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Coulomb Correlations in 4d and 5d Oxides from First Principles - or How Spin-Orbit Materials choose their Effective Orbital Degeneracies

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Abstract

The interplay of spin-orbit interactions and Coulomb correlations has become a hot topic in condensed matter theory. Here, we review recent advances in dynamical mean-field theory-based electronic structure calculations for iridates and rhodates. We stress the notion of the *effective degeneracy* of the compounds, which introduces an additional axis into the conventional picture of a phase diagram based on filling and on the ratio of interactions to bandwidth.

1 Introduction

Electronic Coulomb correlations are at the heart of a variety of exotic properties in compounds with partially filled 3d or 4f shells. Prominent examples are found among the 3d transition metal oxides, where unconventional transport behaviors, ordering phenomena or unusual spectroscopic properties are observed [1]. It was argued early on that the comparably weak spatial extension of 3d orbitals leads to large electronic Coulomb interactions, competing with kinetic contributions. Depending on crystal fields, hybridisation, Hund's exchange, and band filling, this interplay can lead to renormalised metallic behavior such as in simple oxides like $SrVO_3$ [2, 3] or iron pnictide compounds [4–9] or induce Mott insulating behavior like in $YTiO_3$ [10] or V_2O_3 [11–14]. According to common belief until recently, such effects would be less dramatic in 4d and even less in 5d compounds, due to the substantially more extended radial wave functions of those shells, as shown in Fig. 1. The discovery of Mott insulating behavior in Sr_2IrO_4 therefore triggered a little revolution in the field [15, 16]. In 5d oxides, spin-orbit coupling acts

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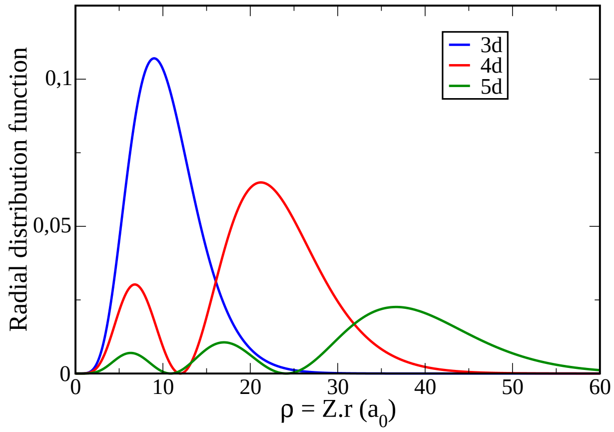

Figure 1: Radial distribution function $r^2 R_{n\ell}(r)^2$ as a function of the distance from the nucleus r expressed in atomic units, for the $3d$, $4d$ and $5d$ orbitals. To ease the comparison between different atoms, we use the renormalized distance $\rho = Z.r$ on the abscissa, where Z is the effective nuclear charge for a given multi-electron atom. As the principal quantum number n increases, Z remains almost constant for d valence electrons and their radial distribution is thus more and more extended.

on an energy scale comparable to the other scales of the system (Coulomb interactions, bandwidths, ligand fields ...), and the electronic state is the result of a complex interplay of Coulomb correlations, spin-orbit splitting and crystal field effects (for recent reviews see [17, 18]). But, as pointed out already earlier [19, 20], also in 4d compounds spin-orbit interactions can influence the electronic properties substantially. In Sr_2RhO_4, for example, the experimentally observed Fermi surface can only be reconciled with experiments when spin-orbit coupling and electronic Coulomb correlations are taken into account [19–22]. Here, we give a review of recent efforts to describe correlated spin-orbit physics from first principles, in a combined density functional and dynamical mean-field theory framework [21].

2 Spin-orbit materials – an incomplete literature review

The term *spin-orbit material* refers to systems where spin-orbit coupling (SOC) and its interplay with other elements of the electronic structure – crystal or ligand fields, Coulomb correlations, magnetism, ... – is essential in determining the physical properties. In many such materials, the physics is largely determined by the geometrical aspects of the cristalline structure, and the electronic properties can be understood by analysing the one-particle band structure. In particular, strong enough spin-orbit coupling can cause band inversions, possibly leading to non-trivial topological effects. The quest for topological materials is nowadays a hot topic of condensed matter physics, and several excellent reviews exist in the literature [23–25].

The scope of the present review is however a different one. Here, we focus on materials, where the interplay of spin-orbit interactions and Coulomb correlations is crucial, and the band picture is at best useful as a starting point for further many-body calculations. Early examples are found among the layered tantalum chalcogenides: TaS_2 [26–28] is Mott insulating thanks to the presence of a lone narrow band resulting from the combined effect of SOC and a charge-density wave instability. The corresponding selenide, $TaSe_2$ [29] displays a surface Mott metal-insulator transition. Nevertheless, the true power of the interplay of spin-orbit interactions was fully appreciated only after the discovery of Sr_2IrO_4: the insulating behavior – despite of moderate Coulomb interactions usually present in 5d compounds – was even more intriguing, as the electronic and crystal structures are otherwise seemingly simple. The interplay of Coulomb correlations and spin-orbit coupling was indeed shown to be essential to drive the system insulating, leading to a state dubbed "spin-orbit Mott insulator" [15, 16]. A flury of further spin-orbit materials have by now been characterized, or known compounds have been reinvestigated in the light of new insights. Iridium-based materials, where several families of compounds have been studied systematically, still hold a privileged position. Tab. 1 summarizes the structural, transport and magnetic properties of a selection of iridates. It is interesting to note that the large majority among them display insulating phases. The Ir^{4+} ($5d^5$) state does not allow for a band insulating state without symmetry breaking, and magnetic order is an obvious candidate for helping in opening the gap. Nevertheless, few compounds have been unambigously characterized as Slater insulators.

Slightly more recently, attention focussed yet onto another class of 5d materials, namely osmium-based compounds. In this class fall for example ferroelectric $LiOsO_3$ [30] as well as the prototypical Slater insulator $NaOsO_3$ [31–36] where the loss of magnetic order with increasing temperature is accompanied by a closure of the insulating gap. It has been realised, however, that SOC can also have notable effects in 4d compounds, with prominent examples among ruthenium- and rhodium-based materials, where most interesting consequences for magnetic excitations have been discussed [37]. Tab. 2 gives an overview of the properties of a selection of osmates, ruthanates and rhodates. In the following discussion, we will restrict ourselves to the prototypical correlated iridate Sr_2IrO_4 and its 4d analog, Sr_2RhO_4.

2.1 Correlated spin-orbit insulators: the example of Sr_2IrO_4

The 5d transition metal oxide (TMO) Sr_2IrO_4 has a tetragonal crystal structure, the symmetry of which is lowered from the K_2NiF_4-type, well-known in Sr_2RuO_4 or La_2CuO_4, by an 11° rotation of its IrO_6 octahedra around the **c**-axis [136]. Each Ir atom accomodates 5 electrons and the standard picture neglecting spin-orbit interactions would give a "t_{2g}^5" ground state. However, this compound exhibits insulating behavior up to the highest measured temperatures, with a strongly temperature-dependent gap. The optical gap at room temperature is about 0.26 eV [137]. Below $T_N = 240\,\mathrm{K}$, a canted-antiferromagnetic (AF) order sets in, with an effective local moment of $0.5\,\mu_B$/Ir, and a saturation moment of $0.14\,\mu_B$/Ir [138]. This phase has triggered much experimental and theoretical work [139–142], highlighting in particular the importance of the SOC.

Here, we focus on the paramagnetic phase, above 240 K, which is most interesting due to the persistance of the insulating nature despite the absence of magnetic order, as shown by transport measurements [15], by scanning tunneling microscopy and spectroscopy experiments [143], by angle-resolved spectroscopy [16, 144], time-resolved spectroscopy [145, 146] or optical conductivity [137].

Resonant Inelastic X-ray spectroscopy (RIXS) experiments [15] have early on proposed a picture in terms of j_{eff}=1/2 and j_{eff}=3/2 states:

Iridium-based spin-orbit materials							
Compound	Crystal Struct.		Transport Property		Magnetic Ordering		Ref.
$CaIrO_3$	post-perovskite	$Cmcm$	Ins.	gap: 0.34 eV	AFM	$T_N = 115$ K	[38–40]
$NaIrO_3$	post-perovskite	$Cmcm$	Ins.	–	None		[41, 42]
$BaIrO_3$	monoclinic	$C2/m$	Ins.	gap: 0.05 eV	FM	$T_C = 180$ K	[43–45]
$SrIrO_3$	monoclinic	$C2/c$	Metal		None		[46–49]
α-Na_2IrO_3	honeycomb monoclinic	$C2/c$	Ins.	gap: 0.35 eV	zig-zag AFM	$T_N = 15$ K	[50–55]
α-Li_2IrO_3	honeycomb monoclinic	$C2/c$	Ins.	–	spiral AFM	$T_N = 15$ K	[56, 57]
β-Li_2IrO_3	hyperhoneycomb	$Fddd$	Ins.	–	unconventional AFM	$T_N = 38$ K	[58, 59]
γ-Li_2IrO_3	stripyhoneycomb	$Cccm$	Ins.	–	unconventional AFM	$T_N = 38$ K	[60]
Ba_2IrO_4	K_2NiF_4-type	$I4/mmm$	Ins.	gap: 0.14 eV	AFM	$T_N = 240$ K	[61–64]
Sr_2IrO_4	distorted K_2NiF_4-type	$I4_1/acd$	Ins.	gap 0.25 eV	canted AFM	$T_N = 240$ K	[15, 16, 21]
Ca_4IrO_6	hexagonal	$R\bar{3}c$	Ins.	–	AFM	$T_N = 13.95$ K	[65–67]
$Y_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 155$ K	[68, 69]
$Pr_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Metal		None		[70, 71]
$Nd_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	MIT	$T = 36$ K	AIAO	$T = 36$ K	[70, 72]
$Sm_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	MIT	$T = 117$ K	AIAO	$T = 117$ K	[70, 72]
$Eu_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	MIT	$T = 120$ K	AIAO	$T = 120$ K	[70, 73–75]
$Gd_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 127$ K	[70]
$Tb_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 130$ K	[70, 76]
$Dy_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 134$ K	[70]
$Ho_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 141$ K	[70]
$Er_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 140$ K	[76]
$Yb_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 130$ K	[77]
$Lu_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Ins.	–	AIAO	$T = 120$ K	[78]
$Bi_2Ir_2O_7$	pyrochlore	$Fd\bar{3}m$	Metal		None		[79, 80]
$Sr_3Ir_2O_7$	monoclinic	$C2/c$	Ins.	gap: 0.1 eV	AFM	$T_N = 285$ K	[46, 81–85]
$Na_4Ir_3O_8$	hyperkagome	$P4_132$	Ins.	–	AFM	$T_N = 6$ K	[86–88]
$Ca_5Ir_3O_{12}$	hexagonal	$P-62m$	Ins.	–	AFM	$T_N = 7.8$ K	[66, 67, 89]
La_2ZnIrO_6	double-perovskite	$P2_1/n$	Ins.	–	FM	$T_C = 7.5$ K	[90]
La_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	gap: 0.16 eV	AFM	$T_N = 12$ K	[90, 91]
Pr_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	gap: 0.2 eV	AFM	$T_N = 14$ K	[91, 92]
Nd_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	–	AFM	$T_N = 12$ K	[92]
Sm_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	–	AFM	$T_N = 15$ K	[92]
Eu_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	–	AFM	$T_N = 10$ K	[92]
Gd_2MgIrO_6	double-perovskite	$P2_1/n$	Ins.	–	None		[92]
Sr_2CeIrO_6	double perovskite	$P2_1/n$	Ins.	gap: 0.3 eV	AFM	$T_N = 21$ K	[93–95]
Ba_2YIrO_6	double perovskite	$Fm\bar{3}m$	Ins.	gap: 0.221 eV	None		[96]
$Ba_3IrTi_2O_9$	hexagonal	$P6_3mc$	Ins.	–	None		[97, 98]
$Ba_3ScIr_2O_9$	hexagonal	$P6_3/mmc$	Ins.	–	None		[99]
$Ba_3YIr_2O_9$	hexagonal	$P6_3/mmc$	Ins.	–	FM	$T = 4$ K	[99]
$Ba_3ZnIr_2O_9$	hexagonal	$P6_3/mmc$	Ins.	–	None		[100]

Table 1: Main structural, transport and magnetic properties of Ir-based spin-orbit materials. In the third column, Ins. refers to *insulator* and MIT to *metal-insulator transition*. The notations AFM, FM and AIAO refer to a *antiferromagnetic*, *ferromagnetic* and *all-in-all-out* magnetic ordering respectively.

$$\begin{aligned}
\left|j_{\text{eff}}=\frac{1}{2}, m_{j_{\text{eff}}}=+\frac{1}{2}\right\rangle &= +\frac{1}{\sqrt{3}}\Big(\left|d_{yz},\downarrow\right\rangle + i\left|d_{xz},\downarrow\right\rangle\Big) + \frac{1}{\sqrt{3}}\left|d_{xy},\uparrow\right\rangle \\
\left|j_{\text{eff}}=\frac{1}{2}, m_{j_{\text{eff}}}=-\frac{1}{2}\right\rangle &= +\frac{1}{\sqrt{3}}\Big(\left|d_{yz},\uparrow\right\rangle - i\left|d_{xz},\uparrow\right\rangle\Big) - \frac{1}{\sqrt{3}}\left|d_{xy},\downarrow\right\rangle
\end{aligned} \tag{1}$$

$$\begin{aligned}
\left|j_{\text{eff}}=\frac{3}{2}, m_{j_{\text{eff}}}=+\frac{1}{2}\right\rangle &= -\frac{1}{\sqrt{6}}\Big(\left|d_{yz},\downarrow\right\rangle + i\left|d_{xz},\downarrow\right\rangle\Big) + \sqrt{\frac{2}{3}}\left|d_{xy},\uparrow\right\rangle \\
\left|j_{\text{eff}}=\frac{3}{2}, m_{j_{\text{eff}}}=-\frac{1}{2}\right\rangle &= +\frac{1}{\sqrt{6}}\Big(\left|d_{yz},\uparrow\right\rangle - i\left|d_{xz},\uparrow\right\rangle\Big) + \sqrt{\frac{2}{3}}\left|d_{xy},\downarrow\right\rangle \\
\left|j_{\text{eff}}=\frac{3}{2}, m_{j_{\text{eff}}}=+\frac{3}{2}\right\rangle &= -\frac{1}{\sqrt{2}}\Big(\left|d_{yz},\uparrow\right\rangle + i\left|d_{xz},\uparrow\right\rangle\Big) \\
\left|j_{\text{eff}}=\frac{3}{2}, m_{j_{\text{eff}}}=-\frac{3}{2}\right\rangle &= +\frac{1}{\sqrt{2}}\Big(\left|d_{yz},\downarrow\right\rangle - i\left|d_{xz},\downarrow\right\rangle\Big)
\end{aligned} \tag{2}$$

Since the quartet of states lies lower in energy than the doublet and the splitting between the j_{eff}=3/2 and j_{eff}=1/2 is large, neglecting any band dispersion would result in a configuration

Compound	Crystal Struct.		Transport Property		Magnetic Ordering		Ref.
Ruthenium-based spin-orbit materials							
$BaRuO_3$	cubic perovskite	$Pm\bar{3}m$	Metal		FM	$Tc = 60\,K$	[101–103]
$CaRuO_3$	perovskite	$Pnma$	Metal		None		[104–106]
$SrRuO_3$	perovskite	$Pnma$	Metal		FM	$T_c = 160\,K$	[104, 106, 107]
Sr_2RuO_4	K_2NiF_4-type	$I4/mmm$	Metal		None		[108, 109]
Ca_2RuO_4	distorted K_2NiF_4-type	$Pbca$ or $P2_1/c$	MIT	$T = 357\,K$	AFM	$T = 110\,K$	[108, 110–112]
Sr_2RuO_6	hexagonal	$P\bar{3}1m$	Ins.	–	AFM	$T_N = 565\,K$	[113, 114]
$Sr_3Ru_2O_7$	orthorhombic	$BBcb$	Metal		None		[106, 115]
$Sr_4Ru_3O_{10}$	orthorhombic	$Pbam$	Metal		FM	$T_c = 105\,K$	[106, 116, 117]
Rhodium-based spin-orbit materials							
Li_2RhO_3	honeycomb	$C2/m$	Ins.	gap: 0.08 eV	None		[118, 119]
Sr_2RhO_4	distorted K_2NiF_4-type	$I4_1acd$	Metal		None		[21, 120, 121]
Sr_4RhO_6	hexagonal	$R\bar{3}c$	Ins.	gap: 0.1 eV	AFM	$T_N = -\,K$	[122]
$Sr_5Rh_4O_{12}$		$P3c1$	Ins.	–	AFM	$T_N = 23\,K$	[66, 123]
Osmium-based spin-orbit materials							
$BaOsO_3$	six-layer hexagonal 6H	$Pm\bar{3}m$	Metal		None		[124, 125]
$CaOsO_3$	perovskite	$Pnma$	Metal		None		[125]
$SrOsO_3$	perovskite	$Pnma$	Metal		None		[125]
$NaOsO_3$	perovskite	$Pnma$	MIT	$T = 410$ K	AFM	$T = 410$ K	[31–36]
$Cd_2Os_2O_7$	pyrochlore	$Fd\bar{3}m$	MIT	$T = 226$ K	AIAO	$T = 226$ K	[126–129]
Ba_2NaOsO_6	double-perovskite	$Fm\bar{3}m$	Ins.	–	FM	$T_c = 6.8\,K$	[130–132]
Ba_2LiOsO_6	double-perovskite	$Fm\bar{3}m$	Ins.	–	AFM	$T_N = 8\,K$	[131]
Ba_2CaOsO_6	double-perovskite	$Fm\bar{3}m$	Ins.	–	FM	$T_c = 50\,K$	[133, 134]
Ba_2YOsO_6	double-perovskite	$Fm\bar{3}m$	Ins.	–	AFM	$T_N = 69\,K$	[134, 135]

Table 2: Main structural, transport and magnetic properties of Ru,Rh and Os-based spin-orbit materials. In the third column, Ins. refers to *insulator* and MIT to *metal-insulator transition*. The notations AFM, FM and AIAO refer to a *antiferromagnetic*, *ferromagnetic* and *all-in-all-out* magnetic ordering respectively.

with one electron in the j_{eff}=1/2 state. The DFT band structure displays a dispersion of width comparable to this splitting, leaving the question *a priori* open again. However, the bandwidth is narrowed due to structural distortions [21], and electronic correlations can then become effective and eventually drive the compound insulating.

Since the discovery of this mechanism, other Ir-based compounds (cf. Tab. 1) have been classified as spin-orbit Mott insulators (Na_2IrO_3, pyrochlores, etc...). Recent theoretical studies predict also some fluoride material [147] to be in this class. The one-orbital nature of insulating Sr_2IrO_4 has contributed to intense activities attempting to dope the compound, with the hope of inducing a superconducting state as in the cuprates. Doping-induced metal-insulator transitions and the properties of the metallic phases have therefore become a hot topic, with studies of various compounds, e.g. Sr_2IrO_4[144, 148], $(Sr_{1-x}La_x)_3Ir_2O_7$ [149], $Ca_{1-x}Sr_xIrO_3$ [150], $Ca_{1-x}Ru_xIrO_3$ [151], $Sr_2Ir_{1-x}Rh_xO_4$ [152, 153], $Sr_2Ir_{1-x}Ru_xO_4$ [154], $Sr_xLa_{11-x}Ir_4O_{24}$ [155].

2.2 Correlated spin-orbit metals: the example of Sr_2RhO_4

It is natural that also in *metallic* 4d or 5d transition metal compounds, SOC can have notable consequences. An example of a “spin-orbit correlated metal” is the end member $SrIrO_3$ of the Ir-based Ruddlesden-Popper $Sr_{n+1}Ir_nO_{3n+1}$ series [46] but also many Ru-,Rh- or Os-based transition metal oxides (TMOs) belong to this class (cf. Tab. 1 and 2). In these compounds, correlations are important enough to renormalize the Fermi surface – albeit in a strongly spin-orbit coupling-dependent way. The respective roles of both effects have been worked out in some details for several compounds, among which $SrIrO_3$ [46–48], Sr_2RuO_4/Ca_2RuO_4 [108, 109, 156] and Sr_2RhO_4[19–21].

We will focus our attention in the following on Sr_2RhO_4 motivated by its structural proximity and isoelectronic nature to Sr_2IrO_4 . Indeed, this TMO is the 4d counterpart of Sr_2IrO_4, both concerning structure and filling. Understanding its Fermi surface requires to include both SOC and correlations [21]. It is composed of three pockets (cf. Fig. 8): a circular hole-like α-pocket around Γ, a lens-shaped electron pocket β_M and a square-shaped electron pocket β_X with a mass enhancement of 3.0, 2.6 and 2.2 respectively [120].

In this review, we will put Sr_2IrO_4 and Sr_2RhO_4 in parallel, shedding light on the spectral properties of these compounds and elaborating on the notion of a reduced effective (spin-orbital) degeneracy that is crucial for their properties.

2.3 Spin-orbit coupling and cubic symmetry: the $\mathbf{j_{eff}}$ picture

Necessary conditions for realising a j_{eff} picture are (1) a strong spin-orbit coupling constant and (2) an important cubic crystal field. These conditions are often met in crystalline structures where IrO_6 octahedra are present (cf. Tab. 1). Similar compounds based on Ru, Rh and Os also show such j_{eff} states (cf. Tab 2). However, not all Ir-based structures belong to this case : we note that neither epitaxial thin films of IrO_2 [157] nor the correlated metal IrO_2 in its rutile structure [158, 159] exhibit such j_{eff}=1/2 state. We will now turn to a more precise description of that picture.

The spin-orbit interaction is one of the relativistic corrections to the Schrödinger-Pauli equation arising when taking the non-relativistic limit of Dirac's equation. It introduces a coupling between the spin $\mathbf{S}$ and the motion – or more precisely the orbital momentum $\mathbf{L}$ in the atomic case – of the electron. In a solid described within an independent-particle picture, spin-orbit coupling has the following general form:

$$H_{SO} = \frac{\hbar}{4m_0^2c^2}\boldsymbol{\sigma}\cdot[\boldsymbol{\nabla}V(\mathbf{r})\times\mathbf{p}] \tag{3}$$

where m_0 is the electron mass, $V(\mathbf{r})$ is the effective Kohn-Sham potential and $\sigma_{i=x,y,z}$ denote the Pauli-spin matrices. Assuming that the potential close to the nucleus has spherical symmetry the mean value of the spin-orbit interaction on the atomic state (n,ℓ) takes the more common form:

$$H_{SO} = \zeta_{SO}(n\ell)\ \mathbf{l}\cdot\mathbf{s} \qquad \text{with} \qquad \zeta_{SO}(n\ell) = \frac{\hbar^2}{2m_0^2c^2}\left\langle\frac{1}{r}\frac{dV}{dr}\right\rangle_{(n,\ell)} \tag{4}$$

where $\mathbf{S}=\frac{1}{2}\boldsymbol{\sigma}$, $\mathbf{L}=\mathbf{r}\times\mathbf{p}=\hbar\,\mathbf{l}$ and $\langle\ldots\rangle_{(n,\ell)}$ denotes the mean value of the radial quantity in the state (n,ℓ). Tab. 3 gives some values of the spin-orbit constant ζ_{SO} for $3d$, $4d$ and $5d$ atoms. The SOC increases with the atomic number, explaining why spin-orbit materials are mostly found in $5d$ and $4d$ TMOs.

Due to the effect of SOC, a multiplet splitting arises in the d-orbitals. Fig. 2 shows the multiplet splitting of d-orbitals due to the spin-orbit coupling as a function of the strength of a cubic crystal field $\Delta = 10Dq$.

In spherical symmetry the fine structure is composed of a six-fold $J = 5/2$ multiplet (in red) and a $J = 3/2$ quartet of lower energy (in blue), following "Landé's interval rule". The presence

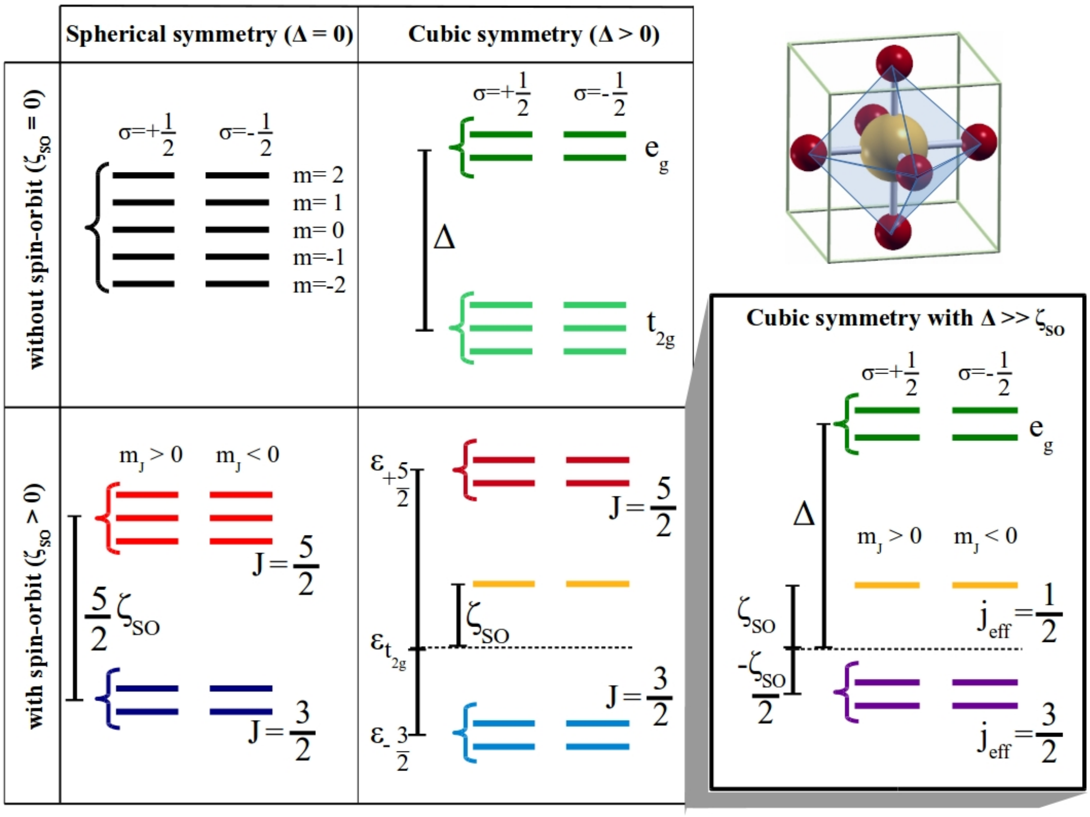

Figure 2: Orbital diagrams for the d-shell of an atom as a function of a cubic crystal field Δ and spin-orbit coupling ζ_{S0}, paramagnetic case. Starting from the d-shell in spherical symmetry, the cubic crystal field splits them into e_g and t_{2g}, while the SOC creates a 6-fold $J = 5/2$ multiplet and a $J = 3/2$ quartet of lower energy. When both parameters are at stake, one gets a new multiplet structure where J remains a good quantum number but not J_z. The initial $J = 5/2$ multiplet splits into a quartet and a doublet of lower energy, while the quartet $J = 3/2$ undergoes some redefinition inside its submanifold. The energetic splitting and the nature of the spin-orbitals depend on the ratio between Δ/ζ_{SO}. An exception is the doublet which is already of the form of the j_{eff}=1/2 . In the limit where $\Delta >> \zeta_{SO}$, as is the case in the compounds of our interest, one gets the celebrated splitting into e_g, j_{eff}=1/2 and j_{eff}=3/2 .

Atom	Z	$\zeta_{SO}(3d)$	Atom	Z	$\zeta_{SO}(4d)$	Atom	Z	$\zeta_{SO}(5d)$
Fe	26	0.050 eV	Ru	44	0.161 eV	Os	76	0.31 eV
Co	27	0.061 eV	Rh	45	0.191 eV	Ir	77	0.4 eV
Cu	29	0.103 eV	Ag	47	0.227 eV	Au	79	0.42 eV

Table 3: Value of the spin-orbit constant ζ_{SO} in the d-valence shells of some transition metals. Data from Landolt-Börnstein database and [160] (3d), from [19, 161] (4d) and from [162] (5d)

of a cubic crystal field splits further the six-fold multiplet. Indeed, the spin-orbit interaction in the cubic basis (e_g and t_{2g} in green and light green respectively in Fig. 2) can be reduced to two five-dimensional submatrices:

$$\left(\begin{array}{ccc|cc} 0 & -i & i & \sqrt{3} & -1 \\ i & 0 & -1 & -i\sqrt{3} & -i \\ -i & -1 & 0 & 0 & -2i \\ \hline \sqrt{3} & i\sqrt{3} & 0 & 0 & 0 \\ -1 & i & 2i & 0 & 0 \end{array}\right) \cdot \frac{\zeta_{SO}}{2} \quad \text{and} \quad \left(\begin{array}{ccc|cc} 0 & i & i & -\sqrt{3} & 1 \\ -i & 0 & 1 & -i\sqrt{3} & -i \\ -i & 1 & 0 & 0 & 2i \\ \hline -\sqrt{3} & i\sqrt{3} & 0 & 0 & 0 \\ 1 & i & -2i & 0 & 0 \end{array}\right) \cdot \frac{\zeta_{SO}}{2} \tag{5}$$

in the bases $\{d_{xz}\uparrow, d_{yz}\uparrow, d_{xy}\downarrow, d_{3z^2-r^2}\downarrow, d_{x^2-y^2}\downarrow\}$ and $\{d_{xz}\downarrow, d_{yz}\downarrow, d_{xy}\uparrow, d_{3z^2-r^2}\uparrow, d_{x^2-y^2}\uparrow\}$ respectively. After diagonalization, the total angular momentum J remains a good quantum number, contrary to j_z/m_j and one gets the following fine structure:

- a first quartet of $J = 5/2$ states (in red) with an energy

$$\varepsilon_{\frac{5}{2}+} = \frac{1}{4}(2\Delta - \zeta_{SO}) + \frac{1}{4}\sqrt{(2\Delta + \zeta_{SO})^2 + 24{\zeta_{SO}}^2}$$

- a doublet of $J = 5/2$ states (in yellow) of energy

$$\varepsilon_{\frac{5}{2}-} = +2\frac{\zeta_{SO}}{2}$$

- a quartet of $J = 3/2$ states (in light blue) with an energy

$$\varepsilon_{\frac{3}{2}} = \frac{1}{4}(2\Delta - \zeta_{SO}) - \frac{1}{4}\sqrt{(2\Delta + \zeta_{SO})^2 + 24{\zeta_{SO}}^2}$$

In the limit of strong crystal field ($\Delta >> \zeta_{SO}$), the $J = 5/2$ doublet (in yellow) remains invariant while the higher-energy quartet will tend to the usual e_g states and the lower-energy $J = 3/2$ quartet will be composed of t_{2g} states only, with an energy of $-\zeta_{SO}/2$.

Since the SOC-matrix restricted to the t_{2g} subspace is exactly the opposite of the SOC-matrix of the p-states of a free atom, one usually labels these latter states by a j_{eff} quantum number in analogy with the $p_{\frac{1}{2}}$ and $p_{\frac{3}{2}}$ multiplets, leading to the expressions given in Eq. (1) and (2). We point out that the j_{eff}=1/2 doublet arises from the interplay of both cubic symmetry and SOC, whatever the strength of the crystal field. The corresponding eigenstates can indeed be written:

$$\begin{aligned} \left|\frac{1}{2}, +\frac{1}{2}\right\rangle &= +\frac{1}{\sqrt{3}}\Big(\left|d_{yz},\downarrow\right\rangle + i\left|d_{xz},\downarrow\right\rangle\Big) + \frac{1}{\sqrt{3}}\left|d_{xy},\uparrow\right\rangle = \frac{i}{\sqrt{6}}\left(\sqrt{5}\left|\frac{5}{2}, -\frac{3}{2}\right\rangle - \left|\frac{5}{2}, \frac{5}{2}\right\rangle\right) \\ \left|\frac{1}{2}, -\frac{1}{2}\right\rangle &= +\frac{1}{\sqrt{3}}\Big(\left|d_{yz},\uparrow\right\rangle - i\left|d_{xz},\uparrow\right\rangle\Big) - \frac{1}{\sqrt{3}}\left|d_{xy},\downarrow\right\rangle = \frac{i}{\sqrt{6}}\left(\sqrt{5}\left|\frac{5}{2}, \frac{3}{2}\right\rangle - \left|\frac{5}{2}, -\frac{5}{2}\right\rangle\right) \end{aligned}$$

(where the right-hand side is written using the J, m_J quantum numbers). This may explain the robustness of this doublet in spin-orbit compounds [163]. However, the splitting between the $j_{\rm eff}$=1/2 and $j_{\rm eff}$=3/2 multiplets follows the inverse Landé interval rule (with the $j_{\rm eff}$=1/2 above the $j_{\rm eff}$=3/2 states) only in the strong crystal field limit.

3 Interplay of spin-orbit interaction and Coulomb correlations from first principles

3.1 DFT+DMFT calculations with spin-orbit coupling

Combined density functional theory (DFT) and dynamical mean-field theory (DMFT), as pioneered in [164, 165] (for a review, see [166, 167]), has made correlated electron systems accessible to first principles calculations. Over the years, various classes of systems ranging from transition metals [168–171], their oxides [11, 172–177], sulphides [178, 179], pnictides [4, 9, 180, 181], rare earths [182–184] and their compounds [185–188], including heavy fermions [189, 190], actinides [191, 192] and their compounds [193, 194] to organics [195], correlated semiconductors [196, 197], and correlated surfaces and interfaces [198–200] have been studied with great success. Besides intensive methodological developments (see e.g. [2, 3, 201–205]), recent research activities continue to extend to new materials classes. In this context, also 4d and 5d oxides have come into focus [21, 22, 62]. In this section, we review the technical aspects related to combined DFT+DMFT calculations in the presence of spin-orbit interactions. Since the applications we later focus on are 4d and 5d oxides in their *paramagnetic* phases, we restrict the discussion to this case.

In DMFT, a local approximation is made to the many-body self-energy which can then be calculated from an effective atom problem, subject to a self-consistency condition (see Fig. 3).

The notion of locality is understood in the sense of many-body theory as a site-diagonal form, with respect to atomic sites after representing the Hamiltonian in an atom-centered Wannier-type basis $|w^{\alpha,\sigma}_{\ell m}\rangle$, where the index α labels the atom in the unit-cell, (ℓ, m) the angular momentum quantum numbers of the atomic orbital and σ the spin degree of freedom. Different choices are possible for the construction of the atom-centered orbitals, and the work reviewed here is based on the construction of projected atomic orbitals subject to a subsequent orthonormalisation procedure [180].

The DMFT self-consistency cycle links the local effective atom problem to the electronic structure of the solid, via the transformation matrix from the Kohn-Sham states $|\psi^{\sigma}_{\mathbf{k}\nu}\rangle$, labelled by their momentum $\mathbf{k}$ their band index ν and their spin σ, to the resulting Wannier-like local orbitals $|w^{\alpha,\sigma}_{\ell m}\rangle$. These key quantities are called *projectors* and denoted $P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k})$.

The main advantage of projector-based implementations of DFT+DMFT (see e.g. [180, 207, 208]) is that not only the DFT-based part of the calculations but also the determination of the local Green's function, used within the DMFT self-consistency condition, can be performed in any convenient basis set, and notably in the one used in the respective DFT code. Since the transformation of the DFT Hamiltonian matrix in that basis into the Kohn-Sham eigenset $|\psi^{\sigma}_{\mathbf{k}\nu}\rangle$ is known, it is sufficient to further determine the projections of the Kohn-Sham eigenstates onto

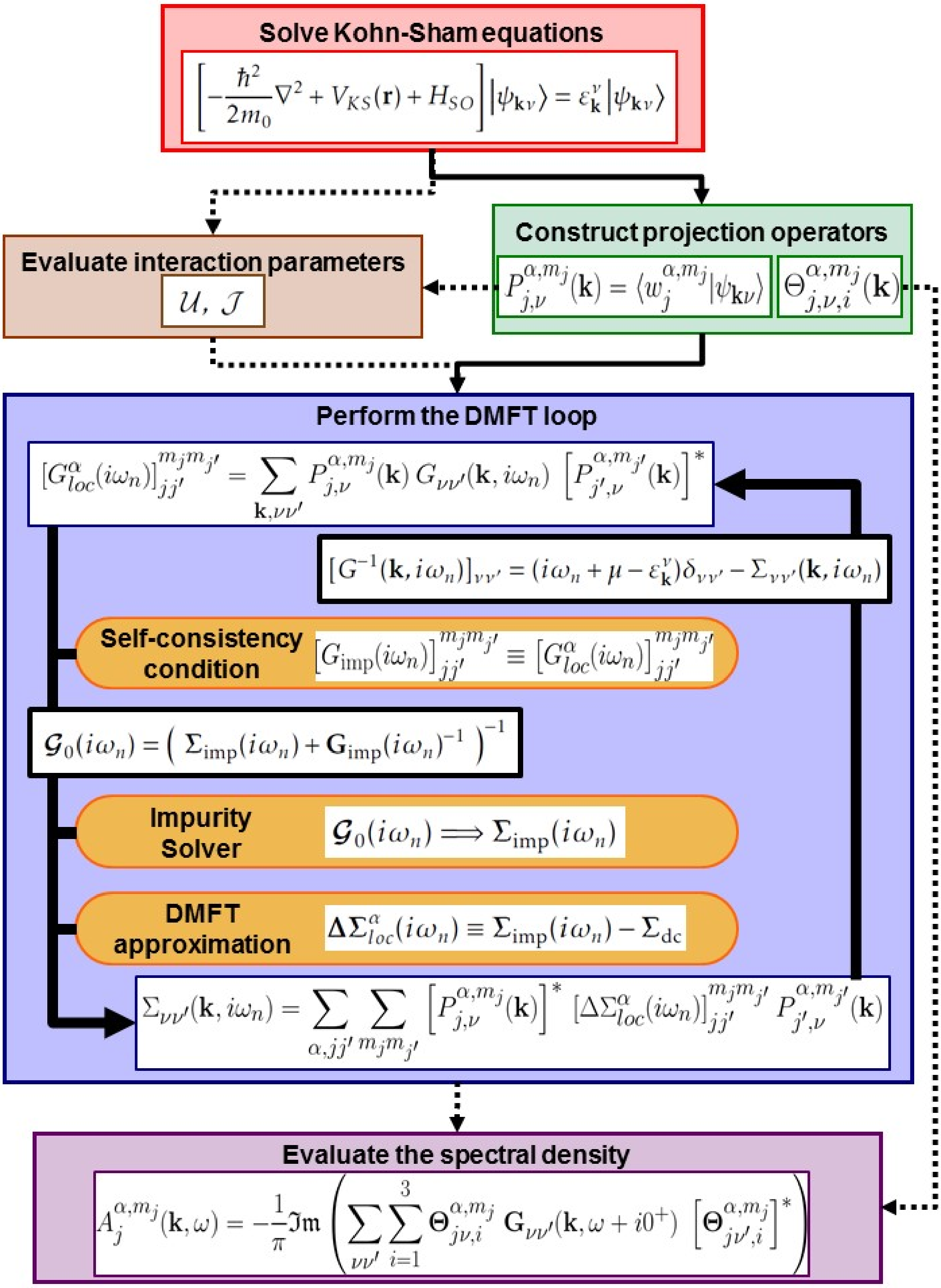

Figure 3: Projector-based implementation of DFT+DMFT for calculations including spin-orbit coupling in the Kohn-Sham equations. Once the Kohn-Sham eigenstates $|\psi_{\mathbf{k}\nu}\rangle$ are known, their projections $P^{\alpha,m_j}_{j,\nu}(\mathbf{k})$ to the correlated Wannier-like orbitals $|w^{\alpha,m_j}_j\rangle$ are calculated. One can then build an effective local many-body atomic problem, subject to a self-consistency condition, which is solved using an impurity solver: this defines the DMFT loop (see Section 3.1). The interaction parameters can also be evaluated consistently using the projectors $P^{\alpha,m_j}_{j,\nu}(\mathbf{k})$ (see [206] and Section 3.3). After convergence of the DMFT cycle, the chemical potential is updated and the spectral function can be evaluated using partial projectors $\Theta^{\alpha,m_j}_{j\nu,i}(\mathbf{k})$ (see Appendix A).

the local orbitals $|w^{\alpha,\sigma}_{\ell m}\rangle$ used in the DMFT impurity problem. This is precisely the role of the projectors.

In [21], this construction was generalised to the case when spin is not a good quantum number any more, and implemented within the framework of the DFT+DMFT implementation of Ref. [180]. Nowadays, it is available within the TRIQS/DFTTools package [209] that links the Wien2k code [210] to DMFT. We give here the main lines of this generalisation of the projector-based DFT+DMFT formalism.

When taking into account SOC, the Kohn-Sham eigenstates $|\psi_{\mathbf{k}\nu}\rangle$ are built out of *both* spin-up and spin-down states – in a similar fashion as the previously introduced j_{eff}=1/2 and j_{eff}=3/2 atomic states. Nevertheless, we can still write them in the following Bloch form:

$$\begin{aligned}\psi_{\mathbf{k}\nu}(\mathbf{r}) &= \left[u^{\uparrow}_{\mathbf{k}\nu}(\mathbf{r}) + u^{\downarrow}_{\mathbf{k}\nu}(\mathbf{r})\right] e^{i\mathbf{k}\cdot\mathbf{r}} \\ &= \phi^{\uparrow}_{\mathbf{k}\nu}(\mathbf{r}) + \phi^{\downarrow}_{\mathbf{k}\nu}(\mathbf{r}). \qquad (6)\end{aligned}$$

where the index ν now runs over *both* spin and band indices. The state $|\phi^{\sigma}_{\mathbf{k}\nu}\rangle$ denotes the projection of the Kohn-Sham state onto its spin-σ contribution and is *not* an eigenstate of the Hamiltonian.

Using this decomposition, we can define the new projectors:

$$P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k}) = \langle w^{\alpha,\sigma}_{\ell m}|\psi_{\mathbf{k}\nu}\rangle = \langle w^{\alpha,\sigma}_{\ell m}|\phi^{\sigma}_{\mathbf{k}\nu}\rangle \qquad (7)$$

We define them in the standard complex basis, but allow for a basis transformation to quantum numbers j, m_j (like j_{eff}=1/2 and j_{eff}=3/2) afterwards by means of a unitary matrix transformation in the correlated ℓ-space:

$$P^{\alpha,m_j}_{j,\nu}(\mathbf{k}) = \sum_{m,\sigma} \mathcal{S}^{m_j,\sigma}_{j,\ell m}\langle w^{\alpha,\sigma}_{\ell m}|\psi_{\mathbf{k}\nu}\rangle = \sum_{m,\sigma} \mathcal{S}^{m_j,\sigma}_{j,\ell m} P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k}) \qquad (8)$$

The main difference with the usual implementation where spin is a good quantum number is that there are now two projectors associated to each band index ν: $P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k})$ with $\sigma =\uparrow,\downarrow$.

Using the decomposition (6) in the formulation of the self-consistency condition relating the lattice Green's function of the solid to the impurity model, the (inverse) Green's function of the solid is given by:

$$[G^{-1}(\mathbf{k}, i\omega_n)]_{\nu\nu'} = (i\omega_n + \mu - \varepsilon^{\nu}_{\mathbf{k}})\delta_{\nu\nu'} - \Sigma_{\nu\nu'}(\mathbf{k}, i\omega_n), \qquad (9)$$

where $\varepsilon^{\nu}_{\mathbf{k}}$ are the (ν-dependent only) Kohn-Sham eigenvalues and $\Sigma_{\nu\nu'}(\mathbf{k}, i\omega_n)$ is the approximation to the self-energy obtained by the solution of the DMFT impurity problem. It is obtained by "mapping" the impurity self-energy to the local self-energy of the lattice and "upfolding" it as:

$$\Sigma_{\nu\nu'}(\mathbf{k}, i\omega_n) = \sum_{\alpha,jj'}\sum_{m_j m_{j'}} \left[P^{\alpha,m_j}_{j,\nu}(\mathbf{k})\right]^* \ [\Delta\Sigma^{\alpha}_{loc}(i\omega_n)]^{m_j m_{j'}}_{jj'} \ P^{\alpha,m_{j'}}_{j',\nu'}(\mathbf{k}). \qquad (10)$$

with

$$[\Delta\Sigma^{\alpha}_{loc}(i\omega_n)]^{m_j m_{j'}}_{jj'} = [\Sigma_{\text{imp}}(i\omega_n)]^{m_j m_{j'}}_{jj'} - [\Sigma_{dc}]^{m_j m_{j'}}_{jj'} \qquad (11)$$

Here, $\Sigma_{\text{imp}}(i\omega_n)$ is the impurity self-energy, expressed in the local orbitals, and Σ_{dc} is the double-counting correction. Consequently, the equations of the DMFT loop (see Figure 3) are formally the same as in the case without SOC but the computations now involve matrices which are double in size.

The local Green's function is obtained by projecting the lattice Green's function to the set of correlated orbitals and summing over the full Brillouin zone,

$$[G^{\alpha}_{loc}(i\omega_n)]^{m_j m_{j'}}_{jj'} = \sum_{\mathbf{k},\nu\nu'} P^{\alpha,m_j}_{j,\nu}(\mathbf{k})\ G_{\nu\nu'}(\mathbf{k},i\omega_n)\ \left[P^{\alpha,m_{j'}}_{j',\nu'}(\mathbf{k})\right]^{*}. \tag{12}$$

In practice, the summation over momenta is done in the irreducible Brillouin zone only, supplemented by a standard symmetrization procedure, using Shubnikov magnetic point groups [211, 212].

The DMFT equations are solved iteratively: starting from an initial local Green's function $G^{\alpha}_{loc}(i\omega_n)$ (obtained from the "pure" Kohn-Sham lattice Green's function using Eq. (12)), the Green's function $\mathcal{G}_0(i\omega_n)$ of the effective environment in the impurity model is constructed. The impurity model is solved, allowing to evaluate the local self-energy of the solid (cf. Eq. 10) and a new lattice Green's function $G(\mathbf{k},i\omega_n)$. The latter can then be projected again onto the correlated subset and the cycle is repeated until convergence is reached.

3.2 Computation of the Wannier projectors within the augmented plane wave framework

The present implementation is within a full-potential linearized augmented plane wave (FLAPW) framework, as realised in the Wien2k package [210]. With respect to the existing DFT+DMFT implementation [180] in this context, the main changes concern the projection technique for building the correlated orbitals: as discussed above, one has to take care of the fact that spin is no longer a good quantum number, leading to the more general construction of localized "spin-orbitals". The necessary modifications in the construction of the projectors are reviewed in the following.

As in the case without SOC, we still use the Kohn-Sham states within a chosen energy window $\mathcal{W}$ to form the Wannier-like functions that are treated as correlated orbitals, and the construction of the Wannier projectors is done in two steps. First, auxiliary Wannier projectors $\widetilde{P}^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k})$ are calculated – separately for each $|\phi^{\sigma}_{\nu\mathbf{k}}\rangle$ term – from the following expression:

$$\begin{aligned}\widetilde{P}^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k}) &= \langle u^{\alpha,\sigma}_{\ell}(E_1\ell)Y^{\ell}_{m}|\psi_{\mathbf{k}\nu}\rangle \\ &= A^{\nu\alpha}_{\ell m}(\mathbf{k},\sigma) + \sum_{n_{LO}=1}^{N_{LO}} c^{\nu,\sigma}_{LO} C^{\alpha,LO}_{\ell m}\mathcal{O}^{\alpha,\sigma}_{\ell m,\ell' m'}.\end{aligned} \tag{13}$$

A description of the augmented plane wave (APW) basis can be found in Ref. [180]. We use the same notations e.g. for the coefficients $A^{\nu\alpha}_{\ell m}(\mathbf{k},\sigma)$ and the overlap matrix $\mathcal{O}^{\alpha,\sigma}_{\ell m,\ell' m'}$ as introduced there.

One performs an orthonormalisation step in order to get the Wannier projectors $P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k})$. The overlap matrix $[O(\mathbf{k})]^{\alpha,\alpha'}_{(m\sigma),(m'\sigma')}$ between the correlated ℓ orbitals is defined by:

$$[O(\mathbf{k})]^{\alpha,\alpha'}_{(m\sigma),(m'\sigma')} = \sum_{\nu=\nu_{min}(\mathbf{k})}^{\nu_{max}(\mathbf{k})} \widetilde{P}^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k}) \widetilde{P}^{\alpha',\sigma'*}_{\ell m',\nu}(\mathbf{k}). \tag{14}$$

leading to the final projectors:

$$P^{\alpha,\sigma}_{\ell m,\nu}(\mathbf{k}) = \sum_{\alpha',m',\sigma'} \left\{[O(\mathbf{k})]^{-1/2}\right\}^{\alpha,\alpha'}_{(m\sigma),(m'\sigma')} \widetilde{P}^{\alpha',\sigma'}_{\ell m'\nu}(\mathbf{k}), \tag{15}$$

which are then further transformed into a j, m_j basis as described above (cf. Eq. (8)).

3.3 Effective local Coulomb interactions from first principles

Hubbard interactions U – obtained as the static ($\omega = 0$) limit of the onsite matrix element $\langle|W^{\text{partial}}|\rangle$ within the "constrained random phase approximation" (cRPA) – have by now been obtained for a variety of systems, ranging from transition metals [213] to oxides [206, 214–217], pnictides [181, 218–220], f-electron elements [221] and compounds [188], to surface systems [222], and several implementations within different electronic structure codes and basis sets have been done, e.g. within linearized muffin tin orbitals [213, 223], maximally localized Wannier functions [214, 219, 224] (as elaborated in [225]), or localised orbitals constructed from projected atomic orbitals [206]. The implementation into the framework of the Wien2k package [206] made it possible that Hubbard U's be calculated for the same orbitals as the ones used in subsequent DFT+DMFT calculations, and, to our knowledge, Ref. [21] was indeed the first work using in this way consistently calculated Hubbard interactions in a DFT+DMFT calculation. Systematic calculations investigating the basis set dependence for a series of correlated transition metal oxides revealed furthermore interesting trends, depending on the choice of the low-energy subspace. In contrast to common belief until then, Hubbard interactions increase for example with the principal quantum number when low-energy effective models encompassing only the t_{2g} orbitals are employed. These trends can be rationalised by two counteracting mechanisms, the increasing extension of the orbitals with increasing principal quantum number and the less efficient screening by oxygen states [206]. We will come back to this point below, in the context of the cRPA calculations for our target compounds.

In the following, we review the specificities involved when determining the Hubbard interactions for our target spin-orbit compounds. We hereby use the same notations as in [206].

We start from the standard Hubbard-Kanamori Hamiltonian H_{int} which allows us to describe the interactions between t_{2g} orbitals within a Hamiltonian restricted to the t_{2g} -space:

$$\begin{aligned} H_{\text{int}} &= \mathcal{U}\sum_{m} n_{m\uparrow}n_{m\downarrow} + \mathcal{U}' \sum_{m<n,\sigma} n_{m\sigma}n_{n\bar{\sigma}} \\ &+ (\mathcal{U}' - \mathcal{J}) \sum_{m<n,\sigma} n_{m\sigma}n_{n\sigma} \\ &- \mathcal{J} \sum_{m<n,\sigma} \left[c^{\dagger}_{m\sigma}c_{m\bar{\sigma}}c^{\dagger}_{n\bar{\sigma}}c_{n\sigma} + c^{\dagger}_{m\sigma}c^{\dagger}_{m\bar{\sigma}}c_{n\sigma}c_{n\bar{\sigma}}\right] \end{aligned} \tag{16}$$

where $\mathcal{U}$ is the intra-orbital Coulomb repulsion term and $\mathcal{U}'$ ($= \mathcal{U} - 2\mathcal{J}$ with cubic symmetry) the interorbital Coulomb interaction which is reduced by Hund's exchange $\mathcal{J}$. (m and n run over the three t_{2g} orbitals and σ stands for the spin).

To draw the link between the cRPA calculations and this model Hamiltonian, the terms $\mathcal{U}$, $\mathcal{U}'$ and $\mathcal{J}$ are understood as the Slater-symmetrized effective interactions in the t_{2g} subspace, related to the Slater integrals F^0, F^2 and F^4 as:

$$\mathcal{U} = F^0 + \frac{4}{49}(F^2 + F^4) \quad \text{and} \quad \mathcal{J} = \frac{3}{49}F^2 + \frac{20}{441}F^4 \tag{17}$$

The last relation $\mathcal{U}' = F^0 - \frac{2}{49}F^2 - \frac{4}{441}F^4$ is redundant since $\mathcal{U}' = \mathcal{U} - 2\mathcal{J}$.

One now transforms H_{int} into the j_{eff} basis using the unitary matrix transformation $\mathcal{S}^{m_j,\sigma}_{j,lm}$. Keeping only density-density terms, H_{int} becomes:

$$H_{\text{int}} = \frac{1}{2}\sum_{j,m_j}\sum_{j',m_{j'}} U^{m_j m_{j'}}_{jj'} n_{j,m_j} n_{j,m_{j'}} \tag{18}$$

Here, the index j is a shortcut notation for the $j_{\text{eff}} = \{3/2, 1/2\}$ quantum number and $m_j = \{\pm 3/2, \pm 1/2\}$. The reduced interaction matrix $U^{m_j m_{j'}}_{jj'}$ has the following form:

$$U^{m_j m_{j'}}_{jj'} = U^{\overline{m}_j \overline{m}_{j'}}_{jj'} = \left(\begin{array}{c|cc} 0 & \mathcal{U} - 2\mathcal{J} & \mathcal{U} - \frac{5}{3}\mathcal{J} \\ \hline \mathcal{U} - 2\mathcal{J} & 0 & \mathcal{U} - \frac{7}{3}\mathcal{J} \\ \mathcal{U} - \frac{5}{3}\mathcal{J} & \mathcal{U} - \frac{7}{3}\mathcal{J} & 0 \end{array}\right) \tag{19}$$

$$U^{m_j \overline{m}_{j'}}_{jj'} = U^{\overline{m}_j m_{j'}}_{jj'} = \left(\begin{array}{c|cc} \mathcal{U} - \frac{4}{3}\mathcal{J} & \mathcal{U} - \frac{8}{3}\mathcal{J} & \mathcal{U} - \frac{8}{3}\mathcal{J} \\ \hline \mathcal{U} - \frac{8}{3}\mathcal{J} & \mathcal{U} - \mathcal{J} & \mathcal{U} - \frac{7}{3}\mathcal{J} \\ \mathcal{U} - \frac{8}{3}\mathcal{J} & \mathcal{U} - \frac{7}{3}\mathcal{J} & \mathcal{U} - \mathcal{J} \end{array}\right) \tag{20}$$

We use the standard convention that $\overline{m}_j$ denotes $-m_j$, as usually done for spin degree of freedom. The ordering of the orbitals $|j, |m_j|\rangle$ is: $|1/2, 1/2\rangle, |3/2, 1/2\rangle, |3/2, 3/2\rangle$, j_{eff}=1/2 and j_{eff}=3/2 blocks are emphasized to ease the reading of the matrices.

3.4 Technicalities of the DMFT calculation

For the solution of the quantum impurity problem we apply the continuous-time quantum Monte Carlo method (CTQMC) in the strong-coupling formulation [226]. We are able to perform calculations at room temperature ($\beta = 1/k_B T = 40$ eV^{-1}) with reasonable numerical effort. In our calculations, we use typically around 16×10^6 Monte Carlo sweeps and 28 $\mathbf{k}$-points in the irreducible Brillouin zone.

Since the CTQMC solver computes the Green's function on the imaginary-time axis, an analytic continuation is needed in order to obtain results on the real-frequency axis. A continuation of the impurity self-energy using a stochastic version of the maximum entropy method [227] yields real and imaginary parts of the retarded self-energy. From those, we calculate the momentum-resolved spectral function $A(\mathbf{k}, \omega)$ using partial projectors introduced in Appendix A.

During the calculations we use the Fully Localized Limit (FLL) expression for the double-counting:

$$\Sigma^{dc}_{j,j'} = \left[U(N_c - \frac{1}{2}) - J(\frac{1}{2}N_c - \frac{1}{2})\right]\delta_{jj'} \tag{21}$$

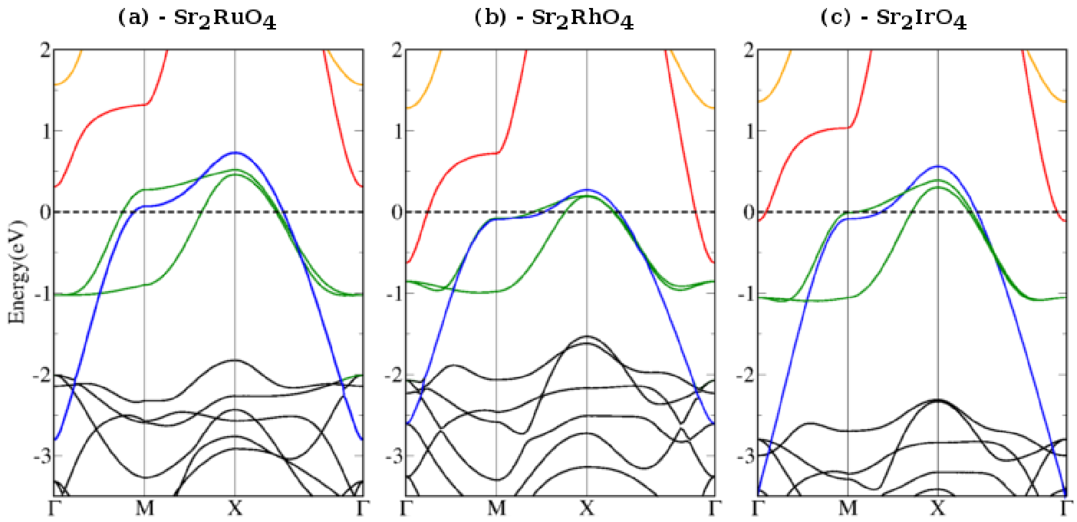

Figure 4: Kohn-Sham band structures of Sr_2RuO_4 (a) Sr_2RhO_4 (b) and Sr_2IrO_4 (c) within LDA (and without spin-orbit coupling), artificially assuming that both Sr_2RhO_4 and Sr_2IrO_4 crystallize in the same K_2NiF_4 structure as their Ru-counterpart. For Sr_2RuO_4, we use the lattice parameters at 300 K given in [230]. The t_{2g}-dominated bands are plotted in green (d_{xy}) and blue (d_{xz} and d_{yz}) while the e_g bands are in red ($d_{x^2-y^2}$) and yellow ($d_{3z^2-r^2}$), the O-2p states in black.

where j and j' run over the j_{eff} states and N_c is the total occupancy of the orbitals. (Since each orbital is doubly degenerate in m_j, $N_c/2$ is used in the term containing J). Moreover, we neglect the off-diagonal terms in the local Green's functions (particularly, we neglect the term between the j_{eff}=1/2 and the j_{eff}=3/2 $|m_j| = 1/2$ which we checked to be two orders of magnitude smaller than the diagonal terms, in the chosen basis).

4 Electronic structure of Sr_2IrO_4 and Sr_2RhO_4

4.1 Electronic structure of Sr_2IrO_4 and Sr_2RhO_4 within DFT-LDA

The Kohn-Sham band structures of Sr_2IrO_4 and Sr_2RhO_4 within the local density approximation and in the presence of spin-orbit coupling (LDA+SO) are represented in Fig. 5-(d) and (e). For Sr_2IrO_4, we use the lattice parameters measured at 295 K in [228], and for Sr_2RhO_4 those measured at 300 K in [229].

The LDA+SO band structures for Sr_2IrO_4 and Sr_2RhO_4 are very similar, as a consequence of both, the structural similarity and the key role of spin-orbit coupling in these compounds. The e_g-states ($d_{x^2-y^2}$ in red and $d_{3z^2-r^2}$ in yellow) start at about 1 to 1.5 eV, and are fully separated from the t_{2g}-manifold which lies around the Fermi level and overlaps at lower energies with the oxygen 2p-states (black). Given the t_{2g}^5 filling and the four-atom unit cell of both compounds, a metallic solution is obtained within LDA for both Sr_2RhO_4 and Sr_2IrO_4 – at variance with experiments for Sr_2IrO_4. Among the t_{2g}-manifold (in green), only the four highest-lying bands, highlighted in blue, cross the Fermi level: this is suggestive of the existence of a separated half-filled j_{eff}=1/2 -derived band, which – within a four-atom unit cell – corresponds to a quartet of bands at each k-point. We stress however that the true picture is much more subtle: in

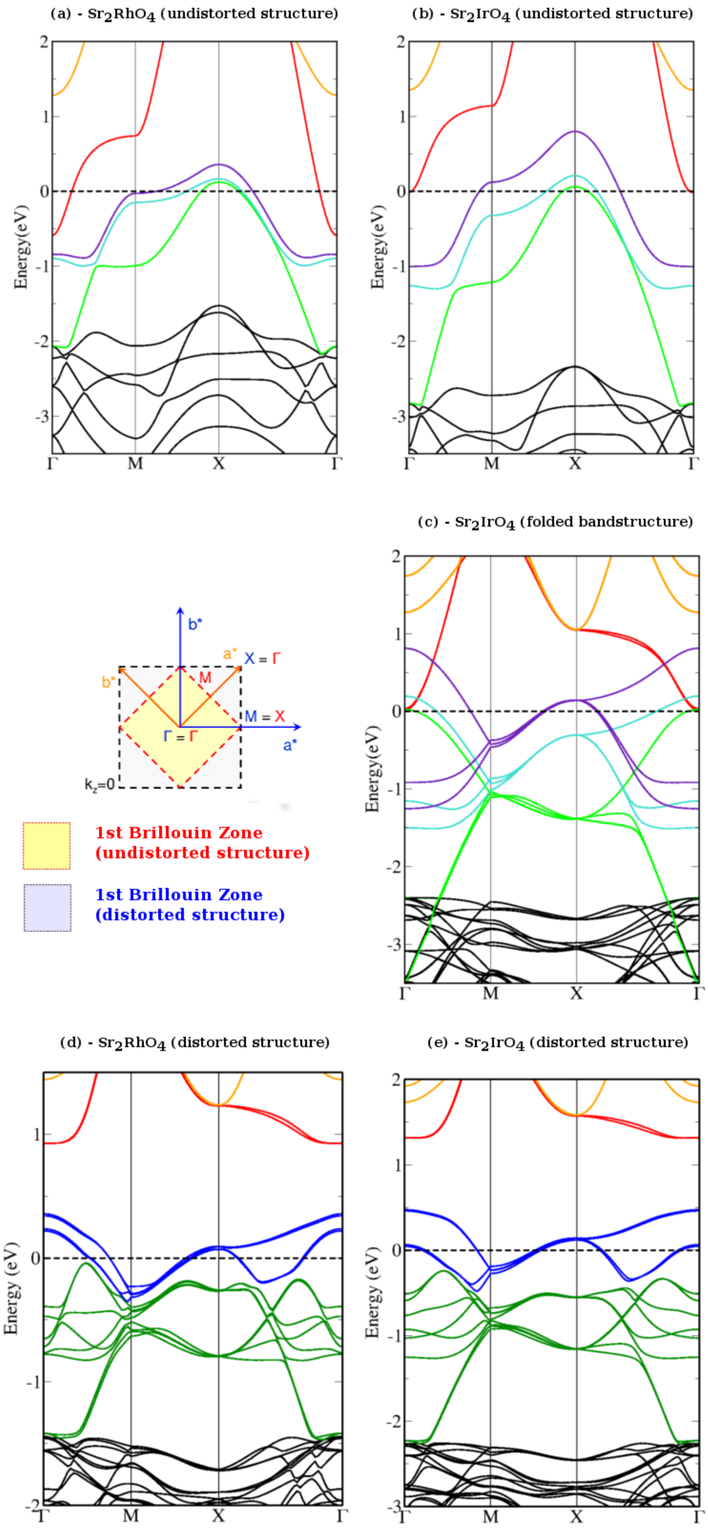

Figure 5: Kohn-Sham band structures within LDA+SO of Sr_2RhO_4 and Sr_2IrO_4 assuming that they crystallize without distortions in a K_2NiF_4 structure (a-b), of Sr_2IrO_4 in a supercell containing 4 "undistorted" unit-cells (c) and of "real" Sr_2RhO_4 and Sr_2IrO_4 (d-e). The reduction of the first Brillouin zone, when the crystal symmetry is lowered, is also shown. The e_g states are in yellow ($d_{3z^2-r^2}$) and red ($d_{x^2-y^2}$). In the t_{2g} manifold, in purple the $j_{\rm eff}$=1/2 in light blue the $j_{\rm eff}$=3/2 $m_j = 3/2$ and in green the $j_{\rm eff}$=3/2 $m_j = 1/2$. In black, the O-2p states.

fact, j_{eff}=1/2 and j_{eff}=3/2 overlap (see the band structure between the Γ and the M-point for instance) and the identification of the upper four bands as the j_{eff}=1/2 states is too simplistic. We will come back to this point below.

To get a better understanding of the Kohn-Sham band structures of Sr_2RhO_4 and Sr_2IrO_4, we study artificial compounds where both the structural distortions and the spin-orbit coupling have been switched off. Fig. 4-(b) and (c) depict the LDA band structure of such "idealized undistorted Sr_2RhO_4 and Sr_2IrO_4". Neglecting the rotation of about 10° of their IrO_6 and RhO_6 octahedra around the c-axis leads to a K_2NiF_4-type crystal structure, like in Sr_2RuO_4, the well-known LDA band structure of which is plotted in Fig. 4-(a).

The similarity of the three band structures is obvious. Around the Fermi level, one distinguishes the three t_{2g} bands. The d_{xy}-band (green) reaches out to lower energies and overlaps with the oxygen 2p-states (black). The e_g-states ($d_{x^2-y^2}$ in red and $d_{3z^2-r^2}$ yellow), higher in energy, cut the Fermi level in both Sr_2IrO_4 and Sr_2RhO_4 due the additional electron remaining in the d-manifold, contrary to Sr_2RuO_4, which has actually a mere t_{2g}^4-filling. The larger extension of the 5d orbitals (cf. Fig 1) explains the wider bandwidth observed for Sr_2IrO_4 in comparison to Sr_2RhO_4: the d_{xy} band reaches the value of -3.5 eV in Γ, while it remains above -3 eV for the 4d counterparts. Another consequence of this wider extension is the stronger hybridization between the 5d states with the oxygen p-states, which are located 1 eV lower in energy in Sr_2IrO_4 than in the 4d-TMOs.

Re-introducing the effects of the spin-orbit coupling in Sr_2RhO_4 and Sr_2IrO_4 (but without considering the structural distortions) modifies these Kohn-Sham band structures to those shown in Fig 5-(a) and (b). The t_{2g} bands are the most affected, while the e_g bands are slightly shifted as a consequence of the topological change in the t_{2g} manifold. A detailed study of the character of these band structures confirms the decoupling between e_g and t_{2g} states (see also Refs. [19, 20]). The cubic crystal field at stake in these compounds is indeed much larger than the energy scale associated to the spin-orbit coupling of about $\zeta_{\text{SO}} \approx 0.4$ eV and $\zeta_{\text{SO}} \approx 0.2$ eV for Sr_2IrO_4 and Sr_2RhO_4 respectively.

The j_{eff} picture is thus justified in both, Sr_2IrO_4 and Sr_2RhO_4: the t_{2g} orbitals split into a quartet of j_{eff}=3/2 states and a higher lying doublet j_{eff}=1/2 . Each state is doubly degenerate in $\pm m_j$ – since we observe the system in its paramagnetic phase at room temperature and the crystal structure has a center of inversion. Therefore we still refer to them as the "j_{eff}=1/2 band" and the two "j_{eff}=3/2 bands" in the following. The three j_{eff} bands can easily be identified: the j_{eff}=1/2 one (light green) lies above the two j_{eff}=3/2 ones ($m_j = 3/2$ in light blue and $m_j = 1/2$ in violet). The three j_{eff} bands are well-separated all along the **k**-path, and more generally in the whole Brillouin zone. Since the spin-orbit coupling is twice smaller in Sr_2RhO_4, the splitting between the j_{eff} bands is reduced by a factor of 2, as one can see for instance at X or Γ.

To draw the link between the "undistorted" band structures and the realistic ones, we plot in Fig 5-(c) the LDA+SO band structure of the *undistorted* Sr_2IrO_4 in a supercell containing four unit cells. Each band is now folded four times and we provide a scheme of the two first Brillouin Zones in the $\mathbf{k}_z = 0$ plane to understand the correspondence between the high-symmetry points of each structure.

Comparing Fig 5-(c) and (e) highlights the key role of the structural distortion in Sr_2IrO_4: an hybridisation between two neighboring Ir d_{xy} and $d_{x^2-y^2}$ orbitals via the in-plane oxygens is now allowed and pushes the t_{2g} and e_g bands apart. Another consequence of the distortions is the general narrowing of the j_{eff} bandwidth, which is of crucial importance to drive the compound insulating, as we will see below.

Finally, comparing Fig 5-(c) and (e) gives more insights into the nature of the four highest-lying bands (blue) of Fig 5-(e). While along the $M-X$ direction, each quartet of j_{eff} bands remain well-separated, j_{eff}=1/2 and j_{eff}=3/2 overlap in the other direction $\Gamma-M$ and $M-X$. As a result, the j_{eff}=3/2 bands cross the Fermi-level closest to the Γ-point, while the other crossings are due to the j_{eff}=1/2 bands. The identification of the upper four bands in Sr_2IrO_4 as "pure" j_{eff}=1/2 states is thus too simplistic, implying the need for a Hamiltonian containing more than one orbital in a realistic calculation.

The same mechanisms are at stake in Sr_2RhO_4 even though we do not display the orbital characters here: the four highest-lying bands, highlighted in blue in Fig. 5-(d) exhibit a mixed character of type j_{eff}=1/2 and j_{eff}=3/2 . Moreover, thanks to the distortions which allow the opening of a gap between t_{2g} and e_g bands, the LDA+SO Fermi surface becomes qualitatively similar to the experimental one: as shown in Fig. 8-(a), they both contain three closed contours : a circular hole-like α-pocket around Γ, a lens-shaped electron pocket β_M and a square-shaped electron pockets β_X. However the striking discrepancies in the size of the pockets point out a subtle deficiency of the LDA for Sr_2RhO_4[19, 20].

4.2 Wannier functions

We have derived the Wannier functions associated to the j_{eff} manifold for both Sr_2IrO_4 and Sr_2RhO_4, using the framework introduced in section 3.1. Because of the mixed character of the four bands that cross the Fermi level in Sr_2IrO_4 and Sr_2RhO_4, the local effective atomic problem used in the DMFT cycle must contain the three j_{eff} orbitals and thus accomodate five electrons. We construct Wannier functions for the j_{eff} orbitals from the LDA+SO band structure of Sr_2IrO_4 and Sr_2RhO_4, using an energy window $[-3.0, 0.5]$ eV for Sr_2IrO_4 and an energy window $[-2.67; 0.37]$ eV for Sr_2RhO_4.

Fig. 6 and 7 depict the projection of these Wannier functions on the LDA+SO band structure. The similarities between Fig 6 and Fig 5-(c) are numerous, thus confirming our previous band character analysis. Tab. 5 gives the decompostion of these local Wannier functions on the t_{2g} manifold and their respective occupation.

To obtain deeper insights into the nature of these Wannier orbitals, Tab. 4 gives the coefficients of the local Wannier orbitals obtained from the LDA+SO band structure of "undistorted" Sr_2IrO_4 using an energy window $[-3.5, 0.8]$ eV. The results agree well with the standard j_{eff} picture (cf. Eq. (1) and (2)) in both modulus and phase. Discrepancies are mostly due to the elongation of the IrO_6 along the c-axis, which introduces an additional tetragonal field between the t_{2g} states. This effect also explains the lifting of the degeneracy of the two j_{eff}=3/2 ($m_j = \pm 1/2$ and $m_j = \pm 3/2$) states and implies the reason why the j_{eff}=1/2 is slightly more than half-filled.

Because of the hydridization between the d_{xy} and $d_{x^2-y^2}$ orbitals in the distorted structures, we had to define in practice "effective j_{eff}=1/2 and j_{eff}=3/2 $|m_j| = 1/2$ states", which remain

"undistorted" Sr_2IrO_4	$\left\lvert\frac{1}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{3}{2}\right\rangle$
$\lvert d_{xy}\uparrow\downarrow\rangle$	$\pm$ 0.6605	$+0.7508$	0
$\lvert d_{xz}\uparrow\downarrow\rangle$	$\pm$ 0.5309 i	$-0.4670i$	-0.7071 i
$\lvert d_{yz}\uparrow\downarrow\rangle$	+ 0.5309	∓ 0.4670	∓ 0.7071
occupation (LDA+SO)	1.20	1.92	1.86

Table 4: Coefficients and occupation of the $j_{\rm eff}$ Wannier orbitals in "undistorted" Sr_2IrO_4. The discrepancy between these coefficients and those given in Eq. (1) and (2) are due to the small elongation of the octahedra along the c-axis.

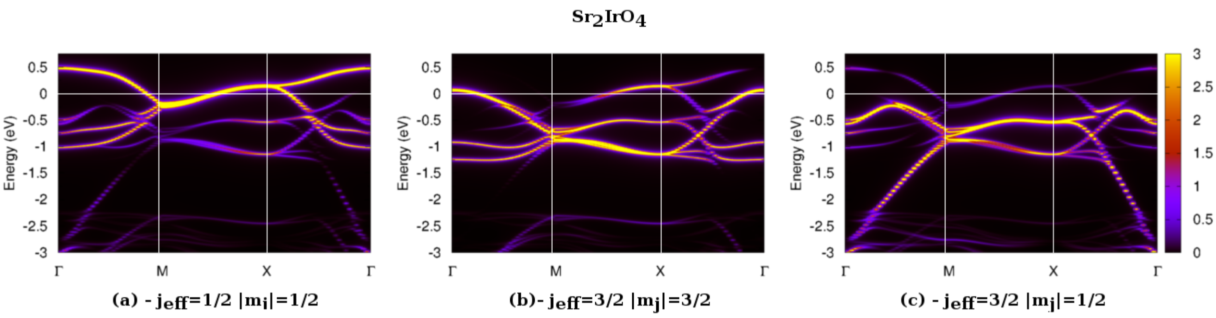

Figure 6: LDA+SO band structure of Sr_2IrO_4, projected on the $j_{\rm eff}$=1/2 (left panel), $j_{\rm eff}$=3/2 $|m_j|$=3/2 (middle), and $j_{\rm eff}$=3/2 $|m_j|$=1/2 (right panel) spin-orbitals.

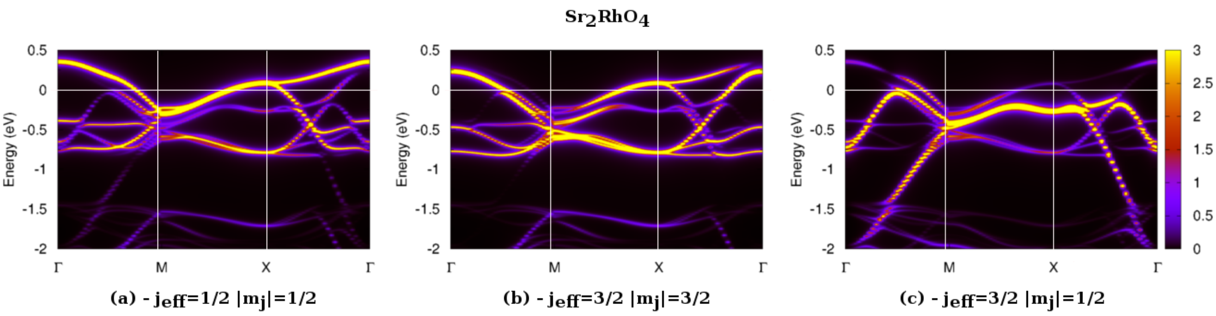

Figure 7: LDA+SO band structure of Sr_2RhO_4, projected on the $j_{\rm eff}$=1/2 (left panel), $j_{\rm eff}$=3/2 $|m_j|$=3/2 (middle), and $j_{\rm eff}$=3/2 $|m_j|$=1/2 (right panel) spin-orbitals.

	Sr_2IrO_4			Sr_2RhO_4		
Wannier orbitals	$\left\lvert\frac{1}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{3}{2}\right\rangle$	$\left\lvert\frac{1}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{1}{2}\right\rangle$	$\left\lvert\frac{3}{2},\pm\frac{3}{2}\right\rangle$
$\lvert \mathrm{d}_{x^2-y^2}\uparrow\downarrow\rangle$	0.0388	0.0766	0	0.0100	0.0302	0
$\lvert \mathrm{d}_{xy}\uparrow\downarrow\rangle$	0.4499	0.8889	0	0.3153	0.9485	0
$\lvert \mathrm{d}_{xz}\uparrow\downarrow\rangle$	0.6309	0.3193	0.7071	0.6710	0.2231	0.7071
$\lvert \mathrm{d}_{yz}\uparrow\downarrow\rangle$	0.6309	0.3193	0.7071	0.6710	0.2231	0.7071
occupation (LDA+SO)	1.16	1.98	1.84	1.42	1.96	1.64
charge (LDA+SO+DMFT)	1.02	2.00	1.98	1.26	1.98	1.76

Table 5: Modulus of the coefficients of the j_{eff} Wannier orbitals in Sr_2IrO_4 and Sr_2RhO_4. The occupation within LDA+SO and within LDA+SO+DMFT of each atomic Wannier orbitals is also provided, showing how electronic correlations enhance the spin-orbital polarization.

close to the atomic j_{eff} picture but take into account a small amount of $d_{x^2-y^2}$ character (cf. Tab. 5). The coefficients have been calculated such that the density matrix of the local atomic problem is closest possible to diagonal form.† In addition to the hybridization, the construction of the "effective j_{eff}" takes also into account the tetragonal crystal field due to the elongation of the octahedra in each crystal structure: this explains the discrepancies with the standard coefficients given in Eq. (1) and (2). We note that the coefficients obtained for the j_{eff}=1/2 state of Sr_2IrO_4 are equivalent to those obtained in the AF phase in Ref. [139].

Finally, comparing the occupation of the orbitals in Tab. 4 and 5 highlights again the role of the hybridisation between the d_{xy} and $d_{x^2-y^2}$ orbitals which pushes the band j_{eff}=3/2 $|m_j| = 1/2$ further below the Fermi level close to Γ : as a result, the four bands that cross the Fermi level are formed only by the j_{eff}=1/2 and j_{eff}=3/2 $|m_j|$=3/2 orbitals and the j_{eff}=1/2 tend to be close to half-filling. Similar conclusions were drawn for the AF phase within a Variational Cluster Approximation (VCA) approach in Ref. [140]. Similar conclusions hold for Sr_2RhO_4.

4.3 Effective Hubbard Interactions from cRPA

After defining the j_{eff} Wannier orbitals, we evaluate the local Coulomb interaction in the effective atomic problem within cRPA [206, 213], as explained in section 3.3. For reasons of computational resources, the cRPA calculations were performed in the case without distortions (without the rotations of the octahedra, hence considering only one formula-unit in a unit-cell) and without SOC. To mimick the effect of the distortions, the e_g states are shifted up to their energetic position in the presence of distortions. We find $\mathcal{U} = 2.54$ eV and $\mathcal{J} = 0.23$ eV for Sr_2IrO_4 and $\mathcal{U} = 1.94$ eV and $\mathcal{J} = 0.23$ eV for Sr_2RhO_4. These parameters lead to the following local

†With the obtained coefficients, the off-diagonal terms remaining in the local Greens functions between the j_{eff}=1/2 and j_{eff}=3/2 $|m_j| = 1/2$ are smaller than 0.05. In practice, the coefficients were chosen real. This can be done in the local problem since only density-density terms were kept for the interaction terms and off-diagonal terms of the density matrix were neglected.

interaction matrices for Sr_2IrO_4:

$$U_{jj'}^{m_j m_{j'}} = \left(\begin{array}{c|cc} 0 & 2.08 & 2.21 \\ \hline 2.08 & 0 & 1.93 \\ 2.21 & 1.93 & 0 \end{array}\right) \quad U_{jj'}^{m_j \overline{m}_{j'}} = \left(\begin{array}{c|cc} 2.25 & 1.98 & 1.90 \\ \hline 1.98 & 2.38 & 2.03 \\ 1.90 & 2.03 & 2.31 \end{array}\right) \tag{22}$$

and for Sr_2RhO_4:

$$U_{jj'}^{m_j m_{j'}} = \left(\begin{array}{c|cc} 0 & 1.48 & 1.66 \\ \hline 1.48 & 0 & 1.29 \\ 1.66 & 1.29 & 0 \end{array}\right) \quad U_{jj'}^{m_j \overline{m}_{j'}} = \left(\begin{array}{c|cc} 1.67 & 1.32 & 1.27 \\ \hline 1.32 & 1.86 & 1.46 \\ 1.27 & 1.46 & 1.71 \end{array}\right) \tag{23}$$

where the values are in eV and the ordering of the $|j, |m_j|\rangle$ orbitals is: $|1/2, 1/2\rangle$,$|3/2, 1/2\rangle$, $|3/2, 3/2\rangle$ and $\overline{m}_j$ denotes $-m_j$. We remind the reader that $U_{jj'}^{m_j m_{j'}} = U_{jj'}^{\overline{m}_j \overline{m}_{j'}}$ and $U_{jj'}^{m_j \overline{m}_{j'}} = U_{jj'}^{\overline{m}_j m_{j'}}$. Since we have used "effective j_{eff}" Wannier orbitals instead of the standard definition given in Eq. (1) and (2), some discrepancies with the formulae given in Eq. (19) and in [22] can be observed. Contrary to common belief, the Hubbard interactions are smaller in the $4d$-TMO than in its $5d$-counterpart. This might seem counterintuitive at first sight, since the $5d$-orbitals are more extended than the $4d$ ones, but finds its explanation in more efficient screening in the $4d$ material: As shown in Fig. 5-(d) and (e), the hybridization between the Rh-$4d$ states and the O-$2p$ is weaker in Sr_2RhO_4 than in Sr_2IrO_4. Correspondingly, the energetic position of the O-$2p$ bands is closer to the Fermi level by about 1 eV, and as a result, the Coulomb interactions are screened more efficiently in Sr_2RhO_4 than in Sr_2IrO_4, explaining the observed trend.

4.4 Correlated electronic structure of Sr_2IrO_4 and Sr_2RhO_4

DFT+DMFT calculations following the procedure described in section 3.1 indeed find an insulating solution for Sr_2IrO_4 and a correlated metal for Sr_2RhO_4[21], in agreement with experiment. The difference in these metallic versus insulating nature of Sr_2RhO_4 and Sr_2IrO_4 can be traced back to the different spin-orbital polarization in the three j_{eff} orbitals, which is enhanced by Coulomb correlations.

The occupations of the j_{eff} Wannier orbitals within LDA+SO and LDA+SO+DMFT are provided in Tab. 5. In Sr_2IrO_4, one detects a considerable spin-orbital polarisation already at the LDA+SO level: the four j_{eff}=3/2 states are almost filled with $n_{3/2,|1/2|}$=1.98 and $n_{3/2,|3/2|}$=1.84 while the j_{eff}=1/2 states thus slightly exceed half-filling with $n_{1/2}$=1.16 (as in the "ideal undistorted" case). Taking into account Coulomb correlations within DMFT opens a gap of about 0.26 eV [21] and enhances the spin-orbital polarisation, such as to fill the j_{eff}=3/2 states entirely, leading to a half-filled j_{eff}=1/2 state. This is thus the celebrated "j_{eff}=1/2 -picture" [16], which comes out here as a result of the calculations, rather than being an input as in most model Hamiltonian calculations.

A different picture emerges for Sr_2RhO_4 according to Tab. 5: while the spin-orbital occupations display some polarisation at the LDA+SO level, the smaller SOC – and thus the smaller effective splitting between the j_{eff} bands – leads to a picture where only the j_{eff}=3/2 $|m_j| = 1/2$ state is entirely filled while both j_{eff}=3/2 $|m_j| = 3/2$ and j_{eff}=1/2 live at the Fermi level. This spin-orbital polarization is enhanced by Coulomb correlations – just as in Sr_2IrO_4 – but this

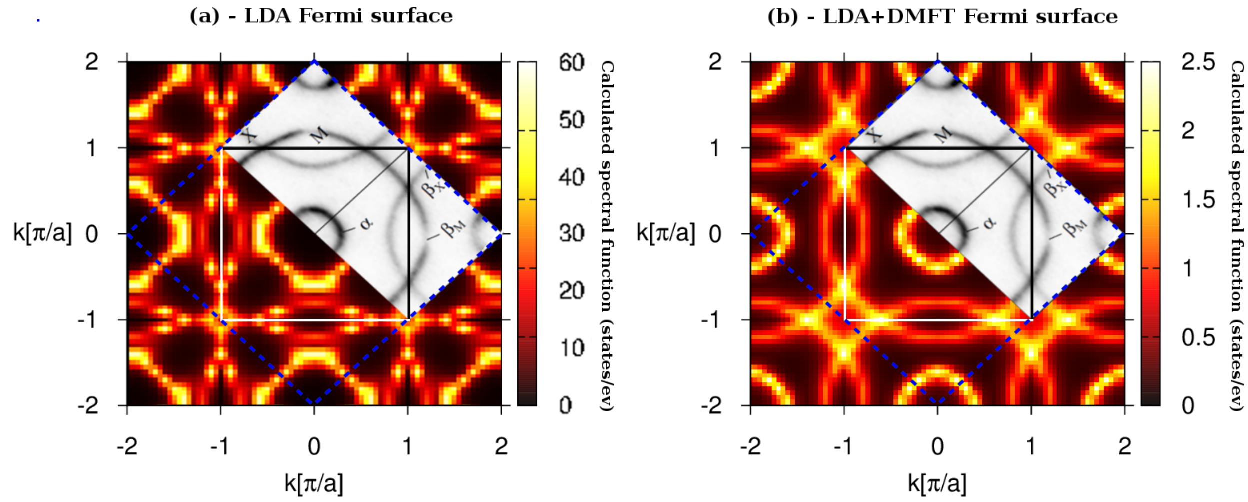

Figure 8: Calculated Fermi surface of Sr_2RhO_4 in the $\mathbf{k}_z = 0$ plane within LDA+SO (left panel) and LDA+SO+DMFT (right panel). Superimposed is the experimentally measured Fermi surface, from Ref. [120].

	α			β_X			β_M		
	LDA	DMFT	Exp.	LDA	DMFT	Exp.	LDA	DMFT	Exp.
FS volume A (% BZ)	18.4	10.1	6.1(4)	4.5	6.2	8.1(5)	10.0	7.6	7.4(4)
$\hbar\overline{v_F}$ (eV.Å)	1.252	0.645	0.41(4)	1.260	0.674	0.55(6)	1.260	0.674	0.61(6)
m^* (m_e)	1.70	2.44	3.0(3)	0.83	1.83	2.6(3)	1.24	2.02	2.2(2)

Table 6: Comparison of the Fermi surface (FS) parameters evaluated within LDA+SO, within LDA+SO+DMFT and ARPES [120]. For each α, β_X and β_M pocket, the FS volume A is defined as a pourcentage of the two-dimensional-BZ volume (using the experimental lattice parameters ($a = 5.45$ Å)). The Fermi velocity $\hbar\overline{v_F}$ is obtained from the slope of the band dispersion at the Fermi level. The cyclotron mass m^*/m_e is calculated using the same method as described in [120]: $m^*\overline{v_F} = \hbar\sqrt{A/\pi}$.

enhancement is not enough to fill both j_{eff}=3/2 states entirely and obtain a half-filled j_{eff}=1/2 state. The higher effective degeneracy together with the smaller value of $\mathcal{U}$ eventually leave Sr_2RhO_4 metallic.

4.5 Spectral properties of Sr_2RhO_4: theory vs. experiment

We now turn to the calculated spectral function of the spin-orbital correlated metal Sr_2RhO_4 that we analyse in comparison to experiment.

Fig. 8 depicts the Fermi surface of Sr_2RhO_4 within LDA+SO (left panel) and LDA+SO+DMFT (right panel) in the $\mathbf{k}_z = 0$ plane, to which we superimpose the experimental measurement from [120]. Tab. 6 gives more quantitative insight to ease the comparison between the different topologies. All three Fermi surfaces, the two theoretical ones and the experimental one, are *qualitatively* similar with three closed contours : a circular hole-like α-pocket around Γ, a lens-shaped electron pocket β_M and a square-shaped electron pockets β_X. These two structures merge in the undistorted tetragonal zone (dashed blue line in Fig. 8) to a large electron-like

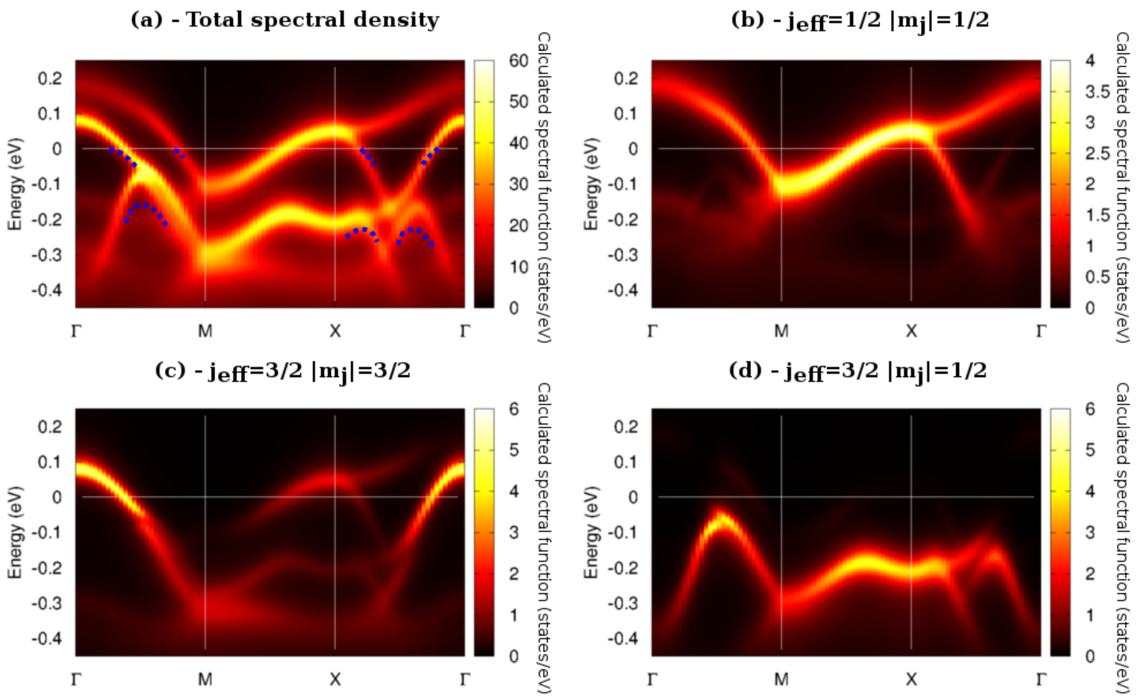

Figure 9: Calculated momentum-resolved spectral function of Sr_2RhO_4 within LDA+DMFT (a) and its orbital-resolved versions for the j_{eff}=1/2 states (b), the j_{eff}=3/2 $|m_j| = 3/2$ (c) and the completely-filled j_{eff}=3/2 $|m_j| = 1/2$ (d). The dashed blue line on panel (a) are the reproduction of the ARPES structure from [121]

pocket β.

Comparing Fig. 8-(a) and (b) highlights the key role of electronic correlations : they decrease the radius of the α pocket from $0.26 - 0.29$ Å^{-1} to 0.21 Å^{-1} and decrease the radius of the large β pocket from $0.69 - 0.72$ Å^{-1} to $0.67 - 0.70$ Å^{-1}, thus enlarging the β_M and β_X pockets such that their volume are well-reproduced within LDA+SO+DMFT (cf. Tab 6). As a result, the agreement between LDA+SO+DMFT data and the experimental measurements becomes even quantitatively excellent.

To go further in the analysis, Fig. 9 depicts the momentum-resolved spectral function, as well as its orbital-resolved version. The completely filled j_{eff}=3/2 $|m_j| = 1/2$ state is visible (panel d), as well as the partially filled character of the j_{eff}=3/2 $|m_j| = 3/2$ (panel c) and j_{eff}=1/2 states (panel b). A detailed comparison with angle-resolved photoemission data from [121] (blue dashed line on the figure) shows that the band dispersion around the Fermi level is well-reproduced, while some discrepancies are observed for the structures experimentally observed along $\Gamma - X$ and $\Gamma - M$ at lower energy. These features, reminiscent of the j_{eff}=3/2 $|m_j| = 1/2$ bands, are indeed located about 0.05 eV higher in energy in our calculated spectral function.

From Fig. 9-(b) and (c), one observes that the Fermi level is crossed by the renormalized j_{eff}=3/2 $|m_j| = 3/2$ band at 0.20 Å^{-1} along $\Gamma - X$ and at 0.21 Å^{-1} along $\Gamma - M$, while the renormalized j_{eff}=1/2 band is responsible for all other crossings. This allows to label the hole-like α-pocket as being of j_{eff}=3/2 $|m_j| = 3/2$ type, whereas the two other pockets β_M and β_X are mostly of type j_{eff}=1/2 . Using the quasiparticle weight of each state ($Z_{1/2} = 0.535$ and $Z_{3/2,|3/2|} = 0.675$), we evaluate the Fermi velocity at each crossing along the path $[\Gamma M X \Gamma]$: we find a huge variation of the values depending on $\mathbf{k}$ and give in Tab. 6 their mean value over the Brillouin zone. Finally, using the same method as described in [120], we evaluate the cyclotron mass m^*/m_e based on the approximate formula used there: $m^*\overline{v_F} = \hbar\sqrt{A/\pi}$. The DMFT results shown in Tab. 6 show a substantial improvement over DFT when compared to experiments.

5 The effective orbital degeneracy as a key quantity determining the correlation strength

In section 4.4, we have identified the spin-orbital polarization as a key factor to explain the different behavior of Sr_2RhO_4 and Sr_2IrO_4.

In Sr_2IrO_4, Coulomb correlations enhance the spin-orbital polarisation, such as to fill the j_{eff}=3/2 states entirely, leading to a half-filled j_{eff}=1/2 one-band picture, while in Sr_2RhO_4 the final situation is an effective two-orbital system containing three electrons. This situation is akin to correlation-induced enhancements of orbital polarisation also observed in other transition metal oxides. In the distorted $3d^1$ perovskites $LaTiO_3$ and $YTiO_3$, for example, it was argued [10] that the interplay of structural distortions and Coulomb correlations leads to a suppression of orbital fluctuations in the t_{2g}-manifold, favoring a particular orbital composition selected by crystal and ligand field effects. At the LDA level, 0.45 [0.88] electrons are found in this particular orbital in $LaTiO_3$ [$YTiO_3$], while Coulomb correlations as described by LDA+DMFT lead to an occupation of 0.88 [0.96] electrons.

In these systems, this reduction of effective orbital degeneracy was shown to be key to their insulating nature since the critical interaction strength needed to localise the single electron is thus effectively determined by the one of a single-orbital system, instead of the one of a three-fold degenerate t_{2g}-manifold. Within DMFT, the critical Hubbard interaction scales with the square-root of the orbital degeneracy N for the lower critical interaction of the phase coexistence region of the first order Mott transition, while the upper critical interaction varies with N [231].

Localising electrons in a single-orbital system therefore needs a critical interaction roughly smaller by a factor of 3 as compared to the degenerate case. This was demonstrated to be crucial for the difference in behaviors in the series of d^1 compounds $SrVO_3$, $CaVO_3$, $LaTiO_3$, $YTiO_3$, where the former are three-fold degenerate metallic systems, whereas the latter realise the single-orbital Mott state.

The situation in the iridates is analogous with the purely formal difference that one is dealing with a one-hole situation instead of one electron. Furthermore, the strong spin-orbit interaction is instrumental for the suppression of the degeneracy, which is the net result of structural distortions, spin-orbit coupling and Coulomb correlations.

This discussion highlights an important aspect of the physics of transition metal oxides, often neglected when considering band filling and interaction strength only: the *effective orbital degeneracy* is a crucial tuning parameter for electronic behavior, suggesting that the popular picture distinguishing filling-controlled and bandwidth-controlled Mott transitions [1] should be complemented by a “third axis” and the notion of degeneracy-controlled Mott behavior.

Crystal and ligand fields together with spin-orbit coupling and the Coulomb correlations themselves are the driving forces for establishing a given effective degeneracy. At the level of the calculations, this effective degeneracy is both an outcome of the calculation and a determining factor of the properties of the given compound.

6 Conclusions and Perspectives

The common belief about electronic Coulomb correlations being less important in 4d and 5d compounds as compared to 3d transition metal oxides, was overruled by insights into the role of spin-orbit coupling for the insulating behavior of iridates [16] and for the Fermi surface topology of Sr_2RhO_4 [19, 20].

Here, we have reviewed recent work on a first principles many-body description of such effects within a dynamical mean-field framework. We have highlighted the notion of the *effective degeneracy* of the system as a crucial parameter determining the physical properties of a system. The effective degeneracy is the result of a complex interplay of structural distortions spin-orbit coupling and Coulomb correlations. We have stressed the analogy of the j_{eff}=1/2 Mott insulating picture for Sr_2IrO_4 with the insulating nature of $LaTiO_3$ and $YTiO_3$ in the “degeneracy-controlled Mott transition” series of d^1 perovskites ($SrVO_3$, $CaVO_3$, $LaTiO_3$, $YTiO_3$).

In Sr_2IrO_4 and Sr_2RhO_4 the difference in degeneracy is itself a consequence of the quantitative aspects of the physics of these two compounds: all three decisive elements – structural distortions, spin-orbit coupling and Hubbard interaction – are smaller in Sr_2RhO_4 than in Sr_2IrO_4 and this quantitative difference in the electronic parameters translates into a qualitative difference of the resulting properties.

We have analysed in detail the spectral properties of Sr_2RhO_4, a spin-orbit correlated 4d metal where the effective degeneracy is reduced by spin-orbit coupling and correlations but not to the point such as to induce a j_{eff}=1/2 Mott insulator. The calculated spectral properties and Fermi surface are in excellent agreement with experimental data. A detailed analysis of the spectral properties of Sr_2IrO_4 is left for future work.

7 Acknowledgments

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Appendix A: Generalized partial Θ-*projectors* and spectral function

In order to calculate quantities for a given atom α and a particular orbital (spin) character j (m_j) – such as the spectral functions $A_j^{m_j\alpha}(\mathbf{k},\omega)$ – , a set of partial projectors called "Θ-*projectors*" was built. Contrary to the previously introduced Wannier projectors $P_{j,\nu}^{\alpha,m_j}(\mathbf{k})$, their definition is not restricted to the correlated orbitals only. The formalism of these partial projectors was initially introduced in [180] and was extended to the case where spin is not a good quantum number anymore in [21].

Inside the muffin-tin sphere associated to an atom α, one can write the spin-σ contribution of the eigenstate $\psi_{\mathbf{k}\nu}(\mathbf{r})$ as:

$$\phi_{\mathbf{k}\nu}^{\sigma}(\mathbf{r}) \;=\; \sum_{\ell=0}^{\ell_{max}}\sum_{m=-\ell}^{+\ell}\Big[A_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)\; u_{\ell m,1}^{\alpha,\sigma}(\mathbf{r}^{\alpha}) + B_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)\; u_{\ell m,2}^{\alpha,\sigma}(\mathbf{r}^{\alpha}) + C_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)\; u_{\ell m,3}^{\alpha,\sigma}(\mathbf{r}^{\alpha})\Big] \tag{24}$$

where the basis $\{u_{\ell m,i}^{\alpha,\sigma}\}_{i=\{1,2,3\}}$ is not orthonormalized as already mentioned in [180]. That is why, to make the calculations easier, one introduces an orthonormal basis set $\{v_{\ell m,j}^{\alpha,\sigma}\}_{\{j=1,2,3\}}$ for each atomic orbital (ℓ,m). These orbitals are defined from the initial basis $\{u_{\ell m,i}^{\alpha,\sigma}\}_{i=\{1,2,3\}}$ as follows:

$$\forall i \quad u_{\ell m,i}^{\alpha,\sigma}(\mathbf{r}^{\alpha}) = \sum_{j=1}^{3} c_{ij} v_{\ell m,j}^{\alpha,\sigma} \quad \text{with} \quad \mathbf{C} = \begin{pmatrix} 1 & 0 & \langle u_{\ell m,1}^{\alpha,\sigma}|u_{\ell m,2}^{\alpha,\sigma}\rangle \\ 0 & \langle u_{\ell m,2}^{\alpha,\sigma}|u_{\ell m,2}^{\alpha,\sigma}\rangle & \langle u_{\ell m,2}^{\alpha,\sigma}|u_{\ell m,3}^{\alpha,\sigma}\rangle \\ \langle u_{\ell m,3}^{\alpha,\sigma}|u_{\ell m,1}^{\alpha,\sigma}\rangle & \langle u_{\ell m,3}^{\alpha,\sigma}|u_{\ell m,2}^{\alpha,\sigma}\rangle & 1 \end{pmatrix}^{\frac{1}{2}}. \tag{25}$$

We can then rewrite (24) as:

$$\psi_{\mathbf{k}\nu}^{\sigma}(\mathbf{r}) = \sum_{\ell=0}^{\ell_{max}}\sum_{m=-\ell}^{+\ell}\sum_{i=1}^{3}\Theta_{\ell m\nu,i}^{\alpha,\sigma}(\mathbf{k}) v_{\ell m,i}^{\alpha,\sigma}(\mathbf{r}^{\alpha}). \tag{26}$$

The matrix elements $\Theta_{\ell m\nu,i}^{\alpha,\sigma}(\mathbf{k})$ are the "Θ-*projectors*", which are thus defined by:

$$\Theta_{\ell m\nu,i}^{\alpha,\sigma}(\mathbf{k}) = \langle v_{\ell m,i}^{\alpha,\sigma}|\phi_{\mathbf{k}\nu}^{\sigma}\rangle = A_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)c_{1i} + B_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)c_{2i} + C_{\ell m}^{\nu\alpha}(\mathbf{k},\sigma)c_{3i}. \tag{27}$$

Contrary to the implementation of [180], there is now a couple of Θ-projectors associated to each band index ν, $\Theta_{\ell m\nu,i}^{\alpha,\sigma}(\mathbf{k})$ with $\sigma=\uparrow,\downarrow$, since spin is not a good quantum number anymore. We have introduced here the Θ-projectors in the complex spherical harmonics basis. As for the Wannier projectors, it is of course possible to get the Θ-projectors in any desired j, m_j basis:

$$\Theta_{j\nu,i}^{\alpha,m_j}(\mathbf{k}) = \sum_{m,\sigma} \mathcal{S}_{j,\ell m}^{m_j,\sigma}\,\Theta_{\ell m\nu,i}^{\alpha,\sigma}(\mathbf{k}) \tag{28}$$

Finally, the spectral function $A(\mathbf{k},\omega)$ which is defined by:

$$A(\mathbf{k},\omega) = -\frac{1}{\pi}\mathfrak{Im}\left[G(\mathbf{k},\omega)\right]. \tag{29}$$

is obtained for a given atom α with orbital character (j, m_j) through the following formula:

$$A_j^{\alpha,m_j}(\mathbf{k},\omega) = -\frac{1}{\pi}\mathfrak{Im}\left[\sum_{\nu\nu'}\sum_{i=1}^{3}\mathbf{\Theta}_{j\nu,i}^{\alpha,m_j}(\mathbf{k})\mathbf{G}_{\nu\nu'}(\mathbf{k},\omega+i0^+)\left[\mathbf{\Theta}_{j\nu',i}^{\alpha,m_j}(\mathbf{k})\right]^*\right] \tag{30}$$

where the band indices ν, ν' run over *both* spin and orbital quantum number.

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