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

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We calculate the electronic structure of Sr$_2$RuO$_4$, treating correlations in the framework of dynamical mean-field theory. The approach successfully reproduces several experimental results 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. 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 other 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 rests on $\hbar \Gamma \sim (kT)^2/E_F \sim (\hbar \omega)^2/E_F$, 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 $T \lesssim 30$K [2], despite the large anisotropy $\rho_{ab}/\rho_{ab} \sim 10^3$. Sr$_2$RuO$_4$ is also an ideal material to investigate the crossover into the incoherent regime. Indeed, at 130K, $\rho_c(T)$ 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 disappear (by broadening and loosing spectral weight) at a temperature close to that where $\rho_c$ 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) [2]. 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 $m^*/m_{LDA}$ 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 [2], the largest mass enhancement is actually observed for the widest ($xy$) band. Furthermore, Ru being a 4$d$ element, the screened on-site repulsion is not expected to be large ($U \lesssim 3$ 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. 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 4$d$ 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. [4]. 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
The interaction matrix is found to be quite isotropic for $U = 2.3$ eV. Other columns: coherence temperatures as defined in the text.

Table I. Mass enhancement of the $xy$ and $xz$ orbitals, as a function of Hund’s coupling, for $U = 2.3$ eV. Other columns: coherence temperatures as defined in the text.

| $J$ [eV] | $m^*/m_{\text{LDA}}_{xy}$ | $m^*/m_{\text{LDA}}_{xz}$ | $T_{\text{xy}}^*$ [K] | $T_{\text{xz}}^*$ [K] | $T_\gamma$ [K] |
|----------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 0.0, 0.1 | 1.7             | 1.7             | >1000           | >1000           | >1000           |
| 0.2      | 2.3             | 2.0             | 300             | 800             | >1000           |
| 0.3      | 3.2             | 2.4             | 100             | 300             | 500             |
| 0.4      | 4.5             | 3.3             | 60              | 150             | 350             |

Figure 1. Temperature-dependence of $\Gamma/kT$, with $\hbar/\Gamma$ the quasiparticle lifetime. The shading indicates the ‘coherent’ regime with long-lived quasiparticles such that $\Gamma \lesssim kT$.

energy. We use the full rotationally invariant interaction appropriate for a correct description of atomic multiplets:

$$H_I = U \sum_n n_{m\uparrow} n_{m\downarrow} + \sum_{m<n,\sigma} [U' n_{m\sigma} n_{n\bar{\sigma}} + (U' - J) n_{m\sigma} n_{n\bar{\sigma}} - J c_{m\sigma}^\dagger c_{m\bar{\sigma}} c_{n\bar{\sigma}} c_{n\sigma}] - J \sum_{m \bar{c}} [c_{m\bar{c}}^\dagger c_{m\bar{c}}^\dagger c_{n\bar{c}} c_{n\bar{c}} + h.c.]$$

(1)

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 [8]. 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 $xz$ orbitals. 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: $m^*/m_{\text{LDA}} = Z^{-1}\big|_{T \to 0}$ with $Z^{-1} = 1 - \partial \text{Im} \Sigma(i \omega_n) / \partial \omega_n |_{\omega \to 0}$. The derivative is extracted by fitting a fourth-order polynomial to the data for the lowest six Matsubara frequencies. 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 reproduce the observed magnitude of mass enhancements and the $xy - xz$ differentiation. 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, by favoring maximal angular momentum, the Hund’s coupling drives the populations of orbitals closer to one another (to 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 oscillations 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 $\Gamma = -\text{Im} \Sigma(0^+)$. At very low temperatures the Fermi-liquid $\Gamma \propto T^2$ behavior is indicated (dashed). We define the coherence scale $T^*$ by $\Gamma(T^*)/kT^* = 1$, but the deviations from $T^2$-law are visible already at lower temperatures. $T^*$ is reported in Table I and also indicated on Fig. [1]. We see that $T^*$ is as low as 60 K for the most correlated $xy$ orbital. At high temperatures $T \gtrsim T_s \sim 400$ K, $\Gamma/kT$ saturates, signaling the ‘incoherent’ regime characterized by a quasilinear temperature dependence $\Gamma \propto kT$. An intermediate crossover region where $\Gamma/kT$ gradually increases connects these two regimes.

How do these regimes reveal themselves when probed by spectroscopic experiments? The left-most panel of Fig. [2] displays an intensity map of the momentum-resolved spectral function demonstrating that our results compare well with ARPES [14]. Panels (b) and (c) display the energy-distribution curves at two specific momenta. In the ‘coherent’ regime, these spectra display sharp peaks corresponding to the Fermi surface crossings. Upon increasing temperature the quasiparticle peaks broaden and above $T_s$, cannot be discerned anymore. Note that in ARPES [3] the peaks disappear already at a somewhat lower temperature, possibly due to the finite momentum resolution in experiment.

The crossover scale $kT_s$ 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 $\text{Re} \Sigma \sim \Sigma(0) + \omega (1 - 1/Z)$, $\text{Im} \Sigma \sim \omega^0 + (\pi T)^2$ appear at an energy scale of order 40 meV $< kT_s$, at which a ‘kink’ [16] is observed in $\text{Re} \Sigma(\omega)$. Such a feature at that energy scale is indeed reported in ARPES (Fig. [2] 15, 17).

The crossover also affects the magnetic response. On Fig. [3(a)] we display the orbitally resolved uniform magnetic susceptibilities and compare them to the NMR Knight shift measurements [18]. Saturation to a Pauli magnetic susceptibility is observed only below $T^*$ (shaded). The stronger temperature depen-
The origin of the larger $xy$ effective mass can be explained by considering the spin-orbit coupling in the system. The spin-orbit coupling mixes the $xy$ and $xz$ orbitals, leading to a splitting of the degenerate states. This mixing is enhanced near the Kondo scale, where the spin and orbital degrees of freedom are strongly coupled. As a result, the effective mass of the $xy$ orbital is increased, while that of the $xz$ orbital remains constant. This agrees with experimental data, where the effective mass is found to be larger for the $xy$ orbital compared to the $xz$ orbital.

The dramatic reduction of coherence scale as a result of the Hund's coupling can also be understood in terms of DMFT. The effective low energy model is a Fermi liquid with a reduced number of degrees of freedom compared to the impurity model. The effective low energy manifold is characterized by a smaller number of states, leading to a lower coherence scale. This is consistent with the experimental observations, where the coherence scale is found to be smaller in the iron pnictides compared to the transition metal oxides.
traced to the proximity of the van-Hove singularity. Higher density of states near the Fermi level implies weaker dispersion and in turn reflects in a lower value of the respective hybridization function $\Delta(\omega)$ at low frequencies (Fig. 1). Indeed, ignoring the self-consistency (i.e. on the first DMFT iteration), $\text{Im}\Delta^{(1)}(i\omega) = -\pi\rho_F/|\text{Re}G_{\text{loc}}(i\omega)|^2 + (\pi\rho_F)^2 \approx -1/(\pi\rho_F)$ 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 [31]. 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 the high-frequency behavior of the hybridization, indeed larger for $xy$ at high-frequency. Note 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.

![Figure 4. The hybridization functions $\Delta(\omega)$ at the initial DMFT step and at self consistency. (Inset) The LDA projected density of states.](image)

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$_2$Ru$_2$O$_4$.

We are grateful to P. Bourges, M. Fabrizio, M. Ferrero, E. Gull, 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 CNERS-LIA program, the Austrian Science Fund (projects J2760, F4103, P18551) and the hospitality of KITP, Santa Barbara (NSF PHY05-51164) and of CPHT (G.K.).

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