

Strongly Correlated Materials from a Numerical Renormalization Group Perspective: How the Fermi-liquid State of Sr_2RuO_4 Emerges

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The crossover from fluctuating atomic constituents to a collective state as one lowers temperature/energy is at the heart of the dynamical mean-field theory description of the solid state. We demonstrate that the numerical renormalization group is a viable tool to monitor this crossover in a real-materials setting. The renormalization group flow from high to arbitrarily small energy scales clearly reveals the emergence of the Fermi-liquid state of Sr_2RuO_4 . We find a two-stage screening process, where orbital fluctuations are screened at much higher energies than spin fluctuations, and Fermi-liquid behavior, concomitant with spin coherence, below a temperature of 25 K. By computing real-frequency correlation functions, we directly observe this spin-orbital scale separation and show that the van Hove singularity drives strong orbital differentiation. We extract quasiparticle interaction parameters from the low-energy spectrum and find an effective attraction in the spin-triplet sector.

Introduction.—Atoms with partially filled shells have a spectrum of many-body eigenstates with degeneracies associated with fluctuating spin and orbital moments. For instance, the isolated ruthenium atom in the Ru^{4+} configuration, subject to an octahedral crystal field, has a nine-fold degenerate groundstate corresponding to spin and orbital quantum numbers $S=L=1$ [1, 2]. In materials with strong electronic correlations, these local fluctuations can be observed at high temperature and energy through, e.g., Curie-Weiss-like spin susceptibilities. In correlated metals, these fluctuations are suppressed as one reaches low temperature and energy. In the Fermi-liquid regime, a nondegenerate collective groundstate is formed, with long-lived coherent quasiparticle excitations and susceptibilities displaying Pauli behavior [3].

How the crossover from fluctuating atomic constituents to a collective state takes place is at the heart of the dynamical mean-field theory (DMFT) description of the solid state [4]. In this theory, each atom is viewed as exchanging electrons with an environment which self-consistently represents the whole solid. The gradual suppression of local fluctuations can be thought of as a self-consistent (multi-stage) Kondo screening process [5] of both spin and orbital moments [6, 7].

The renormalization group (RG) is the appropriate framework to describe and monitor these crossovers as a function of energy scale. Indeed, Wilson’s numerical renormalization group (NRG) [8] has been a tool of choice for solving DMFT equations for lattice models with few orbital degrees of freedom [9], with the additional merit of providing real-frequency properties at any temperature. Following a number of two-particle applications [10–15], recently, even three-orbital studies have become possible [6, 7, 16–19]. Yet, all of these works operated in the

model context. We demonstrate here that NRG can be successfully applied to an actual material, accounting for its electronic structure in a realistic manner using density functional theory (DFT) and DMFT [20].

The material we focus on, Sr_2RuO_4 , is one of the more thoroughly studied quantum materials [21] and an ideal testbed for fundamental developments in quantum many-body theories. Besides the unconventional superconducting state below ~ 1.5 K [22, 23], also the normal, Hund-metal state of Sr_2RuO_4 [2, 7, 24–27] attracts attention, due to text-book Fermi-liquid behavior below $T_{\text{FL}} \approx 25$ K [21, 28–33] (though signatures of quasiparticles are found up to elevated temperatures of ~ 600 K [24]). However, temperatures below T_{FL} could not be reached with controlled computational methods hitherto.

In this Letter, we show that Sr_2RuO_4 undergoes a two-stage Kondo screening process [6, 7, 26], where orbital fluctuations are screened well before the spin degrees of freedom. We determine the associated Kondo temperatures to $T_{\text{orb}} \approx 6000$ K and $T_{\text{sp}} \approx 500$ K, respectively, and show that Fermi-liquid behavior emerges when spin coherence is fully established below a scale of $T_{\text{FL}} \approx 25$ K [34]. With NRG as impurity solver, the entire DMFT calculation is performed on the real-frequency axis [35], and we can compute correlation functions at arbitrarily low energy scales and temperatures. Hence, we are able to go beyond previous Monte Carlo-based DFT+DMFT studies [24, 26, 27, 33, 38–41] and enter deep into the Fermi-liquid regime, even down to $T=0$ [42]. This enables us to explore the counter-intuitive observation that the more itinerant (xy) orbital has the smaller quasiparticle weight [21, 24, 27, 38, 41, 44, 45]. We show that this effect is driven by a van Hove singularity close to the Fermi level, as elaborated in [24].

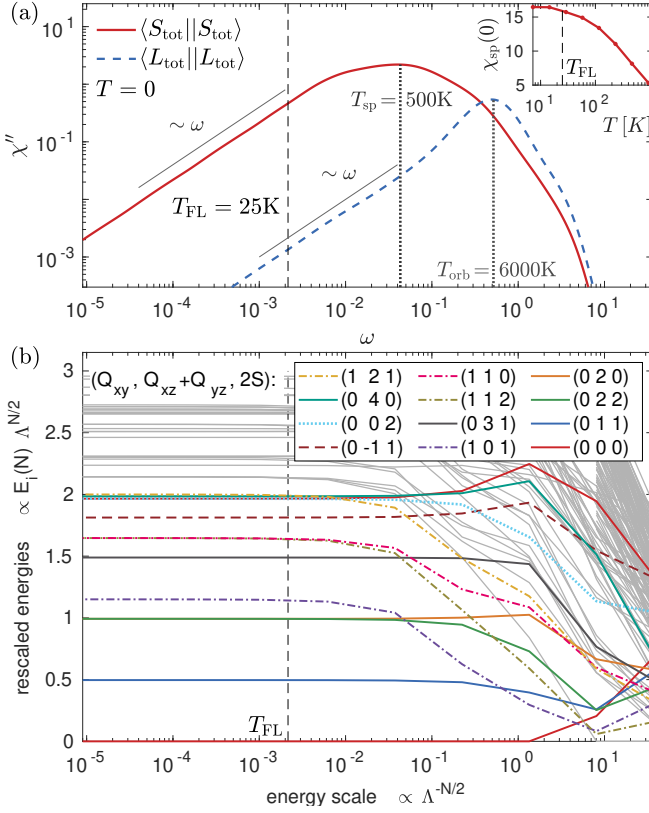


FIG. 1. (a) Dynamic spin and orbital susceptibilities, $\chi''_{\text{sp}}(\omega)$ and $\chi''_{\text{orb}}(\omega)$, showing spin-orbital scale separation. Inset: Static spin susceptibility as a function of temperature. (b) NRG flow diagram, showing the rescaled eigenenergies (with quantum numbers given in the legend) as a function of the energy scale [50], for the impurity model at self-consistency. The spin and orbital Kondo temperatures (maximum of χ'') and the Fermi-liquid scale, T_{FL} , are marked by vertical lines.

Model.—The low-energy structure of Sr_2RuO_4 can be well described by a local basis of three maximally localized Wannier functions [46, 47] with Ru-4d t_{2g} symmetry denoted by $\{xy, xz, yz\}$. The corresponding non-interacting Wannier Hamiltonian is characterized by the density of states (DOS) shown in Fig. 3(a) below, reflecting the quasi-2D tetragonal crystal structure of Sr_2RuO_4 , with quasi-2D xy orbitals and a strongly one-dimensional character of the degenerate xz/yz orbitals. We employ the same Wannier Hamiltonian as in [39, 40, 48] (without spin-orbit coupling) combined with a local Kanamori interaction [2, 49] parametrized by $U=2.3$ and $J=0.4$ [24]. Throughout this work, we use $\text{eV}=1$ as unit of energy if not otherwise indicated. In the Hund-metal phase of Sr_2RuO_4 , the pair-hopping term of the Kanamori interaction is almost inactive and can be neglected to obtain a model with higher symmetry, more tractable for NRG [50].

Spin-orbital separation, Fermi liquid.—Since NRG can reach arbitrarily small energy scales, we are able to directly observe both spin-orbital scale separation and the onset of Fermi-liquid behavior. The zero-temperature real-frequency orbital and spin susceptibilities [50], χ''_{orb}

and χ''_{sp} , exhibit a separation of their maxima by more than one decade, see Fig. 1(a). This spin-orbital separation in Kondo scales, with $T_{\text{orb}} \approx 6000\text{K}$ and $T_{\text{sp}} \approx 500\text{K}$ as found from the maxima of χ'' , is distinctive of correlated Hund metals [2, 6, 7, 19], where the Hund coupling J causes the screening of the respective fluctuations to occur at disparate energy scales. Further, completed screening of fluctuations is signaled by linear behavior, $\chi'' \propto \omega$, found below roughly 1000 K and 25 K for χ''_{orb} and χ''_{sp} , respectively. The fully coherent, Fermi-liquid state thus emerges below an energy scale of 25 K. The Fermi-liquid onset is also seen in the temperature dependence of the static spin susceptibility, $\chi_{\text{sp}}(\omega=0)$, which crosses over from Curie-Weiss- to Pauli-like behavior, saturating below $T_{\text{FL}} \approx 25\text{K}$, see inset of Fig. 1(a). These results clearly establish spin-orbital scale separation in the low-temperature Fermi-liquid state of Sr_2RuO_4 , as proposed by previous studies above T_{FL} [7, 26].

A very direct observation of Fermi-liquid behavior is possible by studying the renormalization group flow diagram of the NRG algorithm [6, 9, 18, 19]. Figure 1(b) shows the NRG-Hamiltonian's (lowest) rescaled eigenenergies, $\Lambda^{N/2}E_i(N)$, depending on the energy scale $\Lambda^{-N/2}$ of the RG flow, where Λ is the NRG discretization parameter and N the length of the Wilson chain [50]. At high energy, the states are pure atomic eigenstates, which are screened by the bath when flowing down in energy. Below T_{FL} , the Fermi liquid is formed. There, the flow reaches a fixed point, where the rescaled eigenenergies become independent of N , $\Lambda^{N/2}E_i(N) = E_i^*$. The Fermi-liquid nature of this fixed point is determined by “towers” [9] of equidistant excitation energies within the same symmetry sector, where each E_i^* is composed of n quasiparticle excitations, $E_i^* = n \cdot E_{\text{qp}}$.

Each eigenstate has the quantum numbers $(Q_{xy}, Q_{xz} + Q_{yz}, 2S)$, with orbital-resolved charge, Q_m , relative to the groundstate, and total spin, S . The most prominent tower of states stems from xz/yz quasiparticles, i.e., eigenstates with quantum numbers $(0, 0, 0)$, $(0, 1, 1)$, $(0, 2, 2)$, $(0, 2, 0)$, $(0, 3, 1)$, etc., see solid lines in Fig. 1(b). States with an additional xy quasiparticle are marked as dash-dotted

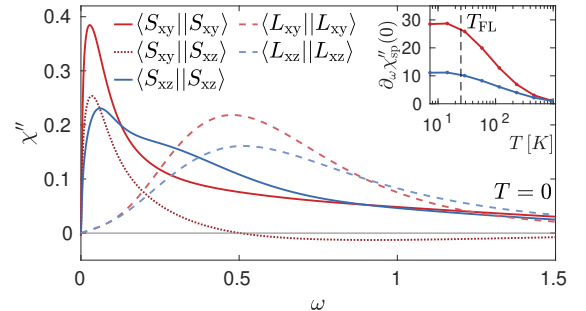


FIG. 2. Orbital-resolved, dynamic spin and angular-momentum susceptibilities, $\chi''(\omega)$. Inset: Temperature dependence of $\partial_\omega \chi''|_{\omega=0}$ in the spin sector, with T_{FL} marked as dashed line.

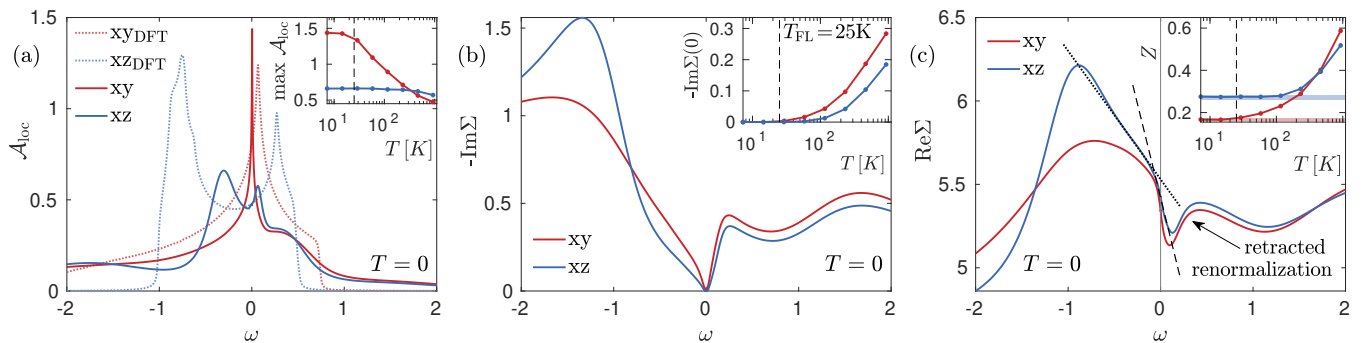


FIG. 3. Main panels: Real-frequency correlation functions at zero temperature. Insets: Characteristic values as function of temperature, converging below T_{FL} (dashed line). (a) Local spectral function, $\mathcal{A}_{\text{loc}}(\omega)$, from DFT+DMFT (solid lines) and DFT (dotted lines). Inset: $\max_{\omega} \mathcal{A}_{\text{loc}}(\omega)$. (b) Imaginary part of the self-energy, $\text{Im}\Sigma(\omega)$. Inset: $\text{Im}\Sigma(\omega=0)$. (c) Real part of the self-energy, $\text{Re}\Sigma(\omega)$, with the two linear regimes for $\omega < 0$ and the low-energy, positive slope for $\omega > 0$ highlighted. Inset: $Z = (1 - \partial_{\omega} \text{Re}\Sigma|_{\omega=0})^{-1}$; thick horizontal lines show the $T=0$ result for Z calculated via renormalized parameters.

lines. The Fermi-liquid scale, T_{FL} , is seen in the RG flow as the point where eigenstates with equal charge but different spin become degenerate, see the pairs $(0, 2, 0)$, $(0, 2, 2)$ and $(1, 1, 0)$, $(1, 1, 2)$. Our direct evidence of the Fermi-liquid scale of Sr_2RuO_4 , which conforms to the $T_{\text{FL}} \approx 25$ K found in experiments [21, 28–31], is one of the main results of this work.

In order to understand how the different orbitals behave regarding spin–orbital scale separation, we investigate in Fig. 2 the orbitally resolved spin and angular-momentum susceptibilities [50]. We find strong orbital differentiation with larger amplitude in the xy than the xz spin response, and generally a shift of spectral weight to lower frequencies in the xy compared to the xz orbital. In nuclear magnetic resonance (NMR) spectroscopy, the inverse nuclear spin-lattice relaxation time, $1/(T_1 T)$, is related to the zero-frequency slope of the electronic spin susceptibility, $1/(T_1 T) \propto \partial_{\omega} \chi''|_{\omega=0}$ (neglecting matrix elements) [62, 63]. Computing the orbitally resolved $\partial_{\omega} \chi''|_{\omega=0}$ as a function of temperature, we find that the xy response is about 2.5 times stronger than the xz response, see inset of Fig. 2, in qualitative agreement with experimental [31, 64, 65] and theoretical works [66]. The temperature dependence changes from linear to constant at T_{FL} , in a similar fashion for *both* orbitals, which we attribute to the strong orbital mixing on the two-particle level [39].

Single-particle spectrum.—Apart from the RG flow and the dynamic susceptibilities, our calculations also provide single-particle spectral information. Although the single-particle properties of Sr_2RuO_4 have been studied extensively [24, 26, 27, 33, 38, 45, 48] using continuous-time quantum Monte Carlo (CTQMC) solvers [67], these calculations have a challenging scaling with inverse temperature β , making it hard to reach the Fermi-liquid regime with $T < 25$ K, i.e., $\beta > 464 \text{ eV}^{-1}$. Additionally, the analytic continuation to real frequencies severely hampers spectral resolution [68]. Here, we go beyond previous works by analyzing Sr_2RuO_4 deep in the Fermi-liquid regime at low temperatures, and even $T=0$, directly on the real-frequency axis.

The local spectral function, $\mathcal{A}_{\text{loc}}(\omega)$, of Sr_2RuO_4 is considerably renormalized compared to the DFT DOS [24, 45, 48], see Fig. 3(a). When accounting for correlations, the spectral features are retained but shifted towards the Fermi level—both for the double peak in the xz/yz orbitals and the narrow xy peak. The latter is generated by the van Hove singularity in the xy orbital, which is shifted towards the Fermi level by electronic correlations. The height of the van Hove peak grows with decreasing temperature and saturates below T_{FL} , see inset of Fig. 3(a).

The imaginary part of the self-energy, $\text{Im}\Sigma(\omega)$, shown in Fig. 3(b), determines the lifetime of excitations. It has larger values at negative compared to positive frequencies, yielding shorter life times for hole excitations. Fermi-liquid behavior only emerges at frequencies below T_{FL} . The real part of the self-energy, $\text{Re}\Sigma(\omega)$, displays linear (Fermi-liquid) behavior on the same small energy scale, see Fig. 3(c). However, at $\omega \approx -100$ meV, it exhibits a “kink” leading to a second linear regime [lines in Fig. 3(c)], while, for ω in the range $+200$ – 400 meV, the slope of $\text{Re}\Sigma(\omega)$ changes sign, “retracting” the renormalization of the quasiparticle dispersion. Hence, in this energy range, the quasiparticle velocity is larger than the bare one [33], as opposed to the usual low-energy reduction due to strong correlations. These single-particle properties are in qualitative agreement with previous Monte Carlo results [24, 26, 27, 33, 38, 45, 48].

The pronounced differentiation between the different orbitals, seen in Figs. 1(b) and 2, is also reflected in the self-energy. The xy orbital shows much stronger correlations than the xz/yz ones, with higher curvature in $\text{Im}\Sigma(\omega)$ and steeper slope in $\text{Re}\Sigma(\omega)$ at $\omega=0$, as visible in Figs. 3(b) and 3(c), respectively. The slope is related to the quasiparticle weight, $Z = (1 - \partial_{\omega} \text{Re}\Sigma|_{\omega=0})^{-1}$, shown in the inset of Fig. 3(c). The zero-temperature values of Z agree with renormalized parameters extracted directly from the spectrum (horizontal lines, see discussion below) and are also consistent with experiments [21, 48]. The low-temperature relation $Z_{\text{xy}} < Z_{\text{xz}}$ contrasts with $Z_{\text{xy}} > Z_{\text{xz}}$ at high temperature. Indeed, when lowering temperature,

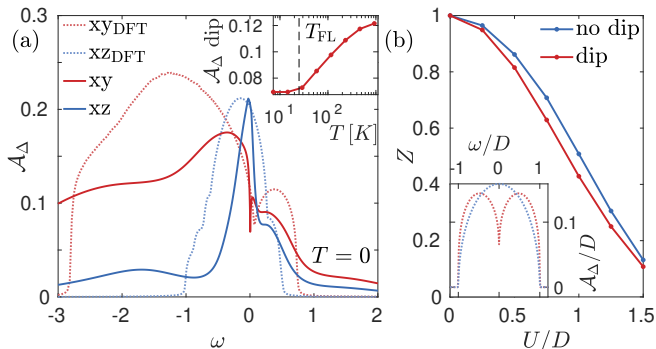


FIG. 4. (a) Spectral function of the hybridization, $\mathcal{A}_\Delta(\omega)$, in DFT+DMFT (solid lines) and DFT (dotted lines). Inset: temperature dependence of the van Hove dip in $\mathcal{A}_{\Delta,xy}(\omega)$. (b) Quasiparticle weight, Z , as a function of Hubbard U , for a simple two-orbital model with identical half bandwidth, D . The hybridization functions at $U=0$ are shown in the inset.

the quasiparticle weights cross at ~ 350 K, and, while Z_{xz} levels off at $T \sim 100$ K, Z_{xy} only saturates below T_{FL} . This shows that the coherence-to-incoherence crossover and the corresponding coherence scales in Sr_2RuO_4 are strongly orbital-dependent [24, 40]. It is only below T_{FL} that *all* orbitals are in the coherent Fermi-liquid regime.

At first sight, the stronger correlation in the xy orbital as compared to the xz/yz orbitals, indicated by $Z_{xy} < Z_{xz}$, is rather counterintuitive. Usually, the ratio between local Hubbard interaction U and the bandwidth W , U/W , is a good estimator for the strength of correlations. However, this clearly does not hold for Sr_2RuO_4 , since the xy orbital has a significantly larger bandwidth, $W_{xy} > W_{xz}$, see Fig. 3(a). In [24], it has been argued that the strong xy correlations result from the proximity of its van Hove singularity to the Fermi level, see Fig. 3(a).

To understand this, we consider the spectral part of the hybridization function, $\mathcal{A}_\Delta(\omega)$, of the self-consistent impurity model. The van Hove singularity in $\mathcal{A}_{\text{loc},xy}(\omega)$ generates a dip in $\mathcal{A}_{\Delta,xy}(\omega)$ [50] close to zero frequency, see Fig. 4(a), which implies a reduction of the effective coupling between impurity and bath at low energies for the xy orbital. The weaker coupling, in turn, increases the correlations and reduces the quasiparticle weight. The temperature dependence of the dip, inset of Fig. 4(a), matches the one of $\max_\omega \mathcal{A}_{\text{loc},xy}$ in the inset of Fig. 3(a).

To disentangle the effect of the van Hove singularity from other factors, we consider a simple, half-filled two-orbital model with both orbitals having the same bandwidth. We choose a semicircular lattice DOS for one orbital and set the second one such that its hybridization function has a dip at zero energy, see Fig. 4(b). Even in this simplified model, we find that Z is smaller in the orbital with a dip in the hybridization. This suggests that the relevant measure of the correlations strength is the Hubbard interaction divided by the effective low-energy hybridization strength, $U/\mathcal{A}_\Delta(\omega=0)$, rather than U/W .

Quasiparticle parameters.—Within the NRG framework, we can extract information about the Fermi liquid

and its quasiparticles not only from correlation functions but directly from the RG flow. To this end, we compute (zero-temperature) renormalized parameters from the low-energy spectrum of the (self-consistent) impurity model [69–71]. The impurity Green’s function has the low-energy expansion

$$G(\omega) \approx Z \cdot (\omega - \tilde{\epsilon} - Z\Delta(\omega))^{-1}, \quad \tilde{\epsilon} = Z \cdot (\epsilon + \Sigma(0)).$$

For a (finite) Wilson chain of length N , $G(\omega)$ has first-order poles at the single-particle excitation energies. Taking the lowest particle- and hole-excitation energy $E_i(N)$ from Fig. 1(b), we have two equations that can be solved for Z and $\tilde{\epsilon}$ and converged in N [69]. The results for Z [and $\tilde{\epsilon}$ or $\Sigma(0)$] are reported in [50] and agree quantitatively with those taken from $\Sigma(\omega)$, see inset of Fig. 3(c).

To go beyond the single-particle picture, we exploit that, at any finite N , there are residual quasiparticle interactions in the form of exponentially small corrections to the equidistant tower of quasiparticle excitations. By comparing two-particle-excitation energies $E_{mm'}^S$, with orbital indices m and m' and spin index S , to two single-particle excitations E_m and $E_{m'}$, the quasiparticle interaction $\tilde{U}_{mm'}^S$ is given by [69]

$$E_{mm'}^S - E_m - E_{m'} = \tilde{U}_{mm'}^S |\psi_m(0)|^2 |\psi_{m'}(0)|^2,$$

where $|\psi_m(0)|^2$ is the quasiparticle density at the impurity [50]. Hence, we are in the unique position to compute quasiparticle interactions $\tilde{U}_{mm'}^S$ as well as the zero-energy real-frequency vertex Γ , related via $\tilde{U}_{mm'}^S = Z_m Z_{m'} \Gamma_{mm'}^S$ [69, 71], for Sr_2RuO_4 . The results, listed in [50], show that the orbital dependence of $\tilde{U}_{mm'}^S$ is governed by Z_m , while $\Gamma_{mm'}^S$ displays only weak orbital dependence. Strikingly, the effective interaction in the spin-triplet sector is *attractive*. We attribute this to the same mechanism as the Hund-metal *s*-wave spin-triplet superconducting instability found in model studies [72, 73].

Conclusion.—By following the NRG flow starting from high and proceeding to the lowest temperature and energy scales, we have analyzed spin-orbital scale separation and the emergence of the Fermi liquid in Sr_2RuO_4 within a real-materials DFT+DMFT setting. Through linear frequency behavior of zero-temperature dynamic susceptibilities and fixed-point analysis of the NRG flow, we provide theoretical evidence for a Fermi-liquid scale, in remarkable agreement with the experimentally observed $T_{FL} \approx 25$ K [21, 28–31]. Characteristic quantities, like χ_{sp} and Z , are found to converge below 25 K. Further, our real-frequency and zero-temperature results substantiate a number of features, such as strongly shifted spectral peaks and the peculiar frequency dependence of the self-energy, previously found from analytically-continued Monte Carlo data [24, 26, 27, 33, 38–41]. We showed that the proximity of the van Hove singularity to the Fermi level drives strong orbital differentiation in Sr_2RuO_4 . Notably, the effect of van Hove singularities on the correlated state is

of importance even in non-transition metal systems like twisted bilayer graphene [74–76]. Finally, the extracted quasiparticle interactions \hat{U}_{mm}^S , reveal attractive coupling in the spin-triplet sector within our *ab initio* analysis. This paves the way towards a complete description of quasiparticles and their interactions in Sr_2RuO_4 , which are of crucial importance for the understanding of the still puzzling superconducting state [23, 77].

Generally, our work demonstrates the potential of DFT+DMFT+NRG as a new computational paradigm for real-material systems to (i) directly access real-frequency properties at arbitrarily low temperatures and (ii) reveal and elucidate the intricate renormalization process that occurs during the dressing of atomic excitations by their solid-state environment.

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Supplemental Material for “Strongly Correlated Materials from a Numerical Renormalization Group Perspective: How The Fermi-liquid State of Sr₂RuO₄ Emerges”

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In this Supplemental Material, we first provide the definitions of the susceptibilities shown in the main text, discuss the Hamiltonians and the neglect of pair hopping, and give some algorithmic details. Next, we list the quasiparticle parameters deduced from the NRG flow. Finally, we benchmark our NRG results at various temperatures against continuous-time Quantum Monte Carlo (CTQMC) [67] data obtained in the hybridization expansion (CTHYB) [51, 67, 81]. Citations refer to the list of references given in the main text.

SUSCEPTIBILITIES

We compute susceptibilities as retarded two-point correlation functions of bosonic operators A, B on the impurity, $\chi(t) = \langle A|B \rangle(t) = -i\Theta(t)\langle [A(t), B] \rangle$. Focusing on their spectral density, we have $\chi = \chi' - i\pi\chi''$ and $\chi''(\omega) = \frac{1}{2\pi} \int dt e^{i\omega t} \langle [A(t), B] \rangle$. Spin susceptibilities are computed via the spin operator in z direction,

$$S_m = \frac{1}{2} \sum_{\sigma\sigma'} d_{m\sigma}^\dagger \tau_{\sigma\sigma'}^z d_{m\sigma'},$$

using the Pauli matrix $\tau^z = \text{diag}(1, -1)$ and the creation operator $d_{m\sigma}^\dagger$ of an electron in orbital m with spin σ on the impurity. Orbital (or angular-momentum) susceptibilities are computed via

$$L_m = \frac{i}{\sqrt{2}} \sum_{\sigma m' m''} \epsilon_{mm'm''} d_{m'\sigma}^\dagger d_{m''\sigma}$$

with the Levi-Civita symbol $\epsilon_{mm'm''}$. The factor of $1/\sqrt{2}$ is chosen for convenience, such that χ''_{sp} and χ''_{orb} have roughly the same integral weight $\int_0^\infty \chi''(\omega) d\omega$. Total susceptibilities are obtained from $S_{\text{tot}} = \sum_m S_m$ and $L_{\text{tot}} = \sum_m L_m$. Finally, the orbital susceptibilities, $\langle L_m || L_m \rangle$, behave very similar to orbital-resolved charge susceptibilities [19], $\langle N_m || N_m \rangle$ with $N_m = \sum_\sigma d_{m\sigma}^\dagger d_{m\sigma}$.

HAMILTONIANS

To construct the non-interacting Hamiltonian, we use maximally localized Wannier functions [46, 47] for the three t_{2g} -like orbitals centered on the Ru atoms, employing the software packages WIEN2K [52] wien2wannier [53], wannier90 [54] and TRIQS/DFTTools [80, 82]; see [48] for further details. The resulting Wannier Hamiltonian, $h_{mm'}(\mathbf{k})$, is nondiagonal in orbital space. However, without spin-orbital coupling, which we neglect in this work, local single-particle quantities are orbital-diagonal due to the crystal symmetry of Sr₂RuO₄. This applies to the impurity energy levels, $\epsilon_d = \sum_{\mathbf{k}} \mathbf{h}(\mathbf{k})$ (momentum sum normalized), the local propagator

$$\mathbf{G}(\omega) = \sum_{\mathbf{k}} [\omega + i0^+ + \mu - \mathbf{h}(\mathbf{k}) - \Sigma(\omega)]^{-1},$$

and the hybridization function $\Delta(\omega)$.

The spectral density of the hybridization is evaluated as $\mathcal{A}_{\Delta,m}(\omega) = \frac{1}{\pi} \text{Im}[G_m^{-1}(\omega) + \Sigma_m(\omega)]$. This already indicates the inverse relation between \mathcal{A}_{Δ} and the spectral function $\mathcal{A} = -\frac{1}{\pi} \text{Im}G$, responsible for producing a dip in the hybridization from a van Hove peak in the spectrum. Indeed, if consider small frequencies ω , where $|\text{Im}\Sigma(\omega)|, |\text{Re}G(\omega)| \ll |\text{Im}G(\omega)|$, we directly get $\mathcal{A}_{\Delta}(\omega) \propto \mathcal{A}^{-1}(\omega)$.

The widely used, local, SO(3)-symmetric Kanamori interaction Hamiltonian [2], consisting of a density-density, spin-flip, and pair-hopping part, is given by

$$\begin{aligned} H_{\text{int}} &= H_{\text{dd}} + H_{\text{sf}} + H_{\text{ph}}, \\ H_{\text{dd}} &= U \sum_m n_{m\uparrow} n_{m\downarrow} + U' \sum_{m \neq m'} n_{m\uparrow} n_{m'\downarrow} \\ &\quad + (U' - J) \sum_{m < m', \sigma} n_{m\sigma} n_{m'\sigma}, \\ H_{\text{sf}} &= -J \sum_{m \neq m'} d_{m\uparrow}^\dagger d_{m\downarrow} d_{m'\downarrow}^\dagger d_{m'\uparrow}, \\ H_{\text{ph}} &= J \sum_{m \neq m'} d_{m\uparrow}^\dagger d_{m\downarrow}^\dagger d_{m'\downarrow} d_{m'\uparrow}, \end{aligned}$$

(a)	Z	$\tilde{\epsilon}$	ϵ	$\Sigma(0)$	(b)	$S=0$ (singlet)				$S=1$ (triplet)	
						xy-xy	xz-xz	xy-xz	xz-yz	xy-xz	xz-yz
xy	0.17	-0.076	-5.80	5.35	\tilde{U}	0.17	0.40	0.27	0.40	-0.085	-0.12
xz	0.26	-0.078	-5.72	5.42	Γ	6.1	5.9	6.1	5.9	-2.0	-1.7

TABLE I. Zero-temperature quasiparticle parameters deduced from the NRG low-energy spectrum. (a) Quasiparticle weight Z , quasiparticle energy level $\tilde{\epsilon}$, bare energy level ϵ , and the resulting self-energy at zero frequency, $\Sigma(0)$. (b) Quasiparticle interactions $\tilde{U}_{mm'}^S$ and the zero-energy vertex $\Gamma_{mm'}^S$, revealing an effective attraction in the spin-triplet sector. The two significant digits in all values give a rough estimate of the numerical accuracy.

with $U' = U - 2J$. The spin-flip term is crucial for the $SU(2)$ spin symmetry and Hund-metal physics. By contrast, we argue in the following that the pair-hopping term is almost inactive in the Hund-metal phase of Sr_2RuO_4 and can be neglected to obtain a model with higher symmetry that is more tractable for NRG (see below).

PAIR HOPPING

Considering the identical prefactor J of the spin-flip and pair-hopping term, it seems *a priori* hardly justified to neglect H_{ph} . However, it is readily understood that the effect of H_{ph} is a high-energy process, requiring states with one fully occupied and one completely empty orbital at the impurity site. At low energies, these contributions are suppressed; the dominant contributions instead have an impurity occupation of four electrons almost equally distributed among the three orbitals in the case of Sr_2RuO_4 .

Furthermore, we can *a posteriori* justify neglecting H_{ph} by evaluating the probability for an empty and doubly occupied orbital in the thermal state ρ . For this, we use the projectors

$$P_{m\uparrow\downarrow} = n_{m\uparrow}n_{m\downarrow}, \quad P_{m0} = (1 - n_{m\uparrow})(1 - n_{m\downarrow}),$$

to find that the probabilities

$$\text{prob}_{\text{ph}, m \rightarrow m'} = \text{Tr}[\rho P_{m\uparrow\downarrow} P_{m'0}]$$

are all on the level of a few percent. We also compared imaginary-time CTHYB results with and without pair hopping and found deviations of similar magnitude.

ALGORITHMIC DETAILS

Combining the quadratic part of the Hamiltonian, with $\Delta_{xy}(\omega) \neq \Delta_{xz}(\omega) = \Delta_{yz}(\omega)$, with the $SO(3)$ -symmetric interaction Hamiltonian, we have a **charge**, **orbital**, and **spin** symmetry of $U(1)_{\text{ch}} \otimes SO(2)_{\text{orb}} \otimes SU(2)_{\text{sp}}$. Computationally, the one-dimensional $SO(2)$ symmetry is rather weak. However, by neglecting the pair-hopping term, we obtain the larger symmetry $U(1)_{\text{ch},xy} \otimes U(1)_{\text{ch},xz} \otimes U(1)_{\text{ch},yz} \otimes SU(2)_{\text{sp}}$.

We employ the full density-matrix (fdm) NRG [55] and exploit these symmetries using the QSpace tensor library [78, 79]. For further efficiency, we interleave [56, 57]

the Wilson chains of all orbitals and thereby artificially break the symmetry between the xz and yz orbitals, but restore it by averaging results for these orbitals at each DMFT iteration. We use an NRG discretization parameter of $\Lambda = 6$ and keep up to 10^5 $SU(2)$ -spin multiplets (roughly $4 \cdot 10^5$ individual states) during the iterative diagonalization. Sufficient resolution at finite energies is obtained by averaging results over four shifted discretization grids [58] and by using an adaptive broadening scheme [59, 60].

In the illustration of the NRG flow in Fig. 1(b), xz and yz contributions are averaged as well. To understand the rescaling of the axes in Fig. 1(b), we recall that the iterative diagonalization with a successively increasing Wilson chain length N sets a characteristic energy splitting of $a\Lambda^{-N/2}$, with a of order unity [9]. The y -axis is thus rescaled $\propto \Lambda^{-N/2}$ to have converged energy levels with convenient values. Further, in fdm-NRG, temperature-dependent quantities are computed unambiguously by including all Wilson shells N (of characteristic energy scale $E_N = a\Lambda^{-N/2}$) with their respective, temperature-dependent weight w_N^T [78]. Typically, w_N^T is maximal close to $E_N \approx T$ [78]. For the x -axis of Fig. 1(b), we fix the prefactor a by actually requiring $(\sum_N w_N^T E_N) / (\sum_N w_N^T) = T$ and thus have a unique assignment of shell index to energy scale and temperature.

Finally, to obtain a smooth hybridization in the DMFT self-consistency iteration, performed entirely on the real-frequency axis, we use a momentum summation with a large number of $4 \cdot 10^6$ \mathbf{k} points in the irreducible Brillouin zone and manually set $-\text{Im}\Sigma_{xy} \geq 0.005$ and $-\text{Im}\Sigma_{xz/yz} \geq 0.01$.

QUASIPARTICLE PARAMETERS

In the main text, we explained that the quasiparticle weight Z and energy level $\tilde{\epsilon}$ can be extracted from the low-energy spectrum. The results are reported in Tab. I(a). The values for Z agree quantitatively with those obtained from the dynamic self-energy via $Z = [1 - \partial_\omega \text{Re}\Sigma(0)]^{-1}$; the same is true when comparing $\tilde{\epsilon} = Z \cdot (\epsilon + \Sigma(0))$ to the zero-frequency value of the dynamic self-energy.

The results for the quasiparticle interaction $\tilde{U}_{mm'}^S$ and the zero-energy vertex $\Gamma_{mm'}^S$ are listed in Tab. I(b). For their computation, we employed the quasiparticle density at the impurity, $|\psi_m(0)|^2$. It is conceptually related to

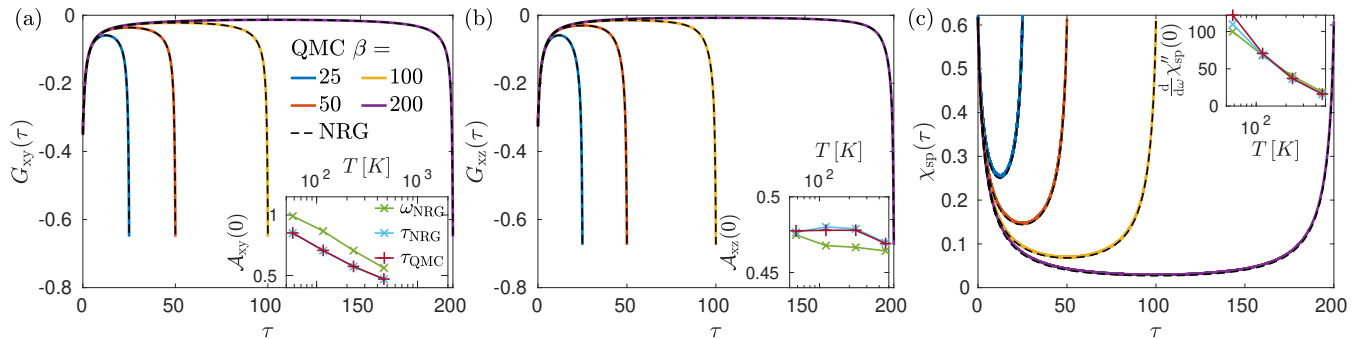


FIG. 5. Benchmark of NRG results against imaginary-time CTHYB data at various temperatures. (a) Single-particle Green's function in the xy orbital. Inset: Spectral weight at zero frequency, $\mathcal{A}(0)$, computed from the NRG real-frequency result (label ω_{NRG}) and the imaginary-time proxy, $-\frac{\beta}{\pi}G(\tau = \beta/2)$, both in NRG and CTHYB (labels τ_{NRG} and τ_{QMC}). (b) Analogous plot for the xz orbital. (c) Total spin susceptibility. Inset: Zero-frequency slope of its spectral density, χ_{sp}'' , computed from the NRG real-frequency result and the imaginary-time proxy, $-(\frac{\beta}{\pi})^2\chi(\tau = \beta/2)$.

the excited state $|E_m\rangle$ and groundstate $|\mathcal{G}\rangle$ of the renormalized impurity model according to $\psi_m(0) = \langle E_m | d_\sigma^\dagger | \mathcal{G} \rangle$, and practically evaluated as

$$|\psi_m(0)|^2 = \text{Res}_{\omega=E_m} G_m(\omega) = \frac{1}{1 - \partial_\omega \Delta(\omega)|_{\omega=E_m}}.$$

BENCHMARKING NRG AGAINST QMC

We benchmark our NRG results against CTHYB imaginary-time data at various temperatures. All results are computed without pair hopping and are converged on their respective DMFT self-consistency cycle. We find very good agreement for the single-particle Green's function G_{xy} and G_{xz} , see Fig. 5(a) and (b). The (total) spin susceptibilities show satisfactory agreement as well, with slightly higher deviations, see Fig. 5(c).

In the insets of Fig. 5, we show the real-frequency

quantities $\mathcal{A}(0)$ and $\frac{d}{d\omega}\chi''|_{\omega=0}$ and their imaginary-time proxies, given by the l.h.s. of the relations

$$-\frac{\beta}{\pi}G(\tau = \beta/2) = \mathcal{A}(0) + \frac{\pi^2}{2\beta^2} \frac{d^2}{d^2\omega} \mathcal{A}(0) + O(\beta^{-4}),$$

$$\left(\frac{\beta}{\pi}\right)^2 \chi(\tau = \beta/2) = \frac{d}{d\omega} \chi''(0) + \frac{\pi^2}{3\beta^2} \frac{d^3}{d^3\omega} \chi''(0) + O(\beta^{-4}).$$

By fitting the corresponding polynomials to the real-frequency curves, we find that the β^{-2} corrections on the r.h.s. amount to roughly 10%, 5%, and 30% for G_{xy} , G_{xz} , χ_{sp} , respectively. This is consistent with the notable deviations between the real-frequency values and their imaginary-time proxies in the insets of Fig. 5. The CTHYB results have been obtained using the TRIQS/CTHYB solver [81] and the TRIQS/TPRF package [61], which are based on the TRIQS library [80].