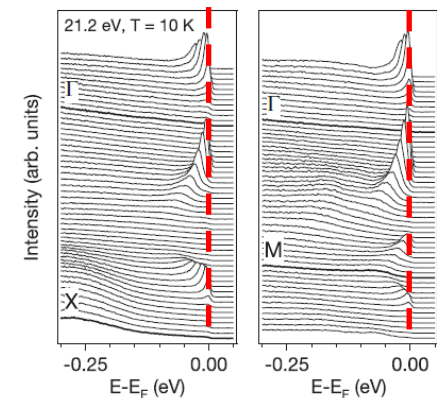
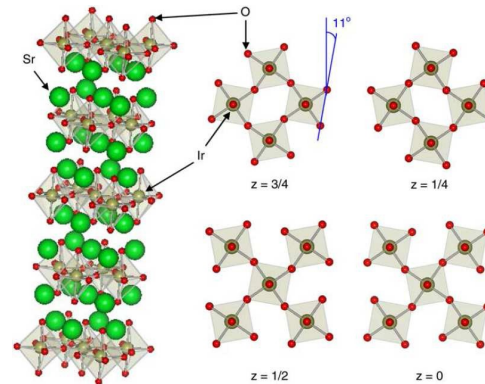
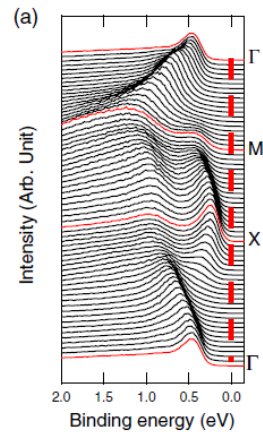
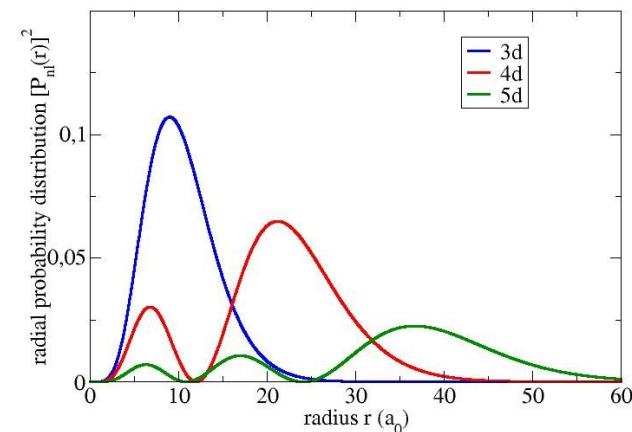
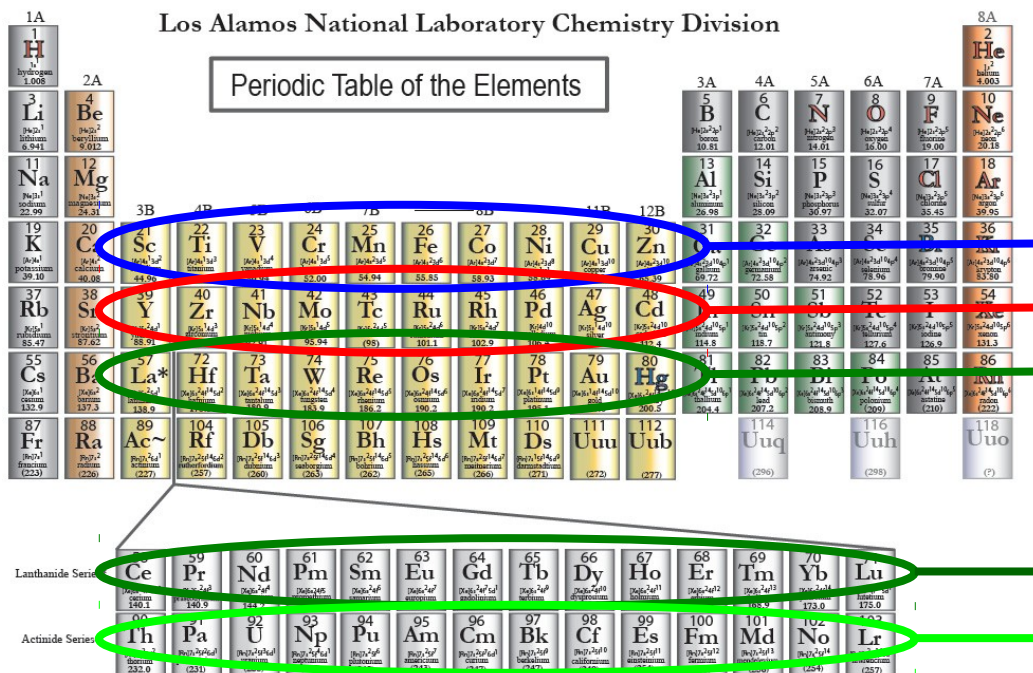


Why is Sr_2IrO_4 insulating and Sr_2RhO_4 is not?

Effects of spin-orbit coupling and structural distortions

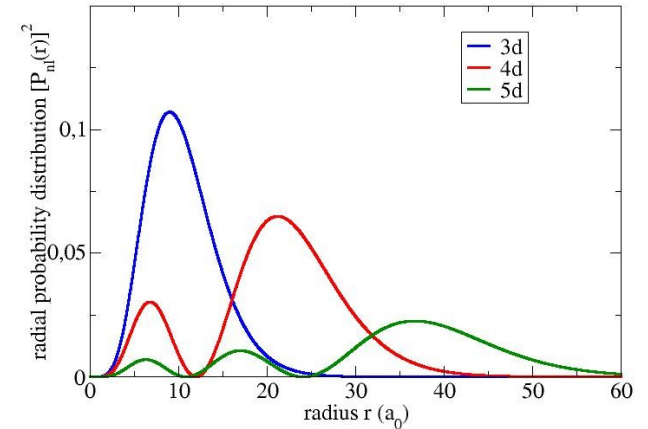




Correlated materials are

- 3d transition metals and their oxides
- Rare earth (Lanthanides 4f) and Actinides (5f)
- 4d transition metal oxides : Sr_2RuO_4 , $SrTcO_3$...

3d	21 Sc [Ar]4s ² 3d ¹ scandium 44.96	22 Ti [Ar]4s ² 3d ² titanium 47.88	23 V [Ar]4s ¹ 3d ⁴ vanadium 50.94	24 Cr [Ar]4s ¹ 3d ⁵ chromium 52.00	25 Mn [Ar]4s ² 3d ⁵ manganese 54.94	26 Fe [Ar]4s ² 3d ⁶ iron 55.85	27 Co [Ar]4s ¹ 3d ⁷ cobalt 58.93	28 Ni [Ar]4s ² 3d ⁸ nickel 58.69	29 Cu [Ar]4s ¹ 3d ¹⁰ copper 63.55	30 Zn [Ar]4s ² 3d ¹⁰ zinc 65.39
4d	39 Y [Kr]5s ² 4d ¹ yttrium 88.91	40 Zr [Kr]5s ² 4d ² zirconium 91.22	41 Nb [Kr]5s ¹ 4d ⁴ niobium 92.91	42 Mo [Kr]5s ¹ 4d ⁵ molybdenum 95.94	43 Tc [Kr]5s ² 4d ⁵ technetium (98)	44 Ru [Kr]5s ¹ 4d ⁶ ruthenium 101.1	45 Rh [Kr]5s ¹ 4d ⁷ rhodium 102.9	46 Pd [Kr]4d ¹⁰ palladium 106.4	47 Ag [Kr]5s ¹ 4d ¹⁰ silver 107.9	48 Cd [Kr]5s ² 4d ¹⁰ cadmium 112.4
5d	57 La* [Xe]5s ¹ lanthanum 138.9	72 Hf [Xe]6s ² 4f ¹⁴ 5d ² hafnium 178.5	73 Ta [Xe]6s ² 4f ¹⁴ 5d ³ tantalum 180.9	74 W [Xe]6s ² 4f ¹⁴ 5d ⁴ tungsten 183.9	75 Re [Xe]6s ² 4f ¹⁴ 5d ⁵ rhenium 186.2	76 Os [Xe]6s ² 4f ¹⁴ 5d ⁶ osmium 190.2	77 Ir [Xe]6s ¹ 4f ¹⁴ 5d ⁷ iridium 190.2	78 Pt [Xe]6s ¹ 4f ¹⁴ 5d ⁹ platinum 195.1	79 Au [Xe]6s ¹ 4f ¹⁴ 5d ¹⁰ gold 197.0	80 Hg [Xe]6s ² 4f ¹⁴ 5d ¹⁰ mercury 200.5



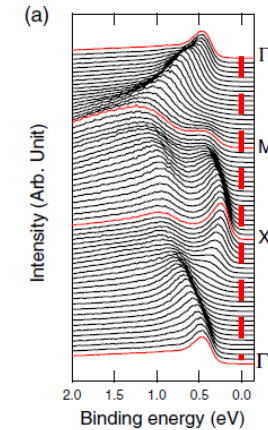
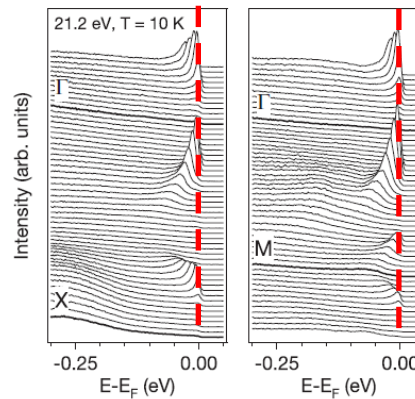
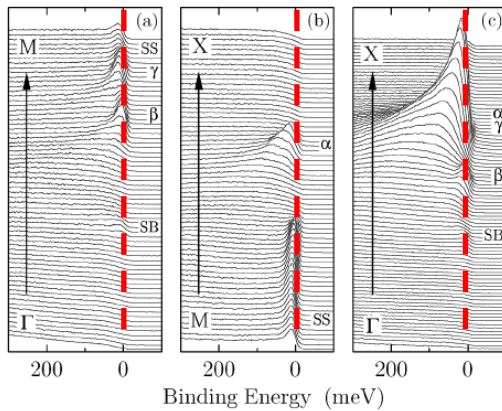
4d states are considered as **less** localized and correlated **than** 3d
 5d states are considered as **less** localized and correlated **than** 4d

Angle Resolved Photo-Emission Spectroscopy (ARPES) spectra for :

Sr2RuO4 (4d⁴)

Sr2RhO4 (4d⁵)

Sr2IrO4 (5d⁵)

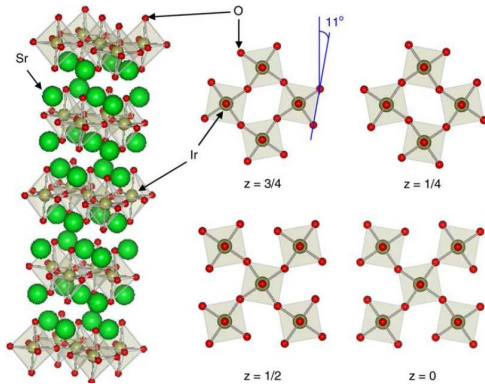


Damascelli et al,
 Phys Rev Lett 85, 5194 (2000)

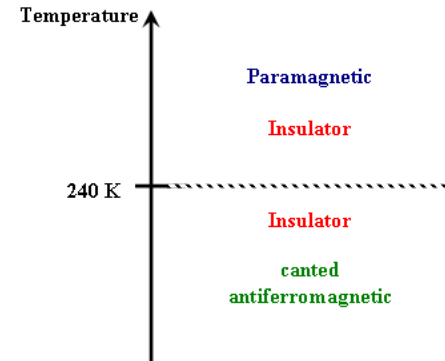
Baumberger et al,
 Phys Rev Lett 96, 246402 (2006)

Kim et al,
 Phys Rev Lett 101, 076402 (2009)

- A structure similar to La_2CuO_4 or Sr_2RuO_4 but with distortions :
 IrO_6 octahedra are rotated around the z axis by about 11° .



Klein & Terasaki, J. Phys.: Cond. Mat. 20 (2008)

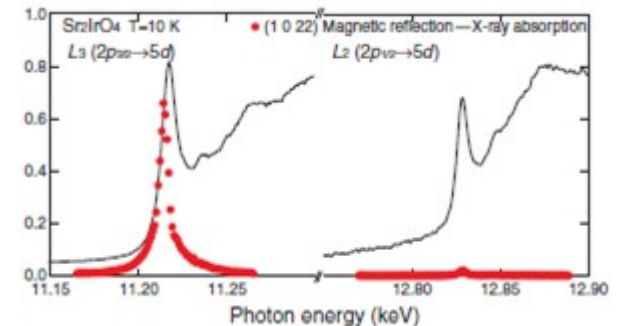


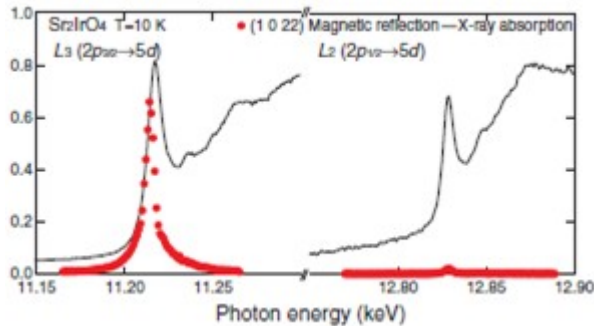
- An insulator at all temperatures but Ir atoms accommodate 5 electrons.

An optical gap at 300 K (room temperature) of 0.26 eV. *Moon et al, Phys Rev B 80, 195110 (2009)*

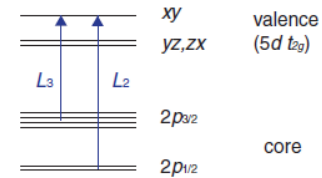
- An important role of the spin-orbit coupling ($\zeta_{SO} \sim 0.4$ eV) according to resonant X-ray scattering.

Kim et al, Science 323, 1329 (2009)





S=1/2 Model

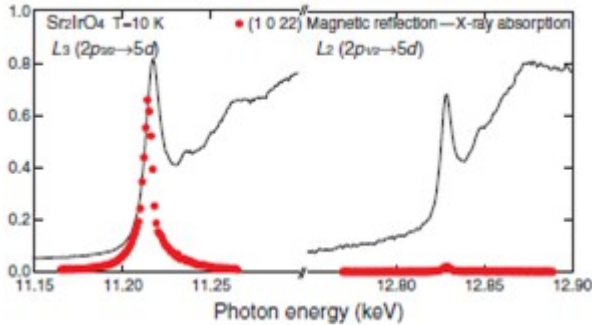
1:1 intensity ratio at L₃ and L₂ $J_{\text{eff}}=1/2$ Modelresonance only at L₃ edge*Kim et al, Phys Rev Lett 101, 076402 (2009)**Kim et al, Science 323, 1329 (2009)*

The spin-orbit interaction is a relativistic correction to the Schrödinger equation.

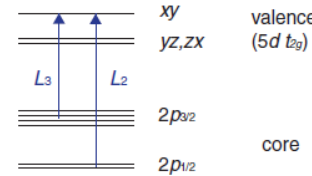
$$H_{SO} = \frac{\hbar^2}{2m_0^2 c^2} \mathbf{S} \cdot [\nabla V(\mathbf{r}) \times \mathbf{p}] \quad \text{and for an atom:} \quad H_{SO} = \frac{1}{2m_0^2 c^2} \frac{1}{r} \frac{dV}{dr} (\mathbf{L} \cdot \mathbf{S}) = \zeta_{SO} \mathbf{L} \cdot \mathbf{S}$$

The energy splitting induced by the spin-orbit coupling scales roughly as Z^4 .

Iron (Fe)	(Z= 26)	$\zeta_{SO} \sim 0.050 \text{ eV}$
Copper (Cu)	(Z= 29)	$\zeta_{SO} \sim 0.103 \text{ eV}$
Ruthenium (Ru)	(Z= 44)	$\zeta_{SO} \sim 0.161 \text{ eV}$
Rhodium (Rh)	(Z= 45)	$\zeta_{SO} \sim 0.191 \text{ eV}$
Iridium (Ir)	(Z= 77)	$\zeta_{SO} \sim 0.4 \text{ eV}$
Bismuth (Bi)	(Z= 83)	$\zeta_{SO} \sim 1.5 \text{ eV}$



S=1/2 Model
1:1 intensity ratio at L₃ and L₂

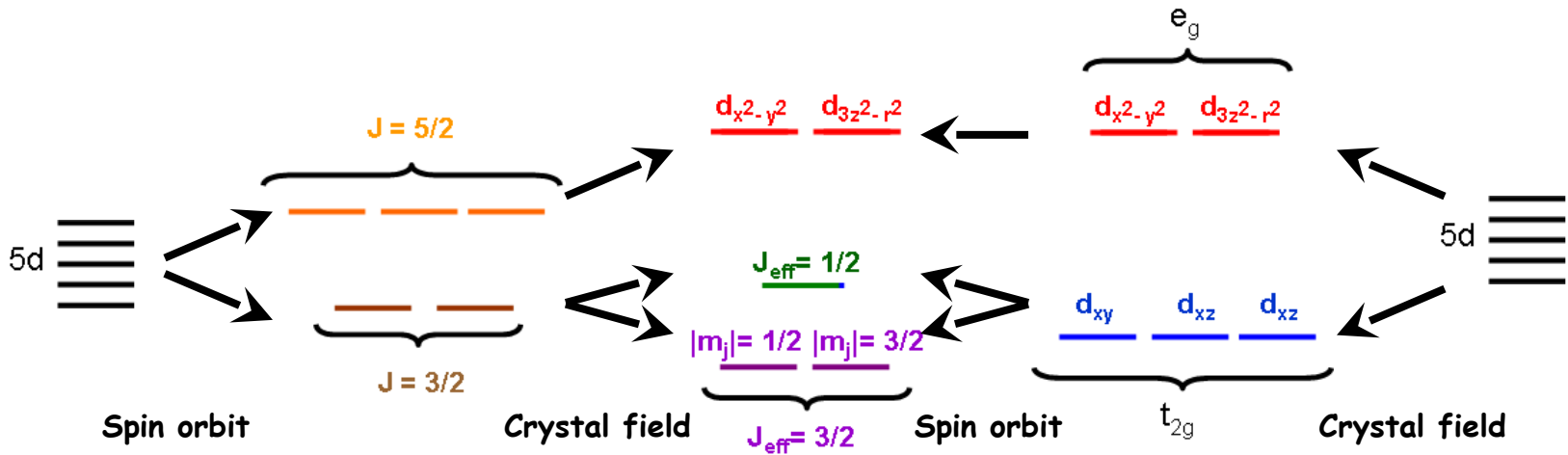


J_{eff}=1/2 Model
resonance only at L₃ edge

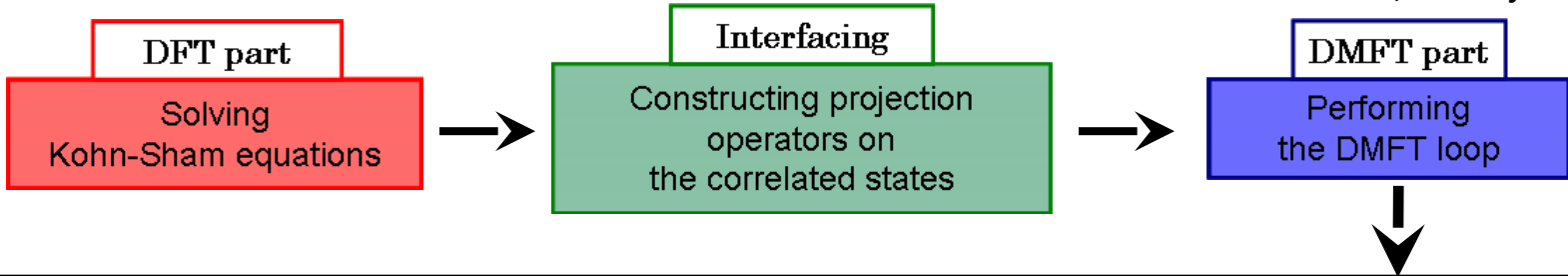


Kim et al, Phys Rev Lett 101, 076402 (2009)
Kim et al, Science 323, 1329 (2009)

Because of the crystal field induced by the oxygen, the 5d states are split into e_g, j_{eff}=1/2 and j_{eff}=3/2 multiplets.



Lichtenstein & Katsnelson, *PRB* 57, 6884 (1998) ; Anisimov et al, *J. Phys. Cond Mat.* 9, 7359 (1997)
 K. Held, *Adv. Phys.* 56, 829 (2007)



Time →

|0> |↑> |↑↓>

Electron reservoir

v_v *v_v*

e_f *e_f*

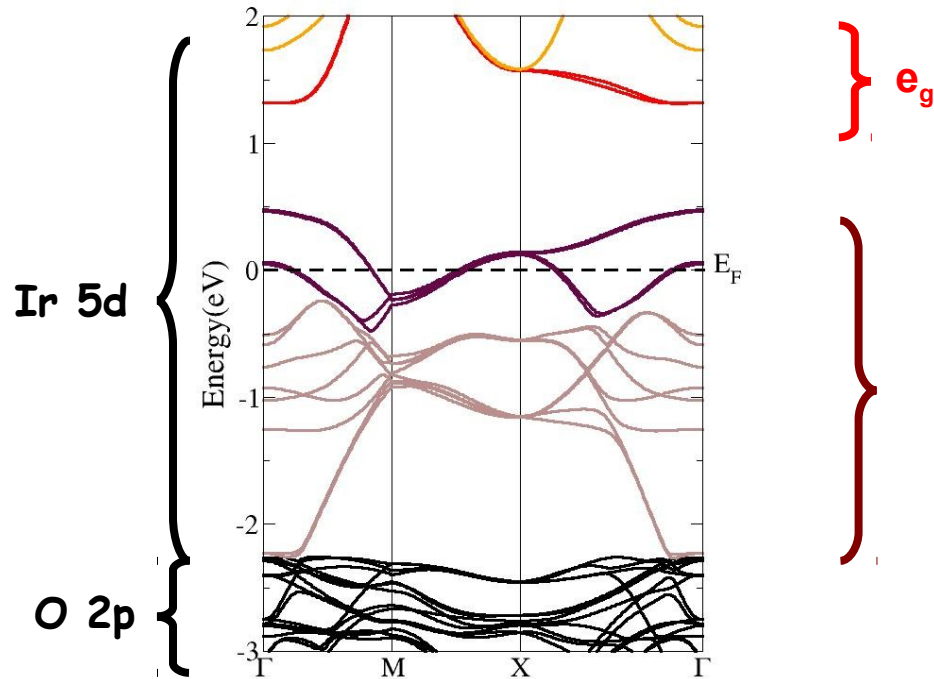
The lattice problem is mapped onto a local impurity problem (AIM).

Solution of the AIM by Continuous Time QMC.
 Parameters from constrained RPA calculations.

Georges et al, Rev Mod Phys 68, 13 (1996)

$$H = U \sum_m n_{m\uparrow} n_{m\downarrow} + U' \sum_{m>n,\sigma} n_{m\sigma} n_{n-\sigma} + (U' - J) \sum_{m>n,\sigma} n_{m\sigma} n_{n\sigma}$$

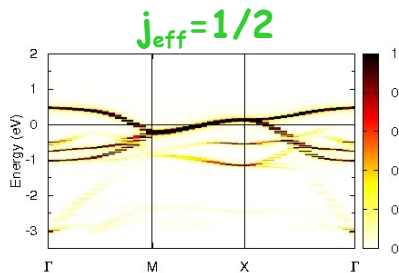
Self consistent implementation in Wien2K: M. Aichhorn et al., *PRB* 2009, *PRB* 2011



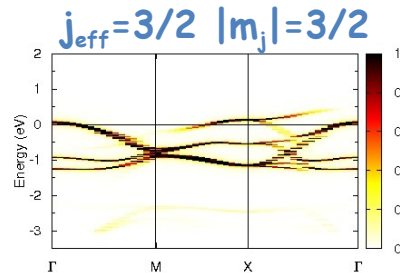
4 bands cross the Fermi level:
a **metallic** Kohn-Sham band structure

$$j_{\text{eff}} = 1/2$$

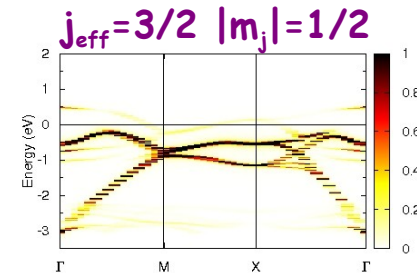
$$j_{\text{eff}} = 3/2$$



$$n = 1.14$$



$$n = 1.96$$



$$n = 2.00$$

Distortions and *SO* coupling produces an effective one-band problem

Interaction parameters estimated by cRPA:

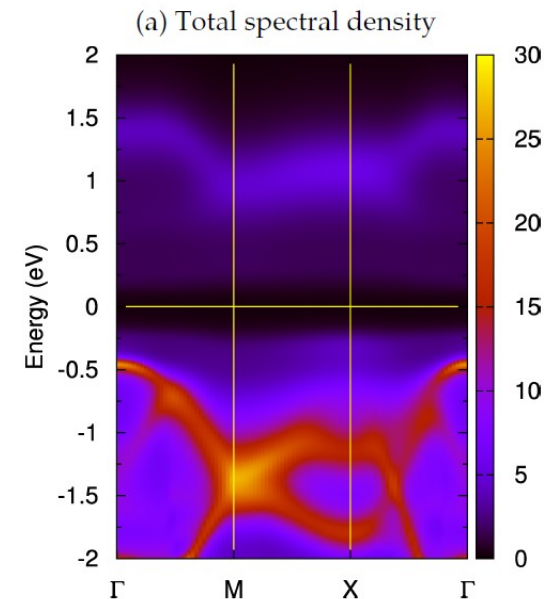
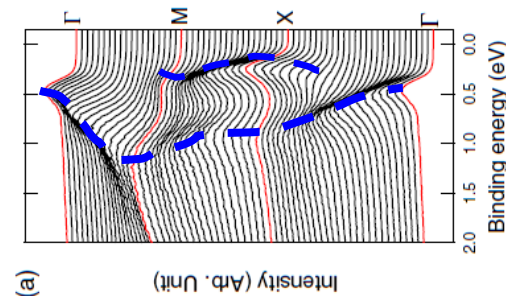
$$U = 2.2 \text{ eV} \quad J = 0.3 \text{ eV}$$



Insulating state with a gap of about 0.26 eV

Comparison with experimental spectral function
obtained with ARPES (at 100K) :

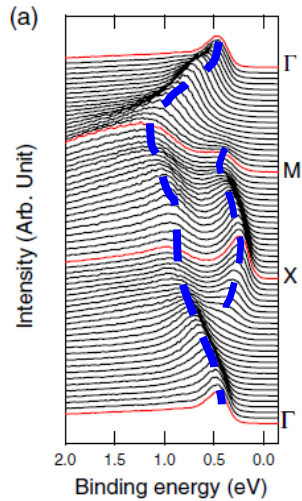
Kim et al, Phys Rev Lett 101, 076402 (2009)



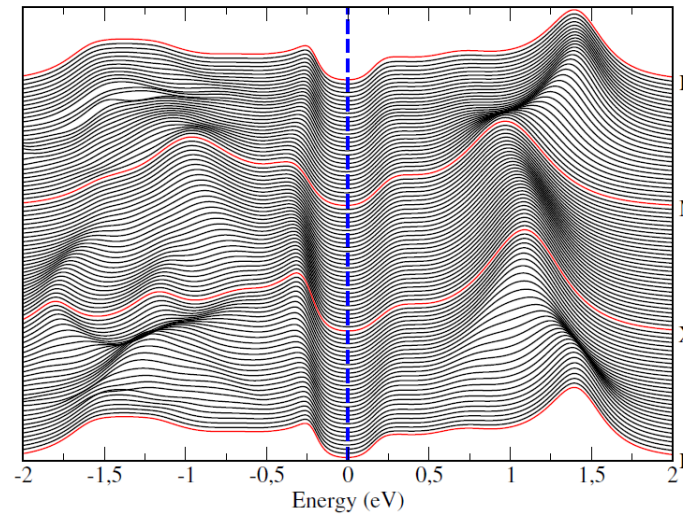
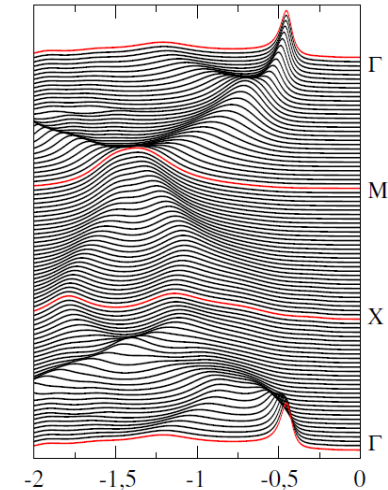
C. Martins, M. Aichhorn, L. Vaugier, and S. Biermann, PRL 107, 266404 (2011)

Experimental spectrum

Kim et al, Phys Rev Lett 101, 076402 (2009)



Calculated orbitally resolved spectral density

Orbital $j_{\text{eff}}=1/2$ Orbital $j_{\text{eff}}=3/2$ 

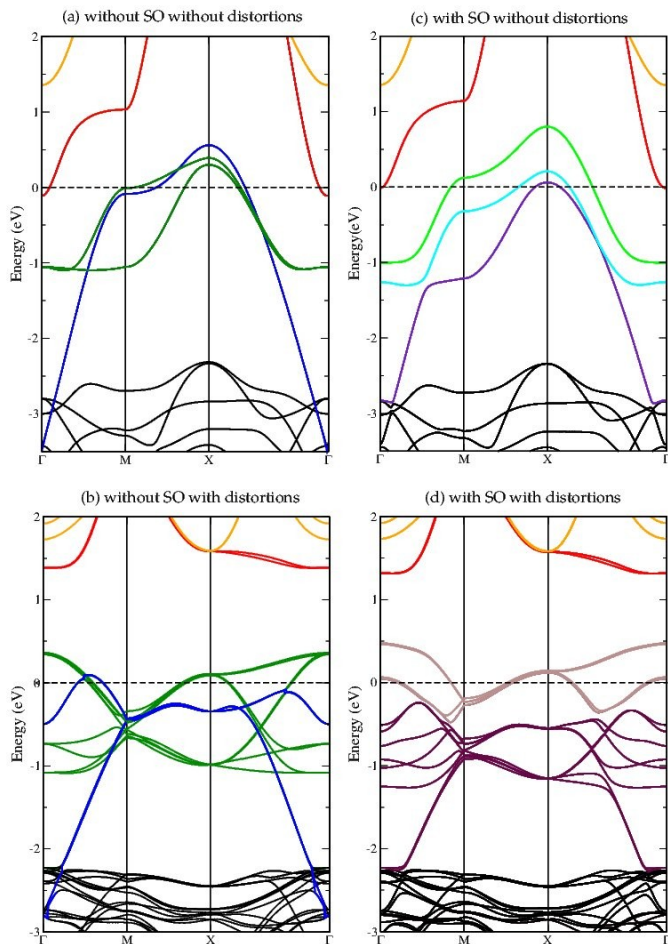
Sr_2IrO_4 is a $j_{\text{eff}}=1/2$ Mott insulator. ($U=2.2$ eV and $J=0.3$ eV)

neither a magnetic order, nor an orbital order but a “spin-orbital order”

The suppression of spin-orbital fluctuations is a consequence of the combined effect of :

- Spin orbit coupling ($\zeta_{\text{SO}} \sim 0.4$ eV)
- the structural distortions (rotation of 11° of the IrO_6 octahedra)

10



Without structural distortions:
A $j_{\text{eff}}=1/2$ Mott insulator is obtained for:

$$U > 3.0 \text{ eV}$$

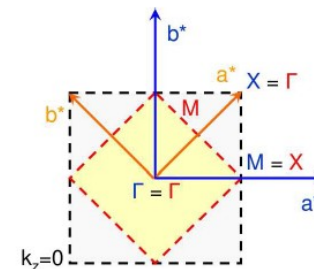
--- dx^2-y^2

--- $d3z^2-r^2$

--- $J_{\text{eff}}=1/2$

--- $J_{\text{eff}}=3/2 \quad |m_j|=3/2$

--- $J_{\text{eff}}=3/2 \quad |m_j|=1/2$

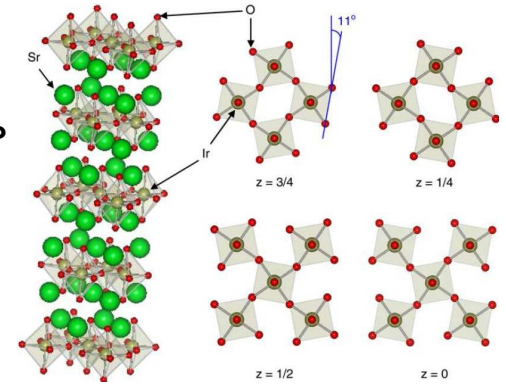


- The same crystal structure as Sr_2IrO_4 :

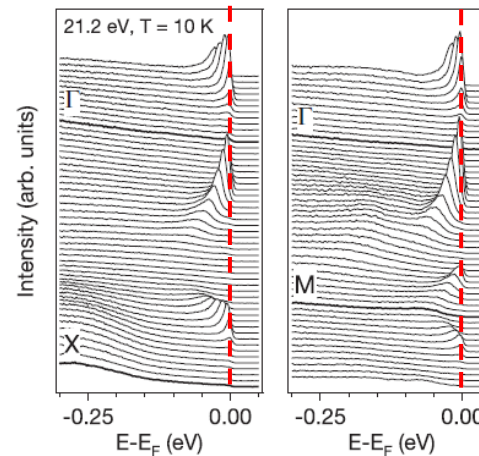
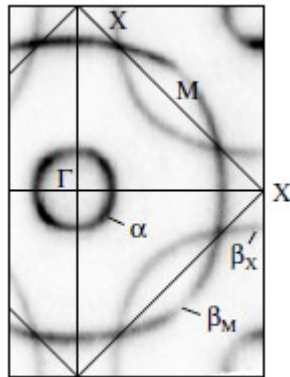
RhO_6 octahedra are rotated around the z axis by about 11°

$$a(\text{Sr}_2\text{RhO}_4) = 5.44 \text{ \AA} ; c(\text{Sr}_2\text{RhO}_4) = 25.75 \text{ \AA}$$

$$a(\text{Sr}_2\text{IrO}_4) = 5.49 \text{ \AA} ; c(\text{Sr}_2\text{IrO}_4) = 25.77 \text{ \AA}$$

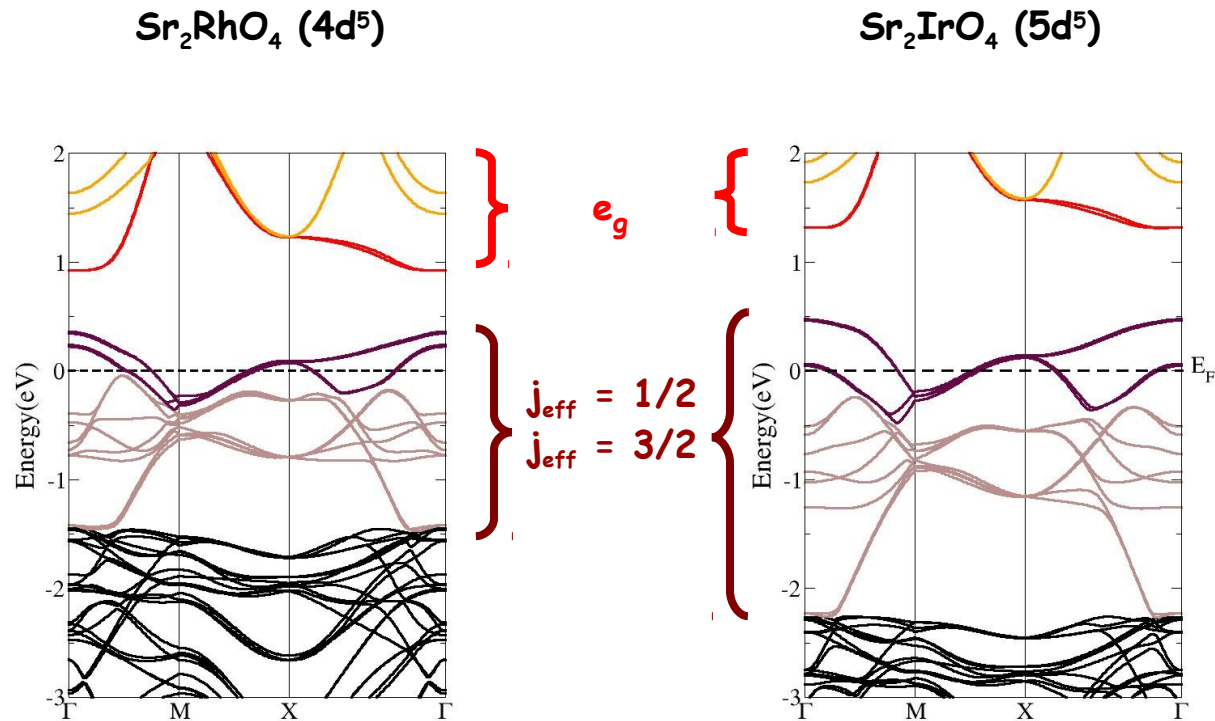


- A paramagnetic metal down to 36 mK. *Moon et al. Phys Rev B 74, 113104 (2006)*



*Baumberger et al.
Phys Rev Lett 96, 246402 (2006)*

- Spin-orbit coupling, electronic correlations and structural distortions are necessary to reproduce the Fermi surface within LDA+SO+U. *Haverkort et al, Phys Rev Lett 101, 026406 (2008)*
Liu et al, Phys Rev Lett 101, 026408 (2008)



- The Kohn-Sham band structure of Sr₂RhO₄ and Sr₂IrO₄ are qualitatively similar.
 - But :
 - the bandwidths of Sr₂RhO₄ are smaller than that of Sr₂IrO₄.
 - the spin-orbit splitting in Sr₂RhO₄ is half of that of Sr₂IrO₄.

Rhodium (Rh)

 $\zeta_{SO} \sim 0.191 \text{ eV}$

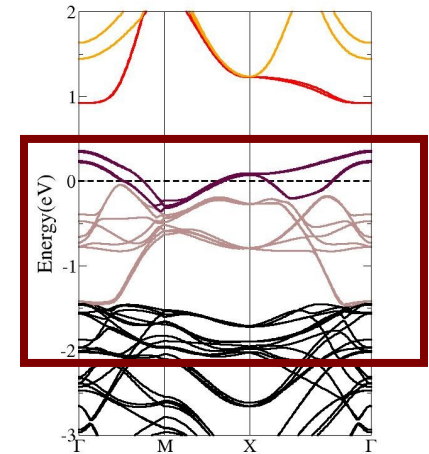
Iridium (Ir)

 $\zeta_{SO} \sim 0.4 \text{ eV}$

Sr_2RhO_4 has identical crystal structure as Sr_2IrO_4

But smaller SO coupling 0.2

Charge repartition between the $j_{\text{eff}}=1/2$ state and the two $j_{\text{eff}}=3/2$ states are different.

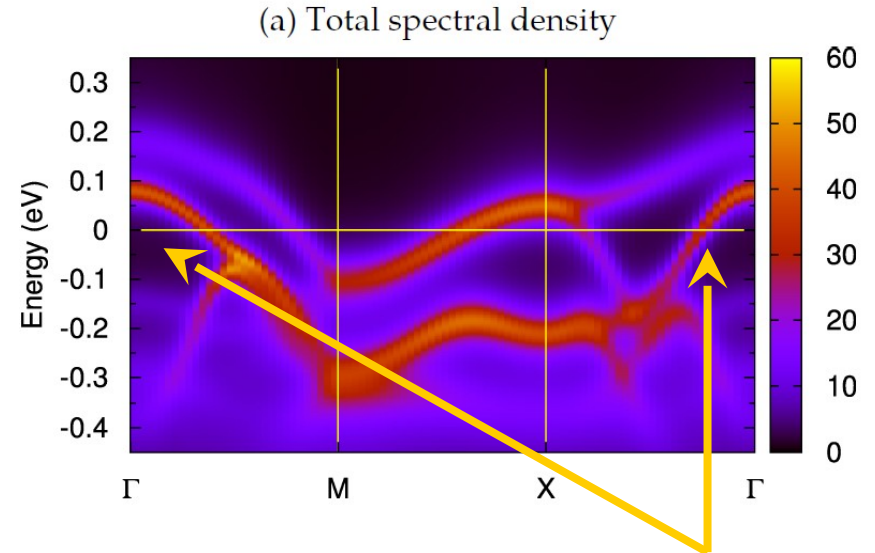
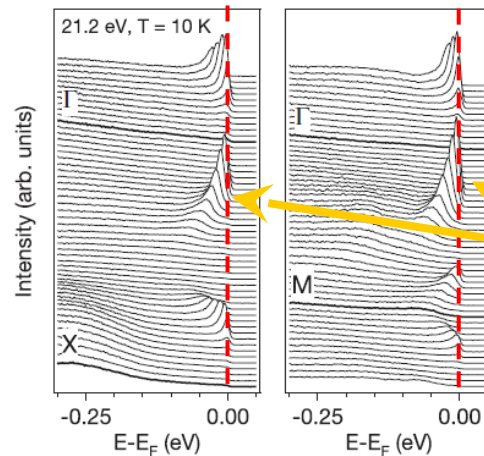
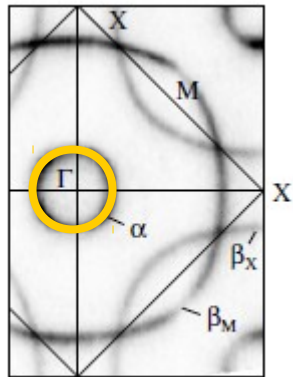


Charge	Sr_2IrO_4	Sr_2RhO_4
$j_{\text{eff}}=1/2$	1.14	1.40
$j_{\text{eff}}=3/2 m_j =1/2$	2.00	1.96
$j_{\text{eff}}=3/2 m_j =3/2$	1.96	1.64

Sr_2RhO_4 is a "three-quarter-filled two-bands" system

Sr₂RhO₄ is a **paramagnetic correlated metal**.
($U = 1.6$ eV and $J = 0.3$ eV)

Comparison with experimental spectral function
obtained with ARPES (at 10K) :



α pocket : $0.18-19 \text{ \AA}^{-1}$

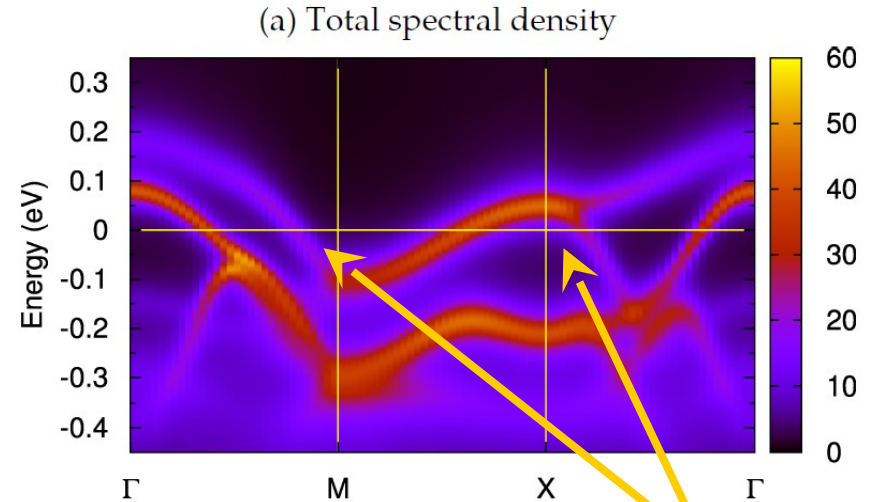
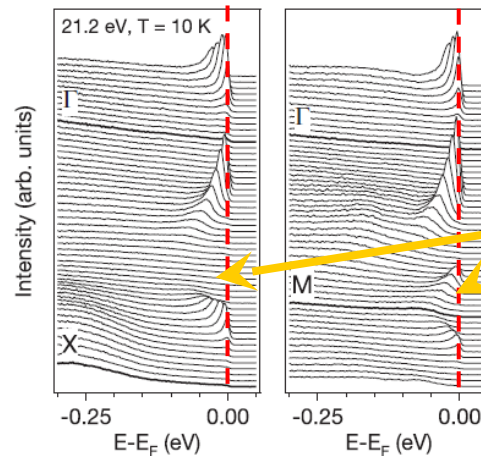
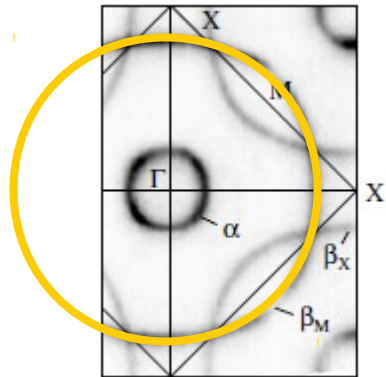
α pocket : 0.17 \AA^{-1}

Baumberger et al., Phys Rev Lett 96, 246402 (2006)

Charge	LDA	LDA+DMFT
$j_{\text{eff}} = 1/2$	1.40	1.26
$j_{\text{eff}} = 3/2 \quad m_j = 1/2$	1.96	2.00
$j_{\text{eff}} = 3/2 \quad m_j = 3/2$	1.64	1.70

Sr₂RhO₄ is a **paramagnetic correlated metal**.
($U = 1.6$ eV and $J = 0.3$ eV)

Comparison with experimental spectral function
obtained with ARPES (at 10K) :



β_M and β_X pocket :
 0.66 \AA^{-1}

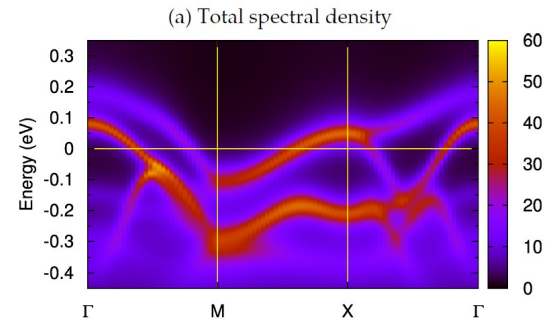
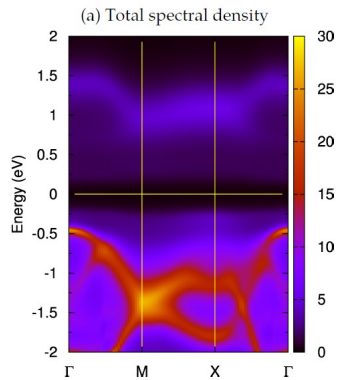
β_M and β_X pocket :
 $0.65\text{-}0.69 \text{ \AA}^{-1}$

Baumberger et al., Phys Rev Lett 96, 246402 (2006)

Charge	LDA	LDA+DMFT
$j_{\text{eff}} = 1/2$	1.40	1.26
$j_{\text{eff}} = 3/2 \quad m_j = 1/2$	1.96	2.00
$j_{\text{eff}} = 3/2 \quad m_j = 3/2$	1.64	1.70

Ab-initio (LDA+SO+DMFT) calculations for Sr_2IrO_4 and Sr_2RhO_4 show that

- Sr_2IrO_4 is a $j_{\text{eff}} = 1/2$ Mott insulator.
- Sr_2RhO_4 is a partially spin-orbital polarized metal.



Difference due to different strength of Spin-Orbit coupling!



Cyril Martins



Loig Vaugier



Silke Biermann