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Orbital selective localization enhancement in $Ca_{2-x}Sr_xRuO_4$

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The discovery of unconventional superconductivity at $T_c = 0.93$ K in Sr_2RuO_4 which is isostructural to High-T_c layered copper-based perovskites [1, 2] motivated the search for a new Ru-based superconductor and gave rise to interest in the investigation of the electronic and magnetic properties of ruthenates. Synthesis and study of a complete series of $Ca_{2-x}Sr_xRuO_4$ alloys with isoelectronic substitution of Sr^{2+} by smaller Ca^{2+} ion was performed by Nakatsuji and Maeno [3, 4]. At concentration $x = 0.5$ the susceptibility shows a Curie form with a $S = 1/2$ moment (not $S = 1$) per Ru ion and the alloy exhibits metallic transport properties. At higher concentrations of Sr, $Ca_{2-x}Sr_xRuO_4$ has a paramagnetic metallic state. At a higher Ca concentration, the alloy remains metallic, but with AFM fluctuations at low temperatures. With a further increase in Ca concentration ($x < 0.2$), the alloy becomes an insulator.

To describe the evolution of the electronic structure at intermediate concentration, the orbital selective Mott transition model was proposed [5]. In this model t_{2g} manifold is divided into two groups of orbitals: degenerate xz, yz-orbitals and in-plane xy-orbital strongly π-hybridized with oxygen $2p$-orbitals. Using LDA results as input parameters for the DMFT method within the NCA computation scheme [6] the authors obtained orbital-selective behavior: the appearance of Mott localization in (xz, yz) subbands, while the broader half-filled xy-band remains itinerant.

In the present paper the DFT + DMFT method was applied to study the electronic structure of $Ca_{2-x}Sr_xRuO_4$ [7, 8]. The non-interacting band structure was calculated using the GGA (generalized gradient approximation) [9] as implemented in the Quantum Espresso package [10]. An effective Hamiltonian was constructed using the basis of atomic-centered Wannier functions as described in [11]. The DMFT impurity problem was solved by hybridization expansion of the continuous-time quantum Monte Carlo method (CT-QMC) [12] as implemented in the AMULET package [13]. QMC calculations were performed for the paramagnetic state at an electronic temperature $\beta = 20\,\mathrm{eV}^{-1}$ (580 K).

Local spin-spin correlation function $\langle \mathbf{S}_z(\tau)\mathbf{S}_z(0)\rangle$ on the imaginary time axis characterizes the lifetime of the local moment. If the magnetic moments are localized, this correlation function is constant: $\langle \mathbf{S}_z(\tau)\mathbf{S}_z(0)\rangle \approx S^2$ and the imaginary time dependence of this correlation function indicates the delocalization of spin moments, which is the Fermi liquid regime. The real part of the local spin-spin correlation function on the real frequency axis can be used to characterize the degree of localization: the half-width of $\langle \mathbf{S}_z(\omega)\mathbf{S}_z(0)\rangle$ peak at a half-height is in inverse proportion to the lifetime of the local moment [14].

The local spin-spin correlation functions $\langle \mathbf{S}_z(\omega)\mathbf{S}_z(0)\rangle$ for different x are shown in Fig. 1 (upper panel). For $x = 2$ the peaks of t_{2g} d_{xy} and $d_{xz,yz}$

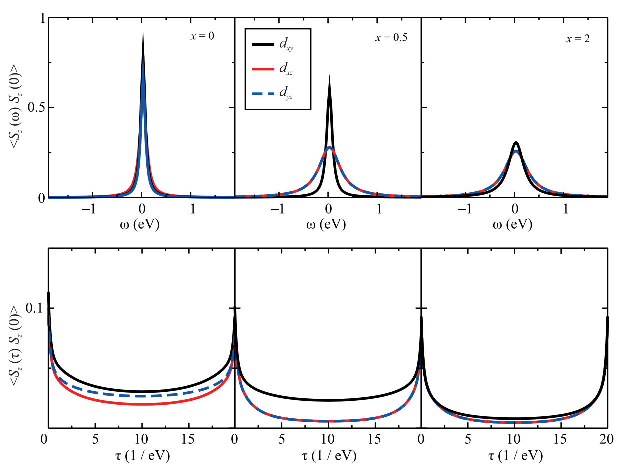

Fig. 1. (Color online) Local spin-spin correlation function $Ca_{2-x}Sr_xRuO_4$ calculated within DFT + DMFT at $\beta = 20$ $U = 1.5$ eV and $J_H = 0.7$ eV for $x = 0$, 0.5 and 2.0 on imaginary time axis (lower panels) and real frequencies (upper panels)

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orbitals have almost equal width and shape which is consistent with small distortion of oxygen octahedra in Sr_2RuO_4. The broad local spin-spin correlation functions correspond to a metallic phase with weakly localized electrons. On the other hand, in the case of Ca_2RuO_4 ($x = 0$) the peaks become sharp and the width reduces by a factor ≈ 2. The $\langle \mathbf{S}_z(\tau)\mathbf{S}_z(0)\rangle$ correlator has substantial value in the whole range $[0,\beta]$ as well as lifting of degeneracy of $d_{xz,yz}$ orbitals due to strong distortion of crystal structure. One can say, that d-electrons in Ca_2RuO_4 are more localized, as expected for Mott insulator regime. The case of $Ca_{1.5}Sr_{0.5}RuO_4$ is much more indicative. Ru t_{2g} orbitals are separated into two subsystems with completely different behavior: while $\langle \mathbf{S}_z(\omega)\mathbf{S}_z(0)\rangle$ curves for $d_{xz,yz}$ orbitals are broad and these states can be treated as itinerant and reminiscent metallic Sr_2RuO_4 the d_{xy} orbital has sharp peak and resembles Ca_2RuO_4. The half-widths of $\langle \mathbf{S}_z(\omega)\mathbf{S}_z(0)\rangle$ peak at a half-height for d_{xy} and $d_{xz,yz}$ orbitals as well as their lifetimes differs in ≈ 6 times. This agrees well with the model proposed in [5]. Variation of J_{H} does not change orbital-selective behavior. The width of $\langle \mathbf{S}_z(\omega)\mathbf{S}_z(0)\rangle$ peaks and hence the lifetime on local moments decreases in ≈ 3 times with increasing J_{H} from 0.5 to 0.9 eV. Note also, that orbital-selective behavior exhibits more distinctly for smaller J_{H}. Most likely, this effect could be explained by the change in the value of mean Coulomb interaction term which is 1.1 eV for $U = 1.5$ eV and $J_{\mathrm{H}} = 0.9$ eV and 1.28 eV for $U = 1.5$ eV and $J_{\mathrm{H}} = 0.5$ eV.

Analysis of the local spin-spin correlation function reveals that the end members of series Sr_2RuO_4 and Ca_2RuO_4 are conventional itinerant metals and correlated insulators, respectively, without any distinct sign of orbital selectivity. In contrast, $Ca_{0.5}Sr_{1.5}RuO_4$ demonstrates a clear picture of orbital selective behavior. The sharp dependence of magnetic properties and degree of localization on the Hund parameter J_{H} are an evident sign that intermediate members of the $Ca_{2-x}Sr_xRuO_4$ series belong to the Hund's metals. Variation of Hund's parameter shows that the localization degree depends on J_{H} while the orbital-selectivity is regulated by average Coulomb interaction value.

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