High-resolution photoemission on Sr$_2$RuO$_4$ reveals correlation-enhanced effective spin-orbit coupling and dominantly local self-energies

A. Tamai, ¹ M. Zingl,² E. Rozbicki,³ E. Cappelli,¹ S. Riccò,¹ A. de la Torre,¹ S. McKeown Walker,¹ F. Y. Bruno,¹ P.D.C. King,³ W. Meevasana,⁴ M. Shi,⁵ M. Radović,⁵ N.C. Plumb,⁵ A.S. Gibbs,⁴⁺ A.P. Mackenzie,⁵,⁺ C. Berthod,² H. Strand,² M. Kim,⁷,⁺ A. Georges,⁹,²,⁶,⁺ and F. Baumberger¹,⁺ ¹Department of Quantum Matter Physics, University of Geneva, 24 Quai Ernest-Ansermet, 1211 Geneva 4, Switzerland ²Center for Computational Quantum Physics, Flatiron Institute, 162 5th Avenue, New York, NY 10010, USA ³SUPA, School of Physics and Astronomy, University of St Andrews, St Andrews, Fife KY16 9SS, United Kingdom ⁴School of Physics, Suranaree University of Technology and Synchrotron Light Research Institute, Nakhon Ratchasima, 30000, Thailand and Thailand Center of Excellence in Physics, CHE, Bangkok, 10400, Thailand ⁵Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland ⁶Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany ⁷Department of Physics and Astronomy, Rutgers, The State University of New Jersey, Piscataway, NJ 08854, USA ⁸Centre de Physique Théorique Ecole Polytechnique, CNRS, Universite Paris-Saclay, 91128 Palaiseau, France ⁹College de France, 11 place Marcelin Berthelot, 75005 Paris, France (Dated: December 18, 2018)

I. INTRODUCTION

The layered perovskite Sr$_2$RuO$_4$ is an important model system for correlated electron physics. Its intriguing superconducting ground state, sharing similarities with superfluid $^3$He [1–3], has attracted much interest and continues to stimulate advances in unconventional superconductivity [4]. Experimental evidence suggest odd-parity spin-triplet pairing, yet questions regarding the proximity of other order parameters, the nature of the pairing mechanism and the apparent absence of the predicted self-energies for the $\beta$ and $\gamma$ sheets are found to display significant angular dependence. By taking into account the multi-orbital composition of quasiparticle states, we determine self-energies associated with each orbital component directly from the experimental data. This analysis demonstrates that the perceived angular dependence does not imply momentum-dependent many-body effects, but arises from a substantial orbital mixing induced by spin-orbit coupling. A comparison to single-site dynamical mean-field theory further supports the notion of dominantly local orbital self-energies, and provides strong evidence for an electronic origin of the observed non-linear frequency dependence of the self-energies, leading to ‘kinks’ in the quasiparticle dispersion of Sr$_2$RuO$_4$.

Quantum oscillation and angle-resolved photoemission spectroscopy (ARPES) measurements [24–34] further reported a strong enhancement of the quasiparticle effective mass over the bare band mass. Theoretical progress has been made recently in revealing the important role of the intra-atomic Hund’s coupling as a key source of correlation effects in Sr$_2$RuO$_4$ [18, 20, 35]. In this context, much attention was devoted to the intriguing properties of the unusual state above $T_{FL}$, which displays metallic transport with no signs of resistivity saturation at the Mott-Ioffe-Regel limit [36]. Dynamical mean-field theory (DMFT) [37] calculations have proven successful in explaining several properties of this intriguing metallic state, as well as in elucidating the crossover from this unusual metallic state into the Fermi-liquid regime [13, 18, 20–23, 38]. Within DMFT, the self-energies associated with each orbital component are assumed to be local. On the other hand, the low-temperature Fermi-liquid state is known to display strong magnetic fluctuations at specific wave-vectors, as revealed, e.g., by neutron scattering [39, 40] and nuclear magnetic resonance spectroscopy (NMR) [41, 42]. These magnetic fluctuations were proposed early on to be an important source of correlations [2, 43, 44]. In this picture, it is natural to expect strong momentum depen-
dence of the self-energy associated with these spin fluctuations. Interestingly, a similar debate was raised long ago in the context of liquid \(^3\)He, with ‘paramagnon’ theories emphasizing ferromagnetic spin-fluctuations and ‘quasilocalized’ approaches à la Anderson-Brinkman emphasizing local correlations associated with the strong repulsive hard-core, leading to increasing Mott-like localization as the liquid is brought closer to solidification (for a review, see Ref. [45]).

In this work, we report on new insights into the nature of the Fermi-liquid state of Sr\(_2\)RuO\(_4\). Analyzing a comprehensive set of laser-based ARPES data with improved resolution and cleanliness, we reveal a strong angular (i.e., momentum) dependence of the self-energies associated with the quasiparticle bands. We demonstrate that this angular dependence originates in the variation of the orbital content of quasiparticle states as a function of angle, and can be understood quantitatively. Introducing a new framework for the analysis of ARPES data for multi-orbital systems, we extract the electronic self-energies associated with the three Ruthenium \(t_{2g}\) orbitals with minimal theoretical input. We find that these orbital self-energies have strong frequency dependence, but surprisingly weak angular (i.e., momentum) dependence, and can thus be considered local to a very good approximation. Our results provide a direct experimental demonstration that the dominant effects of correlations in Sr\(_2\)RuO\(_4\) are weakly momentum-dependent and can be understood from a local perspective, provided they are considered in relation to orbital degrees of freedom.

One of the novel aspects of our work is to directly put the locality ansatz underlying DMFT to the experimental test. We also perform a direct comparison between DMFT calculations and our ARPES data, and find good agreement with the measured quasiparticle dispersions and angular dependence of the effective masses.

The experimentally determined real part of the self-energy displays strong deviations from the low-energy Fermi-liquid behavior \(\Sigma(\omega) \sim \omega(1 - 1/Z) + \cdots\) for binding energies \(|\omega|\) larger than \(\sim 20\) meV. These deviations are reproduced by our DMFT calculations suggesting that the cause of these non-linearities are local electronic correlations. Our results thus call for a revision of earlier reports of strong electron-lattice coupling in Sr\(_2\)RuO\(_4\) [29–31, 46–49]. We finally quantify the effective spin-orbit coupling (SOC) strength and confirm its enhancement due to correlations predicted theoretically [21, 23, 50].

This article is organized as follows. In Sec. II, we briefly present the experimental method and report our main ARPES results for the Fermi surface and quasiparticle dispersions. In Sec. III, we introduce the theoretical framework on which our data analysis is based. In Sec. IV, we use our precise determination of the Fermi surface to reveal the correlation-induced enhancement of the effective SOC. In Sec. V we proceed with a direct determination of the self-energies from the ARPES data. Sec. VI presents the DMFT calculations in comparison to experiments. Finally, our results are critically discussed and put in perspective in Secs. VII, VIII.

II. EXPERIMENTAL RESULTS

A. Experimental methods

The single crystals of Sr\(_2\)RuO\(_4\) used in our experiments were grown by the floating zone technique and showed a superconducting transition temperature of \(T_c = 1.45\) K. ARPES measurements were performed with an MBS electron spectrometer and a narrow bandwidth 11 eV (113 nm) laser source from Lumeras that was operated at a repetition rate of 50 MHz with 30 ps pulse length of the 1024 nm pump [51]. All experiments were performed at \(T \approx 5\) K using a cryogenic 6-axes sample goniometer, as described in Ref. [52]. A combined energy resolution of 3 meV was determined from the width of the Fermi-Dirac distribution measured on a polycrystalline Au sample held at 4.2 K. The angular resolution was \(\approx 0.2^\circ\). In order to suppress the intensity of the surface layer states on pristine Sr\(_2\)RuO\(_4\) [53], we exposed the cleaved surfaces to \(\approx 0.5\) L CO at a temperature of \(\approx 120\) K. Under these conditions, CO preferentially fills surface defects and subsequently replaces apical oxygen ions to form a Ru–COO carboxylate in which the C end of a bent CO\(_2\) binds to Ru ions of the reconstructed surface layer [54].

B. Experimental Fermi surface and quasiparticle dispersions

In Fig. 1 we show the Fermi surface and selected constant energy surfaces in the occupied states of Sr\(_2\)RuO\(_4\). The rapid broadening of the excitations away from the Fermi level seen in the latter is typical for ruthenates and implies strong correlation effects on the quasiparticle properties. At the Fermi surface, one can readily identify the \(\alpha, \beta\) and \(\gamma\) sheets that were reported earlier [25, 27, 28]. However, compared with previous ARPES studies we achieve a reduced line width and improved suppression of the surface layer states giving clean access to the bulk electronic structure. This is particularly evident along the Brillouin zone diagonal (ΓX) where we can clearly resolve all band splittings.

In the following, we will exploit this advance to quantify the effects of SOC in Sr\(_2\)RuO\(_4\) and to provide new insight into the renormalization of the quasiparticle excitations using minimal theoretical input only. To this end we acquired a set of 18 high resolution dispersion plots along radial \(k\)-space lines (as parameterized by the angle \(\theta\) measured from the ΓM direction). The subset of data shown in Fig. 2 (a) immediately reveals a rich behavior with a marked dependence of the low-energy dispersion on the Fermi surface angle \(\theta\). Along the ΓM
high-symmetry line our data reproduce the large difference in Fermi velocity \( v^{\beta,\gamma}_F \) for the \( \beta \) and \( \gamma \) sheet, which is expected from the different cyclotron masses deduced from quantum oscillations [12, 25, 26] and was reported in earlier ARPES studies [33, 55]. Our systematic data, however, reveal that this difference gradually disappears towards the Brillouin zone diagonal (\( \theta = 45^\circ \)), where all three bands disperse nearly parallel to one another. In Sec. IV we will show that this equilibration of the Fermi velocity can be attributed to the strong effects of SOC around the zone diagonal.

To quantify the angle dependence of \( v^{\beta,\gamma}_F \) from experiment, we determine the maxima \( k^{\beta,\gamma}_{\text{max}}(\omega) \) of the momentum distribution curves (MDCs) over the range of 2-6 meV below the Fermi level \( E_F \) and fit these \( k \)-space loci with a second-order polynomial. We then define the Fermi velocity as the derivative of this polynomial at \( E_F \). This procedure minimizes artifacts due to the finite energy resolution of the experiment. As shown in Fig. 2 (c), the Fermi velocities \( v^{\beta,\gamma}_F \) obtained in this way show an opposite trend with azimuthal angle for the two Fermi sheets. For the \( \beta \) band we observe a gentle decrease of \( v_F \) as we approach the \( \Gamma X \) direction, whereas for \( \gamma \) the velocity increases by more than a factor of two over the same range [56]. This provides a first indication for a strong momentum dependence of the self-energies \( \Sigma_{\beta,\gamma}' \), which we will analyze quantitatively in Sec.V. Here, we limit the discussion to the angle dependence of the mass enhancement \( v_{\beta}/v_F \), which we calculate from the measured quasiparticle Fermi velocities of Fig. 2 (c) and the corresponding velocities of a reference Hamiltonian \( H^0 \) defined in Sec. IV. As shown in Fig. 2 (d), this confirms a substantial many-body effect on the anisotropy of the quasiparticle dispersion. Along \( \Gamma M \), we find a strong differentiation with mass enhancements of \( \approx 5 \) for the \( \gamma \) sheet and \( \approx 3.2 \) for \( \beta \), whereas \( v_{\beta}/v_F \) approaches a common value of \( \approx 4.4 \) for both sheets along the Brillouin zone diagonal.

Before introducing the theoretical framework used to quantify the anisotropy of the self-energy and the effects of SOC, we compare our data quantitatively to bulk sensitive quantum oscillation measurements. Using the experimental Fermi wave vectors \( k_F \) and velocities determined from our data on a dense grid along the entire Fermi surface, we can compute the cyclotron masses measured by dHvA experiments, without relying on the approximation of circular Fermi surfaces and/or isotropic Fermi velocities used in earlier studies [33, 49, 55, 57]. Expressing the cyclotron mass \( m^* \) as

\[
m^* = \frac{\hbar^2}{2\pi} \frac{\partial A_{FS}}{\partial \epsilon} = \frac{\hbar^2}{2\pi} \int_0^{2\pi} k_F(\theta) \frac{\partial \epsilon}{\partial k(\theta)} d\theta, \tag{1}
\]

where \( A_{FS} \) is the Fermi surface volume, and using the data shown in Fig. 2 (c), we obtain \( m^*_\beta = 17.3(2.0) m_e \) and \( m^*_\gamma = 6.1(1.0) m_e \), in quantitative agreement with the values of \( m^*_\beta = 16 m_e \) and \( m^*_\gamma = 7 m_e \) found in dHvA experiments [12, 25, 26]. We thus conclude that the quasiparticle states probed by our experiments are
III. THEORETICAL FRAMEWORK

The goal of this work is to extract directly from the ARPES data some key information about quasiparticle properties. We are in particular interested in: (i) a precise determination of the Fermi surface, (ii) the quasiparticle velocities and their renormalization by electronic correlations, and (iii) a direct determination of the electronic self-energy.

In order to define the latter and assess the effect of electronic correlations we need to specify a one-particle Hamiltonian \( \hat{H}^0 \) as a reference point. At this stage, we keep the presentation general, but the particular choice of \( \hat{H}^0 \) will be a focus of Sec. IV. The eigenstates \( |\psi_\nu(k)\rangle \) of \( \hat{H}^0(k) \) at a given quasi-momentum \( k \) and the corresponding eigenvalues \( \varepsilon_\nu(k) \) define the ‘bare’ band structure of the system, with respect to which the self-energy \( \Sigma_{\nu\nu'}(\omega,k) \) is defined from the interacting Green’s function

\[
G_{\nu\nu'}^{-1}(\omega,k) = [\omega + \varepsilon_\nu(k)] \delta_{\nu\nu'} - \Sigma_{\nu\nu'}(\omega,k). \tag{2}
\]

In this expression \( \nu \) and \( \nu' \) label the bands and \( \omega \) denotes the binding energy counted from \( E_F \), which we set to zero throughout the rest of this work. The interacting value of the chemical potential \( \mu \) sets the total electron number. Since \( \mu \) can be conventionally included in \( \hat{H}^0 \), we shall omit it in the following. The electronic spectral function is related to the Green’s function

\[
A_{\nu\nu'}(\omega,k) = \frac{i}{2\pi} [G_{\nu\nu'}(\omega,k) - G_{\nu'\nu}(\omega,k)]. \tag{3}
\]

It is this quantity which is most directly related to the ARPES signal: within the sudden approximation, in the absence of final-state interactions, the ARPES intensity is given by \( \sum_{\nu\nu'} M_{\nu} M_{\nu}' \langle \nu | \hat{A} \hat{A}' | \nu' \rangle f(\omega) \) convolved with a function representing the experimental resolution and assuming an average over the polarization. Here, \( M \) is a matrix element and \( f(\omega) \) is the Fermi function. It is important to note that \( G, \Sigma \) and \( A \) are in general non-diagonal matrices.

The Fermi surface of the system is the locus of zero-energy excitations, and hence corresponds to the momenta \( k_F \) which are the solutions of

\[
\det \left[ \varepsilon_\nu(k_F) \delta_{\nu\nu'} + \Sigma_{\nu\nu'}(0,k_F) \right] = 0. \tag{4}
\]

The dispersion relations of the quasiparticles are obtained as the solutions \( \omega = \omega_0^p(k) \) of

\[
\det \left[ (\omega - \varepsilon_\nu(k)) \delta_{\nu\nu'} - \Sigma_{\nu\nu'}(\omega,k) \right] = 0. \tag{5}
\]

In the above equations \( \Sigma' \) denotes the real part of the self-energy. Its imaginary part \( \Sigma'' \) has been neglected, i.e., we assume that quasiparticles are coherent with a lifetime much longer than \( 1/\omega_0^p \). This assumption gradually breaks down as one departs from the Fermi level or as temperature is increased.

A. Localized orbitals and electronic structure

Let us recall some of the important aspects of the electronic structure of Sr_2RuO_4. As shown in Sec. II B, three bands, commonly denoted \( \nu = \{a, \beta, \gamma\} \), cross the Fermi level. These bands correspond to states with \( t_{2g} \) symmetry deriving from the hybridization between localized Ru-4d \( (d_{xy}, d_{yz}, d_{zx}) \) orbitals and O-2p states. Hence, we introduce a localized basis set of \( t_{2g} \)-like orbitals \( |\chi_m\rangle \), with basis functions conveniently labeled as \( m = \{xy, yz, xz\} \).
In practice, we use maximally localized Wannier functions [59, 60] constructed from the Kohn-Sham eigenbasis of a non-SOC density functional theory (DFT) calculation (see appendix B 1 for details). We term the corresponding Hamiltonian $\hat{H}^{\text{DFT}}$. It is important to note that the choice of a localized basis set is not unique and other ways of defining these orbitals are possible (see, e.g., Ref. [61]).

In the following this set of orbitals plays two important roles. First, they are atom-centered and provide a set of states localized in real-space $|\chi_m(R)\rangle$. Secondly, the unitary transformation matrix to the band basis $|\psi_\nu(k)\rangle$

$$U_{m\nu}(k) = \langle \chi_m(k) | \psi_\nu(k) \rangle,$$

allows us to define an ‘orbital’ character of each band $\nu$ as $|U_{m\nu}(k)|^2$. In the localized-orbital basis the one-particle Hamiltonian is a non-diagonal matrix, which reads

$$\hat{H}_{mm'}^{\text{DFT}}(k) = \sum_\nu U_{m\nu}(k) \varepsilon_\nu(k) U_{m'\nu}(k)^\dagger.$$  

The self-energy in the orbital basis is expressed as

$$\Sigma_{mm'}(\omega, k) = \sum_{\nu\nu'} U_{m\nu}(k) \Sigma_{\nu\nu'}(\omega, k) U_{m'\nu'}(k)^\dagger,$$

and conversely in the band basis as

$$\Sigma_{\nu\nu'}(\omega, k) = \sum_{mm'} U_{m\nu}(k) \Sigma_{mm'}(\omega, k) U_{m'\nu'}(k)^\dagger.$$



B. Spin-orbit coupling

We treat SOC as an additional term to $\hat{H}^{\text{DFT}}$, which is independent of $k$ in the localized-orbital basis, but leads to a mixing of the individual orbitals. The single-particle SOC term for atomic $d$-orbitals projected to the $t_{2g}$-subspace reads [62]

$$\hat{H}_\lambda^{\text{SOC}} = \frac{\lambda}{2} \sum_{mm'\sigma\sigma'} C^{\dagger}_{m\sigma} (I_{mm'} \cdot \sigma_\sigma') C_{m'\sigma'},$$

where $I$ are the $t_{2g}$-projected angular momentum matrices, $\sigma$ are Pauli matrices and $\lambda$ will be referred to in the following as the SOC coupling constant. As documented in appendix B 1, the eigenenergies of a DFT+SOC calculation are well reproduced by $\hat{H}^{\text{DFT}} + \hat{H}_\lambda^{\text{SOC}}$ with $\lambda_{\text{DFT}} = 100$ meV.

IV. ENHANCED EFFECTIVE SPIN-ORBIT COUPLING AND SINGLE-PARTICLE HAMILTONIAN

The importance of SOC for the low-energy physics of Sr$_2$RuO$_4$ has been pointed out by several authors [8, 17, 21, 23, 31, 63–67]. SOC lifts degeneracies found in its absence and causes a momentum dependent mixing of the orbital composition of quasiparticle states, which has non-trivial implications for superconductivity [8, 17, 67]. Signatures of SOC have been detected experimentally on the Fermi surface of Sr$_2$RuO$_4$ in the form of a small protrusion of the $\gamma$ sheet along the zone diagonal [31, 65] and as a degeneracy lifting at the band bottom of the $\beta$ sheet [67]. These studies reported an overall good agreement between the experimental data and the effects of SOC calculated within DFT [31, 65, 67]. This is in apparent contrast to more recent DMFT studies of Sr$_2$RuO$_4$, which predict large but frequency independent off-diagonal contributions to the local self-energy that can be interpreted as a contribution $\Delta \lambda$ to the effective coupling strength $\lambda_{\text{eff}} = \lambda_{\text{DFT}} + \Delta \lambda$ [21, 23], consistent with general perturbation-theory considerations [50].

In the absence of SOC, DFT yields a quasi-crossing between the $\beta$ and $\gamma$ Fermi surface sheets a few degrees away from the zone diagonal, as displayed on Fig. 3 (a). Near such a point we expect the degeneracy to be lifted by SOC, leading to a momentum splitting $\Delta k = \lambda_{\text{eff}}/v$ and to an energy splitting of $\Delta E = Z\lambda_{\text{eff}}$ [23], as depicted schematically in Fig. 3 (e). In these expressions, $v = \sqrt{\beta \gamma Z}$, with $\beta$ and $\gamma$, the bare band velocities in the absence of SOC and correlations, and $Z \equiv \sqrt{\beta \gamma Z}$ involves the quasiparticle residues $Z_{\nu}$ associated with each band (also in the absence of SOC).

It is clear from these expressions that a quantitative determination of $\lambda_{\text{eff}}$ is not possible from experiment alone. Earlier studies on Sr$_2$RuO$_4$ [67] and iron-based superconductors [68], have interpreted the energy splitting $\Delta E$ at avoided crossings as a direct measure of the SOC strength $\lambda_{\text{eff}}$. However, in interacting systems $\Delta E$ is not a robust measure of SOC since correlations can both enhance $\Delta E$ by enhancing $\lambda_{\text{eff}}$ and reduce it via the renormalization factor $Z$. We thus quantify the enhancement of SOC from the momentum splitting $\Delta k$, which is not renormalized by the quasiparticle residue $Z$. The experimental splitting at the avoided crossing between the $\beta$ and $\gamma$ Fermi surface sheets indicated in Fig. 3 (a) is $\Delta k_{\text{QP}} = 0.094(9)$ Å$^{-1}$ whereas DFT predicts $\Delta k_{\text{DFT+SOC}} = 0.046$ Å$^{-1}$. We thus obtain an effective SOC strength $\lambda_{\text{eff}} = \lambda_{\text{DFT}} \Delta k_{\text{QP}} / \Delta k_{\text{DFT+SOC}} = 205(20)$ meV, in quantitative agreement with the predictions in Refs. [21, 23]. We note that despite this large enhancement of the effective SOC, the energy splitting remains smaller than $\lambda_{\text{DFT}}$ as illustrated in Fig. 3 (e). When deviations from linearity in band dispersions are small, the splitting $\Delta E$ is symmetric around the $E_F$ and can thus be determined from the occupied states probed in experiment. Direct inspection of the data in Fig. 3 (f) yields $\Delta E \approx 70$ meV, which is about 2/3 of $\lambda_{\text{DFT}}$ and thus clearly not a good measure of SOC.

The experimental splitting is slightly larger than that expected from the expression $\Delta E = Z \lambda_{\text{eff}}$ and our theoretical determination of $Z_{\beta}$ and $Z_{\gamma}$ at the Fermi surface. This can be attributed to the energy dependence of $Z$, which, in Sr$_2$RuO$_4$, is not negligible over the en-
energy scale of SOC. Note that the SOC-induced splitting of the bands at the \( \Gamma \) point reported in Ref. [67] can also be explained by the competing effects of enhancement by correlations and reduction by the quasiparticle weight as shown in Ref. [23]. We also point out that the equilibration of quasiparticle velocities close to the diagonal, apparent from Figs. 2 (a,c) and 3 (f) is indeed the behavior expected close to an avoided crossing [23].

Including the enhanced SOC determined from this non-crossing gap leads to a much improved theoretical description of the entire Fermi surface [69]. As shown in Fig. 3 (b), our high-resolution experimental Fermi surface deviates systematically from a DFT calculation with SOC. Most notably, \( \hat{H}_{\mathrm{DFT}}+\hat{H}_{\mathrm{SOC}}^{\lambda} \) underestimates the size of the \( \gamma \) sheet and overestimates the \( \beta \) sheet. Intriguingly, this is almost completely corrected in \( \hat{H}_{\mathrm{DFT}}+\hat{H}_{\mathrm{SOC}}^{\lambda+\Delta\lambda} \), with \( \lambda_{\mathrm{DFT}}+\Delta\lambda = 200 \text{ meV} \), as demonstrated in Fig. 3 (c). However, a close inspection shows that the remaining discrepancies between experiment and \( \hat{H}_{\mathrm{DFT}}+\hat{H}_{\mathrm{SOC}}^{\lambda+\Delta\lambda} \) break the crystal symmetry, suggesting that they are dominated by experimental artifacts. A likely source for these image distortions is imperfections in the electron optics arising from variations of the work function around the electron emission spot on the sample. Such distortions can presently not be fully eliminated in low-energy photoemission from cleaved single crystals.

Importantly, the change in Fermi surface sheet volume with the inclusion of \( \Delta\lambda \) is not driven by a change in the crystal field splitting between the \( xy \) and \( xz, yz \) orbitals (see appendix B). The volume change occurs solely because of a further increase in the orbital mixing induced by the enhanced SOC. As shown in Fig. 3 (g), the mixing is not limited to the vicinity of the avoided crossing but extends along the entire Fermi surface. For \( \lambda_{\mathrm{DFT}}+\Delta\lambda \) we find a minimal \( d_{xy} \) and \( d_{xz, yz} \) for the \( \gamma \) and \( \beta \) bands of 20/80% along the \( \Gamma M \) direction with a monotonic increase to \( \approx 50\% \) along the Brillouin zone diagonal \( \Gamma X \). We note that this mixing varies with the perpendicular momentum \( k_z \). However, around the experimental value of \( k_z \approx 0.4 \pi/c \) the variation is weak [70]. The analysis presented here and in Secs. III A and VI is thus robust with respect to a typical uncertainty in \( k_z \). These findings suggest that a natural reference single-particle Hamiltonian is \( \hat{H}_0=\hat{H}_{\mathrm{DFT}}+\hat{H}_{\mathrm{SOC}}^{\lambda+\Delta\lambda} \). This choice ensures that the Fermi surface of \( \hat{H}_0 \) is very close to that of the interacting system. From Eq. 4, this implies that the self-energy matrix approximately vanishes at zero binding energy: \( \Sigma_{\nu'\nu}(\omega = 0, \mathbf{k}) \approx 0 \). We choose \( \hat{H}_0 \) in this manner in all the following. Hence, from now on \( |\psi_\nu(\mathbf{k})\rangle \) and \( \varepsilon_\nu(\mathbf{k}) \) refer to the eigenstates and band structure of \( \hat{H}_0=\hat{H}_{\mathrm{DFT}}+\hat{H}_{\mathrm{SOC}}^{\lambda+\Delta\lambda} \). We point out that although \( \hat{H}_0 \) is a single-particle Hamiltonian, the effective enhancement \( \Delta\lambda \) of SOC included in \( \hat{H}_0 \) is a correlation effect beyond DFT.
V. EXPERIMENTAL DETERMINATION OF SELF-ENERGIES

A. Self-energies in the quasiparticle/band basis

Working in the band basis, i.e., with the eigenstates of $\hat{H}_0$, the maximum of the ARPES intensity for a given binding energy $\omega$ (maximum of the MDCs) corresponds to the momenta $k$ which satisfy (following Eq. 5):

$$\omega - \varepsilon_\nu(k) - \Sigma_\nu(\omega,k) = 0.$$

Hence, for each binding energy, each azimuthal cut, and each sheet of the quasiparticle dispersions, we fit the MDCs and determine the momentum $k_{\nu \text{max}}(\omega)$ at their maximum. Using the value of $\varepsilon_\nu(k_{\nu \text{max}}(\omega))$ at this momentum yields the following quantity

$$\omega - \varepsilon_\nu(k_{\nu \text{max}}(\omega)) = \Sigma_\nu(\omega,k_{\nu \text{max}}(\omega)) \equiv \Sigma_\nu(\omega,\theta).$$

This equation corresponds to the simple construction illustrated graphically in Fig. 4 (b), and it is a standard way of extracting a self-energy from ARPES, as used in previous works on several materials [32, 48, 71–73]. We note that this procedure assumes that the off-diagonal components $\Sigma_{\nu\neq\nu'}(\omega,k)$ can be neglected for states close to the Fermi surface (i.e., for small $\omega$ and $k$ close to a Fermi crossing). This assumption can be validated, as shown in appendix C. When performing this analysis, we only include the $\alpha$ sheet for $\theta = 45^\circ$. Whenever the constraint $\Sigma_\nu(\omega \to 0) \to 0$ on the self-energy is not precisely obeyed, a small shift is applied to set it to zero. We chose this procedure to correct for the minor differences between the experimental and the reference $\hat{H}_0$ Fermi wave-vectors because we attribute these differences predominantly to experimental artifacts.

The determined self-energies for each band $\nu = \alpha, \beta, \gamma$ and the different values of $\theta$ are depicted in Fig. 4 (a,c). For the $\beta$ and $\gamma$ sheets they show a substantial dependence on the azimuthal angle. Around $\Gamma M$ we find that $\Sigma_\gamma$ exceeds $\Sigma_\beta$ by almost a factor of two (at $\omega = -50\text{meV}$), whereas they essentially coincide along the zone diagonal ($\Gamma X$). This change evolves as a function of $\theta$ and occurs via a simultaneous increase in $\Sigma_\beta$ and a decrease in $\Sigma_\gamma$ for all energies as $\theta$ is increased from $0^\circ$ ($\Gamma M$) to $45^\circ$ ($\Gamma X$). In order to better visualize this angular dependence, a compilation of $\Sigma_\nu(\omega)$ for different values of $\theta$ is displayed in Fig. 4 (c).

B. Accounting for the angular dependence: local self-energies in the orbital basis

In this section, we introduce a different procedure for extracting self-energies from ARPES, by working in the orbital basis $|\chi_m(k)\rangle$. We do this by making two key
assumptions:

1. We assume that the off-diagonal components are negligible, i.e., \( \Sigma'_{m\neq m'} \simeq 0 \). Let us note that in Sr₂RuO₄ even a \( \mathbf{k} \)-independent self-energy has non-zero off-diagonal elements if \( \hat{H}^{\text{DFT}} + \hat{H}^{\text{SOC}} \) is considered. Using DMFT, these off-diagonal elements have been shown to be very weakly dependent on frequency in this material [23], leading to the notion of a static correlation enhancement of the effective SOC (\( \Delta \Lambda \)). In the present work, these off-diagonal frequency-independent components are already incorporated into \( \hat{H}^0 \) (see Sec. IV), and thus the frequency-dependent part of the self-energy is (approximately) orbital diagonal by virtue of the tetragonal crystal structure.

2. We assume that the diagonal components of the self-energy in the orbital basis depend on the momentum \( \mathbf{k} \) only through the azimuthal angle \( \theta \): \( \Sigma''_{mm}(\omega, \mathbf{k}) \simeq \Sigma''_m(\omega, \theta_k) \). We neglect the dependence on the momentum which is parallel to the angular cut.

Under these assumptions, the equation determining the quasiparticle dispersions reads

\[
\det \left[ \left( \omega - \Sigma'_m(\omega, \theta_k) \right) \delta_{mm'} - \hat{H}^0_{mm'}(\mathbf{k}) \right] = 0. \tag{12}
\]

In this equation, we have neglected the lifetime effects associated with the imaginary part \( \Sigma''_m \). In order to extract the functions \( \Sigma'_m(\omega, \theta_k) \) directly from the ARPES data, we first determine the peak positions \( k^\text{max}_{\beta}(\omega, \theta) \) for MDCs at a given angle \( \theta \) and binding energy \( \omega \). We then compute (for the same \( \omega \) and \( \theta \)) the matrix \( A_{mn\nu} \equiv \omega \delta_{mn} - \hat{H}^0_{mn\nu}(k^\text{max}_\beta(\omega, \theta_k)) \) and similarly \( B_{mn\nu}, G_{mn\nu} \) for the \( \beta \) and \( \gamma \) band MDCs, \( k^\text{max}_{\beta}(\omega, \theta) \) and \( k^\text{max}_{\gamma}(\omega, \theta) \), respectively. In terms of these matrices, the quasiparticle equations (12) read

\[
\det[A_{mn\nu} - \Sigma'_m(\delta_{mn})] = \det[B_{mn\nu} - \Sigma'_m(\delta_{mn})] = \det[G_{mn\nu} - \Sigma'_m(\delta_{mn})] = 0. \tag{13}
\]

However, when taking symmetry into account, the self-energy has only two independent components: \( \Sigma'_{xy} \) and \( \Sigma'_{xz} = \Sigma'_{yz} \). Hence, we only need two of the above equations to solve for the two unknown components of the self-energy. This means that we can also extract a self-energy in the directions where only two bands (\( \beta \) and \( \gamma \)) are present in the considered energy range of \( \omega \leq 100 \text{meV} \), e.g., along \( \Gamma M \). The resulting functions \( \Sigma'_m(\omega, \theta_k) \) determined at several angles \( \theta \) are displayed in Fig. 4 (d). It is immediately apparent that, in contrast to \( \Sigma''_m \), the self-energies in the orbital basis do not show a strong angular (momentum) dependence, but rather collapse into two sets of points, one for the \( xy \) orbital and one for the \( xz/yz \) orbitals. Thus, we reach the remarkable conclusion that the angular dependence of the self-energy in the orbital basis is negligible, within the range of binding energies investigated here: \( \Sigma'_m(\omega, \theta_k) \simeq \Sigma'_m(\omega) \). This implies that a good approximation of the full momentum and energy dependence of the self-energy in the band (quasiparticle) basis is given by

\[
\Sigma_{\nu\nu'}(\omega, \mathbf{k}) = \sum_m U^*_{m\nu}(\mathbf{k}) \Sigma_m(\omega) U_{m\nu'}(\mathbf{k}). \tag{14}
\]

The physical content of this expression is that the angular (momentum) dependence of the quasiparticle self-energies emphasized above is actually due to the matrix elements \( U_{m\nu}(\mathbf{k}) \) defined in Eq. 6. In Sr₂RuO₄ the angular dependence of these matrix elements is mainly due to the SOC, as seen from the variation of the orbital content of quasiparticles in Fig. 3 (g). In appendix D we show the back-transform of \( \Sigma'_m(\omega, \theta_k = 18^\circ) \) into \( \Sigma'_m(\omega, \theta_k = 0, \pm 18^\circ) \). The good agreement with \( \Sigma'_m \) directly extracted from experiment further justifies the above expression and also confirms the validity of the approximations made throughout this section.

Finally, we stress that expression (14) precisely coincides with the \( \text{ansatz} \) made by DMFT: within this theory, the self-energy is approximated as a local (\( \mathbf{k} \)-independent) object \( \text{when expressed in a basis of localized orbitals} \), while it acquires momentum dependence when transformed to the band basis.

VI. COMPARISON TO DYNAMICAL MEAN-FIELD THEORY

In this section we perform an explicit comparison of the measured quasiparticle dispersions and self-energies to DMFT results. The latter are based on the Hamiltonian \( \hat{H}^{\text{DFT}} \), to which the Hubbard-Kanamori interaction with on-site interaction \( U = 2.3 \text{eV} \) and Hund’s coupling \( J = 0.4 \text{eV} \) [20] is added. For the details of the DMFT calculation and especially the treatment of SOC in this framework, we refer the reader to appendix B. There, we also comment on some of the limitations and shortcomings of the current state of the art for DFT+DMFT calculations in this context. Fig. 5 (a) shows the experimental quasiparticle dispersion extracted from our ARPES data (circles) on top of the DMFT spectral function \( A(\omega, \mathbf{k}) \) displayed as a color-intensity map. Clearly, the theoretical results are in near quantitative agreement with the data: both the strong renormalization of the Fermi velocity and the angular-dependent curvature of the quasiparticle bands are very well reproduced. The small deviations in Fermi wave vectors discernible in Fig. 5 are consistent with Fig. 3(c) and the overall experimental precision of the Fermi surface determination.

In Fig. 6 (a), we compare the experimental self-energies for each orbital with the DMFT results. The overall agreement is notable. At low-energy, the self-energies are linear in frequency and the agreement is excellent. The slope of the self-energies in this regime controls the
angular-dependence of the effective mass renormalisation. Using the local ansatz (14) into the quasiparticle dispersion equation, and performing an expansion around $E_F$, we obtain

$$\frac{v_\theta}{v_\theta F} = \sum_m \frac{1}{Z_m} |U_{m\nu}(\theta)|^2, \quad \sum_m = 1 - \frac{\partial \Sigma_m}{\partial \omega} \bigg|_{\omega=0}. \quad (15)$$

In Fig. 6 (b), we show $v_\theta/v_\theta F(\theta)$ for the $\beta$ and $\gamma$ bands using the DMFT values $Z_{xy} = 0.18 \pm 0.01$ and $Z_{xz/yz} = 0.3 \pm 0.01$ obtained at 29 K (appendix B 2). The overall angular dependence and the absolute value of the $\gamma$ band mass enhancement is very well captured by DMFT, while the $\beta$ band is a bit overestimated. Close to the zone-diagonal ($\theta = 45^\circ$), the two mass enhancements are approximately equal, due to the strong orbital mixing induced by the SOC.

Turning to larger binding energies, we see that the theoretical $\Sigma_{xy}'$ is in remarkable agreement with the experimental data over the full energy range of 2-80 meV covered in our experiments. Both the theoretical and experimental self-energies deviate significantly from the linear regime down to low energies ($\sim 20$ meV), causing curved quasiparticle bands with progressively steeper dispersion as the energy increases (Fig. 5). In contrast, the agreement between theory and experiment for the $xz/yz$ self-energy is somewhat less impressive at binding energies larger than $\sim 30$ meV. Our DMFT self-energy $\Sigma_{xz/yz}'$ overestimates the strength of correlations in this regime (by 20–25%), with a theoretical slope larger than the experimental one. Correspondingly, the quasiparticle dispersion is slightly steeper in this regime than the theoretical result, as can be also seen in Fig. 5.

There may be several reasons for this discrepancy. Even while staying in the framework of a local self-energy, we note that the present DMFT calculation is performed with an on-site value of $U$ which is the same for all orbitals. Earlier cRPA calculations have suggested that this on-site interaction is slightly larger for the $xy$ orbital ($U_{xy} = 2.5$ eV and $U_{xz/yz} = 2.2$ eV) [20] and recent work has advocated the relevance of this for DFT+DMFT calculations of Sr$_2$RuO$_4$ [21]. Another possible explanation is that this discrepancy is actually a hint of some momentum dependent contribution to the self-energy, especially dependence on momentum perpendicular to the Fermi surface. We note in this respect that the discrepancy is larger for the $\alpha$, $\beta$ sheets which have dominant $xz/yz$ character. These orbitals have, in the absence of SOC, a strong one-dimensional character, for which momentum dependence is definitely expected and DMFT is less appropriate. Furthermore, these FS sheets are also the ones associated with nesting and spin-density wave correlations, which are expected to lead to an additional momentum-dependence of the self-energy. We further discuss possible contributions of spin fluctuations in Sec. VIII.

### VII. KINKS

The self-energies $\Sigma'(\omega)$ shown in Figs. 4 and 6 display a fairly smooth curvature, rather than pronounced ‘kinks’. Over a larger range, however, $\Sigma_{xz/yz}'$ from DMFT does show an energy scale marking the crossover from the strongly renormalized low-energy regime to weakly renormalized excitations. This is illustrated in the inset to Fig. 6 (a). Such purely electronic kinks were reported in DMFT calculations of a generic system with Mott-Hubbard sidebands [74] and have been abundantly documented since then [13, 20, 75–78]. In Sr$_2$RuO$_4$ they are associated with the crossover from the Fermi-liquid behavior into a more incoherent regime [18, 20]. The near quantitative agreement of the frequency dependence of the experimental self-energies $\Sigma_m'(\omega)$ and our single-site DMFT calculation provides strong evidence for the existence of such electronic kinks in Sr$_2$RuO$_4$. In addition, it implies that the local DMFT treatment of electronic correlations is capturing the dominant effects.

Focusing on the low-energy regime, we find deviations from the linear form $\Sigma'(\omega) = \omega(1-1/Z)$ characteristic of a Fermi liquid for $|\omega| > 20$ meV, irrespective of the basis. However, this is only an upper limit for the Fermi-liquid energy scale in Sr$_2$RuO$_4$. Despite the improved resolution of our experiments, we cannot exclude an even lower
over, attributing the entire curvature of $\Sigma_m'$ in our data to electron-phonon coupling would result in unrealistic coupling constants far into the polaronic regime, which is hard to reconcile with the transport properties of Sr$_2$RuO$_4$ [10–12]. We also note that a recent scanning tunneling microscope (STM) study reported very strong kinks in the $\beta$ and $\gamma$ sheets of Sr$_2$RuO$_4$ [49], which is inconsistent with our data. We discuss the reason for this discrepancy in appendix A.

VIII. DISCUSSION AND PERSPECTIVES

In this article, we have reported on high-resolution ARPES measurements which allow for a determination of the Fermi surface and quasiparticle dispersions of Sr$_2$RuO$_4$ with unprecedented accuracy. Our data reveal an enhancement (by a factor of about two) of the splitting between Fermi surface sheets along the zone diagonal, in comparison to the DFT value. This can be interpreted as a correlation-induced enhancement of the effective SOC, an effect predicted theoretically [21, 23, 50] and demonstrated experimentally here for this material, for the first time.

Thanks to the high resolution, we have been able to determine the electronic self-energies directly from the ARPES data, using both a standard procedure applied in the band (quasiparticle) basis as well as a novel procedure, introduced in the present article, in the orbital basis. Combining these two approaches, we have demonstrated that the large angular (momentum) dependence of the quasiparticle self-energies and dispersions can be mostly attributed to the fact that quasiparticle states have an orbital content which is strongly angular dependent, due to the SOC. Hence, assuming self-energies which are frequency-dependent but essentially independent of angle (momentum), when considered in the orbital basis, is a very good approximation. This provides a direct experimental validation of the DMFT ansatz, and indeed, the comparison between the ARPES data and DMFT calculations is found to be remarkably accurate. The key importance of atomic-like orbitals in correlated insulators is well established [80, 81]. However, the fact that orbitals retain such considerable physical relevance even in an metal in the low-temperature Fermi-liquid regime is a remarkable fact. Thinking only in terms of quasiparticles associated with a specific Fermi surface sheet is insufficient to unravel the physics of correlated metals such as Sr$_2$RuO$_4$. As the present work demonstrates, hidden simplicity in the nature of correlations is unraveled when instead considering the orbital degrees of freedom, and taking into account the mixed orbital content of each quasiparticle state. Beyond Sr$_2$RuO$_4$, this is an observation of general relevance to metals with strong correlations (see, e.g., Ref. [82, 83] in the context of iron-based superconductors).

Notwithstanding its success, the excellent agreement of the DMFT results with ARPES data does raise puzzling
questions. Sr$_2$RuO$_4$ is known to be host to strong magnetic fluctuations [39–42], with a strong peak in its spin response $\chi(Q)$ close to the spin-density wave (SDW) vector $Q \sim (2\pi/3, 2\pi/3)$, as well as quasi-ferromagnetic fluctuations which are broader in momentum around $Q = 0$. Indeed, tiny amounts of substitutional impurities induce long-range magnetic order in this material, of either SDW of ferromagnetic type [84, 85]. Hence, it is a prominent open question to understand how these long-wavelength fluctuations affect the physics of quasiparticles in the Fermi-liquid state. Single-site DMFT does not capture this feedback, and the excellent agreement with the overall quasiparticle physics must imply that these effects have a comparatively smaller magnitude than the dominant local effect of correlations (on-site $U$ and especially Hund’s $J$) captured by DMFT. A closely related question is how much momentum dependence is present in the low-energy (Landau) interactions between quasiparticles. These effects are expected to be fundamental for subsequent instabilities of the Fermi liquid, into either the superconducting state in pristine samples or magnetic ordering in samples with impurities. Making progress on this issue is also key to the understanding of the superconducting state of Sr$_2$RuO$_4$, for which the precise nature of the pairing mechanism as well as symmetry of the order parameter are still open questions [4].

ACKNOWLEDGMENTS

The experimental work has been supported by the ERC, the Scottish Funding Council, the UK EPSRC and the SNSF. Theoretical work was supported by the European Research Council grant ERC-319286-QMAC and by the SNSF (NCCR MARVEL). The Flatiron Institute is a division of the Simons Foundation. AG and MZ gratefully acknowledge useful discussions with Gabriel Kotliar, Andrew J. Millis and Jernej Mravlje.

Appendix A: Bulk and surface electronic structure of Sr$_2$RuO$_4$

In Fig. 7 we compare the data presented in the main text with data from a pristine cleave taken with $h\nu = 21$ eV at the SIS beamline of the Swiss Light Source. This comparison confirms the identification of bulk and surface bands by Shen et al. [53]. In particular, we find that the larger $\beta$ sheet has bulk character. This band assignment is used by the vast majority of subsequent ARPES publications [29–31, 33, 46, 55, 86, 87], except for Ref. [47], which reports a dispersion with much lower Fermi velocity and a strong kink at 15 meV for the smaller $\beta$ sheet that we identify as a surface band.

Wang et al. [49] have recently probed the low-energy electronic structure of Sr$_2$RuO$_4$ by STM. Analyzing quasiparticle interference patterns along the TX and TM high-symmetry directions, they obtained band dispersions with low Fermi velocities and strong kinks at 10 meV and 37 meV. In Fig. 8 we compare the band dispersion reported by Wang et al. with our ARPES data. Along both high-symmetry directions, we find a clear discrepancy with our data, which are in quantitative agreement with bulk de Haas van Alphen measurements, as demonstrated in the main text. On the other hand, we find a striking similarity between the STM data along TM and the band commonly identified as the surface $\beta$ band. We thus conclude that the experiments reported in Ref. [49] probed the surface states of Sr$_2$RuO$_4$. This is fully consistent with the enhanced low-energy renormalization of the surface bands seen in ARPES [88].

Appendix B: Computational details

1. DFT and model Hamiltonian

We generate our theoretical model Hamiltonian from maximally-localized Wannier function [59, 60] for the three Ru-$4d$ $t_{2g}$ orbitals. These are constructed on a $10 \times 10 \times 10$ $k$ grid based on a non-SOC DFT calculation using WIEN2k [89] with the GGA-PBE functional [90], wien2manner [91] and Wannier90 [92]. The DFT calculation is performed with lattice parameters from Ref. [93] (measured at 100 K) and converged with twice as many $k$-points in each dimension.

The eigenenergies of the resulting Wannier Hamiltonian, $H^{\text{DFT}}$, accurately reproduce the DFT band struc-
ture (Fig. 9 (a)). To take SOC into account, we add the local single-particle term $\hat{H}_{\text{SOC}}^\lambda$, as given in Eq. 10, with coupling constant $\lambda$. In Fig. 9 (b) we show that the eigenenergies of $\hat{H}_{\text{DFT}}^\lambda + \hat{H}_{\text{SOC}}^\lambda$ are in nearly perfect agreement with the DFT+SOC band structure at a value of $\lambda_{\text{DFT}} = 100 \text{ meV}$. Our model Hamiltonian provides the reference point to which we define a self-energy, but it is also a perfect playground to study the change in the Fermi surface under the influence of SOC and the crystal field splitting between the $xy$ and $xz/yz$ orbitals. Concerning the latter, the constructed $\hat{H}_{\text{DFT}}^\lambda$ has already a splitting of $\epsilon_{\text{cf}} = \epsilon_{xz/yz} - \epsilon_{xy} = 85 \text{ meV}$.

In the following, we will confirm that the best agreement with the experimental Fermi surface is found with an effective SOC of $\lambda_{\text{eff}} = \lambda_{\text{DFT}} + \Delta \lambda = 200 \text{ meV}$, but at the same time keeping the crystal field splitting unchanged. We compare in Fig. 10 the experimental Fermi surface (dashed lines) to the one of $\hat{H}_{\text{DFT}}^{\lambda_{\text{DFT}}} + \hat{H}_{\text{SOC}}^{\lambda_{\text{DFT}}}$. The Fermi surfaces for additionally introduced crystal field splittings $\Delta \epsilon_{\text{cf}}$ between $-80$ and $80 \text{ meV}$ are shown with solid lines in different shades of red. In contrast to the Fermi surface without SOC ($\lambda = 0 \text{ meV}$, top left panel), the Fermi surfaces with the DFT SOC of $\lambda = 100 \text{ meV}$ (top right panel) resembles the overall structure of the experimental Fermi surface. However, the areas of the $\alpha$ and $\beta$ sheets are too large and the $\gamma$ sheet is too small. Importantly, the agreement cannot be improved by adding $\Delta \epsilon_{\text{cf}}$. For example, along $\Gamma M$ a $\Delta \epsilon_{\text{cf}}$ of $-40 \text{ meV}$ would move the Fermi surface closer to the experiment, but, on the other hand, along $\Gamma X$ a $\Delta \epsilon_{\text{cf}}$ of $80 \text{ meV}$ would provide the best agreement. The situation is different if we consider an enhanced SOC of $\lambda = 200 \text{ meV}$ (bottom right panel). Then, we find a nearly perfect agreement with experiment without any additional crystal field splitting ($\Delta \epsilon_{\text{cf}} = 0 \text{ meV}$). At an even higher SOC of $\lambda = 300 \text{ meV}$ (bottom left panel) we see again major discrepancies, but with an opposite trend: The $\alpha$ and $\beta$ sheets are now too small and the $\gamma$ sheet is too large. Like in the case of $\lambda = 100 \text{ meV}$ this can not be cured by an adjustment of $\epsilon_{\text{cf}}$. 

2. DMFT

We perform single-site DMFT calculation with the TRIQS/DFTTools [94] package for $\hat{H}_{\text{DFT}}^\lambda$ and Hubbard-Kanamori interactions with a screened Coulomb repulsion $U = 2.3 \text{ eV}$ and a Hund’s coupling $J = 0.4 \text{ eV}$ based on previous works [20, 23]. The impurity problem is solved on the imaginary-time axis with the TRIQS/CTHYB [95] solver at a temperature of 29 K. The employed open-source software tools are based on the TRIQS library [96]. We assume an orbital-independent double counting, and hence it can be absorbed into an effective chemical potential, which is ad-
adjusted such that the filling is equal to four electrons. For the analytic continuation of the self-energy to the real-frequency axis we employ three different methods: Padé approximants (using TRIQS [96]), Stochastic continuation (after Beach [97]) and Maximum Entropy (using TRIQS/maxent [98]). In the relevant energy range obtained with three different analytic continuation methods: Padé approximants (using TRIQS [96]), Stochastic continuation (after Beach [97]) and Maximum Entropy (using TRIQS/maxent [98]). The difference between the analytic continuation methods is smaller than the experimental error.

FIG. 11. Real part of DFT+DMFT self-energy in the considered energy range obtained with three different analytic continuation methods: Padé approximants (using TRIQS [96]), Stochastic continuation (after Beach [97]) and Maximum Entropy (using TRIQS/maxent [98]). The difference between the analytic continuation methods is smaller than the experimental error.
spond to different $k'_{\text{max}}$, which are further defined by the experimental MDCs.

In Fig. 12 (a) we show the result for two selected $k$ points: on the $\beta$ sheet for $\theta = 0^\circ$ and on the $\gamma$ sheet for $\theta = 45^\circ$. For these $k$ points the largest off-diagonal element is $\Sigma'_{\alpha\beta}$, which is about $10-20\%$ of the size of the diagonal elements. A scan performed for the whole $k_z = 0.4 \pi/c$ plane further confirms that $|\Sigma'_{\alpha\neq\beta}|$ is smaller than 20 meV.

However, when neglect the off-diagonal elements it is also important to have a large enough energy separation of the bands. This can be understood by considering a simplified case of two bands $(\nu, \nu')$ and rewriting Eq. 5, which determines the quasiparticle dispersion $\omega^{\text{qp}}_\nu(k)$, as

$$\omega - \varepsilon_\nu(k) - \Sigma'_{\nu\nu}(\omega, k) = 0.$$  \hspace{1cm} \text{(C1)}

Setting the last term to zero, i.e., using the procedure described in Sec. VA to extract $\Sigma'_{\nu\nu}$, is justified at $\omega = \omega^{\text{qp}}_\nu(k)$ as long as

$$\Sigma'_{\nu\nu}(\omega, k) \gg \frac{\Sigma'_{\nu\nu}(\omega, k) \Sigma'_{\nu'\nu}(\omega, k)}{\omega - \varepsilon_{\nu'}(k) - \Sigma'_{\nu'\nu}(\omega, k)}.$$  \hspace{1cm} \text{(C2)}

In this condition the already small off-diagonal elements enter quadratically, but also the denominator is not a small quantity, because the energy separation of the bare bands $(\varepsilon_\nu(k) - \varepsilon_{\nu'}(k))$ is larger than the difference of the diagonal self-energies.

By using the generalized version of Eq. C2 for all three bands, we find that the right-hand side of this equation is indeed less than $1.2\%$ of $\Sigma'_{\nu\nu}(\omega, k'_{\text{max}}(\omega))$ for all experimentally determined $k'_{\text{max}}(\omega)$. This means that for Sr$_2$RuO$_4$ treating each band separately when extracting $\Sigma'_{\nu\nu}$ is well justified in the investigated energy range.

**Appendix D: Reconstruction of $\Sigma'_{\nu\nu}$**

In order to further test the validity of the local ansatz (Eq. 14) and establish the overall consistency of the two procedures used to extract the self-energy in Sec. V, we perform the following ‘reconstruction procedure’. We use the $\Sigma'_{\nu}(\omega, \theta_k)$ (from Sec. VB) at one angle, e.g., $\theta = 18^\circ$, and transform it into $\Sigma'_{\nu\nu}(\omega, k'_{\text{max}}(\omega))$ for other measured angles, using Eq. 14. The good agreement between the self-energy reconstructed in this manner (thin lines in Fig. 12 (b)) and its direct determination following the procedure of Sec. VA (dots) confirms the validity of the approximations used throughout Sec. V. It also shows that the origin of the strong momentum dependence of $\Sigma'_{\nu\nu}$ is almost entirely due to the momentum dependence of the orbital content of quasiparticle states, i.e., of $U_{\nu \nu}(k) = \langle \chi_m(k) | \psi_\nu(k) \rangle$. In Sr$_2$RuO$_4$ the momentum dependence of these matrix elements is mainly due to the SOC.

---

[1] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, Nature 372, 532 (1994).
[2] T. M. Rice and M. Sigrist, J. Phys.: Condens. Matter 7, L643 (1995).
[3] A. P. Mackenzie and Y. Maeno, Rev. Mod. Phys. 75, 657 (2003).
[4] A. P. Mackenzie, T. Scaffidi, C. W. Hicks, and Y. Maeno, npj Quantum Materials 2, 40 (2017).
[5] K. Ishida, H. Mukuda, Y. Kitaoka, K. Asayama, Z. Q. Mao, Y. Mori, and Y. Maeno, Nature 396, 658 (1998).
[6] M. S. Anwar, S. R. Lee, R. Ishiguro, Y. Sugimoto, Y. Tano, S. J. Kang, Y. J. Shin, S. Yonezawa, D. Manske, H. Takayanagi, T. W. Noh, and Y. Maeno, Nat. Commun. 7, 1 (2016).
T. Richter, J. Mudd, M. Basham, L. Pratt, P. Leicester, E. Longhi, A. Tamai, and F. Baumberger, Rev. Sci. Instrum. 88, 013106 (2017).

[53] K. M. Shen, A. Damascelli, D. H. Lu, N. P. Armitage, K. Ronning, D. L. Feng, C. Kim, Z. X. Shen, D. J. Singh, I. I. Mazin, S. Nakatsuji, Z. Q. Mao, Y. Maeno, T. Kimura, and Y. Tokura, Phys. Rev. B 64, 180502 (2001).

[54] B. Stöger, M. Hieckel, F. Mittendorfer, Z. Wang, D. Fobes, J. Peng, Z. Mao, M. Schmid, J. Redinger, and U. Diebold, Phys. Rev. Lett. 113, 116101 (2014).

[55] K. M. Shen, N. Kikugawa, C. Bergemann, L. Balicas, F. Baumberger, W. Meevasana, N. J. C. Ingle, Y. Maeno, Z. X. Shen, and A. P. Mackenzie, Phys. Rev. Lett. 99, 187001 (2007).

[56] We note that radial k-space cuts are not exactly perpendicular to the Fermi surface. This can cause the velocities v_F and v_0 given here to deviate by up to 10% from the Fermi velocity. However, since we evaluate the experimental and theoretical dispersion along the same k-space cut, this effect cancels in Fig. 2 (d) and does not affect the self-energy determination in Sec. V.

[57] F. Baumberger, N. J. C. Ingle, W. Meevasana, K. M. Shen, D. H. Lu, R. S. Perry, A. P. Mackenzie, Z. Hussain, D. J. Singh, and Z.-X. Shen, Phys. Rev. Lett. 96, 246402 (2006).

[58] We attribute the higher Fermi velocities reported in some earlier studies [29, 31, 32, 34] to the lower energy resolution, which causes an extended range near the Fermi level where the dispersion extracted from fits to individual MDCs is artificially enhanced, rendering a precise determination of v_F difficult. The much lower value of v_F given here to deviate by up to 10% from the Fermi velocity. However, since we evaluate the experimental and theoretical dispersion along the same k-space cut, this effect cancels in Fig. 2 (d) and does not affect the self-energy determination in Sec. V.

[59] N. Marzari and D. Vanderbilt, Phys. Rev. B 56, 12847 (1997).

[60] I. Souza, N. Marzari, and D. Vanderbilt, Phys. Rev. B 65, 035109 (2001).

[61] F. Lechermann, A. Georges, A. Poteryaev, S. Biermann, M. Posternak, A. Yamaskii, and O. K. Andersen, Phys. Rev. B 74, 125120 (2006).

[62] By restricting the Hamiltonian to the t_2g-space subspace we neglect the e_g–t_2g coupling terms of H^SOC_3. This approximation is valid as long as the e_g–t_2g crystal field splitting is large in comparison to λ, which is the case for Sr_2RuO_4.

[63] K. K. Ng and M. Sigrist, Europhys. Lett. 49, 473 (2002).

[64] I. Eremin, D. Manske, and K. H. Bennemann, Phys. Rev. B 65, 220502 (2002).

[65] M. Haverkort, I. Elfinov, L. Tjeng, G. Sawatzky, and A. Damascelli, Phys. Rev. Lett. 101, 026406 (2008).

[66] C. M. Puettet and H.-Y. Kee, Europhysics Letters 98, 27010 (2012).

[67] C. N. Veenstra, Z. H. Zhu, M. Raichle, B. M. Ludbrook, A. Nicolau, B. Slomski, G. Landolt, S. Kittaka, Y. Maeno, J. H. Dil, I. S. Elfinov, M. W. Haverkort, and A. Damascelli, Phys. Rev. Lett. 112, 127002 (2014).

[68] S. V. Borisenko, D. V. Evtushinsky, Z.-H. Liu, I. Morozov, R. Kappenberger, S. Wurmehl, B. Büchner, A. N. Yaresko, T. K. Kim, M. Hoesch, T. Wolf, and N. D. Zhigadlo, Nat. Phys. 12, 311 (2015).

[69] Ref. [99] noted that an enhance effective SOC also improves the description of de Haas van Alphen data.

[70] Using the free electron final state approximation, we obtain k_F = (2m_eℏ^2/2)^(1/2) ≈ 0.4(0.12)π/c assuming an inner potential relative to E_F of U = Φ = 8.5(1.0) eV.

[71] M. Hengsberger, D. Purdie, P. Segovia, M. Garnier, and Y. Baer, Phys. Rev. Lett. 83, 592 (1999).

[72] A. Lanzara, P. V. Bogdanov, J. X. Zhou, S. A. Kellar, D. L. Feng, E. D. Lu, T. Yoshida, H. Eissaki, A. Fujimi, K. Kishio, J. I. Shimoyama, T. Noda, S. Uchida, Z. Hussain, and Z. X. Shen, Nature 412, 510 (2001).

[73] A. Tamai, W. Meevasana, P. D. C. King, C. W. Nicholson, A. de la Torre, E. Rozbicki, and F. Baumberger, Phys. Rev. B 87, 075113 (2013).

[74] K. Byczuk, M. Kollar, K. Held, Y. F. Yang, I. A. Nekrasov, T. Pruschke, and D. Vollhardt, Nat Phys 3, 168 (2007).

[75] C. Raas, P. Grete, and G. S. Uhrig, Phys. Rev. Lett. 102, 076406 (2009).

[76] I. Held, R. Peters, and A. Toschi, Phys. Rev. Lett. 110, 246402 (2013).

[77] X. Deng, J. Mravlje, R. Žitko, M. Ferrero, G. Kotliar, and A. Georges, Phys. Rev. Lett. 110, 086401 (2013).

[78] R. Žitko, D. Hansen, E. Perepelitsky, J. Mravlje, A. Georges, and B. S. Shastry, Phys. Rev. B 88, 235132 (2013).

[79] M. Braden, W. Reichardt, Y. Sidis, Z. Mao, and Y. Maeno, Phys. Rev. B 76, 014505 (2007).

[80] K. Kugel and D. Khomski, Sov. Phys. Usp. 25, 231 (1982).

[81] Y. Tokura and N. Nagaosa, Science 288, 462 (2000).

[82] H. Miao, Z. P. Yin, S. F. Wu, J. M. Li, J. Ma, B.-Q. Lv, X. P. Wang, T. Qian, P. Richard, L.-Y. Xing, X.-C. Wang, C. Q. Jin, K. Haule, G. Kotliar, and H. Ding, Phys. Rev. B 94, 201109 (2016).

[83] P. O. Sprau, A. Kostin, A. Kreisel, A. E. Böhm, V. Taufo, P. C. Canfield, S. Mukherjee, P. J. Hirschfeld, B. M. Andersen, and J. C. S. Davis, Science 357, 75 (2017).

[84] M. Braden, O. Friedt, Y. Sidis, P. Bourges, M. Minakata, and Y. Maeno, Phys. Rev. Lett. 88, 197002 (2002).

[85] J. E. Ortman, J. Y. Liu, J. Hu, M. Zhu, J. Peng, M. Matsuda, X. Ke, and Z. Q. Mao, Scientific Reports 3, 2950 (2013).

[86] S. C. Wang, H. B. Yang, A. K. P. Sekharan, H. Ding, J. R. Engelbrecht, X. Dai, Z. Wang, A. Kaminski, T. Valla, T. Kidd, A. V. Fedorov, and P. D. Johnson, Phys. Rev. Lett. 92, 137002 (2004).

[87] T. E. Kidd, T. Valla, A. V. Fedorov, P. D. Johnson, R. J. Cava, and M. K. Haas, Phys. Rev. Lett. 94, 107003 (2005).

[88] T. Kondo, M. Ochi, M. Nakayama, H. Taniguchi, S. Akihe, K. Kuroda, M. Arita, S. Sakai, H. Namatame, M. Taniguchi, Y. Maeno, R. Arita, and S. Shin, Phys. Rev. Lett. 117, 247001 (2016).

[89] P. Blaha, K. Schwarz, G. Madsen, D. Kvasnicka, and J. Luitz, WIEN2k, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties (K. Schwarz, Tech. Univ. Wien, Austria, 2001).

[90] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).

[91] J. Kuneš, R. Arita, P. Wissgott, A. Toschi, H. Ikeda, and K. Held, Comput. Phys. Commun. 181, 1888 (2010).

[92] A. A. Mostofi, J. R. Yates, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, Comput. Phys. Commun. 178,
[93] T. Vogt and D. J. Buttrey, Phys. Rev. B 52, R9843 (1995).

[94] M. Aichhorn, L. Pourovskii, P. Seth, V. Vildosola, M. Zingl, O. E. Peil, X. Deng, J. Mravlje, G. J. Kraberger, C. Martins, M. Ferrero, and O. Parcollet, Comput. Phys. Commun. 204, 200 (2016).

[95] P. Seth, I. Krivenko, M. Ferrero, and O. Parcollet, Comput. Phys. Commun. 200, 274 (2016).

[96] O. Parcollet, M. Ferrero, T. Ayral, H. Hafermann, I. Krivenko, L. Messio, and P. Seth, Comput. Phys. Commun. 196, 398 (2015).

[97] K. S. D. Beach, ArXiv e-prints (2004), arXiv:cond-mat/0403055 [cond-mat.str-el].

[98] “TRIQS/maxent package”, https://triqs.github.io/maxent.

[99] E. J. Rozbicki, J. F. Annett, J.-R. Souquet, and A. P. Mackenzie, J. Phys.: Condens. Matter 23, 094201 (2011).