Sr$_2$MoO$_4$ and Sr$_2$RuO$_4$: Disentangling the Roles of Hunds and van Hove Physics

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Sr$_2$MoO$_4$ is isostructural to the unconventional superconductor Sr$_2$RuO$_4$ but with two electrons instead of two holes in the Mo/Ru-t$_{2g}$ orbitals. Both materials are Hund’s metals, but while Sr$_2$RuO$_4$ has a van Hove singularity in close proximity to the Fermi surface, the van Hove singularity of Sr$_2$MoO$_4$ is far from the Fermi surface. By using density functional plus dynamical mean-field theory we determine the relative influence of van Hove and Hund’s metal physics on the correlation properties. We show that theoretically predicted signatures of Hund’s metal physics occur on the occupied side of the electronic spectrum of Sr$_2$MoO$_4$, identifying Sr$_2$MoO$_4$ as an ideal candidate system for a direct experimental confirmation of the theoretical concept of Hund’s metals via photoemission spectroscopy.

Sr$_2$RuO$_4$ has emerged as an exemplary quantum material, providing fundamental insights into the effect of electronic correlations on material properties [1–14]. The rich electronic properties of Sr$_2$RuO$_4$ are determined by a sophisticated interplay of factors, including the on-site Coulomb repulsion, spin-orbit coupling, and a van Hove singularity, but it is believed that the nontrivial physics of the intra-orbital Hund’s interaction [15–23] is at the heart of the strongly-correlated nature of this material [11, 23–25]. However, unambiguous experimental observation of Hund’s-related physics has been challenging. For example, the presence of a van Hove singularity in the vicinity of the Fermi level impacts electronic correlations, masking the effects of the Hund’s coupling on the quasiparticle mass enhancement [11, 24, 25]. While Hund’s physics has been predicted to produce a characteristic peak in the single-particle spectrum [21, 22, 26], for Sr$_2$RuO$_4$ this peak occurs on the unoccupied side of the spectrum [4, 10, 13]. Thus, a direct experimental observation with conventional photoemission spectroscopy is challenging, though indirect hints have been seen in optical conductivity [4].

In this letter, we use a combination of density functional theory (DFT) and dynamical mean-field theory (DMFT) to argue that Sr$_2$MoO$_4$, a material isostructural to Sr$_2$RuO$_4$ but with a different electron count, provides an ideal platform to study Hund’s physics in quantum materials, while the comparison to Sr$_2$RuO$_4$ provides insight into the role of van Hove singularities. Sr$_2$MoO$_4$ crystallizes in the same tetragonal I4/mmm crystal structure as Sr$_2$RuO$_4$, with a=b and c lattice parameters being slightly larger in Sr$_2$MoO$_4$, as expected from the larger ionic radius of Mo$^{4+}$ in comparison to Ru$^{4+}$ [27–29]. The octahedral oxygen environment surrounding the Ru/Mo atoms leads to an e$_g$-t$_{2g}$ splitting of the Ru/Mo-4d shell with unoccupied e$_g$ orbitals and three t$_{2g}$ orbitals occupied by 2 electrons in Sr$_2$MoO$_4$ and 4 electrons in Sr$_2$RuO$_4$.

Two decades ago, Sr$_2$MoO$_4$ was synthesized in polycrystalline form [27–29], and later, 60 uc single-crystalline films were reported [30]. In contrast to the vast literature on Sr$_2$RuO$_4$, only the basic electronic structure of Sr$_2$MoO$_4$ has been studied with DFT [31]. Since then the material has fallen into oblivion, and we hope that the results presented here will rekindle experimental interest.

Fig. 1 shows the DFT (one electron) electronic structure calculated with WIEN2k [32] using the PBE-GGA [33] exchange-correlation functional and experimental atomic positions [31, 34], along with Wannier bands calculated with Wannier90 [35, 36] and discussed below. The insets of Fig. 1 show the Fermi surfaces, which consist of 3 sheets: two electron-like sheets centered at Γ and one hole-like pocket centered at the M-point. The electron sheets are smaller and the hole pockets are larger in Sr$_2$MoO$_4$ than in Sr$_2$RuO$_4$, due to the lower electron count of Sr$_2$MoO$_4$. Without spin-orbit coupling, the smaller electron sheet and the hole-pockets are of pure $xz/yz$ character (red), whereas the larger electron sheet is of $xy$ orbital character (blue).

The inclusion of spin-orbit coupling, which is slightly smaller in Sr$_2$MoO$_4$ (80 meV) than in Sr$_2$RuO$_4$ (100 meV), leads to a momentum-dependent mixing of the orbital character of the Fermi surface sheets [6, 8, 37]. In contrast to Sr$_2$RuO$_4$, the spin-orbit coupling does not cause a restructuring of the Fermi surface in Sr$_2$MoO$_4$. We discuss the electronic structure with spin-orbit coupling in the supplemental material [34], but we neglect it for most of this work as it is not important for the
Hund’s-related electronic correlations, which are of primary interest here.

To capture the effect of electronic correlations at low energies, we construct a basis of three $t_{2g}$-like maximally-localized Wannier orbitals [38, 39]. As shown in the left panels of Fig. 1, the Wannier states (colored) reproduce the DFT bands (black) very precisely in both materials. We note that for Sr$_2$MoO$_4$ the $t_{2g}$-derived bands around the Fermi energy are separated from the O-$p$ states by more than 1 eV, which makes the selection of a low-energy subspace even more natural. The shape of the Wannier orbital density of states (DOS), Fig. 1 right panels, is a result of the quasi-2D crystal structure, which makes the rather 2D-like $xy$ orbital (blue) different from the more 1D $xz/yz$ ones (red). For Sr$_2$MoO$_4$ the degenerate $xz/yz$ orbitals have a wider band width (2.2 eV) than for Sr$_2$RuO$_4$ (1.5 eV), but the difference in band widths of the $xy$ orbital is less (3.6 eV vs. 3.8 eV). Overall, the band structures and DOS of the two materials are very similar apart from a shift in the Fermi level due to the different electron count. There is one important qualitative difference: For Sr$_2$MoO$_4$ the saddle point of the $xy$-derived band at the X-point, corresponding to a van Hove singularity in the DOS, is at ~1 eV above the Fermi energy, while for Sr$_2$RuO$_4$ it is in close proximity to the Fermi energy. We will see in the following how this key difference in the electronic structure impacts the strength of electronic correlations.

We include the effect of electron-electron correlations by adding local interactions of Hubbard-Kanamori form [45] using a Coulomb repulsion $U = 2.3$ eV and a Hund’s coupling $J = 0.4$ eV [46] and solving the resulting problem within single-site DMFT [47, 48]. We obtain results at non-zero temperatures ranging from 29 to 464 K by employing continuous-time quantum Monte Carlo in the hybridization expansion (CTHYB) [49] as the impurity solver and at effectively zero temperature using a matrix product states (MPS) based solver [40, 41].

We characterize the strength of electronic correlations by the inverse quasiparticle renormalization $Z^{-1} = 1 – \partial \Re \Sigma(\omega \to 0)/\partial \omega$ [34] related, in the single-site DMFT approximation, to the quasiparticle mass enhancement as $m^*/m = Z^{-1}$, shown in Fig. 2. For both materials the calculated low-temperature mass enhancements agree with experimental specific heat measurements, which indicate that the overall mass enhancement of Sr$_2$RuO$_4$ is about 4 [3, 6, 50], while for Sr$_2$MoO$_4$ correlations are weaker and result in a mass enhancement of only around 2 [29, 31]. From the specific heat $c_p \sim \sum_l (m^*/m_l) N_l(E_F)$, where $l \in \{xy, xz, yz\}$ and $N_l(E_F)$ is the bare DOS at the Fermi energy, we obtain a specific heat ratio $c^{SRO}_{SMO} = 2.4$, which is in good agreement with the experimental value of about 2.8 [29].

At zero temperature, the $xz/yz$-orbital mass enhancements of the two materials are approximately in the same ratio as the inverses of the respective band widths. The
FIG. 3. Many-body electronic structure obtained with DMFT for Sr$_2$MoO$_4$ (top panels) and Sr$_2$RuO$_4$ (bottom panels) at $T = 232$ K. Left Panels: Momentum-resolved spectral function $A_k(\omega)$ (false color) along a high-symmetry $k$-path through the Brillouin zone compared to the Wannier bands (dashed blue lines). Insets: Spectral function $A_k(\omega = 0)$ in the $k_z = 0$ plane. Right panels: Momentum-integrated spectral function $A(\omega)$ (per spin) for the $xy$ (blue) and the $xz/yz$ (red) orbitals. Black arrows point to the Hund’s peaks. Note the different range of energy in comparison to Fig. 1.

situation for the $xy$ orbital is different: For Sr$_2$RuO$_4$, in agreement with previous works [24, 25], we find that even though the $xy$ orbital has the larger band width, its mass enhancement is nearly twice as large as the mass enhancement of the $xz/yz$ orbitals. The unusually large $xy$ orbital mass enhancement of Sr$_2$RuO$_4$ has been attributed to the proximity of the van Hove singularity to the chemical potential [24, 25]. Conversely, for Sr$_2$MoO$_4$ the van Hove singularity is far removed from the chemical potential, and the mass enhancements are consistent with the difference in the bare band widths; the $xy$ orbital is substantially less correlated than the $xz/yz$ orbitals. As Sr$_2$RuO$_4$ is cooled, the mass enhancements exhibit a strong temperature and orbital dependence with no sign of saturation above 30 K. This is in accordance with a Fermi liquid temperature of about 25 K [25, 50]. For Sr$_2$MoO$_4$, we observe only a weak temperature dependence of the mass enhancement, and its saturation at about 100 K indicates a much higher Fermi liquid coherence scale than in Sr$_2$RuO$_4$. These findings suggest that the van Hove singularity provokes a suppression of the Fermi liquid temperature in Sr$_2$RuO$_4$ and demonstrate the importance of capturing the interplay of correlation physics and specifics of band structure to understand the quasiparticle properties in strongly-correlated materials.

In contrast to the van Hove singularity, the spin-orbit coupling does not influence the mass enhancements of Sr$_2$RuO$_4$ [13, 40]. However, it is known from theory and experiment that electronic correlations lead to an effective spin-orbit coupling two times larger than its bare value [6, 12, 13, 40, 51]. By using the MPS-based impurity solver for calculations with spin-orbit coupling, we find that the same picture holds in Sr$_2$MoO$_4$, yielding a slightly higher enhancement factor of about 2.5 (see supplemental material [34]). We can therefore conclude that the correlation-enhanced spin-orbit coupling in both materials is mainly a result of local interactions [51] rather than a consequence of van Hove physics.

The materials’ similarities and differences are also evident in the correlated spectral function, shown in the left panel of Fig. 3. We see that for Sr$_2$RuO$_4$, the unoccupied states conform closely to the bare bands, while the occupied bands are shifted substantially towards the chemical potential. For Sr$_2$MoO$_4$, the renormalization is less severe, and the unoccupied states differ considerably from the DFT bands. In the insets of Fig. 3, we show the spectral function at $T = 232$ K and $\omega = 0$ in the $k_z = 0$ plane, practically a many-body version of the Fermi surfaces of Fig. 1 at finite temperature. These many-body Fermi surfaces portray the major differences found in the $xy$ orbitals. While the $xy$ sheet is very sharp in Sr$_2$MoO$_4$, we find it to be broadened in Sr$_2$RuO$_4$. This is caused by the van Hove singularity in Sr$_2$RuO$_4$, which is shifted even closer to the chemical potential due to electronic correlations.

Results for the orbitally-resolved self-energies at $T = 232$ K are presented in the inset of Fig. 2. Note that for Sr$_2$RuO$_4$ what is shown is the negative of the reflection of the self-energy through $\omega = 0$, i.e. $-\Sigma(-\omega)$. The $xz/yz$ self-energies for the two materials have a clear qualitative similarity, showing that for these orbitals Sr$_2$MoO$_4$ is - to a good approximation - indeed the particle-hole dual of Sr$_2$RuO$_4$. The self-energies have a negative slope at $\omega = 0$, corresponding to the usual low-energy reduction of the quasiparticle velocity due to strong correlations. There is, however, an interesting inversion of slope around $\omega = -0.2$ eV, which has been pointed out in several DMFT works on Sr$_2$RuO$_4$ [4, 13, 25, 52]. For Sr$_2$MoO$_4$ we find that the inversion of slope is only present in the $xz/yz$ self-energy.

The inversion of slope occurs still well within the bare band width, and may lead to a ‘retracted’ renormalization of the quasiparticle dispersion. The consequence is an additional side-peak in the spectral function $A(\omega)$ (marked with small arrows in Fig. 3, right panels), which cannot be related to a structure present in the non-interacting DOS. Model system calculations indicate that the inverted slope and the corresponding side-peak in $A(\omega)$ are characteristic signatures of the spin-orbital separation occurring in Hund’s metals [20–22]. For Sr$_2$MoO$_4$, with two electrons in three orbitals, the screening of the orbital degrees of freedom requires binding a conduction band electron to the correlated site, resulting in the formation of a large $S = 3/2$ local mo-
ment [21]. Breaking this composite spin requires the removal of an electron, and thus an excitation corresponding to the energy of this process can be expected in the electron-removal part of the spectrum. Conversely, for Sr$_2$RuO$_4$ with a more than half-filled shell, i.e. four electrons in three orbitals, the screening involves an additional hole, and thus the Hund’s metal side-peak is found at positive energies.

To our knowledge, no photoemission experiment has yet observed this side-peak, probably because most studied Hund’s metals have more than half-filled correlated shells so the Hund’s peak is on the unoccupied side of the spectrum and not observable in photoemission. Crucially, for Sr$_2$MoO$_4$ the Hund’s metal peak is present on the occupied side and therefore observable in photoemission. However, in the momentum-integrated spectral function $A(\omega)$, the Hund’s metal peak is a relatively weak feature. We therefore show here how an exploration of the momentum dependence of the spectral function also reveals the importance of Hund’s physics.

Examination of Fig. 3 shows that for Sr$_2$MoO$_4$ along the $\Gamma$-X path, there are two pronounced spectral features on the occupied side, one at $-0.2\ eV$ corresponding to the renormalized $xz/yz$-derived bands and another corresponding to the strongly dispersing $xy$-derived band. Between these two is an additional spectral weight which corresponds to the Hund’s metal excitation (see also supplemental material [34]). The Hund’s metal spectral weight roughly follows the energy of the lower non-interacting $xz/yz$-derived band. We also see that the occupied side of $A_k(\omega)$ of Sr$_2$MoO$_4$ is very different from that of Sr$_2$RuO$_4$. The latter shows strongly renormalized $xz/yz$-derived bands and a very incoherent $xy$ quasiparticle dispersion only visible around zero energy close to the X-point. For Sr$_2$RuO$_4$ the Hund’s metal physics is responsible for the weight on the unoccupied side above $\sim 0.3\ eV$ on the X-M path.

In Fig. 4 we examine the Hund’s peak physics in more detail by focusing on the energy dependence of the spectrum at two characteristic momentum points. Concentrating first on the $\Gamma$ point, for the parameters believed to be relevant to Sr$_2$MoO$_4$ and Sr$_2$RuO$_4$, a distinct three-peak structure is observed on the occupied side of the spectrum (black line). Following our discussion above, the peak closest to the chemical potential stems from the strong renormalization of the bare $xz/yz$ bands, the peak furthest from the chemical potential results from the $xy$ orbital, and the peak in the middle is a direct consequence of the Hund’s metal nature of Sr$_2$MoO$_4$. Based on our calculations, the three peaks are well separated and the intensity of the Hund’s metal peak is similar to the intensity of the other two peaks.

Changing the Coulomb repulsion or Hund’s coupling away from the physically expected values changes the behavior. At fixed $U$ the Hund’s peak shifts away from the chemical potential with increasing $J$ (right panel), while at fixed $J$ it shifts towards the chemical potential when $U$ is increased (left panel). Increasing $J/U$ will favor the formation of a composite $S = 3/2$ impurity spin, leading to an increased coherence energy scale for the orbital screening process, and thus the Hund’s metal peak likewise moves to higher (negative) energies. Of course, the three-peak structure is only present for parameters within the Hund’s metal regime. For too small $J/U$ the three-peak structure ceases to exist, as is indeed the case for $U = 2.3\ eV$ and $J = 0.2\ eV$, see Fig. 4 right panel. We see similar behavior at the X point (Fig. 4 bottom panels), although the Hund’s side-peak is much less pronounced than at the $\Gamma$ point. For $U = 2.3\ eV$ and $J = 0.4\ eV$ (black lines), we find a small Hund’s side-peak at $\omega \sim 0.5\ eV$. The peak moves closer to the chemical potential for increasing $U$ (left panel), while the peak moves in the opposite direction for increasing $J$. For $J = 0.2\ eV$ the peak disappears entirely.

We remark that the calculated dependence on $U$ and $J$ excludes that the observed peak originates from the atomic multiplet structure because the multiplet splitting would evolve in the opposite way as $J$ is varied. The multiplet structure has been used to determine $J$, for example in the Mott-insulating state of Ca$_2$RuO$_4$, resulting in the same Hund’s coupling as used in this work: $J = 0.4\ eV$ [53]. Our work shows that the position of the Hund’s peak can provide information on the interaction strength in a material which is metallic and where the atomic multiplet structure is not experimentally visible.

In this paper we have presented a study of the correlated electronic structure of Sr$_2$MoO$_4$ in comparison with that of the well understood material Sr$_2$RuO$_4$. The difference in electron density (2 electrons per Mo vs 4 per Ru) leads to similarities (in many respects Sr$_2$MoO$_4$ is the particle-hole dual of Sr$_2$RuO$_4$) but also to pronounced differences in physics, which can be used to gain

![FIG. 4. DMFT spectral function of Sr$_2$MoO$_4$ at the $\Gamma$-point (top row) and the X-point (bottom row) for different values of $U$ at fixed $J = 0.4\ eV$ (left panels) and for different values of $J$ at fixed $U = 2.3\ eV$ (right panels) calculated at $T=232\ K$. Arrows point to the Hund’s peaks.](image-url)
understanding of the interplay between correlated electron physics and band structure aspects. For Sr$_2$MoO$_4$ the van Hove singularity is far from the chemical potential, while for Sr$_2$RuO$_4$ it is very close. A comparison of the two materials therefore provides insight into the importance of van Hove physics in Hund’s metals. Perhaps of more fundamental significance, for Sr$_2$MoO$_4$ the characteristic spectral features theoretically predicted to arise in Hund’s metals occur on the occupied side of the electronic spectrum and should therefore be accessible to photoemission experiments. Single-crystalline Sr$_2$MoO$_4$ thin films have been synthesized [30], allowing for angle-resolved measurements. Sr$_2$MoO$_4$ is thus an ideal system for direct experimental tests of the novel aspects of Hund’s metal physics. Further, an experimental observation of the Hund’s metal peak in Sr$_2$MoO$_4$ would open the intriguing possibility to experimentally determine $J/U$ in a metallic system.

In Sr$_2$MoO$_4$ no superconductivity has been found up to date. Nevertheless, we believe that future studies of this material could bring key insights on the importance of Hund’s physics, spin-orbit coupling, the van Hove singularity, and other band structure details for the emergence of unconventional superconductivity in Sr$_2$RuO$_4$.

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Supplemental Material for

“Sr$_2$MoO$_4$ and Sr$_2$RuO$_4$: Disentangling the Roles of Hunds and van Hove Physics”

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I. METHOD

We perform DFT calculations using WIEN2k [32] with the standard PBE version of the GGA functional [33]. For both materials we use the experimentally determined I4/mmm crystal structure, with $a = b = 3.907$ Å and $c = 12.843$ Å for Sr$_2$MoO$_4$ [31], and $a = b = 3.861$ Å and $c = 12.722$ Å for Sr$_2$RuO$_4$ [60]. The DFT calculations are converged on a $21 \times 21 \times 21$ k-point grid with $\mathcal{R}K_{\text{max}} = 7$. We use wien2wannier [35] and Wannier90 [36] to construct maximally localized Wannier functions [38, 39] of $t_{2g}$ symmetry on a $10 \times 10 \times 10$ k-point grid and employ a frozen energy window from $[-2.0, 1.5]$ eV for Sr$_2$MoO$_4$ and $[-1.8, 3.0]$ eV for Sr$_2$RuO$_4$.

We use Hubbard-Kanamori on-site interactions [45]:

$$H = U \sum_l n_{l\uparrow}n_{l\downarrow} + \sum_{l<\ell',\sigma} [U' n_{l\sigma}n_{\ell'\sigma} + (U' - J) n_{l\sigma}n_{\ell'\bar{\sigma}}]
-J c_{l\sigma}^\dagger c_{\ell'\sigma} c_{\ell'\bar{\sigma}} c_{l\bar{\sigma}} - J \sum_{l<\ell'} [c_{l\uparrow}^\dagger c_{\ell'\uparrow} c_{\ell'\downarrow} c_{l\downarrow} + H.c.]$$

with $l \in \{xy, zz, yz\}$ and $U' = U - 2J$. With the exception of main text Fig. 4, we have assumed the same $U$ and $J$ of 2.3 eV and 0.4 eV for both materials. These values are commonly used for Sr$_2$RuO$_4$ [4, 9, 13, 14, 24, 40]. We note that cRPA estimates for the interaction parameters are about 8% higher for Sr$_2$MoO$_4$ [61].

We perform DMFT calculations using the TRIQS library [42] and the TRIQS/DFTTools application [48], using a very dense $400 \times 400 \times 400$ k-point grid. The calculations are “one-shot” DFT+DMFT, meaning that the DFT density is kept fixed and not updated. We absorb the double counting correcting into the chemical potential, as we purely work in the low-energy subspace defined by the $t_{2g}$-like Wannier orbitals.

For calculations at a set of finite temperatures between 29 and 464 K, we solve the impurity problem using continuous-time quantum Monte Carlo in the hybridization expansion (CTHYB) [43, 49]. To obtain high-quality data we use a total of $\sim 10^9$ measurements in the last iteration. All $T = 0$ K calculations are carried out using a matrix product states (MPS) based impurity solver [40, 41]. Conceptually, the MPS-based solver is equivalent to impurity solvers based on exact diagonalization [59], and thus also allows for the inclusion of SOC. Calculations including SOC with CTHYB are limited to high temperatures due to a severe sign problem. The results of calculations with SOC are discussed in Sec. III and methodological details on the MPS-based solver are provided in Ref. [40] and Sec. II below.

For the calculation of quantities on the real-frequency axis, we use the inversion method [64] for to analytically continue the self-energy. We employ the stochastic analytic continuation, following the approach by Beach [44].

We quantize the strength of correlations with the renormalization factor:

$$Z = \left(1 - \frac{\partial \text{Im} \Sigma(i\omega_n)}{\partial \omega_n} \bigg|_{\omega_n \to 0} \right)^{-1}.$$

We determine $Z$ by fitting a polynomial of 4th order to the lowest 6 points of the Matsubara self-energies and extrapolate Im$\Sigma(i\omega_n \to 0)$, a procedure also used in Refs. [9, 24].

II. MPS-BASED SOLVER

For calculations at $T = 0$ K (with and without SOC), we use the MPS-based impurity solver in imaginary time as introduced in [41] and already successfully applied to Sr$_2$RuO$_4$ [40]. We refer to Refs. [40, 41] for methodological details. The calculations are performed using the SyTen toolkit [62, 63].

For numerical purposes, and to compare to CTHYB calculations, we use a discrete grid of Matsubara frequencies at a (fictitious) inverse temperature $\beta_{\text{fict}} = 200 \text{ eV}^{-1}$. Without the inclusion of spin-orbit coupling, the bath consists of three SU(2) symmetric orbitals with $L_b = 8$ bath sites per spin and orbital. We use five quantum numbers: the occupation parity of each orbital, the particle number, and the spin. Occupation parity is important due to the pair hopping in the Hubbard-Kanamori Hamiltonian, such that odd and even sectors are disconnected, trapping ground state searches; auxiliary small single-particle hopping terms are numerically unreliable and deteriorate the quality of results. Ground state
We add a spin-orbit coupling of strength \( \lambda \), see Refs. [12, 40], to the Wannier Hamiltonian. We determine the SOC strength \( \lambda \) by matching the resulting eigenenergies to the Kohn-Sham bands obtained from DFT calculations with SOC included, see Fig. 5. Using SOC strengths of \( \lambda^\text{SRO}_{\text{DFT}} = 100 \text{ meV} \) for \( \text{Sr}_2\text{RuO}_4 \) and \( \lambda^\text{SMO}_{\text{DFT}} = 80 \text{ meV} \) for \( \text{Sr}_2\text{MoO}_4 \) results in a nearly perfect agreement with DFT. For \( \text{Sr}_2\text{RuO}_4 \), the SOC substantially reshapes the Fermi surface, cf. Figs. 1 and 5, which has been also shown for example in Refs. [6, 12, 13, 37]. In contrast to \( \text{Sr}_2\text{RuO}_4 \), the SOC does not distort the shape of the Fermi surface for \( \text{Sr}_2\text{MoO}_4 \).

With SOC included, the determination of the Green’s functions is numerically much more involved. We consider bath sizes \( L_b = 4 \), shown in Ref. [40] to be sufficiently accurate for \( \text{Sr}_2\text{RuO}_4 \) when SOC is included. The remaining quantum numbers are the particle number and the \( z \)-component of the total angular momentum in the \( J \)-basis. Ground state searches result in bond dimension of 4096, and for the imaginary time evolution two-site TDVP is used up to \( \tau = 100 \text{ eV} \) in time steps of \( \Delta \tau = 0.05 \text{ eV} \) with subsequent linear prediction. The problem of determining a matrix-valued (6 × 6)-dimensional Green’s function (3 bands, 2 non-degenerate spin orientations) is alleviated by rotating to the \( J \)-basis, reducing the problem to two 1 × 1 blocks and two 2 × 2 blocks [40].

To study the effect of SOC, we add a local \( t_{2g} \)-only spin-orbit term, see e.g. in Refs. [12, 40], to the Wannier Hamiltonian. We determine the SOC strength \( \lambda \) by matching the resulting eigenenergies to the Kohn-Sham bands obtained from DFT calculations with SOC included, see Fig. 5. Using SOC strengths of \( \lambda^\text{SRO}_{\text{DFT}} = 100 \text{ meV} \) for \( \text{Sr}_2\text{RuO}_4 \) and \( \lambda^\text{SMO}_{\text{DFT}} = 80 \text{ meV} \) for \( \text{Sr}_2\text{MoO}_4 \) results in a nearly perfect agreement with DFT. For \( \text{Sr}_2\text{RuO}_4 \), the SOC substantially reshapes the Fermi surface, cf. Figs. 1 and 5, which has been also shown for example in Refs. [6, 12, 13, 37]. In contrast to \( \text{Sr}_2\text{RuO}_4 \), the SOC does not distort the shape of the Fermi surface for \( \text{Sr}_2\text{MoO}_4 \).

Additionally, the SOC leads to a mixed \( xy-xz/yz \) orbital character of degenerate and nearly degenerate states. On the Fermi surface, the mixed orbital character is especially important along the diagonal direction \( \Gamma-X \), see inset of Fig. 5. For \( \text{Sr}_2\text{RuO}_4 \), this results in states on the Fermi surface with 50:50 mixing of \( xy \) and \( xz/yz \) characters, while for \( \text{Sr}_2\text{MoO}_4 \) the mixing of the orbital character is much less pronounced.

For \( \text{Sr}_2\text{RuO}_4 \) it has been predicted theoretically [12, 13, 51] and confirmed experimentally [6] that electronic correlations lead to an enhancement of the effective SOC. To be precise, the off-diagonal elements of the self-energy have the same structure as the SOC term and are found to be almost frequency independent. Hence, the physics can be described - to a good approximation - by an effectively enhanced SOC term \( (\lambda^\text{eff} = \Delta\text{DFT} + 2\Sigma_{\text{off}}(\omega = 0)) \), following the definition of \( \lambda \) in Ref. [40]) and a purely diagonal self-energy. For \( \text{Sr}_2\text{RuO}_4 \), the MPS-based impurity solver yields a correlation-enhanced effective SOC of \( \lambda^\text{SRO} = 192 \text{ meV} \) and \( \lambda^\text{SMO} = 179 \text{ meV} \) [40], which is
nearly two times larger than $\lambda_{\text{DFT}}$. For Sr$_2$MoO$_4$, we find similar correlation-enhanced SOC of $\lambda_{\text{SMO}}^{\text{xy}} = 204$ meV and $\lambda_{\text{SMO}}^{\text{yz}} = 182$ meV, see Fig. 6, while the bare SOC is 20 meV smaller than for Sr$_2$RuO$_4$. Thus, at the same interaction values of $U$ and $J$, electronic correlations enhance the SOC by a factor of about 2.5, which is even more than what is found for Sr$_2$RuO$_4$.

On the other hand, the diagonal elements are nearly unchanged by the inclusion of SOC, as shown in Fig. 7 for Sr$_2$MoO$_4$ and in Ref. [40] for Sr$_2$RuO$_4$. This implies that the mass enhancements of both materials are not influenced by the SOC.

IV. IMAGINARY PART OF SELF-ENERGY

Fig. 8 shows the imaginary part of the real frequency self-energy (corresponding to the real part in Fig. 2) at $T = 232$ K. For Sr$_2$RuO$_4$ what is shown is the reflection of the self-energy through $\omega = 0$, i.e. $\Sigma(-\omega)$. Like the real part, the imaginary part of the Sr$_2$RuO$_4$ $xz/yz$-orbital self-energy has a similar structure to that of Sr$_2$MoO$_4$. We also note that the Sr$_2$MoO$_4$ $xy$-orbital self-energy has a smaller imaginary part at $\omega = 0$ than the $xz/yz$ orbitals, opposite to what we find for Sr$_2$RuO$_4$. In the latter, the substantial electron-electron scattering in the $xy$ orbital and the presence of the van Hove singularity at the chemical potential leads to a broadening of the $xy$-derived many-body Fermi surface sheet at finite temperature, see insets of Fig. 3. For Sr$_2$MoO$_4$, where the van Hove singularity is not in proximity to the chemical potential, the $xy$ orbital is much more coherent with a smaller imaginary part of the self-energy at $\omega = 0$, resulting in a very sharp many-body Fermi surface.

V. QUASIPARTICLE DISPERSIONS

To better understand how the peculiar structure in the real part of the self-energies leads to an additional peak in the spectral function, we look at the quasiparticle dispersions, given by det $\left[ \omega + \mu \delta_{l,l'} - H_{l,l'}(k) - \text{Re} \left[ \Sigma_l(\omega) \delta_{l,l'} \right] \right] = 0$. Note that without SOC the self-energy in the orbital basis is diagonal. We focus only on the $\Gamma$ and $X$ points and take into account that for the studied materials at those points this equation simplifies to $\omega - \epsilon_l(k) = \text{Re} \left[ \Sigma_l(\omega) \right] - \mu$ due to $H_{l,l'}(\Gamma,X) = \epsilon_l(\Gamma,X)\delta_{l,l'}$. The left hand side, $\omega - \epsilon_l(k)$, of the equation is shown as straight lines in Fig. 9 and the intersections with $\text{Re} \left[ \Sigma_l(\omega) \right] - \mu$ give the solutions of the quasiparticle equation, which yield peaks in the corresponding spectral functions. There are also peaks that can emerge when the quasiparticle equation is almost satisfied, given that the imaginary part of the self-energy introduces a large enough broadening of the peaks. For example, at the $\Gamma$ point this leads to a peak at about $-0.2$ eV in the $xz$ and $yz$ orbitals. We emphasize that this peak would prevail even without the inversion of slope in the self-energy. On the contrary, the second peak at about $-0.8$ eV is a direct consequence of the inverted slope (together with the fact that it stays flat at further negative frequencies), and hence this peak is a signature of the Hund’s metal physics governing Sr$_2$MoO$_4$. 

![Fig. 7. Comparison of Matsubara self-energies for Sr$_2$MoO$_4$ obtained with the MPS-based impurity solver at $T = 0$ with (orange) and without (green) SOC compared to CTHYB results obtained without SOC at $T = 58$ K. Interaction parameters are $U = 2.3$ eV, and $J = 0.4$ eV and we substracted the chemical potential from the real parts.](image1)

![Fig. 8. Imaginary part of the real-frequency self-energy for both materials at $T = 232$ K using CTHYB as impurity solver [40] with $U = 2.3$ eV and $J = 0.4$ eV and analytic continuation [44] to the real-frequency axis. For Sr$_2$RuO$_4$ (dashed lines) we show the reflection of the self-energies through $\omega = 0$, i.e. $\Sigma(-\omega)$. The real parts are shown in the inset of main text Fig. 2.](image2)
FIG. 9. Relationship of the inverted slope in the real part of the self-energy to the peaks in the spectral functions for Sr$_2$MoO$_4$ at $T = 232$ K. The orbitally resolved spectral functions (blue lines) are shown together with the real parts (orange lines) and imaginary parts (red lines) of the self-energy and the linear function $\omega - \epsilon_l(k)$ (see text for further details). The top row shows the $\Gamma$ point and the bottom row the $X$ point, and the columns correspond to the different orbitals.