Fermi Surface of Sr$_2$RuO$_4$: Spin-Orbit and Anisotropic Coulomb Interaction Effects

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The topology of the Fermi surface of Sr$_2$RuO$_4$ is well described by local-density approximation calculations with spin-orbit interaction, but the relative size of its different sheets is not. By accounting for many-body effects via dynamical mean-field theory, we show that the standard isotropic Coulomb interaction alone worsens or does not correct this discrepancy. In order to reproduce experiments, it is essential to account for the Coulomb anisotropy. The latter is small but has strong effects; it competes with the Coulomb-enhanced spin-orbit coupling and the isotropic Coulomb term in determining the Fermi surface shape. Its effects are likely sizable in other correlated multiorbital systems. In addition, we find that the low-energy self-energy matrix—responsible for the reshaping of the Fermi surface—sizable differs from the static Hartree-Fock limit. Finally, we find a strong spin-orbital entanglement; this supports the view that the conventional description of Cooper pairs via factorized spin and orbital part might not apply to Sr$_2$RuO$_4$.

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Sr$_2$RuO$_4$ has attracted a lot of attention as a possible realization of a spin-triplet superconductor [1–4] and, at the same time, as a very peculiar strongly correlated metal [5–13]. Understanding the details of its Fermi surface (FS) is key to unraveling the nature of quasielectrons in the normal phase and can cast light on the mechanisms and the symmetry of the superconducting order parameter. It is thus not surprising that the Fermi surface of Sr$_2$RuO$_4$ has been intensively investigated, both experimentally [14–20] and theoretically [21–23]. Although the main features are nowadays well understood, the effects of the interplay between correlations, spin-orbit, and crystal structure have not been fully disentangled yet.

Sr$_2$RuO$_4$ is a tetragonal layered perovskite (space group $I4/mmm$ [24]) with the Ru $4d^5$ ($t_{2g}^3e_g^0$) electronic configuration and Ru atoms at sites with $D_{4h}$ symmetry; due to the layered structure the Ru $t_{2g}$, $xz$ and $yz$ bands are almost one dimensional and very narrow, with a bandwidth $W_{xz} = W_{yz}$ about half as large as that of the two-dimensional Ru $xy$ band, $W_{xy}$. Experimentally, the Fermi surface of Sr$_2$RuO$_4$ has been studied via both the de Haas–van Alphen technique [14–16] and angle-resolved photoemission spectroscopy (ARPES) [17–20]. It is made (Fig. 1) by three sheets, the electronlike $\gamma$ ($xy$ band) and $\beta$ ($xz$, $yz$ bands) sheets and the holelike $\alpha$ sheet ($xz$, $yz$ bands). Theoretically, \textit{ab initio} calculations based on the local-density approximation (LDA) qualitatively reproduce the FS topology, provided that the spin-orbit (SO) interaction is taken into account [22,23]. Indeed, several experiments point to a sizable SO coupling [1,25,26]. These calculations fail, however, in describing the relative size of the sheets, suggesting that perhaps many-body effects play a key role. The relevance of the Coulomb interaction for the electronic properties of Sr$_2$RuO$_4$, as well as its interplay with bands of different width, was shown early on via model many-body studies [21]. More recently, LDA+DMFT (local-density approximation + dynamical mean-field theory) calculations have emphasized the interplay of Coulomb interactions and spin-orbit coupling. In this Letter, we show that, by accounting for the anisotropic Coulomb interaction, all important features of Sr$_2$RuO$_4$’s FS are reproduced. In addition to the SO interaction and the Coulomb renormalization, we find that the low-energy self-energy matrix is strongly anisotropic and responsible for the reshaping of the Fermi surface.

FIG. 1. Fermi surface ($k_z = 0$) of Sr$_2$RuO$_4$ from (a) LDA, (b) LDA + SO, (c) LDA + DMFT, and (d) LDA + SO + DMFT calculations performed with $O(3)$-symmetric Coulomb matrix, $(U, J) = (3.1, 0.7)$ eV, $T \to 0$ limit. (Light lines) $\alpha$ and $\beta$ sheets. (Dark lines) $\gamma$ sheet. (Gray density maps) Experimental data taken from Ref. [17].
interaction and $t_{2g}$ crystal field (CF) [9,27], and the role of the Hund’s rule coupling [10]. LDA + slave-boson calculations point to SO effects on the correlated bands [28]. It remains, however, unclear to what extent many-body effects actually modify the Fermi surface, and how they compete with other effects.

In this Letter, by using the LDA + DMFT method with SO interaction, we investigate, for the first time, the interplay between Coulomb repulsion, spin-orbit, and symmetry at the Fermi surface in a realistic setting. We show that, surprisingly, the standard isotropic Coulomb interaction alone [$O(3)$ symmetry] does not improve (or even worsens) the agreement between theoretical and experimental Fermi surface. The agreement of independent Coulomb parameters increases to six.

First, let us analyze the LDA results without SO interaction. For a free atom the Coulomb interaction tensor for $d$ states can be written in terms of the three Slater integrals $F_0$, $F_2$, and $F_4$. For $t_{2g}$ states the essential terms [38] are the direct $[U_{mm'm''} = U_{m,m'} = U - 2J(1 - \delta_{m,m'})]$, and the exchange $[U_{mm'm''m'''} = J]$ screened Coulomb interaction, the pair-hopping $[U_{mm'm''m''''} = J]$ and the spin-flip term $[U_{mm'm''m''''} = J]$; here $U = F_0 + \frac{4}{7}(F_2 + F_4)$ and $J = \frac{1}{2}(3F_2 + \frac{8}{7}F_4)$. For site symmetry $D_{4h}$ the number of independent Coulomb parameters increases to six.

Here we will discuss, in particular, the effect of $\Delta U = U_{xy,xy} - U_{xz,xz}$ and $\Delta U' = U_{xy,xy} - U_{xz,xz}$. We solve (1) with DMFT using the CT-INT QMC method. We work with a $6 \times 6$ self-energy matrix $\Sigma_{mm',m''}(\omega) = \Sigma'_{mm',m''}(\omega) + i\Sigma''_{mm',m''}(\omega)$ in spin-orbital space, extending the solver of Ref. [9] to deal explicitly with the SO term; $\Sigma'$ is the real and $\Sigma''$ the imaginary part of the self-energy. The calculations with SO coupling are performed in the basis $|m\rangle_\sigma = \tilde{T}|m\rangle_\sigma$, where the unitary operator $\tilde{T}$ is chosen such that the local imaginary-time Green function matrix is real. In the rest of the Letter, for calculations with SO coupling, the elements of the self-energy matrix are given in the $|m\rangle_\sigma$ basis; since $\tilde{T}$ only changes the phases [39] but does not mix orbitals, we rename for simplicity $|m\rangle_\sigma$ as $|m\rangle_\sigma$.

First, let us analyze the LDA results without SO interaction [Fig. 1(a)]. Our results agree very well with previous theoretical works [22,23,40,41]. Compared with ARPES data, LDA describes well the $\alpha$ and $\gamma$ sheets, and in particular the region around the $M$ point of the $\gamma$ sheet. There are two major discrepancies. First, the LDA $\beta$ and $\gamma$ sheets cross, differently than in ARPES. Second, the area enclosed by the $\beta$ sheet is larger in LDA than in ARPES.
Once the SO interaction is switched on three relevant changes occur, as Fig. 1(b) shows. The $\beta - \gamma$ crossing becomes an anticrossing due to the SO coupling $\lambda_{xy}$; the $\beta$ sheet shrinks and the $\gamma$ sheet expands. These effects improve the overall agreement [22,23,26] with ARPES results; however, the $\beta$ sheet remains too large with respect to experiments.

The next step consists in incorporating the Coulomb interaction via LDA + DMFT [see Fig. 1(c)]. First, we perform standard calculations with no SO term and $O(3)$-symmetric Coulomb tensor. We use two sets of parameters: $(U, J) = (3.1, 0.7)$ eV, as obtained via constrained LDA [42], and $(U, J) = (2.3, 0.4)$ eV, as obtained via constrained random-phase approximation [43] (CRPA). These sets of values yield spectral functions with Hubbard bands in line with available experiments [7,42,44,45]. Low-energy many-body effects change the splitting $\epsilon_{\text{CF}}$ into $\epsilon_{\text{CF}} + \Delta\epsilon_{\text{CF}}$, with [46]

$$
\Delta\epsilon_{\text{CF}} = \frac{1}{2} \sum_{\sigma\gamma\sigma'\gamma'} \langle 0 | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | 0 \rangle + \frac{1}{2} \sum_{\sigma\gamma\sigma'\gamma'} \langle 0 | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | 0 \rangle.
$$

The shift $\Delta\epsilon_{\text{CF}}$ turns out to be positive [21]; at $T = 290$ K we find $\Delta\epsilon_{\text{CF}} = 108$ meV for $(U, J) = (3.1, 0.7)$ eV and 80 meV for $(U, J) = (2.3, 0.4)$ eV. As a consequence, with respect to LDA, the $\alpha$ and $\gamma$ sheets expand whereas the $\beta$ sheet shrinks. This is shown in Fig. 1(c) for $(U, J) = (3.1, 0.7)$ eV; the LDA + DMFT Fermi surface deviates from ARPES in particular around the $M$ point ($\gamma$ sheet), which approaches the boundary of the first Brillouin zone. For $(U, J) = (2.3, 0.4)$ eV the effect is smaller and the FS remains closer to the LDA one [47].

In Fig. 1(d) we show the effect of including the SO term (LDA + SO + DMFT). We find a $\Delta\epsilon_{\text{CF}}$ slightly smaller than for $\lambda_{i} = 0$; the SO couplings are, however, sizably enhanced with respect to LDA, i.e., $\lambda_{i} \rightarrow \lambda_{i} + \Delta\lambda_{i}$, with

$$
\Delta\lambda_{\gamma} = -\sum_{\gamma} \langle \gamma | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | \gamma \rangle + \sum_{\gamma} \langle \gamma | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | \gamma \rangle,
$$

$$
\Delta\lambda_{xy} = \frac{1}{2} \sum_{\sigma} \langle \sigma | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | \sigma \rangle - \sum_{\sigma} \langle \sigma | \hat{\Sigma}_{\gamma\sigma,\gamma'\sigma'}(0) | \sigma \rangle.
$$

At 290 K we obtain $\Delta\lambda_{xy} \simeq 96$ meV and $\Delta\lambda_{\gamma} \simeq 88$ meV. For the FS, with respect to LDA + DMFT, the agreement worsens for the $\gamma$ sheet and it improves for the $\alpha$ and $\beta$ sheets. The change can be ascribed to the enhanced SO couplings. Comparing Figs. 1(b) and 1(c) with Fig. 1(d) it appears that the combined effect of Coulomb and SO interaction results in the $\gamma$ sheet approaching the boundary of the first Brillouin zone. For $(U, J) = (2.3, 0.4)$ eV the effects of the SO coupling are qualitatively similar [47]. These results point to the existence of an important mechanism neglected so far.

We identify the missing mechanism in low-symmetry Coulomb terms. Because of the elongation of the RuO bond in the $c$ direction, the $e_{g}$ ($xz, yz$) Wannier orbitals have a larger spread than $xy$ orbital [48], suggesting positive $\Delta U$ and $\Delta U'$. This is in line with the results of CRPA, $\Delta U \sim 0.3$ eV [43]. To study the effect of the Coulomb anisotropy we perform two additional sets of LDA + SO + DMFT calculations, the first with $0 < \Delta U < 0.6$ eV and $\Delta U' = 0$ and the second with $0 < \Delta U' < \Delta U/3 < 0.2$ eV [49]. The most significant results are shown in Fig. 2 for $T = 290$ K [50]. We find that both $\Delta\lambda_{\gamma}$ and $\Delta\lambda_{xy}$ are weakly dependent on $\Delta U$. Instead, $\Delta\epsilon_{\text{CF}}$ decreases linearly with $\Delta U$ and changes sign at a quite small $\Delta U \sim 0.25$ eV; at this value the effective CF has the LDA value [51]. As a consequence, the area enclosed by the $\gamma$ sheet decreases as well. In Fig. 3 we present the same quantities shown in Fig. 2, however, as a function of the temperature $T$; we find that the tetragonal SO splitting $|\Delta\delta_{z}|$ increases on lowering $T$, while $|\Delta\epsilon_{\text{CF}}|$ decreases slightly. In comparison with the strong dependence of $\Delta\epsilon_{\text{CF}}$ with $\Delta U$, all parameters change weakly on lowering $T$ [52].

Remarkably, these effects are to a large extent dynamical in nature [47,53]. The zero-frequency crystal-field enhancement is given by $\Delta\epsilon_{\text{CF}} = \Delta\Sigma'(\infty) + (1/\pi) \int d\omega \Delta\Sigma''(\omega) / \omega$. The term $\Delta\Sigma'(\infty)$ can be obtained via the static mean-field Hartree-Fock method; in the $\Delta U = \Delta U' = 0$ case one can show that $\Delta\Sigma'(\infty) \sim \frac{1}{2} (U - 5J)p$, where $p = n_{xz} - n_{yz}$ is the orbital polarization. Because of the bandwidth mismatch [9] the LDA total polarization is $p \sim -0.17$, i.e., negative, despite of the positive CF splitting; in LDA + DMFT it becomes basically zero, hence $\Delta\Sigma'(\infty) \sim 0$ as well. The enhancement $\Delta\epsilon_{\text{CF}} > 0$ comes thus essentially from the second term; it turns out, by analyzing the integrand $\Delta\Sigma''(\omega) / \omega$, that it has large contributions from the lower Hubbard bands. The SO interaction

![FIG. 2. Many-body corrections of the on-site parameters at the Fermi energy as a function of $\Delta U$, with $\Delta U' = \Delta U/3$. The LDA + SO + DMFT calculations are done at $T = 290$ K and for $(U, J) = (3.1, 0.7)$ eV. (Top) Spin-orbit couplings corrections, $\Delta\lambda_{\gamma}$ and $\Delta\lambda_{xy}$. (Bottom) Crystal-field splitting correction, $\Delta\epsilon_{\text{CF}}$. The LDA values $\lambda_{\gamma}$, $\lambda_{xy}$, and $\epsilon_{\text{CF}}$ are indicated by arrows. QMC error bars are shown.](106402-3)
does not affect much the CF splitting, but it slightly increases the initial orbital polarization, from \( p = -0.17 \) (LDA) to \( p = -0.19 \) (LDA + SO); furthermore, it couples the \( ej \) and \( b_{2g} \) orbitals, yielding a negative SO polarization \( p_j \sim -0.10 \), with \( p_j = n_{3/2} - n_{1/2} \), where \( n_j \) is the average occupation of an orbital with total angular momentum \( j \); switching on the Coulomb interaction reduces the orbital polarization \( p \sim 0 \) and slightly increases \( p_j \sim -0.12 \). Finally, when \( \Delta U > 0 \), electrons are transferred from the \( xy \) to the \( xz \) and \( yz \) bands as \( \Delta \epsilon_{CF} \) decreases, yielding a negative orbital polarization \( p \sim -0.11 \) for \( \Delta U = 0.45 \text{ eV} \) \([54]\).

Returning to the FS, we find that the agreement between calculations and experiments can only be recovered if both low-symmetry Coulomb terms and correlation-enhanced SO couplings are included in the calculations. To show this and test the robustness of our conclusion, in addition to LDA + SO + DMFT calculations for \( \Delta U = 0.3 \text{ eV} \) (CRPA estimate) we perform a series of model calculations. For the latter we take \( \Delta \epsilon_{CF} \) in the interval \([-0.08, -0.02] \text{ eV} \), \( \Delta \lambda_{xy} \) and \( \Delta \lambda_z \) in the intervals \([0.10,0.16] \text{ eV} \) and \([0.04,0.08] \text{ eV} \) \([55]\). These intervals estimate the possible input parameters variations and are chosen around the results in Fig. 2 for \( 0.3 \text{ eV} \leq \Delta U \leq 0.45 \text{ eV} \). In this realistic parameter range the theoretical FS is in very good agreement with experiments \([56]\), as shown in a representative case in Fig. 4.

Our results have consequences concerning the nature of Cooper pairs. It is often assumed that Cooper pairs can be classified as singlets or triplets \([1,3,4]\). Recently, it was pointed out that in \( \text{Sr}_2\text{RuO}_4 \) this scenario might break down due to the SO interaction \([26]\). Indeed, already in LDA the SO coupling is comparable with the crystal-field splitting. Turning on the Coulomb interaction, for \( \Delta U > 0 \) we find that the ratio \( \lambda_\gamma/\epsilon_{CF} \) becomes even larger than \( \lambda_\alpha/\epsilon_{CF} \). This points to a strong spin-orbital entanglement, which should not be neglected in studying the nature of Cooper pairs, as suggested in Refs. \([26,57]\).

In conclusion, we investigate in a realistic setting how different mechanisms affect the topology of the Fermi surface of \( \text{Sr}_2\text{RuO}_4 \). LDA calculations with spin-orbit effects describe well the topology of the Fermi surface, but not the relative size of the Fermi sheets. We show that adding alone the effects of the standard isotropic Coulomb interaction via dynamical mean-field theory does not improve (or even worsens) the agreement with experiments. It is essential to also include the small anisotropic part of the Coulomb interaction. Remarkably, we find that (small) low-symmetry Coulomb terms have a large effect at the Fermi surface. The standard isotropic Coulomb interaction enhances the crystal-field splitting and the spin-orbit coupling. The Coulomb-enhanced spin-orbit coupling shrinks the \( \beta \) sheet and extents the \( \gamma \) sheet. The low-symmetry Coulomb term \( \Delta U \) reduces the Coulomb crystal-field enhancement, modifying correspondingly the \( \alpha \) and \( \gamma \) sheets. To reproduce the experimental Fermi surface all these interactions are essential. Our results support the recent suggestions of strong spin-orbital entanglement for Cooper pairs. These mechanisms could be at work also in other multiorbital correlated systems: other layered metallic ruthenates, iridates, or iron-based superconductors.

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[36] Projectors select Wannier functions of $t_{2g}$ symmetry. A similar procedure is used in R. Sakuma, Phys. Rev. B 87, 235109 (2013). Since the occupied O$p$ states have been integrated out, the $t_{2g}$ Wannier functions have $p$ tails of the proper symmetry on the neighboring O sites.
[37] For $H_\Delta$, we adopt an around mean-field-like form, a typical choice for correlated metals.
[39] For Sr$_2$RuO$_4$, the transformation merely amounts to an extra $(-1)^n\pi$ phase for the $|xz\rangle$ orbital.
[46] We extrapolate the zero-frequency limit (shown in Figs. 2 and 3) from the values of the self-energy at the first Matsubara frequency and by analytical continuation of the self-energy; the two approximations yield similar results.
[48] The spread $\langle |r|^2 \rangle$ is $3.79$ Å$^2$ for the $xz$ and $3.55$ Å$^2$ for the $xy$ orbital.
[49] The effect of $\Delta U'$, which modifies interorbital Coulomb terms, is weaker than that of the intraorbital correction $\Delta U$, hence the results of the two sets of calculations are similar. Thus, only results for $\Delta U' = \Delta U/3$ are shown.
[50] For $(U, J) = (2.3, 0.4)$ eV the changes with respect to calculations with isotropic $U$ are smaller.
[51] For isotropic Coulomb interaction, $H_{dc}$ is a mere shift of the chemical potential and has no effect on the parameters.
Instead, for finite $\Delta U$, $H_{dc} \propto \Delta U$. A positive (negative) $\delta H_{dc}$ yields correspondingly a positive (negative) $\delta \Delta \epsilon_{CF}$; the other parameters are little affected by $\delta H_{dc}$. For realistic $H_{dc}$ the change $\delta \Delta \epsilon_{CF}$ is, however, small; an increase of $H_{dc}$ of, e.g., 9%, yields for $\Delta U = 3\Delta U'' = 0.45$ eV a $\Delta \epsilon_{CF} \sim -57$ meV (instead of $-83$ meV). Thus, the main effect of a $\delta H_{dc} \sim \pm 9\% H_{dc}$ is equivalent to that of a shift of a few tenths of meV on the right or left (depending on the sign of $\delta H_{dc}$) of the value of $\Delta U$ for which the best agreement with experiments is reached. For comparison, $|\delta H_{dc}| \sim 12\% H_{dc}$ is the difference between the around-mean-field limit and the fully localized limit with $n_{xy} = 1, n_{xz} + n_{yz} = 3$. Hence, our conclusions remain unaffected by $\delta H_{dc}$ unless $H_{dc}$ becomes unrealistically large, yielding a large orbital polarization, to the best of our knowledge never reported experimentally.

[52] This, together with the Luttinger theorem, allows the $T \to 0$ extrapolation. The strongest temperature dependence is perhaps shown by $\Delta \delta \epsilon_{CF}$.

[53] For the CF, model calculations [21] yield a similar conclusion. For the SO couplings, a dynamical $\Delta \delta \epsilon_{CF}$ has been obtained for the hybridization function of insulating Sr iridates, see H. Zhang, K. Haule, and D. Vanderbilt, Phys. Rev. Lett. 111, 246402 (2013).

[54] Thus, neither the SO interaction nor the crystal field splitting dominate in determining the most occupied states.

[55] In line with our LDA + SO + DMFT results, we assume that also for the model calculations the system is a Fermi liquid satisfying the Luttinger theorem.

[56] The agreement remains good provided that $\Delta \epsilon_{CF}$ is at least 100 meV smaller than the value obtained with isotropic $U$ and that the SO couplings are correspondingly sufficiently enhanced by Coulomb repulsion. It starts to visibly deteriorate for, e.g., $\Delta \epsilon_{CF} \sim +0.03$ eV.