Pairing symmetry and dominant band in Sr$_2$RuO$_4$

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We study the superconductivity pairing mechanism in Sr$_2$RuO$_4$ in the limit of small interaction by extending a renormalization group calculation developed by Raghu et al. [Phys. Rev. B 81, 224505 (2010)] to include spin-orbit coupling and multi-band effects. We show these effects to be crucial to discriminate between the possible order parameters. In contrast to previous results and without the necessity of fine-tuning, we obtain pseudo-spin triplet gaps of the same order of magnitude on the two-dimensional $\gamma$ band and the quasi-one-dimensional $\alpha$ and $\beta$ bands. The ratio of the gap amplitude on the different bands varies continuously with the interaction parameter. The favoured pairing symmetry is shown to be chiral when $\gamma$ is slightly dominant and helical when $\alpha$ and $\beta$ are slightly dominant.

Strontium ruthenate [1–3] is a layered perovskite material exhibiting a transition at 1.5 K from a well-behaved Fermi liquid to a superconducting phase. Strong experimental evidence points towards an odd-parity order parameter (OP) [4–7]. Based on multiple experiments [7–12], the prevailing candidate for the symmetry of the OP has been the chiral p-wave state, $d = (p_x \pm ip_y)\hat{z}$, which breaks time-reversal symmetry (TRS), hosts topologically protected chiral edge states and is analogous to superfluid $^3$He-A [13, 14] ($d$ is defined below).

On the other hand, this state is supposed to carry edge currents at sample edges and domain walls, which have been elusive so far despite intense scrutiny [15, 16]. As a result, other OP symmetries have been considered theoretically [17–20], including the helical states, $d = p_x\hat{x} \pm p_y\hat{y}$ and $d = p_y\hat{x} \pm p_x\hat{y}$. These phases can be viewed as time-reversal invariant versions of chiral superconductors. Their edges host two counter-propagating Majorana modes of opposite spin whose net charge current is zero. Since most of the interest in Sr$_2$RuO$_4$ is driven by its potential use as a resource towards topological quantum computation, the identification of its topological properties is of prime importance.

Another controversy has arisen recently regarding the band(s) on which the superconducting instability is dominant. The Fermi surface (FS) of Sr$_2$RuO$_4$ is made of three cylindrical sheets: the $\gamma$ band is mainly derived from the Ru $4d_{xy}$ orbital and is fairly isotropic in the basal plane, while the $\alpha$ and $\beta$ bands are mainly derived from the Ru $4d_{xz}$ and $4d_{yz}$ orbitals and are quasi-one-dimensional (see Fig. 1).

The prevailing assumption in the field has been that $\gamma$ is the active band, due to its proximity to a Van Hove singularity. This assumption was based on specific heat data [21] and backed by several calculations [22–26] that predicted a dominant gap on $\gamma$ and a sub-dominant, near-nodal gap on $\alpha$ and $\beta$.

This scenario was challenged recently. First, Raghu et al. [27] (see also [28]) showed that, in absence of band coupling and in the weak coupling limit, $\alpha$ and $\beta$ are the active bands. Second, Firmo et al. [29] reported a phenomenological model with a gap amplitude of similar size on the three bands but slightly larger on $\alpha$ and $\beta$ than on $\gamma$ that is consistent with specific heat and scanning tunnelling microscopy (STM) measurements.

In this letter, we report the extension of Raghu et al.’s renormalization group (RG) scheme [30–34] to include spin-orbit coupling and multi-band effects. This enables us to study the orientation of $d$ at a microscopic level and determine the gap on the three bands. We find similarly sized gaps on the three bands without the necessity of fine-tuning. Depending on the interaction parameter, we find two OPs that are compatible with the thermodynamic data: either a chiral gap whose amplitude is slightly larger on $\gamma$, or a helical gap whose amplitude is slightly larger on $\alpha$ and $\beta$.

The three bands of strontium ruthenate are reproduced using the following tight-binding Hamiltonian for electrons hopping on a square lattice [35]

$$H = \sum_{k,s} \psi_s^\dagger(k) \hat{H}_s(k) \psi_s(k)$$

(1)

where $\psi_s(k) = [c_{k,A,s}; c_{k,B,s}; c_{k,C,-s}]^T$ with $s = 1$ (-1) for up (down) spins. The matrix $\hat{H}_s(k)$ is given by

$$\hat{H}_s(k) = \begin{pmatrix} E_A(k) & g(k) - i\eta & in \hline g(k) + i\eta & E_B(k) & -s\eta \hline -i\eta & -s\eta & E_C(k) \end{pmatrix}$$

(2)
where \( E_A(k) = -2t \cos(k_x) - 2t' \cos(k_y) - \mu \), \( E_B(k) = -2t' \cos(k_x) - 2t \cos(k_y) - \mu \), \( E_C(k) = -2t'' \cos(k_x) + 4t' \cos(k_y) - 4t'' \cos(k_x) \cos(k_y) - \mu_c \) and \( g(k) = -4t'' \sin(k_x) \sin(k_y) \). \( A, B, C \) stand for the Ru orbitals 4d_{xz}, 4d_{yz}, 4d_{xy} on each lattice site. The spin-orbit coupling (SOC) parameter is \( \eta \) and the inter-orbital hopping term is \( g(k) \). The parameters were chosen to fit the electronic band structure calculated with LDA \( ^{36,37} \): in dimensionless units, \( (t, t', t'', \mu, \mu_c, \mu_v, \eta) = (1, 0.1, 0.8, 0.3, 1.0, 0.1, 0.01, 0.1) \).

After diagonalization, we obtain three pairs of degenerate pseudo-spin bands:

\[
H = \sum_{\mathbf{k}, \alpha, \sigma} \epsilon_{\mathbf{k},\alpha,\sigma} \hat{c}_{\mathbf{k},\alpha,\sigma}^\dagger \hat{c}_{\mathbf{k},\alpha,\sigma} \tag{3}
\]

with \( \sigma = 1 \) (1) for + (-) pseudo-spin and \( \alpha = \alpha, \beta, \gamma \). Roman indices refer to spin and orbital space while Greek indices refer to pseudo-spin and band space.

We study the multi-orbital on-site Coulomb interaction \( ^{20} \):

\[
H_{\text{int}} = \sum_{i,a} U n_{ia\uparrow} n_{ia\downarrow} + \sum_{i,a \neq b} \frac{V^\dagger_{ia\uparrow}}{2} \sum_s n_{ias} n_{ibs} \nonumber \\
+ \sum_{i,a \neq b} \frac{V^\dagger_{ia\uparrow}}{2} \sum_s n_{ias} n_{ibs} - \sum_{i,a \neq b} \frac{J}{2} \sum_s \hat{c}_{ias} \hat{c}_{ia\sigma} \hat{c}_{ibs} \hat{c}_{ib\bar{\sigma}} \nonumber \\
- \sum_{i,a \neq b} \frac{J}{2} \sum_s \hat{c}_{ias} \hat{c}_{ia\sigma} \hat{c}_{ibs} \hat{c}_{ib\bar{\sigma}} \tag{4}
\]

where \( i \) is the site index, \( a = A, B, C \) is the orbital index, \( s \equiv -s, V^\dagger = U - 2J, V = U - 3J \) and \( n_{ias} \equiv \hat{c}_{ias}^\dagger \hat{c}_{ias} \).

Following Raghu et al. \( ^{30} \), we treat the weak-coupling limit, which corresponds to \( U, J \ll W \) where \( W \) is the bandwidth and \( J/U \) a finite constant that fully parametrizes the interaction. Despite the fact that \( U \) and \( J \) are actually of the same order as \( W \), considering an RG which is controlled in this limit is justified by the fact that the superconducting instability in \( \text{Sr}_2\text{RuO}_4 \) emerges out of a well-behaved Fermi liquid, and at a much smaller scale since \( T_c \ll E_f \). We integrate out all the modes with energies greater than an artificial cutoff to derive the effective particle-particle interaction in the Cooper channel \( V(k_a, q_\beta) \), where \( \epsilon_{\alpha}(k_a) \) lies below the cut-off.

The effective interaction \( V(k_a, q_\beta) \) corresponds to the diagram depicted in Fig. 2(a). Its pseudo-spin dependence is left implicit for now. Besides the bare vertex and its ladder, which give a trivial repulsive contribution, the effective interaction at one-loop order is made of the three diagrams shown in Fig. 2(b). These diagrams are expressed in terms of the static susceptibility of the non-interacting system and correspond to the celebrated “Kohn-Luttinger” physics \( ^{38,39} \). The different bare vertices given in Eq. (4) are represented diagrammatically by a unique dashed line that corresponds to a matrix in spin and orbital space. As the external propagators are in pseudo-spin and band space, the diagram expressions are supplemented by form factors from the unitary transformation going from spin and orbital to pseudo-spin and band space.

The second stage of the weak-coupling analysis is the calculation of the RG flow \( ^{30} \). Each eigenmode of the effective interaction flows independently under the evolution of the running cutoff. These eigenmodes are solutions of

\[
\sum_{\beta} \int_{SF} \frac{dq_\beta}{S_F} g(k_a, q_\beta) \psi(q_\beta) = \lambda \psi(k_a) \tag{5}
\]

where

\[
g(k_a, q_\beta) = \sqrt{\rho \frac{v_{F,a}}{v_F(k_a)}} V(k_a, q_\beta) \sqrt{\rho \frac{v_{F,\beta}}{v_F(q_\beta)}}, \tag{6}
\]

\( S_F \) is the “area” of the FS, \( \rho \) is the density of states (DOS) of the band \( \alpha \) at the Fermi level and the average of the norm of the Fermi velocity is given by

\[
\frac{1}{v_{F,\alpha}} = \int d\mathbf{k}_a \frac{1}{v_F(k_a)} v_F(k_a)^{-1}. \tag{7}
\]

Since \( k_a \) and \( q_\beta \) are constrained to lie on their respective FS, Eq. (5) is solved in matrix form once the FSs are discretized.

The energy scale at which the perturbative treatment of the interaction breaks down corresponds to the critical temperature and is given by \( ^{30} \)

\[
T_c \sim W \exp \left( \frac{-1}{|\lambda|} \right). \tag{8}
\]

The gap is proportional to the eigenvector\( ^{30} \):

\[
\Delta(k_a) \sim \left( \frac{v_F(k_a)}{v_{F,\alpha} \rho_\alpha} \right) \psi(k_a). \tag{9}
\]

The pseudo-spin dependence of the order parameter is written in matrix form:

\[
\Delta(k_a) = \begin{pmatrix} \Delta_{++} & \Delta_{+-} \\ \Delta_{-+} & \Delta_{--} \end{pmatrix} = \begin{pmatrix} -d_x + id_y & d_z + \Delta_x \\ d_z - \Delta_x & d_x + id_y \end{pmatrix}, \tag{10}
\]
which defines a scalar order parameter $\Delta_x$ for the singlet case and a vectorial order parameter $\mathbf{d}$ for the triplet case. Since they are respectively even and odd under inversion, these two cases are mutually exclusive. The direction of $\mathbf{d}$ defines the normal to the plane in which the electrons are equal pseudo-spin paired.

The order parameter has to be in a given irreducible representation of the crystal symmetry group $D_{4h}$. The odd-parity representations can be split into two groups: the chiral state $\mathbf{d} = (p_x + ip_y)\hat{z}$ and the helical states $\mathbf{d} = p_x\hat{x} \pm p_y\hat{y}$ and $\mathbf{d} = p_y\hat{x} \pm p_x\hat{y}$. The symbols $p_{x,y}$ stand for any function of momentum that has the same properties as $\sin(k_{x,y})$ under the symmetry operations of $D_{4h}$. The unit vectors $\hat{x}, \hat{y}, \hat{z}$ are the directions $a [100], b [010]$ and $c [001]$. The representation with the most negative pairing eigenvalue $\lambda$ corresponds to the favoured state.

Since there is no consensus regarding the value of the interaction parameters, we will study a priori the whole acceptable range of $J/U$ and then compare predictions with experiments to infer its possible value. The singlet case appears only for $J/U > 0.29$ and can be discarded based on multiple measurements [4–7]. While, for $J/U < 0.065$, the chiral state is favoured in agreement with the most prevailing assumption in the field, the helical state $\mathbf{d} = p_x\hat{x} \pm p_y\hat{y}$ takes over for $0.065 < J/U < 0.29$. The helical state is the 2D equivalent of superfluid $^3$He-B [10].

The TRS obeyed by the helical state is in contradiction with muon spin relaxation [8] and optical Kerr effect [10] experiments but the interpretation of these experiments appears to conflict with the absence of edge currents [14,17]. The absence of spin susceptibility decrease below $T_c$ for both in and out-of-plane fields measured by NMR Knight shift experiments [14,15] has been interpreted as evidence in favour of a weakly pinned $\mathbf{d} \parallel c$ that can be rotated to the plane by a field $h \parallel c$ smaller than 20 mT. We emphasize that a helical state with a weakly pinned $\mathbf{d} \perp c$ that would be rotated by a field $h \parallel ab$ smaller than 150 mT would also be consistent with these experiments.

Furthermore, the helical state would provide a simple explanation for the presence of edge states [44] but the absence of edge currents [13,16]. It would also explain the emergence of out-of-plane spin fluctuations in the superconducting state [45], which require in-plane fluctuations of $\mathbf{d}$. The disappearance of these fluctuations under an in-plane magnetic field would also be consistent with the expulsion of $\mathbf{d}$ from the plane under such a field. Half-quantum vortices, measured recently in a mesoscopic sample of Sr$_2$RuO$_4$ [10], correspond to a spatially dependent rotation of $\mathbf{d}$ in order to accommodate a half-integer flux. They require a freeing of $\mathbf{d}$ from its intrinsic direction imposed by SOC and their existence is therefore equally plausible in the chiral and the helical state. Given these contradictory experimental results, we will study these two states on an equal footing.

Once the mode with the most negative eigenvalue is identified, its eigenvector provides valuable information regarding the gap. The gap scale is too small to be measured directly by angle-resolved photoemission spectroscopy (ARPES) but specific heat measurements have revealed properties of the order parameter [29,47]. In Fig. 3 we compare the measured [48] critical jump in specific heat $\Delta C/C$ with its value calculated using BCS theory on the gap functions obtained from the RG technique. The two highlighted regions correspond to a prediction for $\Delta C/C$ in agreement with experiments: the chiral OP at $J/U \approx 0.06$ and the helical OP at $J/U \approx 0.08$. The departure of $\Delta C/C$ from its well-known BCS maximal value of 1.43 measures the anisotropy of the gap over the three FS. A large difference between the scale of the gap amplitudes on the different bands corresponds to a $\Delta C/C$ that is smaller than experiments, as can be seen on Fig. 3. Accordingly, the two predicted OPs in agreement with specific heat data have gaps of the same order on the three bands. The slightly dominant band is different in the two cases: the chiral state has a gap approximately two times larger on $\gamma$ than on $\alpha$ and $\beta$, while the ratio of the helical gap amplitude on $\gamma$ over the one on $\alpha$ and $\beta$ is approximately 0.7. We checked that both these states give rise to a $T$ linear dependence of $C/T$ below $T_c$, in agreement with experiments [48]. By tuning $J/U$ towards smaller values, it is possible to obtain a largely dominant gap on $\gamma$ like previously reported [22,26].

As shown on Fig. 4, the gaps on $\alpha$ and $\beta$ present near-nodes near the direction $[110]$ in both cases. The incommensurate peak $\mathbf{Q}$ in the antiferromagnetic fluctuation

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig3}
\caption{Critical specific heat jump $\Delta C/C$ and ratio of the maxima of the gap amplitudes over the different bands $R = \max\left|\frac{\Delta c}{\Delta s}\right|$. The vertical line separates the stability regions of the chiral and helical OPs. The curve for a given OP is drawn in full width only in the OP’s stability region. The horizontal lines delimit the range of $\Delta C/C$ estimated from experiments: $\Delta C/C = 0.75 \pm 0.05$ [29,48]. The braces indicate the range of $J/U$ for which the prediction is in agreement with experiments.}
\end{figure}
The rationale behind the association of the chiral state with \( \gamma \) and the helical state with \( \alpha \) and \( \beta \) is the anisotropy of the normal-state spin dynamics. The chiral (helical) state has an out-of-plane (in-plane) \( d \) and is therefore driven by in-plane (out-of-plane) magnetic fluctuations. Due to SOC, the incommensurate peak \( Q \) is larger for the out-of-plane component of the susceptibility \([50]\), thereby favouring a helical state on the quasi-1D bands. On the other hand, the (ferromagnetic) long wave-length part of the spectrum is larger for the in-plane component, which favours a chiral state on \( \gamma \).

By a microscopic accounting of multi-band and SOC effects, our model reconciles the two distinct scenarios of 2D superconductivity on \( \gamma \) versus quasi-1D superconductivity on \( \alpha \) and \( \beta \) inside one framework. The presence of similarly sized gaps on all three bands requires no fine tuning and, depending on the interaction parameter, the balance can be slightly tilted one way or another. As shown on Fig. 4, this result is true for both the chiral and the helical state and is therefore robust regardless of the favored pairing symmetry.

We now discuss experiments probing the relative size of the gaps on the different bands. Recently, out-of-plane STM \([29]\) has exhibited the presence of a near-nodal gap of 0.350 meV on \( \alpha \) and \( \beta \). We find a position for the near-nodes on \( \alpha \) and \( \beta \) that is consistent with their phenomenological model, and we could reproduce their experimental tunnelling DOS curves based on our gap. Due to orbital anisotropy, the gap on \( \gamma \) cannot be measured with such an experiment. The fact that the measured gap size corresponds to \( 2\Delta/T_c \approx 5 \), which is close to the BCS value, was interpreted as evidence that \( \alpha \) and \( \beta \) are dominant. A gap 0.7 times smaller on \( \gamma \) was then inferred from the specific heat jump value, in agreement with our findings for the helical state.

On the other hand, the conductance of in-plane tunneling junctions \([44]\) has been reported to present a two-step peak shape that is consistent with a dominant gap of 0.93 meV on \( \gamma \) and a subdominant gap of 0.28 meV on \( \alpha \) and \( \beta \). The relative sizes of the gap amplitude on the different bands would then point towards the chiral scenario.

Since they quantify the SOC and the inter-orbital hopping, the inclusion of the parameters \( \eta \) and \( t'' \) are crucial in the modelling of \( \text{Sr}_2\text{RuO}_4 \) \([37] \ [51]\). In this perspective, we checked the robustness of our conclusions: a 50\% increase or decrease of these parameters does not alter the results qualitatively.

Finally, we emphasize the need for new experiments that would make it possible to discriminate between the two proposed states. In-plane STM could be one of them since it could also measure the gap on \( \gamma \) unlike in the out-of-plane case. Experiments probing the phase of the order parameter, including quasi-particle interference and Josephson tunnelling spectroscopy \([7] \ [11] \ [12]\), could be discriminating but their interpretation is non-trivial given the reported convoluted dependence of that phase on the in-plane orientation. Methods to detect helical edge modes have also been proposed recently \([52]\).
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[51] Since both these parameters create repulsion between the bands, there is some freedom in their choice. We chose them so as to fit the band structure from LDA without SOC for η = 0 and LDA with SOC for η ≠ 0 [36]. Our value of t′′′ is therefore smaller than in calculations with-
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