The coherence-incoherence crossover and the mass-renormalization puzzles in Sr$_2$RuO$_4$

Jernej Mravlje,1,2 Markus Aichhorn,3,1 Takashi Miyake,4,5
Kristian Haule,6 Gabriel Kotliar,6 and Antoine Georges1,7,5

1Centre de Physique Théorique, École Polytechnique, CNRS, 91128 Palaiseau Cedex, France
2Jožef Stefan Institute, Jamova 39, Ljubljana, Slovenia
3Institute of Theoretical and Computational Physics, TU Graz, Petersgasse 16, Graz, Austria
4Nanostystem Research Institute, AIST, Tsukuba 305-8568, Japan
5Japan Science and Technology Agency, CREST, Kawaguchi 332-0012, Japan
6Physics Department and Center for Materials Theory, Rutgers University, Piscataway NJ 08854, USA
7Collège de France, 11 place Marcelin Berthelot, 75005 Paris, France

We calculate the electronic structure of Sr$_2$RuO$_4$, treating correlations in the framework of dynamical mean-field theory. The approach successfully reproduces and explains the key properties of this material: the anisotropic mass renormalization of quasiparticles and the crossover into an incoherent regime at a low temperature. The results agree well with quantum oscillations, NMR and photoemission measurements. While the orbital differentiation originates from the proximity of the van Hove singularity, strong correlations are caused by the Hund’s coupling. The generality of this mechanism for a wide class of correlated materials is pointed out.

Fermi-liquid theory describes the low-energy excitations of metals in terms of quasiparticles, which carry the quantum numbers of a bare electron but have a renormalized mass $m^*$. Quasiparticles have infinite lifetime on the Fermi surface and at temperature $T = 0$, but otherwise acquire a finite lifetime $\hbar/\Gamma$. They carry only a fraction $Z$ of the total spectral weight associated with all single-particle excitations, as encoded in the spectral function $A(k, \omega)$. A hallmark of strong correlations is that some of these interaction-induced renormalizations ($m^*, Z^{-1}, \Gamma$) become large.

The concept of a quasiparticle is meaningful only as long as its inverse lifetime is smaller than the typical excitation or thermal energy $\hbar \Gamma \lesssim \hbar \omega, kT$. The internal consistency of Fermi-liquid theory is insured by the fact that $\hbar \Gamma \sim \langle kT \rangle^2 / E_F^* \sim \langle \hbar \omega \rangle^2 / E_F^*$, due to phase-space constraints. For temperatures larger than a coherence scale $T^* (\sim E_F^*/k)$, quasiparticles become short-lived and the Landau Fermi-liquid description no longer applies. Due to strong correlations, $T^*$ can be much lower than the bare electronic scale $E_F/k$. The description of the incoherent regime $T > T^*$ and of the associated crossover is a major challenge which requires new concepts and techniques.

Of all transition metal oxides, the layered perovskite Sr$_2$RuO$_4$ is undoubtedly the one in which the Fermi liquid regime has been most studied 1. Resistivities obey accurately a $T^2$ law for $T \lesssim 30\,\text{K}$ 2, despite the large anisotropy $\rho_{ab}/\rho_{cd} \sim 10^3$. Sr$_2$RuO$_4$ is also an ideal material to investigate the crossover into the incoherent regime. Indeed, at 130K, $\rho_{cd}(T)$ reaches a maximum and decreases as temperature is further increased, while the $T$-dependence of $\rho_{ab}$ remains metallic. ARPES studies indicate that quasiparticle peaks at the Fermi surface disappear (by broadening and loosing spectral weight) at a temperature close to that where $\rho_{cd}$ reaches its maximum 3. 4.

The 3-sheet Fermi surface of this material has been accurately determined by quantum oscillation experiments 1 and is reasonably well described by electronic structure calculations in the local density approximation (LDA) 5. On the other hand, the measured masses are not reproduced by the LDA. Three bands of mainly $t_{2g}$ character cross the Fermi surface. The broadest (3.5 eV) band of $xy$ character gives rise to a two-dimensional Fermi surface sheet $\gamma$. The degenerate $xz$ and $yz$ orbitals give rise to narrower (1.5 eV) bands with quasi one-dimensional Fermi surface sheets $\alpha$ and $\beta$. Experimentally, large and anisotropic mass enhancements $m^*/m_{\text{LDA}}$ are found, namely (3, 3.5, 5.5) for sheets $\alpha$, $\beta$, $\gamma$, respectively 1.

These experimental findings raise several puzzles, unresolved to this day. The large effective masses and the low coherence scale indicate that Sr$_2$RuO$_4$ is a strongly correlated material. Surprisingly, as stressed recently by Konik and Rice 6, the largest mass enhancement is actually observed for the widest ($xy$) band. Furthermore, Ru being a $4d$ element, the screened on-site Coulomb interaction is not expected to be large ($U \lesssim 3\text{ eV}$, somewhat smaller than the bandwidth). In a nutshell, these puzzles can be loosely summarized by the question: why is Sr$_2$RuO$_4$ strongly correlated?

In this letter, we answer these questions in terms of the electronic structure of the material and of a first-principle determination of the Coulomb interaction parameters. Treating correlation effects within dynamical mean-field theory (DMFT), we achieve quantitative agreement with experiments. At a qualitative level, our explanation relies on the Hund’s coupling $J$ and the proximity of the van Hove singularity for the $xy$ band. These key elements of our picture, especially the Hund’s coupling, have general relevance to $4d$ transition-metal oxides, as well as to other materials in which strong correlation effects are observed but are not due to a strong Hubbard $U$ or the...
proximity to a Mott insulator.

The calculations use the full potential implementation of LDA+DMFT as presented in Ref. [1]. The framework of Ref. [8] gives very similar results. Wannier-like $t_{2g}$ orbitals are constructed out of Kohn-Sham bands within the energy window $[-3, 1]$ eV with respect to the Fermi energy. We use the full rotationally invariant interaction appropriate for a correct description of atomic multiplets:

$$H_1 = U \sum_m n_m^{\uparrow} n_m^{\downarrow} + \sum_{m < n, \sigma} [U' n_m^{\sigma} n_n^{\bar{\sigma}} + (U' - J)n_m^{\sigma} n_n^{\bar{\sigma}} - Jc_m^{\dagger \sigma} c_n^{\dagger \bar{\sigma}} c_n^{\sigma} c_m^{\bar{\sigma}}]$$

where $J$ is the Hund’s coupling constant, $U' = U - 2J$ and $m, n$ run over $t_{2g}$ orbitals. Ru $e_g$ and O $p$ orbitals are not explicitly included. The importance of correlations leading to charge transfer among the orbitals, mass renormalizations and satellites was recognized in earlier studies [1]. Here we use the strong-coupling continuous-time Monte-Carlo impurity solver [10] in order to reach the low-temperature regime where the coherence-incoherence crossover takes place [11].

We calculated the interaction parameter $U$ from first-principles using constrained-RPA [12]. The interaction matrix is found to be quite isotropic with $U = 2.5$ eV for $xy$ and $U = 2.2$ eV for $xz$ orbital. The stronger mass enhancement of the $xy$ orbital can thus not be explained by an anisotropy of the interactions [6].

We now turn to results. In table I we report the mass enhancements of each orbital, given within DMFT by: $m^*/m_{LDA} = Z^{-1}_{\text{coherent}}$ with $Z^{-1} = 1 - \partial \text{Im} \Sigma(i\omega)/\partial \omega |_{\omega = 0}$. In order to reveal the key role of the Hund’s coupling, we report results for several values of $J$. The derivative is extracted by fitting a fourth-order polynomial to the data for the lowest six Matsubara frequencies. The result converges below 50K. The calculated mass enhancements for $U = 2.3$ eV, $J = 0.4$ eV (used in the remainder of the paper [13]) are found to be close to the experiment [1].

Table I demonstrates that the Hund’s coupling is essential to yield the $xy - xz$ differentiation ($m^*_{xy} > m^*_{xz}$), and correct magnitude of mass enhancements at sensible values of the Coulomb interaction $U$. A comparable mass enhancement (but without $xy - xz$ differentiation) occurs at $J = 0$ only for the unphysically large $U = 5$ eV. In addition, we find that the Hund’s coupling by favoring maximal angular momentum brings the populations of the different orbitals closer to each other (1.29 and 1.36, for $xy$ and $xz$, respectively) in comparison to the LDA value (1.23, 1.39), hence improving the agreement with quantum oscillation experiments ($\sim 1.33, 1.33$).

To understand the coherence-incoherence crossover, we look at the inverse quasiparticle lifetime, presented in Fig. 1 as $\Gamma/kT$ vs. $T$, with $\Gamma = -Z \text{Im} \Sigma(0^+)$.
momentum resolution in experiment.

The crossover scale \( kT_c \) manifests itself also in the dependence of the self-energy on frequency, displayed in the rightmost panels of Fig. 2. We observe that deviations from the low-frequency Fermi liquid regime \( \text{Re}\Sigma(\omega) - \Sigma(0) + \omega(1 - 1/2Z) \), \( \text{Im}\Sigma(\omega) \sim \omega^2 + (\pi T)^2 \) appear at an energy scale of order 40 meV~\( kT_c \), at which a ‘kink’ [10] is observed in \( \text{Re}\Sigma(\omega) \). Such a feature at that energy scale is indeed reported in ARPES [15] and compared to ARPES [14]. (b,c) Spectral lineshapes at wavevectors \( \mathbf{k}_1, \mathbf{k}_2 \) compared to ARPES [3]. Also indicated (cross, dotted line) are the low-\( \omega \) behavior from a polynomial fit.

In Fig. 2(b) we display \( \lim_{\omega \to 0} \sum_q \text{Im}\chi(q,\omega)/\omega \) and compare to the NMR data for 1/\( T/T_c \) (for hyperfine couplings we used values from Refs. [21, 22]). There, the data saturate only well below \( T^* \), illustrating that the Fermi liquid behavior in two-particle properties is more fragile than in single-particle ones. Indeed, in the well known Kondo problem the Kondo resonance persists at temperatures up to 2\( T_K \) while the magnetic susceptibility saturates to a Pauli form only below 1\( T_K/5 \).

Having demonstrated that the LDA+DMFT results agree with experimental data, we extract qualitative theoretical insights from this method. In DMFT the local physics is revealed by solving an impurity problem with local interactions given by Eq. (1). This can be rewritten as \( H_I = (U - 3J)\mathbf{n}(n - 1)/2 - 2JS^2 - (J/2)T^2 \), where \( S \) is the total spin and \( T \) is the total angular momentum at the atom [23]. The four-electrons subspace separates into five \( T = 2, S = 0 \) states, a single \( T = 0, S = 0 \) state and nine \( T = 1, S = 1 \) states. At \( J = 0 \) all these states are degenerate and constitute a 15-dimensional representation of an \( SU(6) \) symmetry group. This high degeneracy results in a very high coherence scale \( \sim 0.5eV \) and small mass renormalizations (see Table 4). The Hund’s coupling \( J \) lowers the \( SU(6) \) symmetry down to \( SU(2)_{\text{spin}} \times SU(2)_{\text{orbit}} \) with the 9-fold degenerate atomic multiplet \( S = 1, T = 1 \) having lowest energy. The ground state of the impurity model is non-degenerate with \( S = 0, T = 0 \) corresponding to exact screening of this atomic multiplet [22]. Thus Sr\(_2\)RuO\(_4\) is a Fermi liquid at low temperatures. The Hund’s coupling projects the spin degrees of freedom to a low energy
manifold characterized by a reduced Kondo coupling, resulting in a suppressed Kondo scale \cite{24,25}. The effective low energy model is in our case a $S = 1$ Kondo model. Indeed, the inset of Fig. 3(a) demonstrates that at low $T$ the LDA+DMFT result for $\chi_{\text{loc}}$ is fit well by the $S = 1$ Kondo model Bethe ansatz curve \cite{26}.

The dramatic reduction of coherence scale as a result of the Hund’s coupling has been noted before in impurity models \cite{24,25,27}, DMFT studies of model Hamiltonians \cite{28} and for iron pnictides \cite{29}. Such reduction can be expected whenever multiplet correlations persist while the on-site $U$ is strongly screened (due to the large spatial extension of the correlated orbital as in $4d$ transition metal oxides, or the large polarizability of screening orbitals in pnictides).

The origin of larger $xy$ effective mass can be traced to the proximity of the van-Hove singularity and associated peak in the density of states for that orbital. This implies a smaller dispersion of the $xy$ band for states near the Fermi level, and in turn reflects in a lower value of the low-frequency hybridization function for that orbital (Fig. 3). Indeed, ignoring the self-consistency (i.e. on the first DMFT iteration), the low-frequency hybridization is given by: $\text{Im}\Delta (i\omega) = -\pi \rho_F/\left[\text{Re}G_{\text{loc}}(i\omega)^2 + (\pi \rho_F)^2\right] \approx -1/(\pi \rho_F)$ with $\rho_F$ the LDA density of states at the Fermi level. The large value of $\rho_F$ thus corresponds to a suppressed low-energy effective hopping \cite{30}. In contrast, the full bandwidth is larger for the $xy$, and so is the LDA kinetic energy ($0.27eV$ for $xy$, 0.20eV for $xz$). This reflects in high-frequency behavior of the hybridization, indeed larger for $xy$ at high-frequency. We thus see that the degree of correlation cannot be guessed from the kinetic energy or bandwidth of each band, which would naively suggest a smaller mass for $xy$, in contrast to observations.

In summary, we have demonstrated that several experimental results for Sr$_2$RuO$_4$ are well reproduced by the LDA+DMFT method. We have shown that the suppression of the coherence scale is due to the Hund’s coupling, and pointed out the generality of this mechanism. We have also shown that the orbital differentiation and larger $xy$ mass is due to the difference in low-energy hybridization properties of each orbital, caused by their orientation-dependent bonding properties in this anisotropic material. This is expected to be relevant to other layered perovskites, most notably to the metal-insulator transition in Ca$_2$Zr$_2$RuO$_4$.

We are grateful to P. Bourges, M. Fabrizio, M. Ferrero, E. Guill, L. de Leo, A. Mackenzie, Y. Sidis and M. Sigrist for useful discussions. We acknowledge the support of the NSF-materials world network (NSF DMR 0806937), the Partner University Fund, the CNRS-LIA program, the Austrian Science Fund (project J2760) and the hospital-}

[11] We use $>10^7$ Monte Carlo steps per iteration and, at low temperatures, up to $10^9$ steps for the thermalization.
[13] Extracting $J$ from the reduced $J_{\text{mat}}$ matrix calculated by constrained RPA yielded $J = 0.25eV$, although a somewhat larger value is expected to be obtained when considering the full $U_{m1m2m3m4}$ matrix.
[20] The plotted NMR estimates (dots) are thus multiplied by 1.3, to ease the comparison of the temperature dependence.


A single band calculation with the orbitally projected density of states at filling $4/3$ gives $m^*/m \sim 2.2(1.8)$ and $T^* = 0.05(0.1)eV$ for xy (xz). See also R. Žitko, J. Bonča, and T. Pruschke, Phys. Rev. B 80, 245112 (2009); S. Schmitt, ibid. 82, 155126 (2010).