

# Spin-orbit coupling within Dynamical Mean-Field Theory: Coulomb correlations in 5d and 4d transition metal oxides

C. Martins <sup>1,2,\*</sup>, M. Aichhorn <sup>3</sup>, S. Biermann <sup>1,2</sup>

<sup>1</sup> Centre de Physique Théorique, Ecole Polytechnique, CNRS, 91128 Palaiseau Cedex, France

<sup>2</sup> Japan Science and Technology Agency, CREST, Kawaguchi 332-0012, Japan

<sup>3</sup> Institute of Theoretical and Computational Physics, TU Graz, Petergasse 16, Graz, Austria

\* New address : CEA, DAM, DIF, F-91297, Arpajon, France

Phys. Rev. Lett. 107, 266404 (2011)



energie atomique • énergies alternatives

## Abstract

The concept of spin-orbit coupling has been known for more than half a century, and its importance in magnetic materials has been recognized early on. Still, the interplay of spin-orbit interactions with electronic Coulomb interactions in paramagnetic materials remains a largely unexplored field.

We discuss here the notions of spin-orbital polarization and ordering, and address their consequences in transition metal oxides. By extending the combined density functional theory (DFT) and dynamical mean-field theory (DMFT) scheme as implemented in [1] to the case where spin-orbit interactions are important, we investigate the electronic excitations of the paramagnetic phases of Strontium Iridate ( $\text{Sr}_2\text{IrO}_4$ ) and Strontium Rhodate ( $\text{Sr}_2\text{RhO}_4$ ).

## Strontium Iridate ( $\text{Sr}_2\text{IrO}_4$ ) and Strontium Rhodate ( $\text{Sr}_2\text{RhO}_4$ )

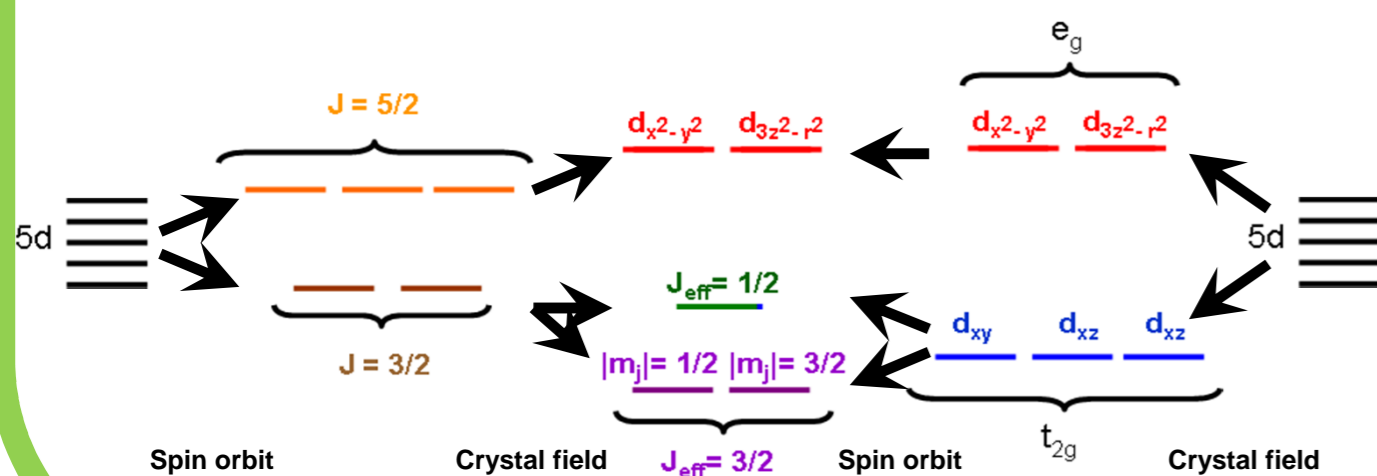
A structure similar to  $\text{La}_2\text{CuO}_4$  or  $\text{Sr}_2\text{RuO}_4$  but with distortions:

$\text{IrO}_6$  and  $\text{RhO}_6$  octahedra are rotated around the z axis by about 11°.  
 $a(\text{Sr}_2\text{RhO}_4) = 5.44 \text{ \AA}$ ;  $c(\text{Sr}_2\text{RhO}_4) = 25.75 \text{ \AA}$   
 $a(\text{Sr}_2\text{IrO}_4) = 5.49 \text{ \AA}$ ;  $c(\text{Sr}_2\text{IrO}_4) = 25.77 \text{ \AA}$

Structure of  $\text{Sr}_2\text{IrO}_4$  or  $\text{Sr}_2\text{RhO}_4$  → From [2]

Each Ir and Rh atom accommodates 5 electrons.

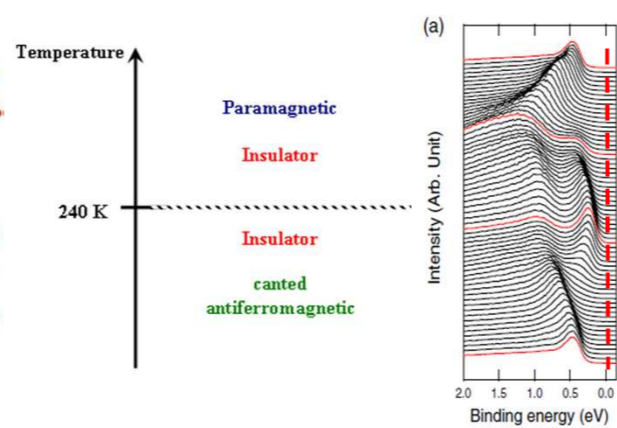
Effect of the spin-orbit coupling (SOC) in both compounds:



Because of the crystal field induced by the oxygen, the d states are split into  $e_g, j_{\text{eff}}=1/2$  and  $j_{\text{eff}}=3/2$  multiplets.

$\text{Sr}_2\text{IrO}_4$ : an insulating 5d transition metal oxide

At 300 K, an optical gap of 0.26 eV [5].



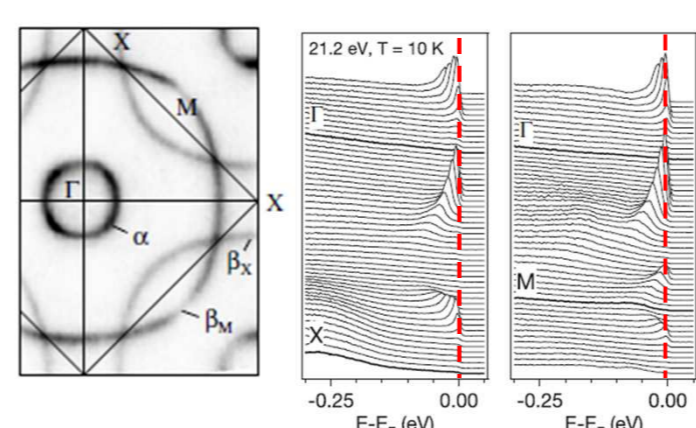
ARPES spectrum for  $\text{Sr}_2\text{IrO}_4$  (measures performed at 100 K). From [3]

Spin-orbit in  $\text{Sr}_2\text{IrO}_4$ :  $\zeta_{\text{SO}} \sim 0.4 \text{ eV}$

According to resonant X-ray scattering, SOC plays an important role in  $\text{Sr}_2\text{IrO}_4$  [7]. The antiferromagnetic phase is well-described in LDA+U calculations only when SOC is taken into account. [8]

$\text{Sr}_2\text{RhO}_4$ : a metallic 4d transition metal oxide

A paramagnetic metal down to 36 mK [6].

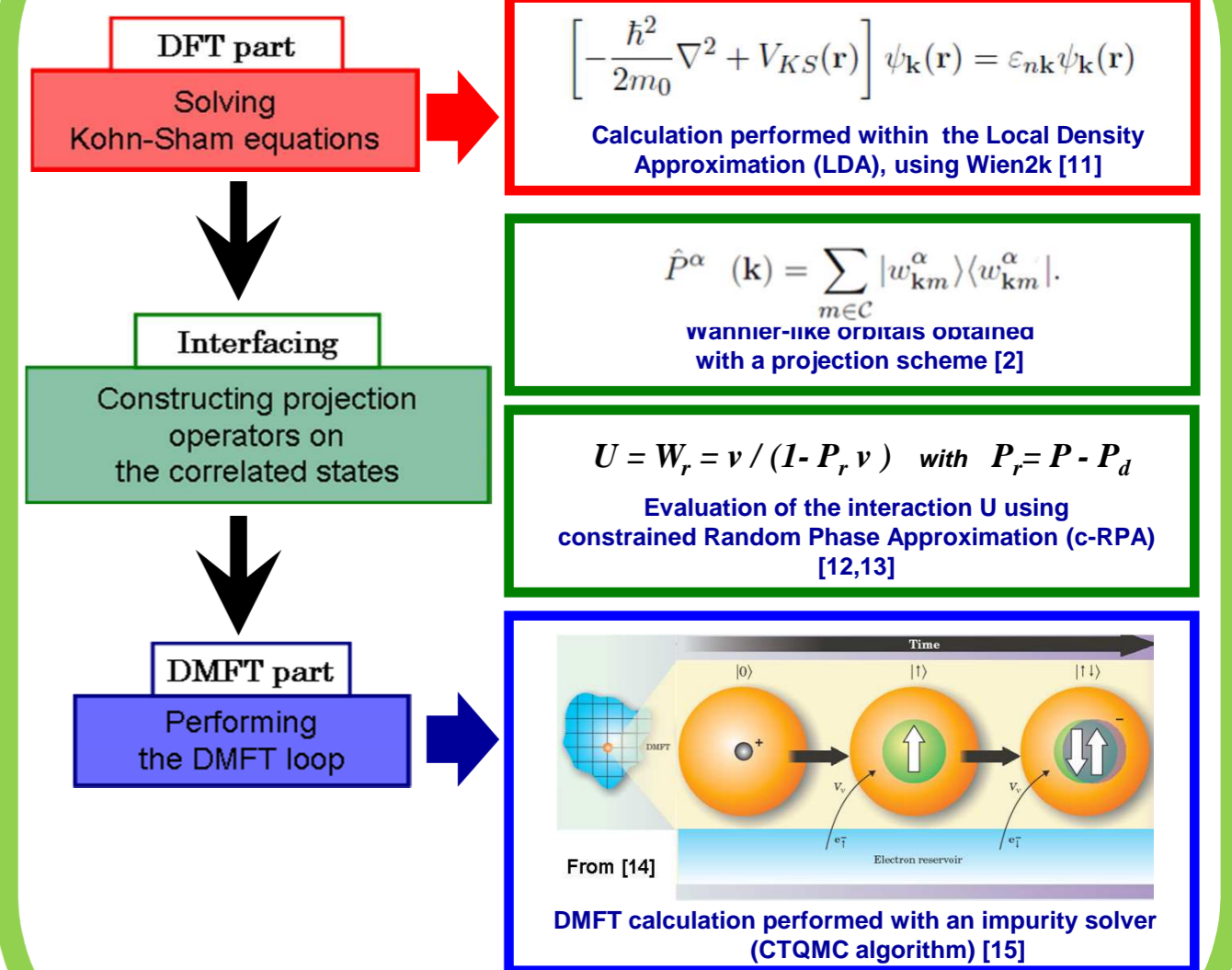


Fermi surface in  $\text{Sr}_2\text{RhO}_4$  by ARPES (measures performed at 10 K). From [4]

Spin-orbit in  $\text{Sr}_2\text{RhO}_4$ :  $\zeta_{\text{SO}} \sim 0.191 \text{ eV}$

The Fermi surface of  $\text{Sr}_2\text{RhO}_4$  is reproduced only if SOC is taken into account, according to recent LDA+U calculations. [9,10]

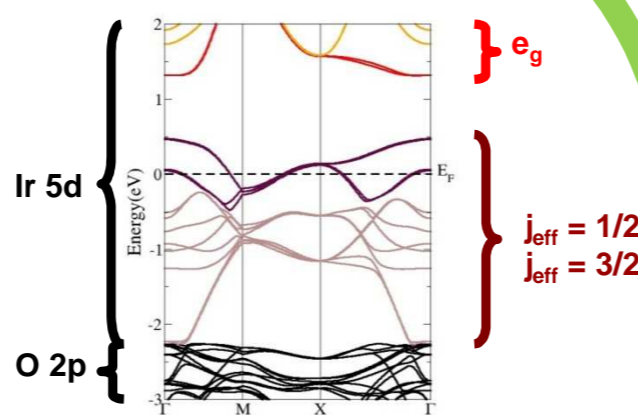
## The method : LDA+SO+DMFT



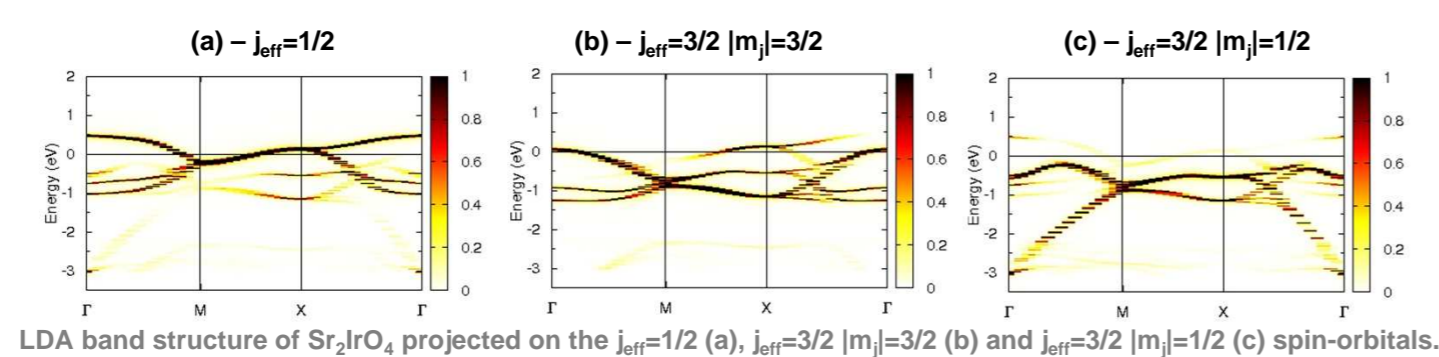
## Paramagnetic $\text{Sr}_2\text{IrO}_4$ within LDA+SO+DMFT

DFT part  
Solving Kohn-Sham equations

At the LDA level, 4 bands cross the Fermi level: a metallic Kohn-Sham band structure.  
Necessity to treat better the correlations between electrons.



Interfacing  
Constructing projection operators on the correlated states

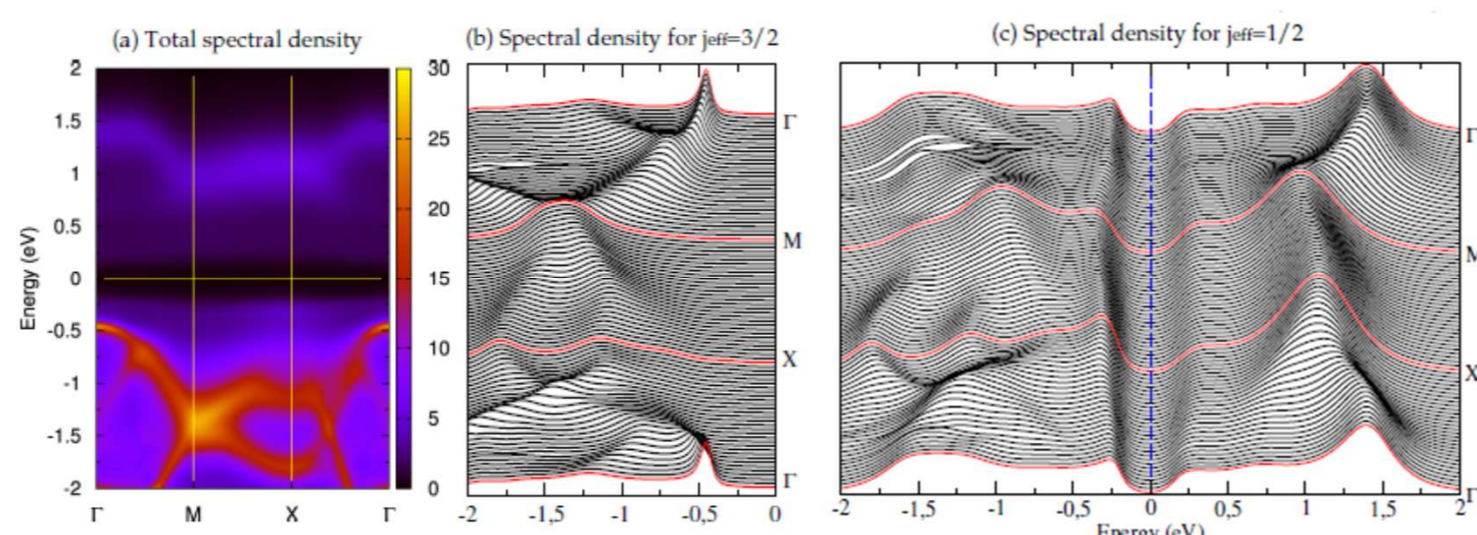


The  $j_{\text{eff}}=1/2$  state is half-filled  
The  $j_{\text{eff}}=3/2$  states are almost completely filled.

Charge	$\text{Sr}_2\text{IrO}_4$
$j_{\text{eff}}=1/2$	1.14
$j_{\text{eff}}=3/2  m_j =1/2$	2.00
$j_{\text{eff}}=3/2  m_j =3/2$	1.96

Within constrained RPA, we estimate  $U = F^0 = 2.2 \text{ eV}$  and  $J = (F^2 + F^4)/14 = 0.3 \text{ eV}$  for our model.

DMFT part  
Performing the DMFT loop



$\text{Sr}_2\text{IrO}_4$  is a Mott insulator with one hole in the  $j_{\text{eff}}=1/2$  spin-orbital. The size of the gap is  $\sim 0.25 \text{ eV}$ .

There is neither a magnetic order, nor an orbital order but a "spin-orbital order".

$\text{Sr}_2\text{IrO}_4$  is a " $j_{\text{eff}}=1/2$  Mott insulator".

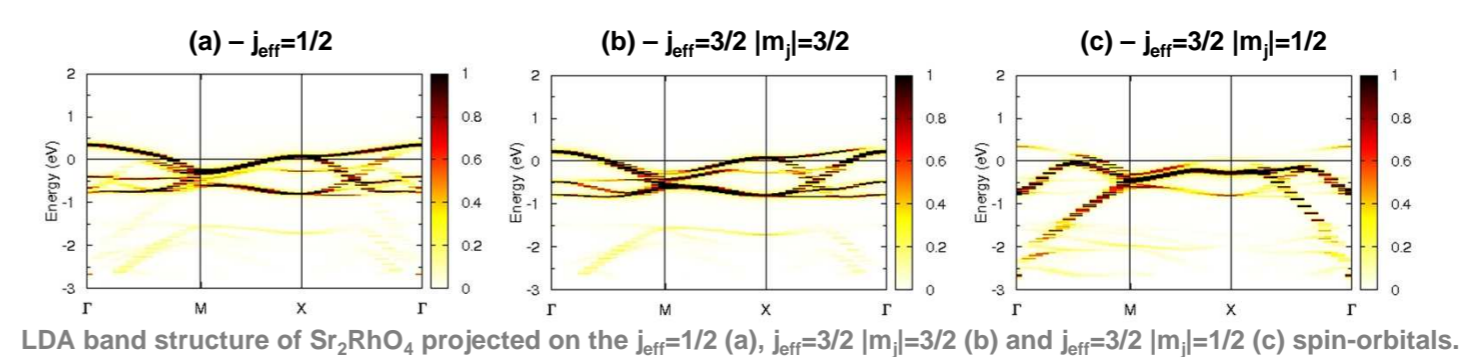
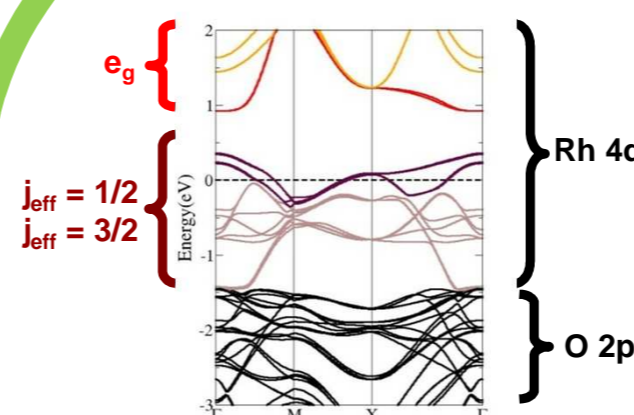
The suppression of spin-orbital fluctuations is a consequence of the combined effect of SOC and the structural distortions.

## Paramagnetic $\text{Sr}_2\text{RhO}_4$ within LDA+SO+DMFT

DFT part  
Solving Kohn-Sham equations

The Kohn-Sham band structures of  $\text{Sr}_2\text{RhO}_4$  and  $\text{Sr}_2\text{IrO}_4$  are similar.

But:  
 ❖ The bandwidths in  $\text{Sr}_2\text{RhO}_4$  are smaller than that in  $\text{Sr}_2\text{IrO}_4$ .  
 ❖ The O-2p bands are 1 eV higher in energy in  $\text{Sr}_2\text{RhO}_4$ .



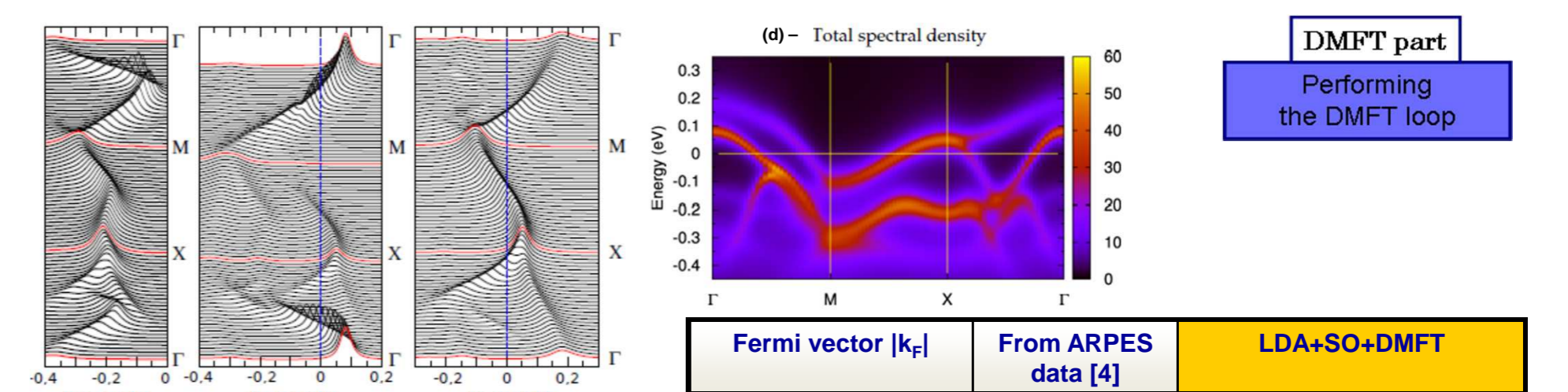
Charge	$\text{Sr}_2\text{RhO}_4$
$j_{\text{eff}}=1/2$	1.40
$j_{\text{eff}}=3/2  m_j =1/2$	1.96
$j_{\text{eff}}=3/2  m_j =3/2$	1.64

$\text{Sr}_2\text{RhO}_4$  is a "three-quarter-filled two-bands" system.

Within constrained RPA, we estimate  $U = F^0 = 1.6 \text{ eV}$  and  $J = (F^2 + F^4)/14 = 0.3 \text{ eV}$  for our model.

Because the screening by the O-2p bands is more efficient in  $\text{Sr}_2\text{RhO}_4$ .

Interfacing  
Constructing projection operators on the correlated states



Fermi vector $ k_F $	From ARPES data [4]	LDA+SO+DMFT
$\alpha$ pocket	$0.17 \text{ \AA}^{-1}$	$0.18-19 \text{ \AA}^{-1}$
$\beta$ pocket ( $\beta_M$ and $\beta_X$ )	$0.66 \text{ \AA}^{-1}$	$0.65-0.69 \text{ \AA}^{-1}$

$\text{Sr}_2\text{RhO}_4$  is a partially spin-orbital polarized correlated metal.  
Our LDA+SO+DMFT calculations are in excellent agreement with ARPES spectra obtained at 10 K [4].

## Conclusion

❖ We have performed complete *ab initio* calculations for  $\text{Sr}_2\text{IrO}_4$  and  $\text{Sr}_2\text{RhO}_4$ .

$\text{Sr}_2\text{IrO}_4$  is a  $j_{\text{eff}}=1/2$  Mott insulator ( $U = 2.2 \text{ eV}$ ,  $J = 0.3 \text{ eV}$ ) and  $\text{Sr}_2\text{RhO}_4$  is a partially spin-orbital polarized metal ( $U = 1.6 \text{ eV}$ ,  $J = 0.3 \text{ eV}$ ).

❖ We have emphasized the interplay of the spin-orbit coupling with electronic Coulomb interactions in 4d and 5d transition metal oxides.

❖ We have performed LDA+SO+DMFT calculations.

Application to doped  $\text{Sr}_2\text{IrO}_4$  or  $\text{Ba}_2\text{IrO}_4$  (search for superconductivity) and, more generally, to other 5d transition metal oxides or compounds.

[1] – Aichhorn et al, Phys Rev B 80, 085101 (2009)  
 [2] – Klein & Terasaki, J. Phys.: Cond. Mat. 20 (2008)  
 [3] – Kim et al, Phys Rev Lett 101, 076402 (2009)  
 [4] – Baumberger et al, Phys Rev Lett 96, 246402 (2006)  
 [5] – Moon et al, Phys. Rev. B 80, 195110 (2009)

[6] – Moon et al, Phys. Rev. B 74, 113104 (2006)  
 [7] – Kim et al, Science 323, 1329 (2009)  
 [8] – Jin et al, Phys. Rev. B 80, 075112 (2009)  
 [9] – Haverkort et al, Phys. Rev. Lett. 101, 026406 (2008)

[10] – Liu et al, Phys. Rev. Lett. 101, 026408 (2008)  
 [11] – Blaha et al, « Wien2k, An Augmented Plane Wave+ Local Orbitals Program for Calculating Crystal Properties » (Karlheinz Schwarz, Tech. Univ. Wien, Austria, 2001)

[12] – Aryasetiawan et al, Phys Rev B 70, 195104 (2004)  
 [13] – Vaugier et al, to be published.  
 [14] – Kotliar & Vollhardt, Physics Today 57, 53 (2004)  
 [15] – Werner et al, Phys. Rev. Lett. 97, 076405 (2006)