Low-energy electronic structure of the itinerant metamagnet $Sr_3Ru_2O_7$

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Abstract. We have investigated the low-energy electronic structure of $Sr_3Ru_2O_7$ single crystals via synchrotron-based angle resolved photoemission spectroscopy measurements performed at 1 K, focusing in particular on the band dispersion in the vicinity of the Γ and X points in the first Brillouin zone. Our results show the existence of two strongly renormalised bands - one centred around each high-symmetry point - attributed to Ru 4*d* states, whose heavy quasiparticles are located within the first 6.5 meV from the Fermi level, and show a remarkably sharp line width (about 6 meV). Such flat bands give rise to van Hove singularities in the near-Fermi-energy region whose energy scales are compatible with theoretical models for the appearance of itinerant metamagnetism in this system.

1. Introduction

The discovery of the unconventional spin triplet superconductivity in the single layer ruthenate Sr_2RuO_4 [1] attracted interest to the Ruddlesden-Popper (RP) series of strontium ruthenates $Sr_{n+1}Ru_nO_{3n+1}$. This family of metallic compounds turned out to exhibit remarkably different physical properties tuned by the number n of RuO_2 octahedra layers in the unit cell, ranging from superconductivity ($n = 1$), through anisotropic metamagnetic ordering ($n = 2, 3$) [2, 3, 4, 5], to itinerant electron ferromagnetism [2, 3, 4, 5] ($n > 3$). The contribution of the Sr states is non-negligible [6], with the electronic structure of the series being dominated by the RuO_2 octahedra layers. In particular, as Ru always appears to be in an oxidation state +4, the low energy electronic properties of ruthenates are determined by the electrons in the Ru t_{2g} manifold consisting of the nearly degenerate d_{xy}, d_{yz} and d_{zx} orbitals, which strongly hybridise with O $2p$ states [6].

In this study we focus only on the second member of the RP series, $Sr_3Ru_2O_7$. The ground state of $Sr_3Ru_2O_7$ is a paramagnetic Fermi liquid very close to a ferromagnetic instability at zero field [7]. The value of its electronic specific heat is $\gamma = 110$ mJ/(Ru mol K^2), which is among the largest in any oxide indicating strong electronic correlations [8]. This metal exhibits anisotropic itinerant metamagnetism with critical fields ranging from 5.5 T (for H $\parallel$ ab plane) to 7.7 T (for H $\parallel$ c-axis) [5]. This metamagnetic transition ends in a quantum critical point [5, 9, 10] and it is accompanied by the formation of a novel ordered quantum phase known as a "nematic phase" [11, 12] with broken rotational symmetry [11] and heavy d-electron masses [13].

A lot of effort has been so far devoted into understanding the nature of the microscopic origin of metamagnetism in $Sr_3Ru_2O_7$ and has presented a significant challenge within condensed

matter physics [14, 15, 16]. The underlying idea compatible with the Stoner model is that metamagnetism is caused by the presence of narrow peaks in the electronic density of states (called van Hove singularities, vHS) in the vicinity of the Fermi energy level E_F [14]. Such vHS can be found in regions of the reciprocal space where the gradient of the electronic bands is zero, i.e. where the bands are flat or show a saddle point in the dispersion. According to the above-mentioned model, the application of an external magnetic field would cause the vHS corresponding to spin up and spin down electrons into opposite directions, and one of these would be brought closer to the Fermi level - and even on the opposite side of it - producing a discontinuous transition in the magnetisation known under the name of metamagnetism [17].

In this study, we report detailed angle resolved photoemission spectroscopy (ARPES) measurements of $Sr_3Ru_2O_7$ around the high symmetry points X and Γ in the first Brillouin zone, reporting direct evidence of flat Ru 4*d* bands close to the Fermi energy, giving rise to vHS located within a few meV below E_F, the energy scale relevant for metamagnetism. Our results for the band dispersion around the X point are in agreement with that reported by Tamai and coworkers [18]. Finally, the line shape of the quasiparticle peaks can be described within the "peak-dip-hump" model, showing a fingerprint of strong electronic correlations in this system.

2. Experimental Method

$Sr_3Ru_2O_7$ single crystals were grown by the floating zone technique with Ru self-flux in the Department of Physics at the University of Salerno (Italy) [19]. Samples were characterized by means of X-ray diffraction, resistivity, energy dispersive microscopy and electron backscattering diffraction, confirming the phase purity and the high quality of the crystals. ARPES experiments were carried out at the 1^3 end-station installed at the beamline UE112-lowE-PGM-b of the synchrotron BESSY II in Berlin (Germany) using a Scienta R4000 electron energy analyser, which allowed us to simultaneously measure many energy distribution curves (EDC) in an angular range of 28°. Samples were cleaved *in situ* on the manipulator of the experimental chamber at 40 K, and then cooled down to the measurement temperature of 1 K (temperature that was kept constant throughout the experiment). The spectra presented here were measured with photon energies of 20 eV and 25 eV and with various incident photon polarisations. The overall energy and angular resolutions were $\sim$ 5 meV (as confirmed by the broadening of the Fermi level) and 0.15°, respectively.

3. Results and Discussion

Panels (a) and (b) in Figure 1 show the band dispersion of $Sr_3Ru_2O_7$ acquired around the *X* point with horizontally and vertically polarised light (HP and VP, respectively). The position of the dispersing quasiparticle peak as a function of $k_{||}$ has been determined by fitting the experimental EDC with a Gaussian line shape, and it has been plotted in Figure 1 (c) for four different light polarisations. From this plot it can be seen that the dispersion of the quasiparticle peak is very narrow and it is confined within a bandwidth of $\sim$ 2.5 meV for all polarisations, in a binding energy range that goes from 3 to 6.8 meV, and that the central part of the band centred around the X point is basically flat, i.e. it does not show any dispersion at all, and it is centred at an average binding energy of 5 meV. Such a flat band gives rise to a very high DOS below E_F, compatible with the presence of a vHS located within a few meV of the Fermi energy.

Further information regarding the symmetry of this vHS can be obtained by comparing the results obtained with HP and VP. In fact, changing the light polarisation in an ARPES experiment is an established methodology that allows to take advantage of the effect of the photoemission (PE) matrix elements associated to a specific electronic band. This fact results in the enhancement or suppression of the PE intensity from the band, from which one can gain information about the symmetry of those states. According to the experimental geometry exploited in this work, the electric field of vertically polarised light lays entirely in the *ab* plane of

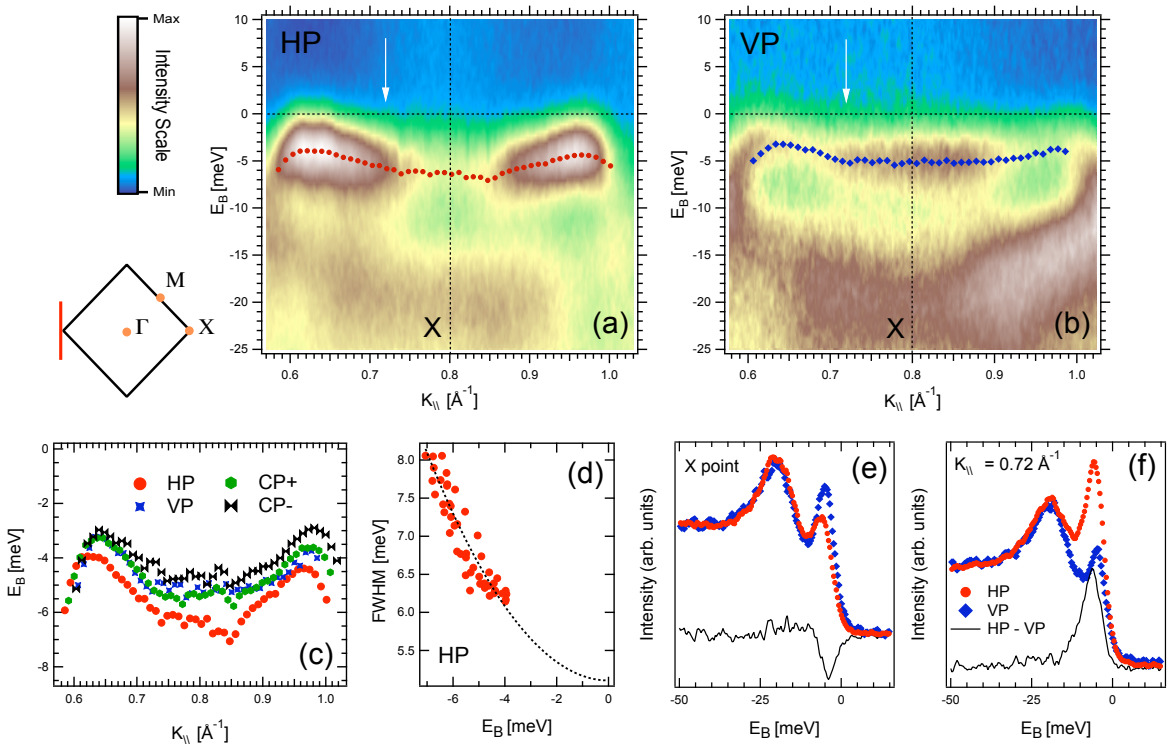

Figure 1. Panels (a) and (b): ARPES two-dimensional cuts acquired in the vicinity of E_F around the X point. These were taken along the direction indicated in the sketch of the first Brillouin zone and acquired at a temperature of 1 K and a photon energy hν = 20 eV with horizontal (HP) and vertical (VP) polarisation, respectively. White and blue colours correspond to high and low photoemission intensity, respectively, as indicated by the colour intensity scale. The fitted binding energy versus momentum curves (markers) have been plotted on top of the bands. (c) Momentum dependence of the quasiparticle peak binding energy fitted from momentum-energy cuts at different polarisations. The peak positions are found by fitting the energy distribution curves (EDCs) taken from (a) and (b) with a Gaussian function (the momentum-energy cuts for circular polarisations are not shown in this work). (d) Binding energy dependence of the full-width half-maximimum (FWHM) of the quasiparticle peak for the momentum-energy cut acquired shown in (a). The obtained experimental FWHMs are well fitted with a parabolic function (dashed black line), which is a fingerprint of Fermi liquid behaviour of this heavy quasiparticle in $Sr_3Ru_2O_7$ [20]. Panels (e) and (f): Comparison of HP and VP EDCs extracted from (a) and (b) at the X point and at $k_{||} = 0.72\,\text{Å}^{-1}$ (indicated by white arrows in (a) and (b)). The difference between HP and VP spectra is also shown in black.

the sample, while for horizontally polarised light it is almost completely parallel to the c axis of the sample. We can therefore conclude that, because of the dipole selection rules, VP enhances in-plane states while HP enhances out-of-plane states. The effect of PE matrix elements is evident in our data in panels (a) and (b) of Figure 1. The same band has an enhanced intensity in its central non-dispersing part (centred around the X point) in VP, while the use of HP seems to favour the emission from the dispersing wings of the band approaching E_F. This suggests that the non-dispersing part of the band has a majority in-plane character, while the dispersing part of the band has a majority out-of-plane character. This is also reflected in Figure 1 (e) and (f) where the difference between the EDCs in HP and VP shows how VP enhances the PE intensity close to E_F around the X point while it suppresses the intensity away from X. The in-plane character of the vHS close to the X point is in agreement with the theoretical models cited in the introduction [14, 15, 16] according to which the vHS close to the X point has d_{xy} orbital character [18].

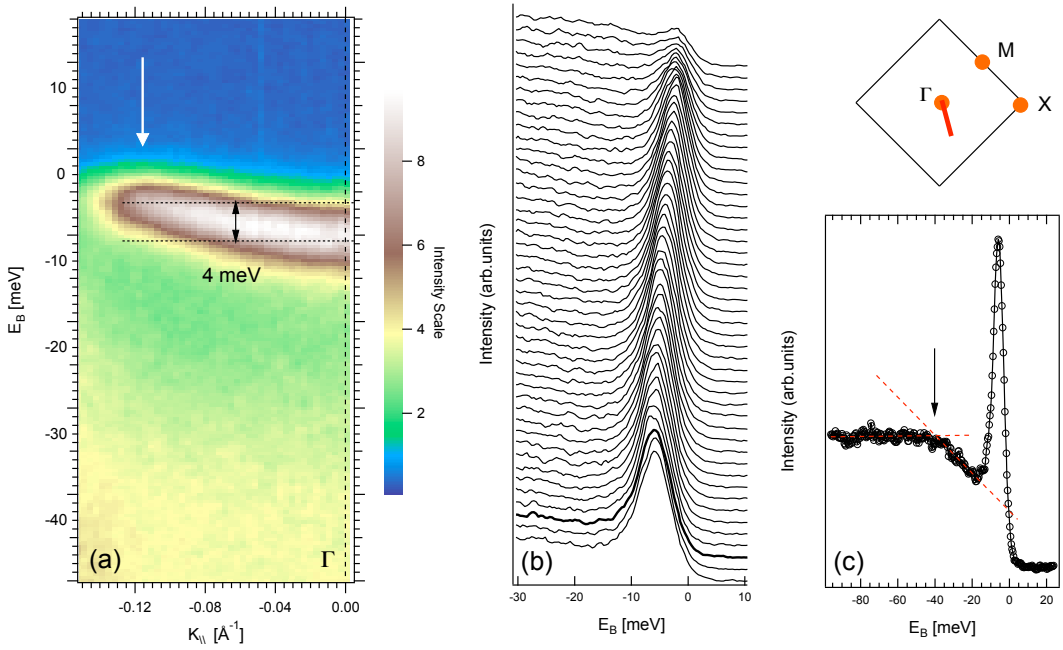

Figure 2. (a) ARPES two-dimensional cut acquired at a temperature of 1 K and a photon energy hν = 25 eV with left circularly polarised light in the vicinity of E_F around the Γ point. The red line in the schematic representation of the Brillouin zone shows the direction along which the cut was measured. (b) Stack of the EDC spectra constituting the cut shown in (a). (c) EDC spectrum at the Γ point. The arrow indicates the position of the hump.

Another promising Fermi surface sheet in terms of vHS is the so-called δ electron pocket whose dispersion is centred around the Γ point. Figure 2 (a) and (b) show the experimental quasiparticle dispersion of this feature, from where it can be seen that its bandwidth is limited to $\sim$ 4 meV. By integrating the spectral weight of the δ pocket in the relevant $k_{||}$ region, one can see that this feature gives rise to a vHS (inset in Figure 2 (a)) located at 2 meV from the Fermi level, again in an energy scale which is relevant to the formation of metamagnetism in $Sr_3Ru_2O_7$.

It is worth mentioning at this point that the bandwidth of the δ pocket is reduced by a factor 25 with respect to the bare band dispersion predicted by LDA calculations [21] due to strong correlation effects. Signatures of strong correlation effects are also evident in the line shape of the quasiparticle peak itself, as shown in Figure 2 (c) for the Γ point EDC, where some of the spectral weight of the coherent quasiparticle has been pushed to higher binding energies to form the characteristic 'peak-dip-hump' line shape of correlated systems. Such a line shape was observed for the first time in high-T_c superconductors [22, 23] and then in other correlated systems [24], and there is agreement of it being due to the interaction between electrons and a sharp collective mode, which alters the shape of the self energy resulting in the appearance of an incoherent peak. Following Campuzano *et al.* [23], the position of the hump is located where the spectrum changes slope as highlighted by the intersecting straight lines in (c). The hump is located 34 meV below the Fermi level. Further investigation on the dispersion and the origin of the hump are beyond the scope of this work.

In conclusion, we have presented low-temperature electronic structure data for $Sr_3Ru_2O_7$, revealing the presence of heavy quasiparticles associated to two very flat bands centred around the X and Γ point in the first Brillouin zone. Such bands give rise to vHS in the close vicinity of

the Fermi level, which can be interpreted as responsible for the appearance of metamagnetism in this system.

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