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Multi-orbital nature of the spin fluctuations in Sr_2RuO_4

Lewin Boehnke[1], Philipp Werner[1] and Frank Lechermann[2]

[1] *Department of Physics, University of Fribourg - 1700 Fribourg, Switzerland*
[2] *I. Institut für Theoretische Physik, Universität Hamburg - D-20355 Hamburg, Germany*

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Abstract – The spin susceptibility of strongly correlated Sr_2RuO_4 is known to display a rich structure in reciprocal space, with a prominent peak at $\mathbf{Q}_i = (0.3, 0.3, 0)$. It is still debated if the resulting incommensurate spin-density-wave fluctuations foster unconventional superconductivity at low temperature or compete therewith. By means of density functional theory combined with dynamical mean-field theory, we reveal the realistic multi-orbital signature of the (dynamic) spin susceptibility beyond existing weak-coupling approaches. The experimental fluctuation spectrum up to 80 meV is confirmed by theory. Furthermore, the peak at $\mathbf{Q}_i$ is shown to carry nearly equal contributions from each of the Ru($4d$)-t_{2g} orbitals, pointing to a cooperative effect resulting in the dominant spin fluctuations.

Introduction. – The Ruddlesden-Popper series of the strontium ruthenates $Sr_{n+1}Ru_nO_{3n+1}$, with n labeling the layers of corner-sharing RuO_6 octahedra separated by SrO rocksalt layers, poses a formidable correlated-electron problem in a realistic scenario. Intricate competitions between Fermi-liquid, Mott-critical and superconducting behavior are accompanied by a complex metallic magnetism. While the perovskite $SrRuO_3$ ($n \to \infty$) as series end member is ferromagnetic (FM) below $T_C \sim 165\,K$, a ferromagnetic-to-paramagnetic transition occurs by lowering the number of layers n. This transition takes place between the orthorhombic $n = 3$ and $n = 2$ systems. The $Sr_4Ru_3O_{10}$ compound is verified to be FM [1], whereas bilayer $Sr_3Ru_2O_7$ remains paramagnetic (PM) down to lowest temperatures. However, the bilayer material is prone to FM order [2,3] with puzzling metamagnetic (MM) behavior in an applied field below $T_{MM} \sim 1\,K$ (see ref. [4] for a review).

The tetragonal $n = 1$ compound Sr_2RuO_4 stands out, since it displays unconventional superconductivity (most probably triplet pairing), below $T_C \sim 1.5\,K$ [5–7]. De Haas-van Alphen measurements [8] and angle-resolved photoemission spectroscopy (ARPES) [9–11] documented the quasi–two-dimensional (quasi-2D) electron structure for this single-layer ruthenate. The strongly correlated nature is furthermore proven by a large mass renormalization [12], originating from the cooperation between less-screened Coulomb interactions and the actual filling of the t_{2g} manifold within the Ru($4d$) shell. More specifically, by combining band structure and dynamical mean-field theory (DMFT) it was shown that the intriguing interplay between the intra-orbital Hubbard interaction U and the inter-orbital Hund's exchange J_H gives rise to a so-called Hund's metal [13,14]. Though paramagnetic above the superconducting temperature, Sr_2RuO_4 exhibits substantial magnetic correlations, either interlinked or competing with the pairing instability [15]. A prominent peak in the $\mathbf{q}$-dependent spin susceptibility at finite $\mathbf{Q}_i = (0.3, 0.3)$ in the $\mathbf{q}_z = 0$ plane [16,17] renders it obvious that antiferromagnetic(-like) spin fluctuations must still be an essential feature of the puzzling ruthenate physics.

In this letter we advance on the understanding of the pecularities of Sr_2RuO_4 by connecting the strong correlation specification to the spin-fluctuations phenomenology. From the merger of density functional theory (DFT) with DMFT, we have a realistic many-body theory at hand that goes beyond existing studies [16,18–20] within the weak-coupling random phase approximation (RPA). We introduce a means of investigation of fluctuation characteristics by a rigorous eigenanalysis of the full orbital dependence of the complete susceptibility tensor. From that it is shown that multi-orbital correlations known to

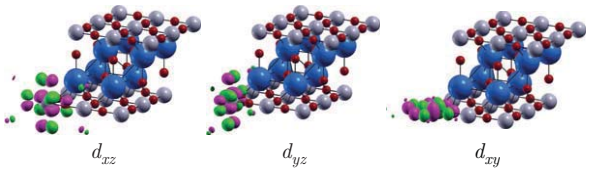

Fig. 1: (Color online) Maximally localized Ru(t_{2g})-like Wannier orbitals for Sr_2RuO_4 in the $I4/mmm$ crystal structure. Sr: blue; Ru: grey; O: red.

be relevant for the one-particle spectral function are also vital in the two-particle response functions such as the spin susceptibility $\chi(\mathbf{q},\omega)$. Namely, albeit the contribution of the Ru($4d$)-t_{2g} orbitals with nominal total four-electron filling from the Ru^{4+} oxidation state differs in $\mathbf{q}$-space, they equally take part in the formation of the dominant peak at $\mathbf{Q}_i$. This points to the relevance of including the contributions of all three t_{2g} orbitals when accounting for spin fluctuations in Sr_2RuO_4.

Calculational procedure. – Single-layer Sr_2RuO_4 has ideal tetragonal symmetry (space group $I4/mmm$) with fourfold rotation C_4^z around the c-axis. Our first-principles many-body calculations are based on the structural data of Walz and Lichtenberg [21]. A mixed-basis pseudopotential scheme is used for the band structure part within the local density approximation (LDA) [22]. To account for strong electronic correlations in the DMFT treatment, a correlated subspace is constructed from the derived Ru $4d(t_{2g})$-like maximally localized Wannier functions [23,24] (see fig. 1). Therein a three-orbital Hubbard model in rotationally invariant Slater-Kanamori paramaterization with $U = 2.3\,\mathrm{eV}$ and $J_\mathrm{H} = 0.4\,\mathrm{eV}$ is constructed [13]. The multi-orbital DMFT impurity problem is solved by the continuous-time quantum Monte Carlo method in the hybridization-expansion formulation [25–29]. As in previous DFT+DMFT studies [13,30], effects of spin-orbit coupling are not included in the present work.

In order to access the two-particle response, we compute the complete three-orbital particle-hole susceptibility tensor $\chi^{\sigma\sigma'}_{mm'm''m'''}(\mathbf{q},\omega)$, with $m,m'm'',m''' = 1,2,3$ and $\sigma,\sigma' = \uparrow,\downarrow$, at finite temperature, with full generality in the frequency-dependent structure [27], employing the numerically beneficial orthogonal polynomial basis for the fermionic degrees of freedom. The full $\mathbf{q}$-dependence is obtained from the DMFT two-particle Green's function by inverting the Bethe-Salpeter equation, assuming the locality of the irreducible vertex [31,32]. For the bosonic dynamic ω-dependence of the susceptibility a further numerically exact optimization was employed, which alters the shift in the fermionic transformation depending on the bosonic frequency [33].

To cope with the multi-orbital character, we focus on the eigenvalues/modes of the susceptibility tensor $\chi_{\kappa\kappa'}$ in the superindices $\kappa = \{\sigma m m'\}$ and $\kappa' = \{\sigma' m''' m''\}$ [34].

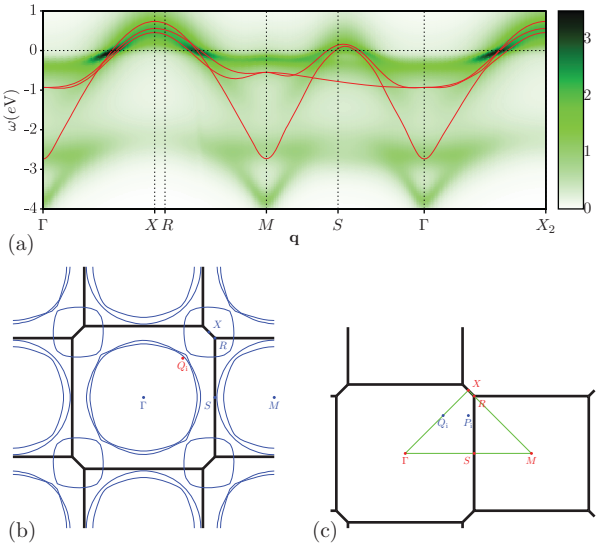

Fig. 2: (Color online) Correlated electronic structure of Sr_2RuO_4 from DFT+DMFT. (a) One-particle spectral function (full lines: LDA data) and (b) Fermi surface in the ($k_z = 0$)-plane. (c) High-symmetry points in reciprocal space. The X_2 point is located above X on the same level as the upper surface of the Brillouin zone.

Specifically

$$\chi^{(l)}(\mathbf{q},\omega=0) = \left\langle \mathcal{T}_\tau \sum_\kappa \hat{v}^{(l)}_\kappa(\mathbf{q}) \sum_{\kappa'} \hat{v}^{\star(l)}_{\kappa'}(\mathbf{q}) \right\rangle = \left\langle \mathcal{T}_\tau \hat{V}^{(l)}(\mathbf{q}) \hat{V}^{\star(l)}(\mathbf{q}) \right\rangle \quad (1)$$

is the l-th eigenvalue with $\hat{V}^{(l)} = \sum_{\sigma m m'} v^{(l)}_{\sigma m m'} c^{\dagger\sigma}_m c^\sigma_{m'}$ as the corresponding fluctuating eigenmode ($v^{(l)}_\kappa$ is the l-th eigenvector of $\chi_{\kappa\kappa'}$).

Results. – Based on our DFT+DMFT calculations, we first provide some results on the Sr_2RuO_4 correlated electronic structure consistent with previous theoretical work [13,30]. Figure 2(a) displays the one-particle spectral function $A(\mathbf{k},\omega)$ along high-symmetry lines in the corresponding Brillouin zone (BZ) (cf. fig. 2(c)). Four electrons reside in the low-energy t_{2g}-like manifold with LDA bandwidth $W \sim 3.4\,\mathrm{eV}$. Electron-electron interactions lead to significant band renormalization, with additional spectral-weight transfer to $\sim -3\,\mathrm{eV}$. The wider low-energy band is of dominant d_{xy} character, while the two narrower bands are mainly formed by d_{xz}, d_{yz}. The resulting Fermi surface has three sheets: α centered around X and β centered around Γ, both stemming from the narrower quasi–one-dimensional (quasi-1D) bands, as well as γ (also around Γ) derived from the quasi-2D d_{xy}-like band (see fig. 2(b)).

Due to the possibility of producing very clean samples for single-layer ruthenate, inelastic neutron scattering (INS) experiments elucidated the magnetic excitations in great detail [17,35–37]. Though it remains difficult to reach the experimental very low-T regime with quantum

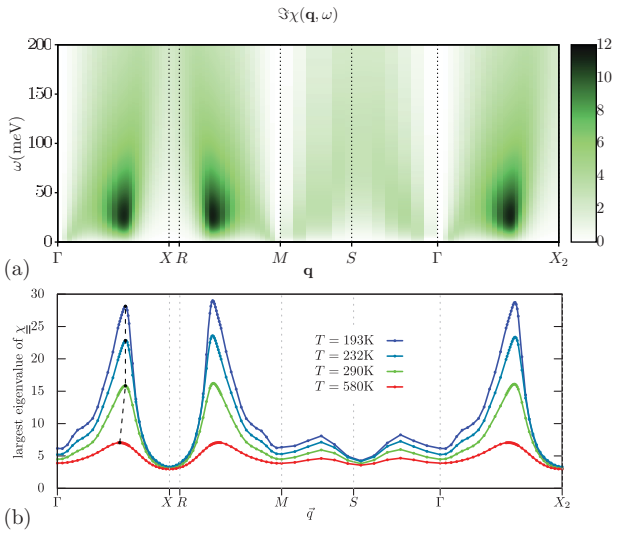

Fig. 3: (Color online) Data on the spin susceptibility along high-symmetry lines in reciprocal space. (a) Dynamic spin susceptibility $\chi_s(\mathbf{q},\omega)$ at $T = 290\,\mathrm{K}$. (b) Dispersion of the dominant eigenvalue of the spin-susceptibility tensor for selected temperatures.

Monte Carlo, the computed room temperature dynamic spin susceptibility $\chi_s(\mathbf{q},\omega)$ (see fig. 3(a)) agrees very well with available data. Note that also recent INS reported sizeable fluctuations even up to that temperature range [37]. The location of the incommensurate peak $\mathbf{Q}_i$ as well as its position in energy $\omega_{\mathbf{Q}} \sim 27\,\mathrm{meV}$ is consistent with INS [36,37]. Moreover, the experimental finding of a fluctuation regime up to 80 meV [37] is confirmed by our calculations.

To identify the dominant particle-hole fluctuation in an unbiased way, the dominant eigenvalues of the susceptibility tensor $\chi^{\sigma\sigma'}_{mm'm''m'''}$ are monitored in the static limit $\omega = 0$. The maximum eigenvalue in Sr_2RuO_4 corresponds to a mode that is diagonal in the orbital components throughout the BZ. The eigenmode is indeed related to S^z and exhibits the form $V^{\max}(\mathbf{q}) \sim v_{xz}(\mathbf{q})S^z_{xz} + v_{yz}(\mathbf{q})S^z_{yz} + v_{xy}(\mathbf{q})S^z_{xy}$, where the normalization $\Sigma_\kappa v^2_\kappa(\mathbf{q}) = 1/2$ holds. Note that the orbital diagonality is not trivial and there are other cases with a more complex structure [34]. The dispersion of this dominant eigenvalue, plotted in fig. 3(b), shows a significant temperature dependence. Whereas the peak close to $\mathbf{Q}_i$ appears already at rather high T (with notable shifting in q-space when lowering the temperature) a clear peak shoulder along ΓX towards Γ sets in only below room temperature. That shoulder is again in very good agreement with experiment [35]. Directly at Γ the eigenvalue grows with T, but a large response pointing to an obvious FM contribution does not show up.

A full 2D mapping of the dominant-eigenvalue intensity within the irreducible wedge of the BZ readily marks $\mathbf{Q}_i$ as the hot spot for spin-like fluctuations (see fig. 4(a)). Most interesting in this context is the variation of the respective orbital contributions v_{xz}, v_{yz}, v_{xy} throughout

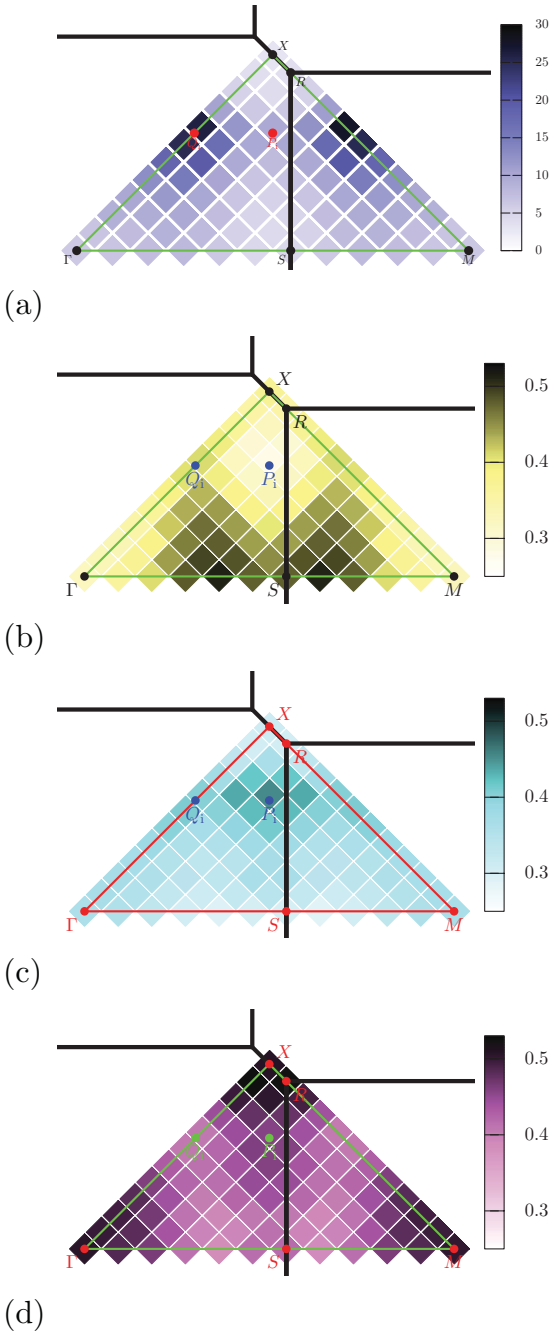

Fig. 4: (Color online) Data extracted from the orbital-resolved spin-susceptibility tensor at $T = 193\,\mathrm{K}$. (a) Dominant eigenvalue of the susceptibility tensor throughout the irreducible wedge of reciprocal space. (b)–(d) q-dependent orbital contributions to the dominant eigenmode: (b) v_{xz}, (c) v_{yz} and (d) v_{xy}.

the BZ, depicted in fig. 4(b)–(d). For instance, the eigenmode at Γ is characterized by $v_{xz}(\mathbf{0}) = v_{yz}(\mathbf{0}) = 0.34$ and $v_{xy}(\mathbf{0}) = 0.52$. Thus, the d_{xy} orbital makes the major contribution to the spin fluctuations at Γ. Within the irreducible wedge, the regions of dominant weight of the different t_{2g} orbitals are mutually exclusive. As expected, d_{xz} and d_{yz} contribute strongly along the original 1D-like Fermi-surface directions, while the d_{xy} weight is largest

around the high-symmetry points Γ, X and M. Quite surprisingly, right at the spin-fluctuation peak position $\mathbf{Q}_i$ *all* the orbitals have an *equal* share, *i.e.*, $v_{xz}(\mathbf{Q}_i) = v_{yz}(\mathbf{Q}_i) = v_{xy}(\mathbf{Q}_i) = 0.41$. Hence a straightforward d_{xz}-d_{yz} nesting scenario cannot explain the $\mathbf{Q}_i$-peak. Spin flucutations at the latter q-point have a manifest multi-orbital character.

Summary. – Our realistic many-body approach based on DFT+DMFT allows for an extended study of the multi-orbital electronic properties in the single-layer Ruddlesden-Popper compound Sr_2RuO_4. By means of generic lattic susceptibilities at strong coupling, *i.e.*, including vertex contributions, we examine the spin-fluctuation spectrum depending on wave vector, frequency and temperature. Good agreement with available INS data is obtained, the experimental findings concerning the relevant energy and temperature scales are confirmed.

From an eigensystem analysis of the generic particle-hole susceptibility tensor it is indeed possible to designate a magnetic eigenmode proportional to S^z as the dominant one. The orbital contributions to that mode vary over the Brillouin zone, describing some anisotropy in the spin response. Importantly, at the major peak position $\mathbf{Q}_i = (0.3, 0.3)$ in the $\mathbf{q}_z = 0$ plane the latter has an intrinsic multi-orbital nature, involving fluctuations in all three t_{2g} orbitals equally, in contrast to a combination of 1D-nesting pictures. This finding is important not only for a deeper understanding of the magnetism in Sr_2RuO_4, but also for theoretically approaching the pairing mechanism at very low temperatures. Therefore, advancing the present scheme towards an assessment of the particle-particle susceptibility would be promising. Investigating the influence of spin-orbit coupling [38] is an additional important aspect.

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