# Absence of orbital-selective Mott transition in $Ca_{2-x}Sr_xRuO_4$

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Quasi-particle spectra of the layer perovskite $Sr_2RuO_4$ are calculated within Dynamical Mean Field Theory for increasing values of the on-site Coulomb energy $U$. At small $U$ the planar geometry splits the $t_{2g}$ bands near $E_F$ into a wide, two-dimensional $d_{xy}$ band and two narrow, nearly one-dimensional $d_{xz,yz}$ bands. At larger $U$, however, the spectral distribution of these states exhibit similar correlation features, suggesting a common metal-insulator transition for all $t_{2g}$ bands at the same critical $U$.

$Sr_2RuO_4$ has attracted considerable interest during the recent years because of a variety of fascinating properties. It exhibits unconventional p-wave superconductivity [1, 2] and is in fact the only known layered perovskite without copper that becomes superconducting in the absence of doping. The ground state is non-magnetic whereas the volume perovskite $SrRuO_3$ is a strong ferromagnet. Substitution of Sr with Ca makes $Sr_2RuO_4$ undergo a transition to an antiferromagnetic Mott insulator [3, 4]. In contrast, doping with La leads to a ferromagnetic instability accompanied by a breakdown of Fermi-liquid behavior [5]. Spin fluctuations in $Sr_2RuO_4$ are believed to have both ferromagnetic and antiferromagnetic components [6]. Moreover, inelastic neutron scattering data indicate a soft phonon mode associated with in-plane rotations of oxygen octahedra [7]. Clearly $Sr_2RuO_4$ is close to structural and magnetic instabilities. Substitution of Sr by the smaller Ca ions also causes a crystallographic distortion consisting of tilting and rotation of oxygen octahedra. These modifications reduce the effective hopping between Ru $4d$ orbitals via O $2p$ levels. The resulting narrowing of the $4d$ bands is believed to be the driving mechanism for the metal-insulator transition observed in $Ca_{2-x}Sr_xRuO_4$ for $x = 0.2$ [3, 4, 8].

The layer perovskite $Sr_2RuO_4$ can be regarded as a prototype of a strongly correlated, highly anisotropic multi-orbital transition metal oxide with coexisting wide and narrow bands near the Fermi level. Due to the planar crystal structure the partially filled $t_{2g}$ bands separate into a two-dimensional $d_{xy}$ band with a van Hove singularity (vHs) just above $E_F$ [9], and two nearly one-dimensional $d_{xz,yz}$ bands. In the single-particle picture these bands do not hybridize for symmetry reasons. Local electron-electron interactions in the Ru $4d$ shell, however, are strong and according to angle-resolved photoemission spectra [10] the on-site Coulomb energy lies between the single-particle band widths of the $d_{xz,yz}$ and $d_{xy}$ bands: $W_{xz,yz} < U < W_{xy}$ [11]. Thus, although correlations primarily affect the narrow $d_{xz,yz}$ bands, multi-orbital interactions also influence the wider $d_{xy}$ bands, in particular its singularity above $E_F$. Recent quasi-particle calculations [11] based on the Quantum Monte Carlo (QMC) Dynamical Mean Field Theory (DMFT) [12, 13, 14] showed that on-site correlations give rise to a charge transfer from the narrow bands to the wider $d_{xy}$ band and to a red-shift of the van Hove singularity to within approximately 10 meV of $E_F$. Thus, doping with a few % La moves the Fermi level through the $d_{xy}$ singularity, thereby causing deviations from Fermi liquid behavior [5].

At the surface of $Sr_2RuO_4$, a $\sqrt{2} \times \sqrt{2}$ lattice reconstruction driven by a soft phonon mode involving rotations of O octahedra reduces the effective $d-d$ hopping [15]. As a result of the slightly narrower width of the $d_{xy}$ band in the first layer the van Hove singularity is shifted below $E_F$, converting the $\gamma$ sheet at the Fermi surface from electron-like to hole-like. Thus, photoemission spectra reveal a superposition of bulk bands with $E_{vHs} > E_F$ and surface bands with $E_{vHs} < E_F$ [16]. The bulk bands are then consistent with the Fermi surface derived from de Haas-van Alphen measurements [17].

To investigate the metal-insulator transition induced by iso-electronic substitution of Sr with Ca, Anisimov *et al.* [8] recently performed DMFT calculations for $Sr_2RuO_4$ for increasing values of the local Coulomb repulsion $U$. While in reality the transition is driven by a distortion of O octahedra, giving rise to a narrowing of the Ru $4d$ band without any significant increase in $U$, these authors argued that a qualitative understanding of the Mott transition can be achieved by increasing $U$ and ignoring the modifications of the density of states due to the structural transition. This picture is very interesting from a conceptual point of view since in the anisotropic $t_{2g}$ configuration $U$ is comparable at first to $W_{xz,yz} \approx 1.3$ eV and subsequently to $W_{xy} \approx 3.5$ eV. A remarkable result of these calculations were two successive Mott transitions for the $d_{xz,yz}$ and $d_{xy}$ bands, implying an intermediate region of $U$ or, equivalently, of Ca concentration, where the narrow bands exhibit an excitation gap while the wide band is still metallic. Only at still larger $U$ a gap appeared for the entire $t_{2g}$ complex.

To study this unusual electronic structure in greater detail we have extended our previous DMFT calculations for $Sr_2RuO_4$ [11] to larger values of the on-site Coulomb energy. Surprisingly, we find a behavior that differs fundamentally from the one reported in Ref. [8]. For small $U$, the $d_{xy}$ and $d_{xz,yz}$ quasi-particle spectra indeed have quite different shapes owing to their predominantly two- versus one-dimensional electronic structure, respectively. As $U$ increases, however, these spectral distributions begin to resemble one another, with only minor differences in the weight and shape of the coherent peak near $E_F$

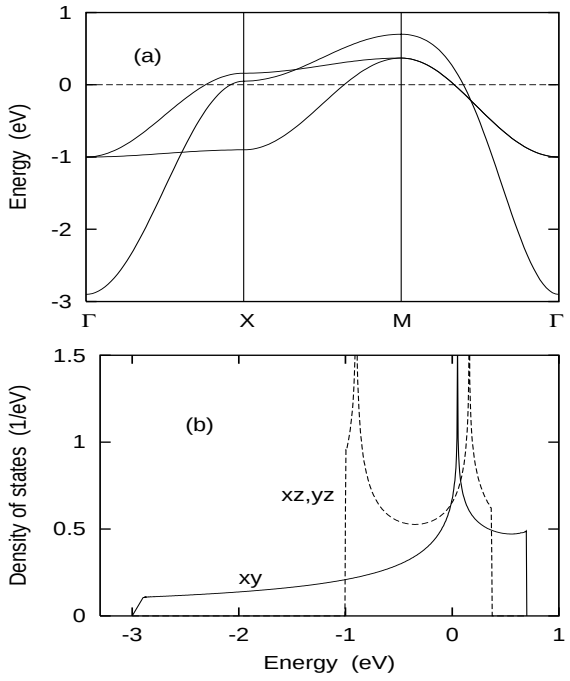


FIG. 1: (a) Tight-binding fit to LDA $t_{2g}$ bands of $Sr_2RuO_4$ ($E_F = 0$); (b) Density of states $\rho_i(\omega)$ of $d_{xy}$ bands (solid curve) and of $d_{xz,yz}$ bands (dashed curve).

and of the lower and upper Hubbard bands. Our results therefore suggest that for sufficiently large $U$ the correlated electronic structure within the $t_{2g}$ shell dominates the one-electron hopping. As a consequence, one common metal-insulator transition exists at an intermediate critical value of $U$ between those of the isolated $d_{xz,yz}$ and $d_{xy}$ bands. The reason for the discrepancy between the two approaches is not clear at present, but could be related to the fact that in Ref. [8] the quantum impurity problem was treated within the non-crossing approximation (NCA) [18, 19] whereas we use the more accurate Quantum Monte Carlo method [13].

Fig. 1 shows the tight-binding fit to the LDA band structure of $Sr_2RuO_4$ [9] in the vicinity of $E_F$ and the density of states. In the bulk the $d_{xy}$ vHs is about 50 meV above $E_F$. All three bands are approximately 2/3 filled. The non-symmetric shape of the two-dimensional $\rho_{xy}(\omega)$ is caused by second-neighbor Ru-Ru hopping terms. The $d_{xz,yz}$ densities are dominated by one-dimensional hopping along the Ru rows.

To take account of local Coulomb interactions we calculate the quasi-particle spectra within the multi-orbital QMC-DMFT approach [11, 20]. Since the $d_{xy}$ and $d_{xz,yz}$ bands do not hybridize, the self-energy is diagonal in orbital space. In the DMFT the elements $\Sigma_i$ ($i = xy, xz, yz$) are functionals of the bath Green's functions $\mathcal{G}_i^{-1} = G_i^{-1} + \Sigma_i$, where the local $G_i$ is given by

$$G_i(i\omega_n) = \int_{-\infty}^{\infty} d\omega \; \frac{\rho_i(\omega)}{i\omega_n + \mu - \Sigma_i(i\omega_n) - \omega} \qquad (1)$$

and $\mu$ is the chemical potential. Most QMC calculations were done for $\beta = 8$ with 64 time slices and several runs using $2x10^5$ sweeps. In the low temperature calculations for $\beta = 40$ 128 time slices were used, with up to $5x10^4$ sweeps. The quasi-particle density of states $N_i(\omega) = -\mathrm{Im}\, G_i(\omega)/\pi$ is obtained via maximum entropy reconstruction [21].

As shown in Ref. [11], the angle-resolved photoemission spectra of $Sr_2RuO_4$ can qualitatively be represented by quasi-particle distributions for local Coulomb and exchange energies $U = 1.2$ eV, $J = 0.2$ eV. The $d_{xy}$ and $d_{xz,yz}$ spectra then look like moderately deformed versions of their respective single-particle densities of states. For instance, the lower van Hove singularity of the $d_{xz,yz}$ bands near $-1$ eV is broadened and shifted to about $-0.5$ eV. Larger Coulomb energies would yield too small binding energies for this spectral feature. This result is consistent with the fact that on-site Coulomb energies for $4d$ transition metals are smaller than for $3d$ metals. The $d_{xy}$ vHs is also shifted and lies only about 10 meV above $E_F$. Both bands show very low spectral weight in the energy region of the Hubbard peaks. The effective masses calculated within the QMC-DMFT for these Coulomb energies also agree with experiment.

We now increase the on-site Coulomb energy in order to explore the metal-insulator transition for this anisotropic multiband system. To illustrate the effect of the different widths of the $t_{2g}$ subbands on the quasi-particle spectra we show first in Fig. 2 the results for hypothetical three-fold degenerate bands consisting either of $d_{xy}$ or $d_{xz,yz}$ character. The total filling is 4 as in actual $Sr_2RuO_4$. For $U = 3.0$ eV the wide $d_{xy}$ band is dominated by the strong quasi-particle peak at $E_F$. Since $U < W_{xy}$ it is only moderately affected by correlations and shows only weak shoulders in the range of the Hubbard bands. On the other hand, since $U >> W_{xz,yz}$ the narrow band derived from the $d_{xz,yz}$ density of states exhibits clear signs of a Mott transition. The upper and lower Hubbard peaks are the dominant spectral features. Because of the finite temperature used in the QMC-DMFT calculation, the remaining quasi-particle peak at $E_F$ obscures the gap between the Hubbard bands. Nevertheless, according to the temperature dependence of similar features found in one-band systems [13, 22] the value of $U$ used in the spectra shown in Fig. 2 should be very close to the critical value for a metal-insulator transition within the $d_{xz,yz}$ bands.

The quasi-particle spectra for the actual $t_{2g}$ bands of $Sr_2RuO_4$ are shown in Fig. 3 (a) for the same on-site energies. Evidently, as as result of the strong inter-orbital Coulomb and exchange interactions both $d_{xy}$ and $d_{xz,yz}$ spectral distributions have nearly lost their original density of states character and look remarkably similar: with coherent peaks at $E_F$ and upper and lower Hubbard

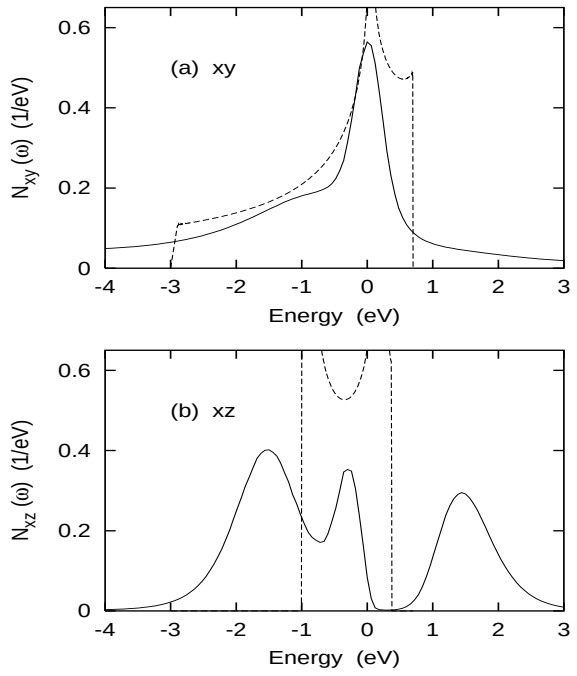


FIG. 2: Quasi-particle density of states $N_i(\omega)$ of $Sr_2RuO_4$ derived from DMFT for $T = 1450$ K, assuming independent three-fold degenerate (a) $d_{xy}$ and (b) $d_{xz,yz}$ bands (solid curves; see text). $U = 3.0$ eV, $J = 0.2$ eV; dashed curves: bare densities.

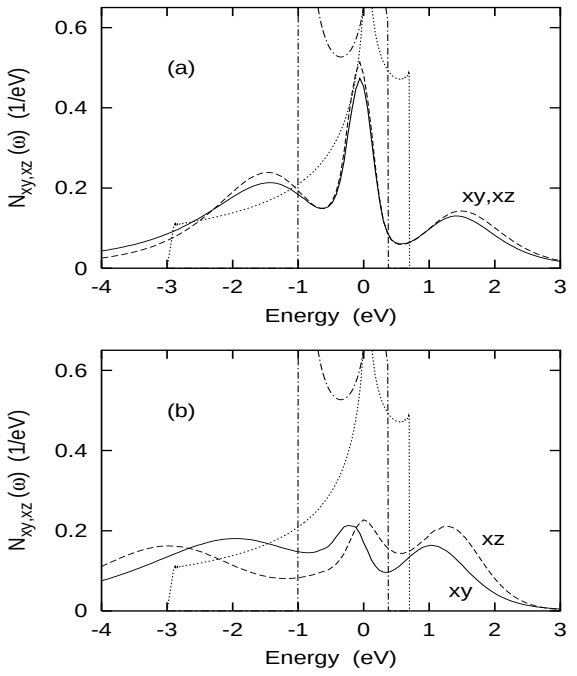


FIG. 3: Quasi-particle density of states $N_i(\omega)$ of $Sr_2RuO_4$ derived from DMFT for $T = 1450$ K. (a) $U = 3.0$ eV, $J = 0.2$ eV; (b) $U = 4.0$ eV, $J = 0.7$ eV. Solid curves: $d_{xy}$ states; dashed curves: $d_{xz,yz}$ states; dotted and dash-dotted curves: corresponding bare densities.

bands of about the same intensity, position and shape. These spectra suggest a common Mott transition for the $t_{2g}$ bands at a value of $U$ only slightly larger than 3 eV. We point out that the local correlations for $U = 3$ eV induce a charge transfer of about 0.05 electrons from the narrow $d_{xz,yz}$ bands to the $d_{xy}$ band. This would imply that the $d_{xy}$ van Hove singularity has shifted below $E_F$ and that the $\gamma$ sheet of the Fermi surface is hole-like rather than electron-like as observed in de Haas-van Alphen measurements [17]. This supports our previous choice of a smaller $U$ for the actual $Sr_2RuO_4$ $t_{2g}$ bands [11].

Fig. 3 (b) illustrates the effect of increasing the exchange energy to $J = 0.7$ eV. In order to keep the average Coulomb energy $\bar{U}$ unchanged $U$ is increased to 4 eV. (For a $t_{2g}$ complex $\bar{U}$ coincides with the inter-orbital Coulomb energy $U' = U - 2J$ [8]. Thus, $\bar{U} = 2.6$ eV in Figs. 3 (a) and (b).) Although the $d_{xy}$ and $d_{xz,yz}$ spectra are not quite as similar as in Fig. 3 (a), they have coherent and incoherent peaks of about the same intensity. Only the positions of these features are shifted. Extrapolation to lower temperatures does not seem to be quite as straightforward as in the previous case. Nevertheless, qualitatively both $d_{xy}$ and $d_{xz,yz}$ spectra are equally correlated and therefore also suggest a common Mott transition.

Judging from the results discussed so far for $U$ in the critical region it does not seem likely that upon lowering the temperature the $d_{xy}$ and $d_{xz,yz}$ spectra would begin to lose their similarity again and diverge towards two separate Mott transitions. To test this behavior we have performed multiband QMC-DMFT calculations at about 300 K which are computationally much more costly. Fig. 4 shows the $d_{xy}$ and $d_{xz,yz}$ quasi-particle distributions for $U = 3.0$ eV, $J = 0.2$ eV. The peak near $E_F$ exhibits a fine structure which is the remnant of the upper and lower edge singularities of the $d_{xz,yz}$ bands. The $d_{xy}$ spectrum also reveals this structure as a result of the strong orbital interactions. The important point is that both $d_{xy}$ and $d_{xz,yz}$ spectra are similarly correlated: the coherent peak has about the same weight and the upper Hubbard bands are almost identical. The main difference is that the lower Hubbard $d_{xz,yz}$ satellite is split off from the main peak by a gap while the $d_{xy}$ satellite lies in the lower tail of the $d_{xy}$ density of states and therefore forms a continuum with the main peak. Leaving aside the uncertainties caused by the maximum entropy reconstruction, such a gap does not imply that the spectral weight at the Fermi level vanishes at a different $U_{\rm crit}$ or $T_{\rm crit}$ than in the case of the $d_{xy}$ states. On the contrary, the comparable strength of the coherent peaks indicates that the $d_{xy}$ and $d_{xz,yz}$ bands undergo a common Mott transition.

The above results differ qualitatively from the ones obtained by Anisimov *et al.* [8]. The NCA in principle should be applicable also to multiband materials. On the

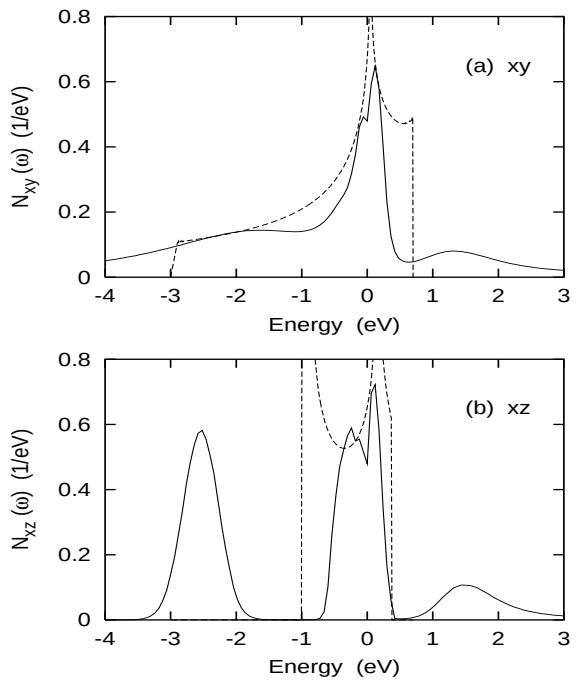


FIG. 4: Quasi-particle density of states $N_i(\omega)$ of $Sr_2RuO_4$ derived from DMFT for $T = 300$ K, $U = 3.0$ eV, $J = 0.2$ eV. Solid curves: (a) $d_{xy}$ states; (b) $d_{xz,yz}$ states; dashed curves: bare densities.

other hand, even in one-band cases it is known to be unreliable in certain situations and even violate Fermi-liquid behavior [13, 14, 20]. It would be desirable to perform numerical renormalization group calculations for this system at very low temperatures. Unfortunately, multiband calculations within this scheme are computationally not yet within reach.

In summary, we have used the QMC-DMFT to calculate quasi-particle spectra of $Sr_2RuO_4$ in the range of on-site electron-electron interactions appropriate for a possible metal-insulator transition. Qualitatively, increasing $U$ might be a way to simulate the reduced $t_{2g}$ band width caused by distortions of O octahedra when Sr is replaced by Ca. At low Coulomb energies, the spectral distributions of the $d_{xy}$ and $d_{xz,yz}$ bands differ strongly because of the planar geometry of this perovskite material. Despite this anisotropy, at sufficiently large $U$ these spectra resemble one another in the sense that they are equally strongly affected by local correlations. In particular, they exhibit similar upper and lower Hubbard bands and a coherent peak of similar strength. These results suggest that there is a common Mott transition for the $t_{2g}$ complex, in contrast to the squential orbital-selective Mott transitions found in Ref. [8]. Thus, in the critical region of $U$ local Coulomb interactions appear to dominate the anisotropic one-electron hopping.

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