Shadowed Triplet Pairings in Hund’s Metals with Spin-Orbit Coupling

Jonathan Clepkens,1 Austin W. Lindquist,1 and Hae-Young Kee1,2

1Department of Physics and Center for Quantum Materials, University of Toronto, 60 St. George St., Toronto, Ontario, M5S 1A7, Canada
2Canadian Institute for Advanced Research, Toronto, Ontario, M5G 1Z8, Canada

Hund’s coupling in multi-orbital systems allows for the possibility of even-parity orbital-antisymmetric spin-triplet pairing which can be stabilized by spin-orbit coupling (SOC). While this pairing expressed in the orbital basis is uniform and spin-triplet, it appears at the Fermi surface (FS) as a pseudospin-singlet, with the momentum-dependence determined by the SOC and the underlying triplet character remaining in the form of interband pairing active away from the FS. Here, we examine the role of momentum-dependent SOC (k-SOC) in generating non-trivial pairing symmetries at the FS, as well as the hidden triplet nature associated with inter-orbital pairing, which we dub a “shadowed triplet”. Applying this concept to Sr$_2$RuO$_4$, we first derive several forms of k-SOC with d-wave form-factors from a microscopic model and subsequently show that for a reasonable range of SOC parameters, a pairing state with $s + id_{xy}$ symmetry at the FS can be stabilized. Such a pairing state is distinct from pure spin-singlet and -triplet pairings due to its unique character of pseudospin-singlet pairing on the FS with underlying pseudospin-triplet nature away from the FS. We discuss experimental probes to differentiate the shadowed triplet pairing from pure spin-triplet and -singlet pairings.

I. INTRODUCTION

A key feature of superconductivity (SC) is the antisymmetric-wavefunction of the Cooper pair under the exchange of two electrons. This has limited the focus to even-parity spin-singlet and odd-parity spin-triplet pairings in a single Fermi surface system. In multi-orbital systems, additional possibilities arise due to the orbital degree of freedom, such as even-parity spin-triplet, or odd-parity spin-singlet pairings. For example, it was shown earlier that Hund’s coupling in multi-orbital systems allows for an even-parity spin-triplet pairing that is orbitally antisymmetric between two orbitals. However, the pairing is fragile, because the SC is formed by pairing two electrons with different orbitals and momenta $k$ and $-k$, yet these momenta occur at different energies due to the different electronic dispersions of the orbitals.

Thus to stabilize such an orbital antisymmetric pairing, significant orbital degeneracy is required throughout momentum space near the FS, because splitting of the orbital degeneracy suppresses the SC. Furthermore, hybridization between different orbitals, when allowed by symmetry, can further weaken the prospect of inter-orbital pairing in some cases, by splitting the degeneracy where it occurs. However, significant spin-orbit mixing such as spin-orbit coupling (SOC) can help to stabilize the inter-orbital pairing. It was shown that SOC indeed enhances even-parity orbital-antisymmetric spin-triplet pairing.

Another important aspect of the orbital antisymmetric spin-triplet pairing is that the Hund’s coupling, significant in transition metal systems, acts as an attractive pairing interaction. Similar to the attractive Hubbard model which allows for BEC pairing, the Hund’s coupling allows a strong Cooper pair to form between two orbitals with spin-triplet character. However, when the electron motion is introduced, i.e., the kinetic term in the Hamiltonian, the pairing is drastically weakened, because the electrons comprising the Cooper pair with momenta $k$ and $-k$ in different orbitals occur at different energies, as discussed above.

However, the story becomes more interesting when atomic SOC is introduced. Note that purely spin-singlet and -triplet pairings are no longer well-defined, since spin and orbital quantum numbers are mixed. Thus one defines Cooper pairs in the total angular momentum (pseudospin) basis. When the SOC is strong, this forms the basis for SC in the heavy fermion superconductors, such as UPt$_3$, leading to odd-parity pseudospin-triplet SC. When the SOC is intermediate, i.e., comparable to the bandwidth of the relevant orbitals, spin and orbital characters vary continuously with $k$, and one can define the pairing in either the orbital or band basis. The SOC not only supports the orbital-antisymmetric spin-triplet pairing as mentioned earlier, but also transforms pure spin-triplet pairing into both pseudospin-singlet and pseudospin-triplet pairings.

Furthermore, the form of the SOC is not limited to atomic SOC, i.e., $L \cdot S$, and the precise form can determine the momentum dependence of the superconducting state. For example, $s$-wave SC proximate to a topological insulator or strong Rashba SOC with broken inversion symmetry leads to an effective $p + ip$ SC, which is odd-parity and a spinless triple. For materials with inversion symmetry, there is still the possibility of even-parity momentum-dependent SOC (k-SOC), which has been discussed in the context of the unconventional superconductor, Sr$_2$RuO$_4$, based only on symmetry. Similar to a Rashba-SOC generated $p + ip$ SC, the inclusion of k-SOC in a microscopic model with $s$-wave pairing is reflected in an intraband pairing with the same symmetry as the k-SOC at the FS.

Here we study a microscopic route to k-SOC and how SOC transforms even-parity inter-orbital spin-triplet
SC into pseudospin-singlet and -triplet SC in a Hund’s metal with multiple orbitals and strong Hund’s coupling. We illustrate how the symmetry of this pairing on the FS is dictated by the SOC, while away from the FS, pseudospin-triplet interband pairing remains, dubbed a “shadowed triplet”. While this SC behaves like a singlet in response to low energy excitations, its hidden identity shows up at finite magnetic fields, and can be tested when the field strength reaches an appreciable percentage of the superconducting gap size.\(^{16}\)

Below we first consider a simple but general two-orbital model to show how even-parity spin-triplet pairing arises in Sec. II. This includes the stability conditions and how SOC transforms this pairing into a pseudospin singlet and triplet in the Bloch band basis. The SOC in the shadowed triplet not only plays an essential role in enhancing the pairing, but also determines the pairing symmetry. In Sec. III we investigate microscopic routes to enhancing the pairing, but also determines the pairing symmetry. In Sec. III we investigate microscopic routes to enhance the pairing, but also determines the pairing symmetry. In Sec. IV, we apply the shadowed triplet pairing scenario to the prominent unconventional superconductor, Sr\(_2\)RuO\(_4\), for which the SOC has been shown to be important\(^{2,6,16,18}\) and discuss the leading instabilities within a realistic three orbital model.

## II. TWO-ORBITAL MODEL

We consider a mean-field (MF) Hamiltonian consisting of a generic tight binding model with two orbitals, SOC and a pairing term,

\[
H = \sum \Psi^\dagger_k (H_0(k) + H^z_{SOC}(k) + H_{pair}(k)) \Psi_k
\]

\[
H_0(k) = \rho_3 \left( \frac{\xi^+_{k\uparrow}}{2} \sigma_0 \tau_0 + \frac{\xi^+_{k\downarrow}}{2} \sigma_0 \tau_3 + t_k \sigma_0 \tau_1 \right)
\]

\[
H_{SOC}(k) = -\lambda_k \rho_3 \sigma_3 \tau_2 \\
H_{pair}(k) = -\Delta^z \rho_3 \sigma_3 \tau_2.
\]

(1)

Here \(\Psi^\dagger_k = (\psi_k^\dagger, T \psi_k^\dagger T^{-1})\), where \(T\) indicates time-reversal and \(\psi_k = (c_{k\uparrow}^a, c_{k\downarrow}^b, c_{k\uparrow}^b, c_{k\downarrow}^a)\) consists of electron operators creating an electron in one of the two orbitals \(a, b\) with spin \(\sigma = \uparrow, \downarrow\). We have introduced the Pauli matrices plus identity matrix, \(\rho_i, \sigma_i, \tau_i, (i = 0, \ldots, 3)\) in the particle-hole, spin and orbital spaces respectively, where the direct product between them is implied. The tight-binding contribution is given by the sum of the two orbital dispersions \(\xi^\pm_k\) and the difference between them, or orbital polarization \(\xi^z_k\), which are defined by

\[
\xi^\pm_k = \xi^a_k \pm \xi^b_k, \text{ as well as the orbital hybridization of the two orbitals, } t_k.
\]

The pairing Hamiltonian includes the even-parity inter-orbital spin-triplet MF order parameter of interest for Hund’s metals, with the \(d\)-vector aligned with the \(z\)-direction,

\[
\Delta^z = \frac{-(3J_H - U)}{4N} \sum_{k\sigma\sigma'} [i \sigma^\beta \sigma'][c^a_{-k\sigma'} \epsilon^b_{-k\sigma'} - c^b_{-k\sigma'} \epsilon^a_{-k\sigma'}],
\]

(2)

where \(N\) is the number of sites and for which the attractive channel is governed by the Hund’s coupling \(J_H\) and on-site Hubbard repulsion \(U\). While these two contributions to the Hamiltonian are sufficient in principle for pairing to occur, it is the presence of the SOC that will be crucial for our discussion.

The importance of the atomic SOC, \(2\lambda \sum_i \mathbf{L}_i \cdot \mathbf{s}_i\), for this form of inter-orbital pairing has been emphasized before\(^{2,6,18}\), where in the basis of \(t_{2g}\) orbitals, \(d_{xz}(d_{yz})\) is coupled to \(d_{xy}\) through the \(x(y)\) component and the quasi-1d orbitals \(d_{yz}/d_{xz}\) are coupled through the \(z\) component. However, \(k\)-SOC is also possible, with one notable example being Rashba SOC in systems with inversion-symmetry breaking. Without inversion-symmetry breaking there are still many possibilities for \(k\)-SOC, yet its interplay with even-parity spin triple pairing has received little attention except for several recent studies, mostly related to the unconventional superconductor Sr\(_2\)RuO\(_4\), in which such \(k\)-SOC has been introduced only from a symmetry perspective with no microscopic derivation\(^{15,16}\). Here we consider this by including in our model a term denoted by \(H_{SOC}\), which we take to be of the form \(L_z S_z\), but with general momentum-dependence \(\lambda_k\) to illustrate the effects of the SOC. Later, we derive several possible \(k\)-SOC from a microscopic perspective, which can differ from the \(L_z S_z\) form included here, in Sec. III. This model, incorporating a general kinetic Hamiltonian, SOC and even-parity spin-triplet pairing allows for a systematic study of the microscopic components relevant to such pairing in multi-orbital systems and is easily generalized to systems with three-orbitals.

To begin, we aim to understand the stability of the SC state within our model. Let us consider the relationship between the various components and how this affects the quasi-particle (QP) dispersion. With zero orbital polarization and hybridization, i.e., \(\xi^z_k, t_k = 0\), and without SOC, the orbitals are completely degenerate, providing maximal inter-orbital pairing. As either of \(\xi^z_k, t_k\) become non-zero, the degeneracy of the orbitals is reduced, leading to a reduction of the phase space for pairing at the FS and thus reducing the gap formed. These pairing effects are revealed by the commuting behavior with the pairing term\(^{21,22}\). Conversely, the SOC term anti-commutes with the pairing term and generally enhances the pairing state. The anti-commutators are given by

\[
\{ \frac{\xi^z_k}{2} \rho_3 \sigma_0 \tau_3, H_{pair} \} = \xi^z_k \Delta^z \rho_1 \sigma_3 \tau_1 \\
\{ t_k \rho_3 \sigma_0 \tau_1, H_{pair} \} = -2t_k \Delta^z \rho_1 \sigma_3 \tau_3 \\
\{ -\lambda_k \rho_3 \sigma_3 \tau_2, H_{pair} \} = 0.
\]

(3)
These effects are reflected in the QP dispersion, given by

\[ E_k = \pm \frac{1}{2} \left[ \xi_k^2 - \xi_k^2 + 4\left(t_k^2 + (\Delta^2)^2 + \lambda_k^2\right) \right] \]
\[ \pm 2\sqrt{\xi_k^4 \left(\xi_k^2 + 4t_k^2 + 4\lambda_k^2\right)^2 + 4(\Delta^2)^2\left(\xi_k^2 + 4t_k^2\right)^2} \].

(4)

The general equation of the FS is \( \xi_k^2 = \xi_k^2 - 4(t_k^2 + \lambda_k^2) \), from which it can be seen that if \( \lambda_k = 0 \) and the orbitals are degenerate, i.e., \( \xi_k = 0 \), we recover the conventional BCS result for the gap energy on the FS: \( \pm \Delta^2 \). In the general case where these terms are non-zero, assuming \( \Delta^2 \) is small, the gap energy is

\[ E_k^{\Delta}(FS) \approx \pm \sqrt{\frac{(\Delta^2)^4 + 16\lambda_k^2(\Delta^2)^2}{4(\xi_k^2 + 4(t_k^2 + \lambda_k^2))}}. \]

(5)

From this, it is clear that increasing \( \xi_k^- \) and \( t_k \) decreases the overall gap energy. The detrimental effects on the gap energy, signified by the commuting nature of both \( t_k \) and \( \xi_k^- \) with the pairing Hamiltonian, are a result of shifting apart in energy the bands being paired, resulting in the gap moving away from the FS. However, turning on the SOC significantly enhances the gap energy\(^2\), and as the SOC strength becomes comparable to the dispersion terms, the gap energy can be restored to the order of \( \Delta^2 \). This enhancement of the SC state is accomplished by providing a non-zero intraband pseudospin-singlet pairing on the FS, even for significant orbital polarization, as we show below.

With the aim to further understand the stability of the SC state and the nature of the pairing at the FS, we diagonalize the kinetic Hamiltonian with a unitary transformation and study how the pairing transforms to the Bloch band basis, labeled by band indices \( \alpha, \beta \) and pseudospin \( s = (+, -) \). The transformation is given by

\[ \begin{pmatrix} a_k^a \\ b_k^b \\ c_k^c \\ d_k^d \end{pmatrix} = \begin{pmatrix} \eta_{\sigma} f_k - \eta_{\sigma}^{-1} g_k & -g_k \\ -\eta_{\sigma}^{-1} f_k & \eta_{\sigma} g_k \end{pmatrix} \begin{pmatrix} \alpha_k \sigma \\ \beta_k \sigma \end{pmatrix} \]

(6)

where \( \eta_{\sigma} = +1(-1) \) for \( \sigma = \uparrow (\downarrow) \) and \( s = (+(-). \) The coefficients of the transformation are given by

\[ f_k = -\frac{2\eta_{\sigma}^2}{\sqrt{\xi_k^2 + 4\lambda_k^2}} \] and \( g_k = -\frac{1}{\sqrt{2\left(1 - \frac{\xi_k^2}{\sqrt{\xi_k^2 + 4\lambda_k^2}}\right)}} \), where \( f_k \) is chosen to be complex with the same phase as \( \gamma_k = t_k + i\lambda_k \) and \( \eta_{\sigma} \) is real and \( |f_k|^2 + |g_k|^2 = 1 \). Applying this transformation on \( H_{\text{pair}} \) results in the pairing in the band basis, where we have also defined the Pauli matrices \( \hat{\rho}_k, \hat{\sigma}_k, \hat{\tau}_k \) in the Nambu, pseudospin and band spaces with the basis \( \hat{\Phi}_k = (\phi_k^1, T\phi_k^2 T^{-1}) \) and \( \phi_k^2 = (\alpha_k^+, \beta_k^+, \alpha_k^-, \beta_k^-) \). We obtain

\[ H_{\text{pair}}(k) = \hat{\rho}_k \hat{\sigma}_0 (\Delta^s(k) \hat{\tau}_3 - \Delta_{\alpha\beta}^s(k) \hat{\tau}_1) \]
\[ -\Delta_{\alpha\beta}^s(k) \hat{\rho}_2 \hat{\sigma}_3 \hat{\tau}_2, \]

(7)

where \( \Delta^s(k), \Delta_{\alpha\beta}^s(k) \) denote pseudospin-singlet intraband and interband pairings respectively and \( \Delta_{\alpha\beta}^s(k) \) is a pseudospin-triplet interband pairing. The intraband and interband nature of these pairings becomes more apparent from the operator form,
dependence as a function of momentum as pseudospin-singlet pairing which acquires the same sign weak-coupling instability in realistic systems, since the pseudospin singlet forming on the FS that leads to a mental inter-orbital spin-triplet MF, it is the intraband

\[ \Delta^s(k) = -2\Delta^2 \text{Im}(f_k)g_k = -\frac{-2\Delta^2 \lambda_k}{\sqrt{\xi_k^2 + 4(t_k^2 + \lambda_k^2)}} \]

\[ \Delta^s_{\alpha\beta}(k) = -\Delta^2 \text{Im}(f_k^\dagger) = -2\Delta^2 |f_k|^2 t_k^\dagger \lambda_k t_k + \lambda_k^2 \]

\[ \Delta^t_{\alpha\beta}(k) = \Delta^2 (g_k^2 + \text{Re}(f_k^\dagger)) = \Delta^2 (g_k^2 + |f_k|^2 t_k^2 - \lambda_k^2). \]

While the orbital MF is s-wave and contains no explicit momentum-dependence, transforming to the band basis generates potentially complex momentum dependence from SOC and orbital hybridization. The orbital MF spin-triplet character carries over to the interband pseudospin-triplet which, in the limit of zero SOC, becomes equal to \( \Delta^s \), with both the intraband and interband pseudospin singlets vanishing. However, an important feature for \( \lambda_k \neq 0 \) is the presence of the intraband pseudospin-singlet pairing which acquires the same sign dependence as a function of momentum as \( \lambda_k \). While the intraband pseudospin triplet is a signature of the fundamental inter-orbital spin-triplet MF, it is the intraband pseudospin singlet forming on the FS that leads to a weak-coupling instability in realistic systems, since the interband pairing will generally be negligible on the FS such that the gap is purely given by \( |\Delta^s| \). Considering the QP dispersion in terms of the band pairings,

\[ E_k = \pm \left[ \frac{(\xi_k^2)^2 + (\xi_k^\beta)^2}{2} + (\Delta^s)^2 + (\Delta^s_{\alpha\beta})^2 + (\Delta^t_{\alpha\beta})^2 \right]^{1/2} \]

and evaluating this both the \( \alpha \) or \( \beta \) FS, for only \( \Delta^s \) intraband pairing, we obtain the gap energy \( \pm |\Delta^s| \) as expected. For either only \( \Delta^s_{\alpha\beta} \) or \( \Delta^t_{\alpha\beta} \) interband pairing, a gap of \( \pm \Delta^s_{\alpha\beta} \) or \( \pm \Delta^t_{\alpha\beta} \) forms at \( \xi_k^\beta \) where \( \xi_k^\beta = -\xi_k^\alpha \).

This corresponds to an energy gap on the FS only where \( \xi_k^\alpha = \xi_k^\beta = 0 \), which is not a generic feature but rather requires fine-tuning to achieve.

To illustrate this, we now study the inter-orbital pairing on the FS in a systematic way within our two-orbital model by taking orbital dispersions resembling the \( d_{xz}, d_{zx} \) orbitals, \( \xi_k^\alpha = -2t_1 \cos k_y - 2t_2 \cos k_x - \mu \) and for \( \xi_k^\beta \) we take \( x \leftrightarrow y \). The orbitals are coupled through the SOC, for which we take two cases: the atomic SOC denoted by \( \lambda_0 \) and a \( d \)-wave SOC given by \( \lambda_k = \lambda_d (\cos k_x - \cos k_y) \), as well as the orbital hybridization, which we take as \( t_k = -4t_{ab} \sin k_x \sin k_y \).

With these dispersions, the orbital polarization is given by \( \xi_k^\beta = 2t^- (\cos k_x - \cos k_y) \), where \( t^- = t_1 - t_2 \) and all parameters are given in units of \( t_1 \). The results are summarized in Fig. 1 for which the gap over the FS is shown for four cases: (a) zero SOC and zero orbital polarization as \( t_{ab} \) is increased from zero to 0.1, (b) keeping zero SOC with \( t_{ab} = 0.1 \) as \( \xi_k^\beta \) is increased by tuning \( t^- \) from zero to 0.13, (c) both \( t_{ab} \) and \( t^- \) are kept the same as \( \lambda_0 \) is increased from zero to 0.48 and (d) the same as (c) but with the \( d \)-wave SOC, instead of \( \lambda_0 \), increased by tuning \( \lambda_d \) from zero to 0.48.

Beginning with zero SOC and the orbital dispersions completely degenerate, i.e., \( t_1 = t_2 \), we see that the gap is non-zero everywhere over the two identical bands, as shown by the middle contour in Fig. 1(a). As the strength of the hybridization is increased from zero, the energy separation of the two orbital dispersions increases wherever \( t_k \neq 0 \) and the gap arising from interband pairing disappears at the FS, except where \( t_k \) vanishes along the \( k_x/y = 0 \) lines, where the two-orbital dispersions remain degenerate. Starting from there with zero interband pairing over most of the FS, Fig. 1(b) demonstrates the effect of the orbital polarization in further reducing the interband pairing to zero everywhere on the FS, due to the absence of phase space for zero-momentum pairing. From this, Fig. 1(c) reveals how turning on the SOC revives the SC state by allowing for an interband pairing on the FS. As the SOC is increased from zero, the interband gap becomes non-zero over the entire FS, even where the orbital polarization is large. Additionally, the sign of the gap function is opposite on the two-bands, matching the \( \Delta^s(k) \) \( z \)-dependence in Eq. 7 and Eq. 8, but uniform on each band due to the lack of momentum dependence of the atomic SOC. In contrast to this, Fig. 1(d) displays the \( d \)-wave dependence of the gap arising from the \( d \)-wave SOC. Thus with the introduction of the orbital hybridization/polarization and subsequently the SOC, the pairing at the FS is transformed from an interband spin triplet to a purely intraband pseudospin singlet with the same
momentum dependence as the associated SOC, while the pseudospin triplet is active away from the FS.

While the model introduced in this section is simple, its generality gives insight into the role of the orbital polarization, hybridization and SOC for multi-orbital systems in dictating the stability of the even-parity spin triplet SC state and is easily extended to three-orbital descriptions. Furthermore, we have seen that in the band basis, the intraband pairing at the FS takes on the momentum-dependence of the SOC, allowing for a rich collection of pairing symmetries unexpected from the original s-wave orbital MF and in contrast to other forms of momentum-dependent SC that arise from non-local interactions. However, the possible pairing symmetries will depend on the forms of k-SOC that can be obtained from microscopic considerations. Therefore, we now turn to a study of how various forms of k-SOC can arise microscopically.

III. MICROSCOPIC ROUTE TO MOMENTUM-DEPENDENT SOC

Here, we take as a specific microscopic example the layered perovskite Sr$_2$RuO$_4$, which has the tetragonal space group I4/mmm and point group D$_{4h}$, for which the Ru 4d $t_{2g}$ orbitals are the relevant low-energy degrees of freedom. With this, we study how the various forms of k-SOC with different d-wave form factors such as a) an in-plane $d_{xy}$ SOC in the B$_{2g}$ representation, b) in-plane $d_{x^2-y^2}$ SOC in the B$_{1g}$ representation and c) interlayer $\{d_{xz},d_{yz}\}$ SOC in the E$_g$ representation can arise microscopically, going beyond a purely symmetry based analysis.

A. in-plane B$_{2g}$

We begin with the in-plane k-SOC in the B$_{2g}$ representation, which does not require any hopping processes involving a different layer of Ru-O octahedra and should therefore be expected to be the leading contribution beyond the atomic SOC. Such an in-plane k-SOC has a $d_{xy}$ form factor and can be obtained by considering hopping between next-nearest neighbour Ru atoms in a given plane, utilizing either the top or bottom apical oxygen of the octahedra as intermediate sites and including the oxygen p-orbitals’ atomic SOC. This hopping process results in an electron hopping from the $d_{xy}$ orbital with spin $\sigma$ to either a $d_{xz}$ or $d_{yz}$ orbital with spin $-\sigma$, where the former case is shown in Fig. 2(a). We can construct the effective Hamiltonian, involving only the Ru sites, with

$$ H_{SOC}^{B_{2g}} = \sum_{p \pm} \frac{H_0^{p \pm}|p \pm\rangle\langle p \pm|H_0}{E_d - E_{p \pm}}, $$

(11)

where $H_0$ denotes the hopping Hamiltonian involving both d and p orbitals, the sum runs over the interme-diate oxygen states for an oxygen site $r$, which are eigenstates of the oxygen SOC $|j,m,\sigma,r\rangle$, and we consider up to the 2nd order of the perturbation theory for this process. For instance, considering a hopping process between a $d_{xy}$ state with spin $\uparrow$ and a spin $\downarrow$ $d_{xz}$ state, we have $|p_+\rangle = -\frac{1}{\sqrt{2}}(|p_{x\downarrow}\rangle+i|p_{y\downarrow}\rangle) + \frac{\sqrt{2}}{\sqrt{3}}|p_{z\uparrow}\rangle$ and $|p_-\rangle = -\frac{1}{\sqrt{2}}(|p_{x\downarrow}\rangle+i|p_{y\downarrow}\rangle) - \frac{1}{\sqrt{3}}|p_{z\uparrow}\rangle$. The energy denominator is given for $|p_+\rangle$ and $|p_-\rangle$ by $E_{pd} + \frac{\Lambda_{pd}}{2}$ and $E_{pd} - \Lambda_{pd}$ respectively, where $E_{pd}$ is the difference in the on-site atomic potentials and $\Lambda_{pd}$ is the oxygen SOC constant. Since only the $p_z$ orbital has finite overlap with $d_{xz}(d_{yz})$ in the $z$ direction, to obtain a spin-flip hopping at 2nd order, we are restricted to considering hopping between the $d_{xy}$ and $p_z$ orbitals. The spin flip then arises from the mixing of the $p_z$ orbital with the opposite spin state of the $p_x$ or $p_y$ orbital due to the SOC. The intermediate hopping amplitudes for the $d_{xy}/d_{xz}$ process are shown schematically in the top view of Fig. 2(b), where the lobe of the $p_z$ orbital closest to the plane of hopping is shown.

Considering the hopping amplitude between the $d_{xy}$ orbital with spin $\sigma$ at site $R$ denoted by $|xy,\sigma,R\rangle$ and the opposite spin state of the $d_{xz}$ orbital at site $R'$, $|xz, -\sigma, R'\rangle$ we obtain,

$$ \langle xz, -\sigma, R'|H_{SOC}^{B_{2g}}|xy, \sigma, R\rangle = \eta_{\sigma} \sum_{r} \text{sgn}(t_{pd}^{xz})\text{sgn}(t_{pd}^{xy})\lambda_{soc}, $$

(12)

FIG. 2. Hopping processes generating the in-plane B$_{2g}$ d-wave k-SOC, with effective hopping amplitude $t_{soc}^{xy}$ indicated by the solid line. a) The relevant $p$ and $d$ orbitals are shown in one layer of the three-dimensional (3D) crystal structure where the top layer is removed for clarity and the dashed lines indicate the intermediate hopping processes. The process involving the $d_{xz}$ orbital is shown as an example, since the alternative process involving $d_{yz}$ is related by a C$_4$ rotation. b) Schematic top view of the hopping process, where the bottom lobes of the $p_z$ orbitals are shown and the dashed lines within the $d_{xz}$ orbitals represent the $p_z$ orbitals.
where \( t_{B_2g}^{\text{soc}} = \frac{\lambda_p |t_{xy,y}^{xy}||t_{xy,x}^{xy}|}{(E_{pd} + \frac{\lambda_p}{2})(E_{pd} - \lambda_p)} \), \( \eta_\sigma = \pm 1 \) for \( \sigma = \uparrow \) (\( \downarrow \)) and the sum over \( r \) runs over the top and bottom apical oxygen sites, which leads to a factor of two due to the reflection symmetry about the \( xy \) plane. From this hopping amplitude and Fig. 2 it is also clear that the momentum-dependence will appear with a \( d_{xy} \) form factor since, while the sign of \( t_{xy,y}^{xy} \) doesn’t change throughout the \( xy \) plane, the sign of \( t_{xy,x}^{xy} \) matches the sign of the \( d_{xy} \) orbital. Therefore, summing over the Ru site and spin indices and performing the Fourier transform, we obtain the SOC Hamiltonian for this process

\[
H_{SOC,X}^{B_2g} = -4i t_{B_2g}^{\text{soc}} \sum_{k} \sin k_x \sin k_y \sigma_\sigma' \eta_\sigma |_{k_\sigma}\rangle \langle_{k_\sigma'} + h.c.,
\]

where \( X, Y \) indicate the orbital combinations \( xz/xy \) and \( yz/xy \) respectively.

If we now instead consider the equivalent process between the \( d_{yz} \) and \( d_{yz} \) orbitals, which is related by a \( C_4 \) rotation, the only difference is that the hopping amplitude \( t_{yz,y}^{yz} \) is replaced by \( t_{yz,x}^{yz} \), which is however identical in both magnitude and sign. We obtain the following Hamiltonian for the hopping process between the \( d_{yz} \) and \( d_{xy} \) orbitals,

\[
H_{SOC,Y}^{B_2g} = 4i t_{B_2g}^{\text{soc}} \sum_{k} \sin k_x \sin k_y \sigma_\sigma' \eta_\sigma |_{k_\sigma}\rangle \langle_{k_\sigma'} + h.c.,
\]

and \( H_{SOC}^{B_2g} = H_{SOC,X}^{B_2g} + H_{SOC,Y}^{B_2g} \) is the total Hamiltonian for the effective \( B_2g \) SOC.

### B. in-plane \( B_{1g} \)

We now consider a microscopic route to obtaining an in-plane k-SOC in the \( B_{1g} \) representation, which has a \( d_{xz,-yz} \) form factor and which now requires a different layer of Ru-O octahedra. Let us consider hopping between nearest neighbour Ru atoms in a given plane, utilizing the out of plane apical oxygen sites as intermediate states and once again including the oxygen \( p \)-orbitals’ atomic SOC. An example of this process is shown in Fig. 3 (a), for which the hopping between the \( d_{xy} \) orbital with spin \( \sigma \) state and the opposite spin state of the \( d_{xy} \) orbital is indicated and we consider the overlap of the \( d_{xy} \) orbital with both the \( p_x \) and \( p_y \) orbitals as shown. The hopping amplitudes we consider are shown schematically in Fig. 3 (b) for both the \( +\hat{x} \) or \( +\hat{y} \) directions, where for the \( p_z \) orbital, the lobe closest to the plane containing the effective hopping is shown and the \( p_x/p_y \) orbitals are shown in separate squares for clarity. We denote the hopping between the \( d_{xz} \) orbital and either \( p_x \) or \( p_y \) as \( t_{pd}^{xy} \) since they have the same magnitude due to the \( C_4 \) rotational symmetry. The hopping between either of the \( d_{yz}/d_{xz} \) orbitals and \( p_z \) is denoted by \( t_{pd} \), where the two are also of equal magnitude.

Using Eq. (11), let us consider the hopping amplitude at 2nd order, between an electron in the \( d_{xy} \) orbital with spin \( \sigma \) at site \( R \) to an electron occupying the \( d_{yz} \) orbital with spin \( -\sigma \) at site \( R' \). The intermediate oxygen states must contain \( |p_z,-\sigma\rangle \) to have non-zero overlap with the \( |y_z,-\sigma\rangle \) state, leading to the same set of intermediate states as before, however with the time-reversed versions contributing for a given spin \( \sigma \). Therefore, evaluating the sum over the possible intermediate states for a given apical oxygen site, we obtain

\[
\langle y_z,-\sigma,R'|H_{SOC}^{B_{1g}}|xy,\sigma,R\rangle = -\frac{t_{B_{1g}}^{\text{soc}}}{4} \sum_{r} \text{sgn}(t_{pd}^{xy,y}) \langle \eta_\sigma \text{sgn}(t_{pd}^{xy,x}) + \text{sgn}(t_{pd}^{xy,y}) \rangle,
\]

where we have defined \( t_{B_{1g}}^{\text{soc}} = 2\frac{|t_{pd}^{xy,y}||t_{pd}^{xy,x}|}{(E_{pd} + \frac{\lambda_p}{2})(E_{pd} - \lambda_p)} \) and \( \text{sgn}(t_{pd}^{xy,y}) \) is the sign of the hopping between the \( d_{xy} \) and \( p_z/p_y \) orbital. With this general form, let us first consider hopping along the positive \( x \) direction, for which we only have to consider the two intermediate apical oxygen sites above the plane in the \( \pm \hat{y} \) direction, due to the mirror symmetry about the \( xy \) plane, where the apical oxygen above the plane in the \( +\hat{z} + \hat{y} \) direction is shown in Fig. 3. Reflecting in the \( xz \) plane containing the \( d_{xy} \) orbital, \( d_{yz} \), \( d_{xz} \) and \( p_y \) change sign while \( p_z \) and \( p_x \) are invariant, leading to a cancellation of the imaginary term in the sum over the two oxygen sites, since \( \text{sgn}(t_{pd}^{xy,y}) \text{sgn}(t_{pd}^{xy,y}) \rightarrow -\text{sgn}(t_{pd}^{xy,y}) \text{sgn}(t_{pd}^{xy,y}) \), while the real
term is invariant. The hopping in the $-\hat{z}$ direction gives the same result, since $\text{sgn}(t_{\lambda}^{xx})\text{sgn}(t_{\lambda}^{yy,x})$ is even with respect to a reflection in the $yz$ plane containing the $d_{xy}$ orbital. Therefore, after summing over the possible intermediate sites along the $\pm \hat{t}$ direction in Eq. (15), we obtain $\langle yz,-\sigma,R \pm \hat{t}|H_{\text{SOC}}^{B_{1y}}|xy,\sigma,R \rangle = -t_{\lambda}^{B_{1y}}\delta_{\sigma}$. For the hopping along the $\pm \hat{y}$ direction, the procedure is repeated, yielding the same result after summing over the intermediate sites, however there is a relative minus sign due to $t_{\lambda}^{y}$ changing sign as shown in the top square of Fig. 3(b). This relative sign leads to the $d_{x}-y$ dependence, shown below in the effective Hamiltonian for the hopping between the $d_{yz}$ and $d_{xy}$ orbitals in $k$-space,

$$H_{\text{SOC},Y}^{B_{1y}} = 2it_{\lambda}^{B_{1y}} \sum_{\kappa \sigma \sigma' \eta} \sigma y_{\sigma \sigma'} \langle \cos k_{x} - \cos k_{y} \rangle \epsilon_{\kappa \sigma}^{y z} \epsilon_{\kappa \sigma'}^{y z} + h.c.$$  

(16)

It is simple to subsequently obtain the equivalent contribution from the process involving the $d_{xz}$ orbital, either via a $C_{4}$ rotation, or by noting that the effective hopping amplitude is identical to Eq. (15), with the only difference occurring in the summation over the intermediate sites. Therefore we have for the SOC between $d_{xz}$ and $d_{xy}$,

$$H_{\text{SOC},X}^{B_{1y}} = 2it_{\lambda}^{B_{1y}} \sum_{\kappa \sigma \sigma' \eta} \sigma x_{\sigma \sigma'} \langle \cos k_{x} - \cos k_{y} \rangle \epsilon_{\kappa \sigma}^{x z} \epsilon_{\kappa \sigma'}^{x z} + h.c.,$$  

(17)

where it can be noted that combining these terms leads to an overall contribution similar to $\lambda(L_{x}S_{x} + L_{y}S_{y})$, but with $\lambda$ replaced with a $d_{x^{2}-y^{2}}$ form factor.

C. interlayer $E_{g}$

We now proceed to consider an interlayer $k$-SOC as well as interlayer inter-orbital hopping between the $d_{xy}$ and either of the $d_{xz}/d_{yz}$ orbitals. The $k$-SOC is the spin-dependent part of this hopping process that occurs between states with the same spin in adjacent layers, via the apical oxygen sites as shown in Fig. 4(a), which displays the hopping processes for the $d_{xz}/d_{xy}$ case. A schematic illustration of the relevant hopping amplitudes is shown in Fig. 4(b), where here, $t_{\lambda}^{xy}$ is the hopping amplitude between the $d_{xy}$ and $p_{x}$ or $p_{y}$ orbitals in different layers and $t_{\lambda}^{x,y}$ is the hopping amplitude occurring purely in the $\hat{z}$ direction between the $d_{xz}$ orbital and the $p_{x}$ at the apical oxygen site.

Let us consider the matrix element, at 2nd order, between an electron in the $d_{xz}$ orbital with spin $\sigma$ at site $R$ to the $d_{xy}$ orbital with the same spin at site $R'$. The intermediate states that contribute to this amplitude must now contain $[p_{x}, \sigma]$ to have finite overlap with $[xz, \sigma]$ in the $\hat{z}$ direction. After evaluating the sum over the intermediate states, we obtain

$$\langle xy, \sigma,R'|H_{\text{SOC}}^{E_{g}}|xz,\sigma,R \rangle =$$

$$i\text{sgn}(t_{\lambda}^{x y}) \left[ \text{sgn}(t_{\lambda}^{x y, x})(2E_{p} - \lambda_{y}) - i\eta_{y}\text{sgn}(t_{\lambda}^{x y, y})\lambda_{y} \right],$$  

(18)

where we have defined $\tilde{i} = \frac{|t_{\lambda}^{x y, x}|}{2(E_{p} + \frac{\lambda_{y}}{2})(E_{p} - \lambda_{y})}$ and from which we see that the real part gives a spin-independent hopping between the orbitals and the imaginary part gives a spin-dependent hopping. By comparing the relative phases of the wavefunctions in Fig. 4(b), we can see for example that both real and imaginary parts are odd in $z$ since $\text{sgn}(t_{\lambda}^{x y, y})$ is odd with respect to reflection about the $xy$ plane due to the sign change of $d_{xz}$, while $p_{x}$ is even. Furthermore, since the real and imaginary parts are proportional to $\text{sgn}(t_{\lambda}^{x y, x})$ and $\text{sgn}(t_{\lambda}^{x y, y})$ respectively, the real part will be even(odd) in $x(y)$ with the opposite signs for the imaginary part. Therefore, performing the Fourier transform, we obtain the effective Hamiltonian between the $d_{xz}$ and $d_{xy}$ orbitals,

$$H_{\text{SOC},X}^{E_{g}} = -8it_{\lambda}^{x z} \sum_{\kappa \sigma} \frac{\cos k_{x}}{2} \sin \frac{k_{y}}{2} \sin \frac{k_{z}}{2} \epsilon_{\kappa \sigma}^{x z} \epsilon_{\kappa \sigma'}^{x z}$$

$$+ 8it_{\lambda}^{x z} \sum_{\kappa \sigma \sigma'} \frac{k_{x}}{2} \sin \frac{k_{y}}{2} \cos \frac{k_{z}}{2} \epsilon_{\kappa \sigma}^{x z} \epsilon_{\kappa \sigma'}^{x z} + h.c.,$$  

(19)

where it is only the second term that gives an effective
SOC due to the spin dependence, with the first being a spin-independent interlayer hopping. The effective hopping amplitudes are

\[ t_{ij}^{int} = \frac{|t_{ij}^{pd}|^2}{2(\lambda_p + \lambda_d)(\lambda_p - \lambda_d)} \]

\[ t_{soc}^{E_g} = \frac{|t_{ij}^{pd}|^2}{2(\lambda_p + \lambda_d)(\lambda_p - \lambda_d)} \lambda_p. \]

To obtain the equivalent Hamiltonian involving the \( d_{x^2-y^2} \) orbital instead of \( d_{xz} \), it is sufficient to apply a \( C_4 \) rotation. Alternatively, the same procedure can be repeated with the only distinction being that instead of the hopping amplitude \( t_{pd}^{y} \) between \( d_{xz}/p_x \), we have a hopping amplitude \( t_{pd}^{x} \) of the same magnitude between \( d_{yz}/p_y \). This leads to an effective hopping amplitude similar to Eq. (18), but with the sign of the hopping between the \( d_{xy} \) and \( p_x(p_y) \) orbitals controlling the sign of the spin dependent(independent) component. The result is an opposite even/odd momentum dependence with respect to the \( x, y \) directions as for the previous case:

\[ H_{SOC,Y}^{E_g} = -8t_{soc}^{E_g} \sum_{k\sigma} \sin \frac{k_x}{2} \cos \frac{k_y}{2} \sum_{\sigma\sigma'} \sigma' \langle \sigma | k_\sigma c_{k\sigma}^\dagger c_{k_{\sigma'}} | \sigma \rangle \]

\[ + 8i t_{soc}^{E_g} \sum_{k\sigma} \sum_{\sigma\sigma'} \sigma' \langle \sigma | k_\sigma c_{k\sigma}^\dagger c_{k_{\sigma'}} | \sigma \rangle \] (21)

with the total \( E_g \) SOC Hamiltonian therefore given by

\[ H_{SOC}^{E_g} = H_{SOC,X}^{E_g} + H_{SOC,Y}^{E_g}. \]

With a microscopic understanding of these three forms of \( k \)-SOC, we next turn to incorporating them into a more realistic three-orbital model and study the pairing instabilities that arise in \( Sr_2RuO_4 \) when the on-site interactions are included.

IV. APPLICATION TO \( Sr_2RuO_4 \)

Let us therefore discuss the application of the shaded triplet pairing scenario to the unconventional superconductor \( Sr_2RuO_4 \) by performing numerical calculations within MF theory for a realistic three orbital model. While there are various possible order parameters that have been recently proposed to account for the diverse set of experimental data pertaining to \( Sr_2RuO_4 \), there has been a growing body of evidence suggesting that any viable order parameter must be a time-reversal symmetry breaking (TRSB) multi-component order parameter with an appropriate symmetry that will lead to a jump in the \( c_{66} \) elastic modulus at \( T_c \) and lead to a substantial reduction of the nuclear magnetic resonance (NMR) Knight shift for an in-plane field. Two even parity proposals are the multi-component \( \{ d_{xz}, d_{yz} \} \) order parameter in the two-dimensional \( E_g \) representation and the \( d_{x^2-y^2} + ig_{xy}(x^2-y^2) \) order parameter which relies on an accidental degeneracy with components from both the \( B_{1g} \) and \( A_{2g} \) representation. Another possibility which has not received as much consideration however is an order parameter of the form \( s + id_{xy} \), which is a combination of the \( A_{1g} \) and \( B_{2g} \) representations. Such an order parameter is also capable of generating a jump in the shear \( c_{66} \) elastic modulus but not \( (c_{11} - c_{12})/2 \), similar to the \( d_{x^2-y^2} + ig_{xy}(x^2-y^2) \) order parameter. This is in contrast to an order parameter in the \( E_g \) representation, for which there is a jump also in the \( (c_{11} - c_{12})/2 \) elastic modulus, which has not been observed experimentally.

Within a microscopic theory including the local on-site Kanamori interactions, the atomic SOC and the \( B_{2g} \) and \( E_g \) \( k \)-SOCs derived in Sec. III, it is possible to stabilize both order parameters of the \( s + id_{xy} \) or \( d_{xz} + id_{yz} \) type at the FS, depending on the relative size of the \( E_g \), \( B_{2g} \) and atomic SOC strengths. While we did not consider a g-wave \( k \)-SOC in Sec. III, it is straightforward to derive by considering higher order hoppings, which however would lead to a SOC strength smaller than all of the others we have derived and is therefore not considered within our model. As discussed in Sec. II, these order parameters will appear as intraband pseudospin singlets at the FS, but with underlying inter-orbital triplet character originating from the orbital MFs, given more generally here by

\[ \Delta_{ab} = \frac{1}{4N} \sum_{k\sigma \sigma'} \sum_{l \neq a} |a_{\sigma\sigma'}^{l} \rangle \langle a_{\sigma'\sigma}^{l} | \langle c_{a_{\sigma\sigma'}^l \sigma} | c_{b_{\sigma\sigma'}^l \sigma'}^\dagger - c_{b_{\sigma\sigma'}^l \sigma} c_{a_{\sigma\sigma'}^l \sigma'}^\dagger \rangle, \]

with \( l \) indicates the direction of the \( d \)-vector and \( a \neq b \) are the orbital indices representing the \( t_{2g} \) orbitals. The atomic SOC will stabilize MFs of the form \( \Delta_{z2,xy} = \Delta_{x2,xy} > \Delta_{x2,zx} \). The \( B_{2g} \) SOC which is given by Eq. (13) and Eq. (14) will favour an order parameter that appears with \( d_{xy} \) symmetry at the FS, with underlying \( \Delta_{z2,xy} \) and \( \Delta_{x2,xy} \) triplet character, while the \( E_g \) SOC, given by Eq. (19) and Eq. (21), will favour a multi-component order parameter that appears as \( d_{xz} + id_{yz} \) at the FS and \( d_{xz} + id_{yz} \) and have underlying \( \Delta_{z2,xy} \) and \( \Delta_{x2,xy} \) character. By including both the atomic and \( B_{2g} \) SOC, a multi-component order parameter with \( s + id_{xy} \) symmetry at the FS can be stabilized, the relative size of the \( s \) and \( d_{xy} \) components is determined by the relative sizes of the atomic SOC \( \lambda \) and \( t_{soc}^{B_{2g}} \). Furthermore, considering the relative strengths of the intermediate hopping amplitudes making up \( t_{soc}^{B_{2g}} \) and \( E_g \), it is reasonable to expect that the \( s + id_{xy} \) pairing state will be dominant over the \( d_{xz} + id_{yz} \) state. This can be seen by the fact that \( t_{soc}^{E_g} \sim |t_{pd}^{y}| \), which is the hopping between the \( d_{xy} \) orbital in one layer to the apical oxygen’s \( p_x/p_y \) orbitals in a different layer (see Fig. 1), while \( t_{soc}^{B_{2g}} \sim |t_{pd}^{x}| \), which is the hopping between the \( d_{xy} \) orbital to the apical oxygen’s \( p_x \) orbital in the same layer (see Fig. 2).

Thus we propose that an order parameter of the form \( s + id_{xy} \) is plausible from a microscopic viewpoint. We present numerical calculations within our microscopic theory displaying the presence of the \( s + id_{xy} \) order
parameter for reasonable k-SOC parameters, and then
discuss how such an order parameter will differ from the
d_{xz}+id_{yz} state stabilized by the E_g SOC in an
experimental context.

Let us take a three orbital model with a Hamiltonian
$H = H_0 + H_{SOC} + H_{pair}$. The kinetic term, $H_0$, consists
of a tight binding model capable of reproducing the
experimental FS of Sr$_2$RuO$_4$ [11] and is given by

$$H_0 = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}^\sigma} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \sum_{\mathbf{k}\sigma} t_{\mathbf{k}} c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \text{h.c.}, \quad (23)$$

with $a$ the orbital index and the orbital dispersions are,
$\epsilon^x_{\mathbf{k}} = -2t_1 \cos k_y a - 2t_2 \cos k_x a - \mu_{1d} - \mu$, $\epsilon^y_{\mathbf{k}} = -2t_3 (\cos k_x + \cos k_y) - \mu_{xy} - \mu$ and $t_{\mathbf{k}} = -4t_{ab} \sin k_x \sin k_y$. The parameters, in units of 2$t_3$, are fixed at $t_1 = 0.45$, $t_2 = 0.05$, $t_3 = 0.5$, $t_4 = 0.2$, $t_{ab} = 0.025$, $\mu_{1d} = 0.35$, $\mu_{xy} = 0.45$ and the chemical potential $\mu$ is adjusted to fix the filling to $\frac{5}{2}$. The pairing term, $H_{pair}$, consists of the interacting part of the Hamiltonian expressed in the terms of the inter-orbital triplet order parameters,

$$H_{pair} = -2N(3J_H - U) \sum_{a \neq b} \hat{\Delta}_{ab}^\dagger \hat{\Delta}_{ab}, \quad (24)$$

where the sum over the orbital indices is a sum over the
unique pairs of two-orbitals. The SOC part of the Hamiltonian includes the atomic SOC, as well as the B$_{2g}$ and E$_g$ SOC terms introduced in Sec. III,

$$H_{SOC} = i\lambda \sum_{\mathbf{k},\sigma,a} \epsilon_{\mathbf{k}^\sigma} c_{\mathbf{k}\sigma}^\dagger b_{\mathbf{k}\sigma}^c + H_{B_{2g}} + H_{E_g}^{SOC}, \quad (25)$$

where $a,b,c$ are orbital indices.

We use a self-consistent MF theory to calculate the
MFs with the attractive interaction $3J_H - U = 0.7$ and the atomic SOC fixed to $\lambda = 0.05$. Focusing first on
the $s + i\Delta_{xy}$ solution, we set the E$_g$ SOC strength to a small
but microscopically reasonable value, $t_{soc} = 0.1\lambda$. With $t_{B_{2g}}^{soc} = 0$, we obtain a purely s-wave solution with
$\Delta_{zz,xy}^x = \Delta_{yz,xy}^y > \Delta_{xz,xy}^z$. As the B$_{2g}$ SOC is turned on, the $d_{yz}$ component becomes non-zero, with purely imaginary MFs $\Delta_{yz,xy}^y = -\Delta_{yz,xy}^y$. This pairing solution is of the form $s + i\Delta_{xy}$ at the FS and corresponds to an under-laying triplet character with a predominantly in-plane d-vector involving pairing mostly between the $d_{xz}$ and $d_{xy}$ as well as $d_{xz}$ and $d_{xy}$ orbitals. For $t_{soc} \approx 0.78\lambda$, the $d_{xy}$ and $s$ components are approximately equal, representing
an accidental degeneracy in the SOC parameter space. Turning to the $d_{xz}+i\Delta_{xy}$ solution in the E$_g$ representation,
with the B$_{2g}$ SOC set to zero, the $d_{xz}+i\Delta_{xy}$ solution
becomes favourable over the s-wave for $t_{soc} \approx 0.3\lambda$. However,
including the B$_{2g}$ SOC by fixing $t_{soc} = 0.78\lambda$, the critical value at which the solution switches from $s + i\Delta_{xy}$ to $d_{xz} + i\Delta_{xy}$ is increased to $t_{soc} \approx 0.4\lambda$. Nearly half of the on-site SOC is large for an interlayer SOC involving the
two-dimensional $d_{xy}$ and $p_x/p_y$ orbitals and given that the B$_{2g}$ SOC is an intralayer process, it is reasonable that $t_{B_{2g}}^{soc}$ is significantly larger than $t_{E_g}^{soc}$.

Therefore, we take a representative set of parameters
where $t_{B_{2g}}^{soc}$ is significantly larger than $t_{soc}$, for which
the $s + i\Delta_{xy}$ solution is stabilized: $t_{soc}^{B_{2g}} = 0.1\lambda$ and $t_{soc}^{E_g} = 0.78\lambda$. This choice for the SOC parameters leads to a FS matching that obtained from the recent ARPES data [22] and the gap at the FS is shown in Fig. 5 along with arrows indicating the nature of the shadowed triplet at select $\mathbf{k}$ points on the FS. The gap is smallest on the $a$ band at the BZ boundaries, on the order of 1% of the maximum gap or $\approx 1\mu$eV, which is already small compared to the bandwidth and the calculated gap will generally be overestimated within MF theory. Furthermore, these deep minima in the gap are robust to changes in the SOC parameters within the region where the $s + i\Delta_{xy}$ solution is stabilized. The direction of the arrows indicates the in-plane direction of the d-vector, with the length indicating the magnitude of the in-plane component; the shorter the arrow, the bigger the c-axis component. Due to the $d_{xy}$ component vanishing along the $a$ and $b$ directions, the pairing solution is composed of $\Delta_{zz,xy}^x$ along the $a$-axis and $\Delta_{yz,xy}^y$ along the $b$-axis, leading to a d-vector that is parallel to the respective axis. The vanishing of the $d_{xy}$ component of the pairing along the $a/b$
directions is illustrated in the inset of Fig. 5.

V. SUMMARY AND DISCUSSION

In summary, we have studied the microscopic mechanisms of $k$-SOC and its importance for even-parity spin-triplet pairing in Hund's metals. By taking a simple two-orbital model, we show how a purely interband $s$-wave triplet pairing in the orbital basis becomes an intraband pseudospin-singlet pairing with non-trivial momentum dependence at the FS. Within the two-orbital model, we have illustrated how the relevant energy scales for this form of pairing are the orbital polarization and hybridization, which disrupt the pairing, as well as the SOC, which if large enough, allows for the pairing to occur for large orbital polarization and hybridization. With Sr$_2$RuO$_4$ as an example, we have derived several forms of $d$-wave $k$-SOC in the $B_{1g}$, $B_{2g}$ and $E_g$ representations, by including the SOC of the oxygen sites within a model consisting of the $t_{2g}$ orbitals. Such an analysis shows that the dominant form of $k$-SOC will be from the next-nearest-neighbour in-plane $B_{2g}$ SOC, which will generate a $d_{xy}$ pairing component at the FS, in addition to the $s$-wave pairing stabilized by the atomic SOC. Subsequently, we have demonstrated the viability of the $s + id_{xy}$ multi-component solution by including the atomic SOC, $B_{2g}$ and $E_g$ SOCs within a realistic three-orbital model; for realistic values of the three SOC parameters, the $s + id_{xy}$ solution with an in-plane d-vector is favourable over the $d_{xz} + id_{yz}$ out-of-plane d-vector solution.

While the pairing solution we have found manifests as a pseudospin-singlet on the FS, the shadowed triplet nature with a predominantly in-plane d-vector will be apparent in the presence of finite field. As discussed previously in Ref. [19], these properties can be confirmed by NMR under uniaxial strain, with an in-plane field applied along both the direction of the strain and perpendicular to it. The $s + id_{xy}$ state we have presented here has essentially the same property that near the $a/b$ directions, where there is mostly $d_{x^2}/d_{yz}$ and $d_{xy}$ orbital character, the d-vector is parallel to the crystal axes due to the $s$-wave component of pairing and the fact that the $d_{xy}$ component vanishes along those directions. Therefore, under uniaxial strain, there should be a rotation of the average d-vector, leading to an anisotropic Knight shift between the $a$ and $b$ directions when the field is a significant fraction of the gap size, which contrasts with the isotropic singlet response for smaller fields.

Going beyond the purely $s$-wave case, an $s + id_{xy}$ pairing naturally explains experiments suggesting a multi-component order parameter with TRSB and the observed jump in the $c_{66}$ elastic modulus but not the $(c_{11} - c_{12})/2$ modulus. The lack of an observed jump in $(c_{11} - c_{12})/2$ already provides evidence in favour of the $s + id_{xy}$ pairing state over the $d_{xz} + id_{yz}$ state. However, these two pairing states can also be distinguished due to their differing triplet character. The leading order MF for the $d_{xz} + id_{yz}$ solution corresponds to an out-of-plane d-vector, which would yield no rotation under uniaxial strain in contrast to the behaviour of the $s + id_{xy}$ state, as discussed above. Such an experiment could provide further support for an $s + id_{xy}$ order parameter arising from the combination of local interactions and $k$-SOC in light of the lack of many other viable multi-component alternatives.

Beyond the NMR Knight shift experiments under uniaxial strain, it will be important going forward for future experiments to clarify whether the putative gap nodes [20,21] are indeed nodes or deep gap minima, which arise naturally in the pairing solution considered here, as well as the precise location in k-space of these nodes.

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