

## Coherence-Incoherence Crossover and the Mass-Renormalization Puzzles in $\text{Sr}_2\text{RuO}_4$

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(Received 27 October 2010; published 2 March 2011)

We calculate the electronic structure of  $\text{Sr}_2\text{RuO}_4$ , treating correlations within 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 above a low temperature scale. 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.

DOI: 10.1103/PhysRevLett.106.096401

PACS numbers: 71.27.+a, 72.15.Qm, 74.20.Pq

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 an 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  $A(\mathbf{k}, \omega)$ . 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  $\hbar\Gamma \sim (kT)^2/E_F^*$ ,  $E_F^* \sim (\hbar\omega)^2/E_F^*$ , 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. Because of 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  $\text{Sr}_2\text{RuO}_4$  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  $\rho_c/\rho_{ab} \sim 10^3$ .  $\text{Sr}_2\text{RuO}_4$  is also an ideal material to investigate the crossover into the incoherent regime. Indeed, at 130 K,  $\rho_c(T)$  reaches a maximum and decreases as temperature is further increased, while the  $T$  dependence of  $\rho_{ab}$  remains metallic. Angle-resolved photoemission spectroscopy (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 three-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  $\text{Sr}_2\text{RuO}_4$  is a strongly correlated material. Surprisingly [6], the largest mass enhancement is actually observed for the widest ( $xy$ ) band. Furthermore, Ru being a  $4d$  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  $\text{Sr}_2\text{RuO}_4$  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  $4d$  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. [7]. 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:

$$H_I = U \sum_m n_{m\uparrow} n_{m\downarrow} + \sum_{m < n, \sigma} [U' n_{m\sigma} n_{n\bar{\sigma}} + (U' - J) n_{m\sigma} n_{n\sigma} - J c_{m\sigma}^\dagger c_{m\bar{\sigma}} c_{n\bar{\sigma}}^\dagger c_{n\sigma}] - J \sum_{m < n} [c_{m\uparrow}^\dagger c_{m\downarrow}^\dagger c_{n\uparrow} c_{n\downarrow} + \text{H.c.}] \quad (1)$$

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 [9]. 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  $m^*/m_{\text{LDA}} = Z^{-1}|_{T \rightarrow 0}$  with  $Z^{-1} = 1 - \partial \text{Im}\Sigma(i\omega)/\partial \omega|_{\omega \rightarrow 0^+}$ . 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 Letter [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

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]	$m^*/m_{\text{LDA}} _{xy}$	$m^*/m_{\text{LDA}} _{xz}$	$T_{xy}^*$ [K]	$T_{xz}^*$ [K]	$T_>$ [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

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 display in Fig. 1 the inverse quasiparticle lifetime, plotted as  $\Gamma/kT$  vs  $T$ , with  $\Gamma = -Z \text{Im}\Sigma(i0^+)$ . 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  $T \geq T_> \sim 400$  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 leftmost 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  $T_>$  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  $kT_>$  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  $\text{Re}\Sigma \sim \Sigma(0) + \omega(1 - 1/Z)$ ,  $\text{Im}\Sigma \sim \omega^2 + (\pi T)^2$  appear at an energy scale of order 40 meV  $\sim kT_>$ , at which a ‘‘kink’’ [15] is observed in  $\text{Re}\Sigma(\omega)$ . Such a feature at that energy scale is indeed reported in ARPES (Fig. 2) [16,17].

The crossover also affects the magnetic response. On Fig. 3(a) we display the orbitally resolved uniform

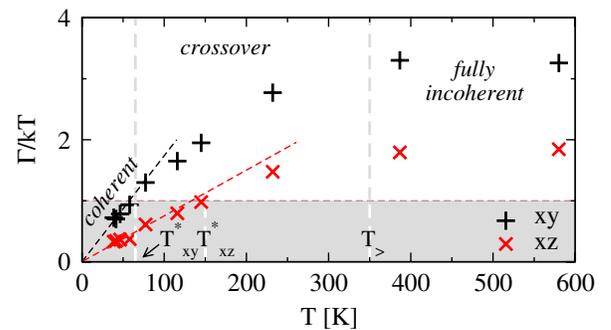


FIG. 1 (color online). Temperature dependence of  $\Gamma/kT$ , with  $\hbar/\Gamma$  the quasiparticle lifetime. The shading indicates the ‘‘coherent’’ regime with long-lived quasiparticles such that  $\Gamma \lesssim kT$ .

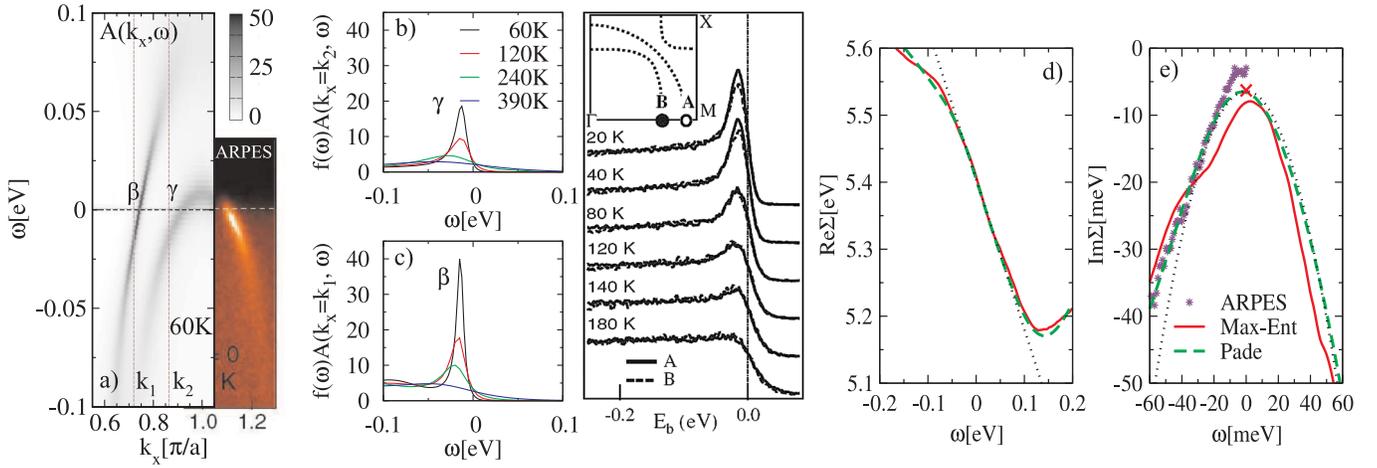


FIG. 2 (color online). (a) Intensity map of the spectral function  $A(k, \omega)$  along  $\Gamma \rightarrow M$  for  $0.55\pi/a \leq k_x \leq 1.05\pi/a$ ,  $k_y = 0$  at  $T = 60$  K compared to ARPES [14]. (b),(c) Spectral line shapes at wave vectors  $k_1, k_2$  compared to ARPES [3]. (d),(e)  $\text{Im}\Sigma(\omega + i0^+)$  and  $\text{Re}\Sigma(\omega + i0^+)$  for  $xz$  orbital at  $T = 60$  K obtained by stochastic maximum entropy (plain line) and Pade approximants (dashed) compared to ARPES [16]. Also indicated (cross, dotted line) are the low- $\omega$  behavior from a polynomial fit.

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 dependence of the  $xy$  orbital related to  $T_{xy}^* < T_{xz}^*$  is reproduced well. The total low-temperature uniform susceptibility (1.2 emu/mol) is within the estimated error ( $\sim 30\%$  [18]) of the thermodynamic measurements (0.9 emu/mol) [19] [20]. We also calculated the local susceptibility (inset) and found that it is larger than the uniform one, especially for the  $xz$  orbital [21]. This signals antiferromagnetic correlations, in agreement with experimental observations [22].

In Fig. 3(b) we display  $\lim_{\omega \rightarrow 0} \sum_q \text{Im}\chi(q, \omega)/\omega$  and compare to the NMR data for  $1/T_1 T$  (we used the values of hyperfine couplings from Refs. [22,23]). 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  $2T_K$  while the magnetic susceptibility saturates to a Pauli form only below  $T_K/5$ .

Having demonstrated that the LDA + DMFT results agree with experimental data, we turn to theoretical insights. In DMFT the local physics is revealed by solving an impurity model (atom + bath) with atomic interactions given by Eq. (1). This can be rewritten as  $H_I = (U - 3J)n(n-1)/2 - 2JS^2 - (J/2)T^2$ , where  $S$  is the total spin and  $T$  is the total angular momentum [24]. 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 form a 15-dimensional representation of an  $SU(6)$  symmetry group. This high degeneracy results in a very high coherence scale  $\sim 0.5$  eV and small mass renormalizations (see Table I). The Hund's coupling  $J$  lowers the  $SU(6)$  symmetry down

to  $SU(2)_{\text{spin}} \times SU(2)_{\text{orbit}}$  with the ninefold degenerate atomic multiplet  $S = 1, T = 1$  having lowest energy. The ground state of the impurity model is nondegenerate with  $S = 0, T = 0$  corresponding to exact screening of this atomic multiplet [24]. Thus  $\text{Sr}_2\text{RuO}_4$  is a Fermi liquid. The Hund's coupling projects the spin degrees of freedom onto a low-energy manifold characterized by a reduced Kondo coupling, resulting in a suppressed Kondo scale [25,26]. 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 [27]. The dramatic reduction of coherence scale as a result of the Hund's coupling has been noted before in impurity models [25,26], DMFT studies of model Hamiltonians [28,29], and for iron pnictides [30]. It occurs whenever multiplet correlations persist while the on-site  $U$  is strongly screened (due

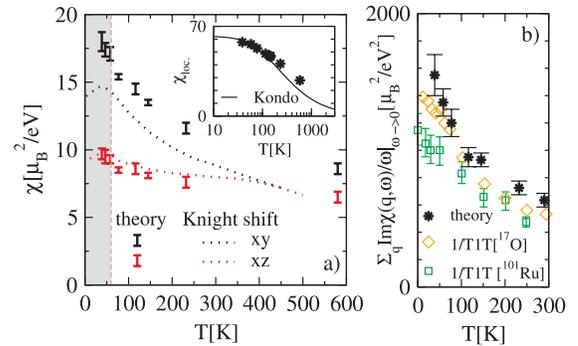


FIG. 3 (color online). (a) The uniform susceptibility  $\chi_m(q = 0)$  for each orbital,  $m = xy, xz$ . (Inset) Total local susceptibility  $\chi_{\text{loc}} = \sum_{qm} \chi_m(q)$  compared to that of the  $S = 1$  Kondo model. (b)  $\lim_{\omega \rightarrow 0} \sum_q \text{Im}\chi(q, \omega)/\omega$  compared to NMR [18].

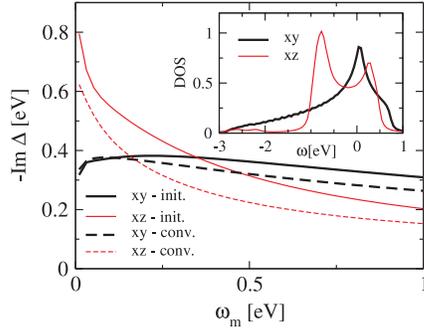


FIG. 4 (color online). The hybridization functions  $\Delta(i\omega)$  at the initial DMFT step and at self-consistency. (Inset) The LDA projected density of states.

to the large spatial extension of the correlated orbital as in  $4d$  transition-metal oxides, or the large polarizability of screening orbitals as in pnictides).

The origin of the larger  $xy$  effective mass can be 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  $\Delta(i\omega)$  at low frequencies (Fig. 4). Indeed, ignoring the self-consistency (i.e., on the first DMFT iteration),  $\text{Im}\Delta^{(1)}(i0^+) = -\pi\rho_F/[\text{Re}G_{\text{loc}}(i0^+)^2 + (\pi\rho_F)^2] \simeq -1/(\pi\rho_F)$  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 (0.27 eV for  $xy$ , 0.20 eV 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.

In summary, we have demonstrated that several experimental results for  $\text{Sr}_2\text{RuO}_4$  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  $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$ .

We are grateful to P. Bourges, M. Fabrizio, M. Ferrero, E. Gull, L. de Leo, A. Mackenzie, Y. Sidis, and M. Sgrist 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 (Projects No. J2760, No. F4103, No. P18551) and the hospitality of KITP, Santa Barbara (NSF PHY05-51164) and of CPHT (G. K.).

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