlet and odd-parity spin-triplet pairings are no longer sufficient. One must also consider Cooper pairs symmetric and antisymmetric in the orbital manifold [49–52]. In addition, care must be taken with the transformation properties of the bilinear pairing operators $c_{m,s}\sigma_i^{mn}c_{n,s'}$, analogously to that of $c^\dagger_{m,s}\sigma_i^{mn}c_{n,s'}$ mentioned above.

Tables I lists 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 $\boldsymbol{k}$ will not mix, because the inversion operation only acts to invert the momentum $\boldsymbol{k}$ 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).

In the following, we develop effective Ginzburg-Landau theories for a few representative irreps. The first example, the A_{1g} irrep, serves to illustrate how the multiple symmetry-equivalent multiorbital pairings in each irrep may be inherently coupled by interorbital hybridization and SOC. In particular, this analysis will make transparent the SOC-induced entanglement of spin-triplet and spin-singlet pairings. We then proceed to the two-dimensional E_g and E_u irreps in view of the signatures of two-component superconducting pairing in Sr_2RuO_4 .

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

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

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

The triplet and singlet components correspond to inter- and intraorbital 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, $f = \hat{\Delta}^\dagger \hat{\Delta}/V + T\sum_l \sum_{\boldsymbol{k},w_n} \mathrm{Tr}[G(iw_n,\boldsymbol{k})\hat{\Delta}\bar{G}(iw_n,\boldsymbol{k})\hat{\Delta}^\dagger]^{2l}/(2l)$, where $G(iw_n,\boldsymbol{k}) = (iw_n - H_{0\boldsymbol{k}})^{-1}$ and $\bar{G}(iw_n,\boldsymbol{k}) = (iw_n + H^*_{0,-\boldsymbol{k}})^{-1}$ are the electron and hole components of the Gorkov Green's funcion. The singlet and triplet pairings are coupled at quadratic order,

$$J_{12} = i\lambda_{12}(\psi_1^*\psi_2 - \psi_2^*\psi_1), \quad (3)$$

with $\lambda_{12} \propto \eta$ a real constant. The complex phase is a consequence of the particular structure of the SOC in Eq. (1). A similar conclusion was reached in Ref. [41]. 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. (2) reads $\hat{\Delta}_{\boldsymbol{k}} \propto [\sigma_y \otimes (\boldsymbol{z}\cdot\boldsymbol{s}) + \epsilon\mathbf{1}] i s_y$, where ϵ 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 $\mathcal{T}\hat{\Delta}_{\boldsymbol{k}}\mathcal{T}^{-1} = \hat{\Delta}_{-\boldsymbol{k}}$. On the contrary, the pairings with relative phases of 0 and π between ψ_1 and ψ_2 are degenerate and violate time-reversal symmetry. It is also worth stressing that, in contrast to a pure spin-triplet state, a mixed singlet-triplet pairing may see a reduced uniform spin susceptibility below T_c.

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

$$\begin{aligned} f_{\mathrm{2nd}} = &\sum_{j=1}^{4} \alpha_j |\psi_j|^2 + i\sum_{j=2}^{4}(\lambda_{1j}\psi_1^*\psi_j - \mathrm{c.c.}) \\ &+ (\lambda_{23}\psi_2^*\psi_3 + \lambda_{24}\psi_2^*\psi_4 + \lambda_{34}\psi_3^*\psi_4 + \mathrm{c.c.}). \end{aligned} \quad (4)$$

All of the α_j and λ_{ij} coefficients are real. Like λ_{12}, the other two coefficients that couple triplet and singlet pairings, λ_{13} and λ_{14}, both depend on SOC. By contrast, the remaining coefficients, λ_{23}, λ_{24}, and λ_{34}, do not rely on SOC. Instead, these three couplings are induced by the σ_x and/or σ_z terms in Eq. (1), with $\lambda_{23} \propto t''/t$, $\lambda_{24} \propto \tilde{t}/t$, and $\lambda_{34} \propto \tilde{t}t''/t^2$. The sign of α_i determines whether an intrinsic Cooper instability exists for the corresponding pairing component. The most negative α_i typically signifies the most dominant component. A component that lacks Cooper instability ($\alpha_i > 0$) 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 $\hat{\psi} = (\psi_1, \psi_2, \psi_3, \psi_4)^T$:

$$f_{\mathrm{2nd}} = \hat{\psi}^\dagger \begin{bmatrix} \alpha_1 & i\lambda_{12} & i\lambda_{13} & i\lambda_{14} \\ -i\lambda_{12} & \alpha_2 & \lambda_{23} & \lambda_{24} \\ -i\lambda_{13} & \lambda_{23} & \alpha_3 & \lambda_{34} \\ -i\lambda_{14} & \lambda_{24} & \lambda_{34} & \alpha_4 \end{bmatrix} \hat{\psi}. \quad (5)$$

In the single most favorable eigenstate, ψ_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

$$\hat{\Delta}_{\boldsymbol{k}} = i\epsilon_1\hat{\Delta}_{1\boldsymbol{k}} + \epsilon_2\hat{\Delta}_{2\boldsymbol{k}} + \epsilon_3\hat{\Delta}_{3\boldsymbol{k}} + \epsilon_4\hat{\Delta}_{4\boldsymbol{k}}, \quad (6)$$

where $(\epsilon_1, i\epsilon_2, i\epsilon_3, i\epsilon_4)$ constitutes the lowest-energy eigenvector of the coupling matrix in Eq. (5). In reality, one or certain subset of the ϵ_i's may dominate, while the rest are induced. For example, since $\hat{\Delta}_{2\boldsymbol{k}}$ and $\hat{\Delta}_{4\boldsymbol{k}}$ both describe intraorbital pairing and since orbital mixing is secondary to the intraorbital hoppings in Sr_2RuO_4 , ϵ_2 and ϵ_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 2D two-orbital model, the simplest E_g pairing taken from Table I is a spin-triplet given by

$$\hat{\Delta}_{\boldsymbol{k}} = (\psi_x \cdot i\sigma_y \otimes \boldsymbol{x}\cdot\boldsymbol{s} + \psi_y \cdot i\sigma_y \otimes \boldsymbol{y}\cdot\boldsymbol{s}) i s_y, \quad (7)$$

where the two order parameters ψ_x and ψ_y form a two-dimensional irrep. This pairing has also been discussed in Ref. [52]. In essence, the two components each describes a spin-triplet interorbital s-wave pairing. A Ginzburg-Landau free energy can be constructed on symmetry basis or through a straightforward gradient expansion, which leads to

$$\begin{aligned} f = &k_1(|\partial_x\psi_x|^2 + |\partial_y\psi_y|^2) + k_2(|\partial_y\psi_x|^2 + |\partial_x\psi_y|^2) \\ &+ \alpha(|\psi_x|^2 + |\psi_y|^2) + \beta(|\psi_x|^4 + |\psi_y|^4) \\ &+ \beta_{xy}|\psi_x|^2|\psi_y|^2 + \beta'[(\psi_x^*\psi_y)^2 + (\psi_y^*\psi_x)^2] + \cdots , \end{aligned} \quad (8)$$

where "$\cdots$" denotes higher-order terms. Note that because ψ_x and ψ_y are both even under spatial transformation $x \to -x$ and $y \to -y$ (or $k_x \to -k_x$ and $k_y \to -k_y$), cross-gradient terms such as $\partial_x\psi_x^*\partial_y\psi_y$ are disallowed. Likewise, $\partial_x\psi_x^*\partial_x\psi_y$, $\partial_y\psi_y^*\partial_y\psi_x$ and their complex conjugates are forbidden, as ψ_x and ψ_y exhibit opposite mirror eigenvalues about the xz (and yz) planes. Dependent on the sign of β', 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 π is favored if $\beta' < 0$, which corresponds to a TRI state.

A TRSB pairing may support spontaneous current at the surface or around defects. Within Ginzburg-Landau theory, it is well understood that the forbidden gradient terms mentioned above would have been crucial for the existence of spontaneous current [54–57]. Thus, unlike the conventional E_g chiral d-wave pairing with $\Delta_{\boldsymbol{k}} \sim (k_x + ik_y)k_z$, the present TRSB E_g pairing (when appears alone) has the salient feature 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 corners of such domain walls carry opposite fractional quantum fluxes analogous to the scenario in a coupled anisotropic XY model [58,59]. The resultant internal field distribution could be detected in μSR measurements. Notably, fractional vortices could still emerge even when the pairing is TRI [60]. A final important remark is that, since this spin-triplet pairing has its d-vector oriented in-plane, the Knight shift shall exhibit a drop under in-plane magnetic fields. This is potentially relevant to the observation in a recent NMR measurement [26].

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

We write down in Table I four of the simplest basis functions belonging to the E_u irrep. Note that, compared to the other terms, the second term, $(\sigma_x \otimes k_y \boldsymbol{z}\cdot\boldsymbol{s},\ \ \sigma_x \otimes k_x \boldsymbol{z}\cdot\boldsymbol{s})$, has the form factors k_x and k_y in reversed order. In this manner, all four terms transform coherently as the basis (k_y, k_x) does. Among the four terms, the first is the only spin-singlet pairing, and the third one is frequently discussed in connection to the proposal of p-wave instability on the quasi-1D bands [32]. A general E_u pairing acquires the form $\hat{\Delta}_{\boldsymbol{k}} = \sum_{i=1}^{4}\sum_{\mu=x,y}\psi_{i\mu}\hat{\Delta}_{i\boldsymbol{k}\mu}$. Following the analysis in the preceding section, we obtain the following free energy:

$$
\begin{aligned}
f_{\text{2nd}} = &\sum_{i=1}^{4}\sum_{\mu=x,y}\alpha_i|\psi_{i\mu}|^2 + \sum_{\mu=x,y}[\lambda_{23}\psi_{2\mu}^*\psi_{3\mu} \\
&+\lambda_{24}\psi_{2\mu}^*\psi_{4\mu} + \lambda_{34}\psi_{3\mu}^*\psi_{4\mu} + \text{c.c.}] \\
&+ i[\psi_{1x}^*(\lambda_{12}\psi_{2x} + \lambda_{13}\psi_{3x} + \lambda_{14}\psi_{4x}) - \text{c.c.}] \\
&- i[\psi_{1y}^*(\lambda_{12}\psi_{2y} + \lambda_{13}\psi_{3y} + \lambda_{14}\psi_{4y}) - \text{c.c.}],
\end{aligned} \tag{9}
$$

where all λ_{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 needs other ingredients such as the interorbital hybridization t''. Finally, the last two lines indicate that the coupling between the singlet and triplet pairings has opposite signs for the x and y components. This has consequences on the phase configuration acquired by the multiple components.

In short, denoting the the two corresponding order parameters $\Psi_{a(b)}$, the E_u gap function is more generally expressed in an alternative two-component form: $\hat{\Delta}_{\boldsymbol{k}} = (\hat{\Delta}_{a\boldsymbol{k}}, \hat{\Delta}_{b\boldsymbol{k}})$, with

$$
\begin{aligned}
\hat{\Delta}_{a\boldsymbol{k}} &= \Psi_a(i\epsilon_1\hat{\Delta}_{1\boldsymbol{k}x} + \epsilon_2\hat{\Delta}_{2\boldsymbol{k}x} + \epsilon_3\hat{\Delta}_{3\boldsymbol{k}x} + \epsilon_4\hat{\Delta}_{4\boldsymbol{k}x}), \\
\hat{\Delta}_{b\boldsymbol{k}} &= \Psi_b(-i\epsilon_1\hat{\Delta}_{1\boldsymbol{k}y} + \epsilon_2\hat{\Delta}_{2\boldsymbol{k}y} + \epsilon_3\hat{\Delta}_{3\boldsymbol{k}y} + \epsilon_4\hat{\Delta}_{4\boldsymbol{k}y}),
\end{aligned} \tag{10}
$$

where $\epsilon_{1,\dots,4}$ are real constants. This leads to the following free energy in powers of Ψ_a and Ψ_b,

$$
\begin{aligned}
f = &\, k_1(|\partial_x\Psi_a|^2 + |\partial_y\Psi_b|^2) + k_2(|\partial_x\Psi_b|^2 + |\partial_y\Psi_a|^2) \\
&+ k_3(\partial_x\Psi_a^*\partial_y\Psi_b + \text{c.c.}) + k_4(\partial_x\Psi_b^*\partial_y\Psi_a + \text{c.c.}) \\
&+ \alpha(|\Psi_a|^2 + |\Psi_b|^2) + \beta(|\Psi_a|^4 + |\Psi_b|^4) \\
&+ \beta_{ab}|\Psi_a|^2|\Psi_b|^2 + \beta'[(\Psi_a^*\Psi_b)^2 + (\Psi_b^*\Psi_a)^2] + \cdots.
\end{aligned} \tag{11}
$$

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. On the other hand, the singlet-triplet mixing shall lead to a suppressed uniform spin susceptibility and therefore a drop in NMR Knight shift under in-plane fields. Further, Ref. [61] will study some peculiar forms of the multiorbital E_u pairing, which exhibits near-nodal excitations consistent with a number of experimental signatures [15,16].

VI. THREE-ORBITAL MODEL

In extending to a full three-orbital model, the Gell-Mann matrices (T_i, $i = 1, \dots, 8$) turn out to be convenient devices. We define $\bar{T}_{11} = (T_0 + \sqrt{3}T_8)/2$ and $\bar{T}_{33} = (T_0 - \sqrt{3}T_8)/4$, where T_0 is a 3×3 identity matrix. Using the orbital spinor basis $(c_{m\uparrow}^\dagger, c_{m\downarrow}^\dagger)$ in the order $m = xz, yz, xy$, to quadratic order in k and with on-site SOC, the Hamiltonian reads

$$
\begin{aligned}
H_{0\boldsymbol{k}} = &\left[t\left(k_x^2 + k_y^2\right) - \mu\right]\bar{T}_{11} + \tilde{t}(k_x^2 - k_y^2)T_3 + t''k_xk_yT_1 \\
&+ \left[t'\left(k_x^2 + k_y^2\right) - \mu_{xy}\right]\bar{T}_{33} \\
&+ \eta(T_2 \otimes s_z + T_5 \otimes s_x - T_7 \otimes s_y),
\end{aligned} \tag{12}
$$

where the t' term and μ_{xy} denote the kinetic energy and chemical potential of the d_{xy} orbital. Note that $T_{1,2,3}$ are equivalent to $\sigma_{x,y,z}$ and $\bar{T}_{11}$ to σ_0. Hence they inherit the transformation properties of the σ_μ operators. $T_{4,5}$ 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_5 and T_7, respects D_{4h}.

Without further elaboration, the gap functions, especially those not involving interorbital pairings with the d_{xy} orbital, can be classified rather straightforwardly following the preceding analyses. Interorbital pairings involving d_{xy} are associated with pairing operators $T_{4,\dots,7}$. As can be checked, (T_4, T_6) [and (T_5, T_7)] transform as E_g (E_u) irrep under D_{4h}. As a consequence, any such pairing must contain both T_4 and T_6 (or T_5 and T_7) in the gap function. This is demonstrated in Table II. As an interesting note, two recent microscopic multiorbital

TABLE II. Representative superconducting basis functions of the interorbital pairing involving the d_{xy} orbital in the two dimensional E_g and E_u irreps. Here T_i and s_i operate respectively in the orbital and spin space, as explained in the text. Note that in the E_g irrep, the order of the two degenerate components is to ensure that each basis function transforms as $(k_y, k_x)k_z$ does, instead of some behaving like $(k_y, k_x)k_z$ and some like $(k_x, k_y)k_z$ and similarly for the E_u basis functions.

Irrep	Basis function
A_{1g}	$T_5 \otimes \boldsymbol{x}\cdot\boldsymbol{s} - T_7 \otimes \boldsymbol{y}\cdot\boldsymbol{s}$
A_{2g}	$T_5 \otimes \boldsymbol{y}\cdot\boldsymbol{s} + T_7 \otimes \boldsymbol{x}\cdot\boldsymbol{s}$
B_{1g}	$T_5 \otimes \boldsymbol{x}\cdot\boldsymbol{s} + T_7 \otimes \boldsymbol{y}\cdot\boldsymbol{s}$
B_{2g}	$T_5 \otimes \boldsymbol{y}\cdot\boldsymbol{s} - T_7 \otimes \boldsymbol{x}\cdot\boldsymbol{s}$
	$(T_4,\ T_6)$
	$[k_{x(y)}^2 T_4,\ k_{y(x)}^2 T_6]$
E_g	$(iT_7 \otimes \boldsymbol{z}\cdot\boldsymbol{s},\ iT_5 \otimes \boldsymbol{z}\cdot\boldsymbol{s})$
	$[iT_7 \otimes k_{x(y)}^2 \boldsymbol{z}\cdot\boldsymbol{s},\ iT_5 \otimes k_{y(x)}^2 \boldsymbol{z}\cdot\boldsymbol{s}]$
	$(T_4 \otimes k_x \boldsymbol{x}\cdot\boldsymbol{s},\ T_6 \otimes k_y \boldsymbol{y}\cdot\boldsymbol{s})$
	$(T_6 \otimes k_y \boldsymbol{x}\cdot\boldsymbol{s},\ T_4 \otimes k_x \boldsymbol{y}\cdot\boldsymbol{s})$
E_u	$(T_4 \otimes k_y \boldsymbol{y}\cdot\boldsymbol{s},\ T_6 \otimes k_x \boldsymbol{x}\cdot\boldsymbol{s})$
	$(T_6 \otimes k_x \boldsymbol{y}\cdot\boldsymbol{s},\ T_4 \otimes k_y \boldsymbol{x}\cdot\boldsymbol{s})$

calculations [62,63] both found noticeable, or even dominant, interorbital E_g or 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 superconducting Sr_2RuO_4 , we explored the possibilities made available by its multiorbital degree of freedom. The superconducting pairings are classified on the basis of the Ru t_{2g} $4d$ 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 Sr_2RuO_4 in due course. As a special note, when a spin-triplet pairing is inherently mixed with spin-singlet pairings due to the presence of SOC, the uniform spin susceptibility, and hence the Knight shift, may exhibit a drop below the superconducting transition.

Our main purpose is not to rule out or identify any pairing for Sr_2RuO_4 but rather to provide a new perspective to further explore the enigmatic superconductivity in this material. Hence we have restricted our study, for simplicity, to in-plane pairings in our symmetry classification of the multiorbital superconductivity. Including out-of-plane couplings, i.e., extending the model to 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 [60,64], 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 [60,64]. 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 [60]. Additionally, models containing out-of-plane pairings have appeared in several other contexts [65–67].

Note added. As this manuscript was being prepared for submission, a preprint appeared on arXiv [68] with a similar idea to exploit the multiorbital nature of the superconductivity in Sr_2RuO_4 . Later, another similar paper also appeared on arXiv [69].

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