Strong Correlations and Spin-Orbit Coupling

From Models to Iridates and Osmates

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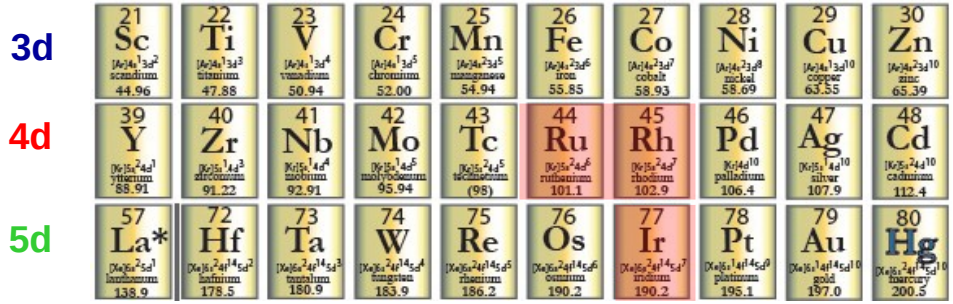
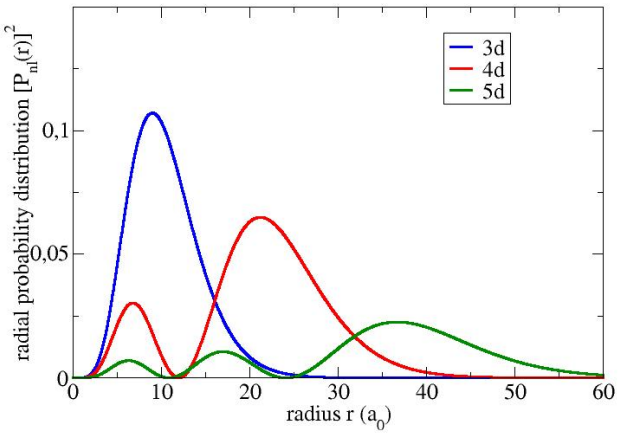
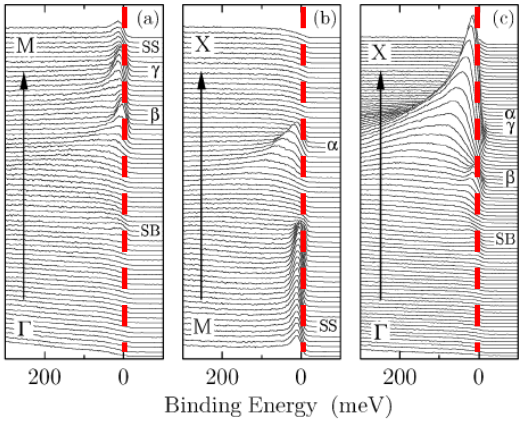


4d states are less localized/correlated than 3d
5d states are less localized/correlated than 4d



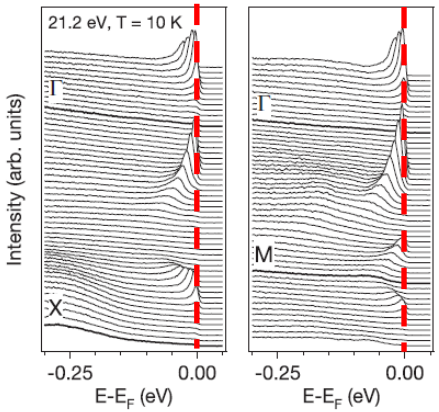
Angle Resolved Photo-Emission Spectroscopy (ARPES):

Sr_2RuO_4 ($4d^4$)



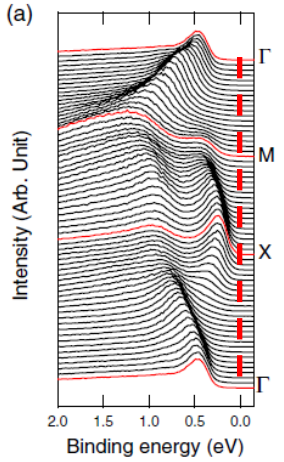
Damascelli *et al,*
Phys Rev Lett 85, 5194 (2000)

Sr_2RhO_4 ($4d^5$)



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

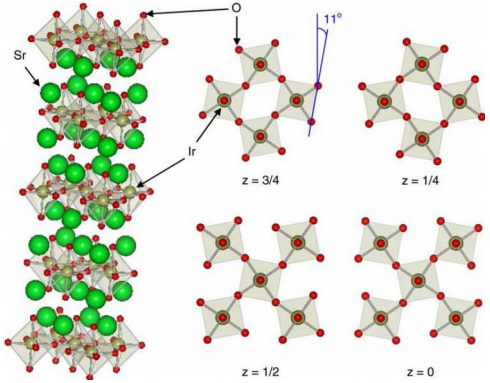
Sr_2IrO_4 ($5d^5$)



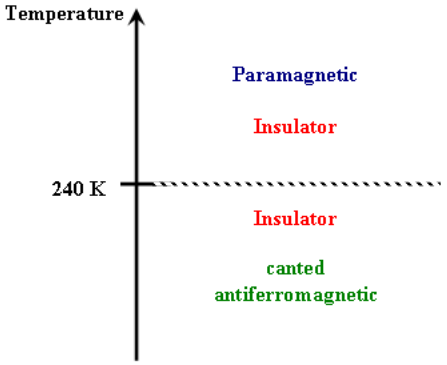
Kim *et al*,
Phys Rev Lett 101, 076402 (2009)

Sr_2IrO_4: Structure similar to La_2CuO_4 but with distortions

IrO_6 octahedra are rotated around the z axis by about 11°



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



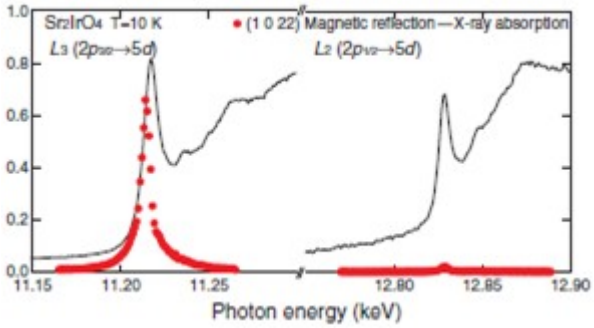
Moon *et al*, Phys Rev B 80, 195110 (2009)

Insulator at all temperatures, but open Ir d^5 shell.

Optical gap at 300 K (room temperature) of 0.26 eV.

Important role of the spin-orbit coupling

$\zeta_{SO} \sim 0.4$ eV

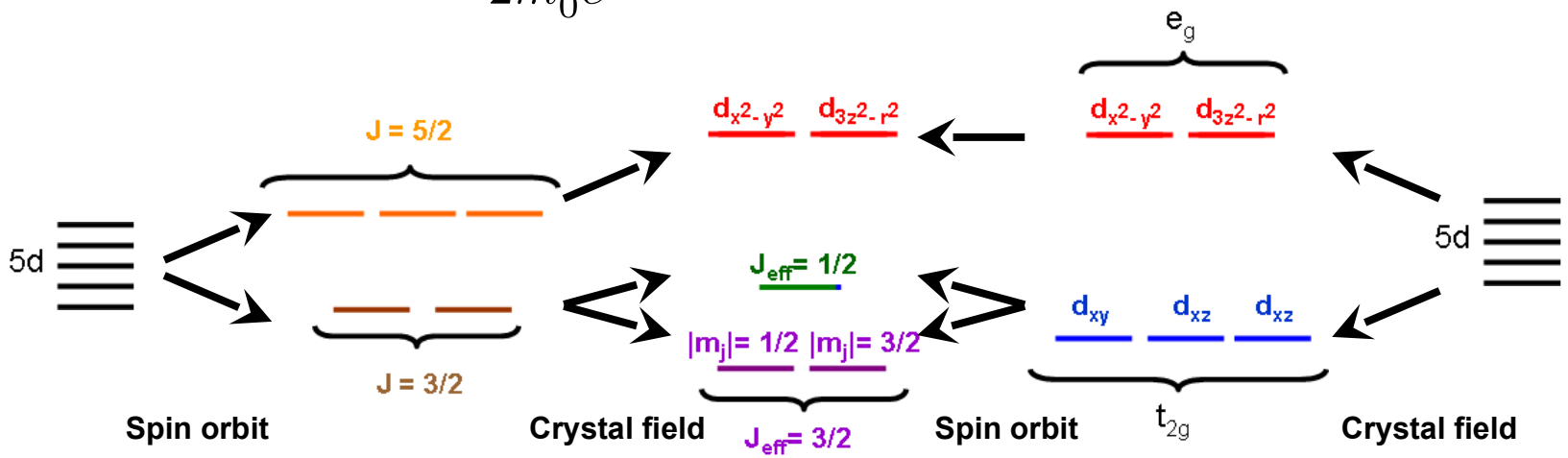


Kim *et al*, Science 323, 1329 (2009)

Spin-Orbit Coupling and local Hamiltonian

Relativistic correction of Schrödinger equation

$$H_{SO} = \frac{\hbar^2}{2m_0^2c^2}\mathbf{S}\cdot[\nabla V(\mathbf{r})\times\mathbf{p}] = \zeta_{SO}(\mathbf{L}\cdot\mathbf{S})$$



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

A model approach: DMFT on a Bethe lattice

Kanamori 3-band Hamiltonian

$$H_I = \sum_i U n_{i\uparrow} n_{i\downarrow} + (U-2J) \sum_{i \neq j} n_{i\uparrow} n_{j\downarrow} + (U-3J) \sum_{i<j,\sigma} n_{i\sigma} n_{j\sigma}$$
$$+ J \sum_{i \neq j} c^\dagger_{i\uparrow} c^\dagger_{j\downarrow} c_{i\downarrow} c_{j\uparrow} + J \sum_{i \neq i} c^\dagger_{i\uparrow} c^\dagger_{i\downarrow} c_{j\downarrow} c_{j\uparrow}$$

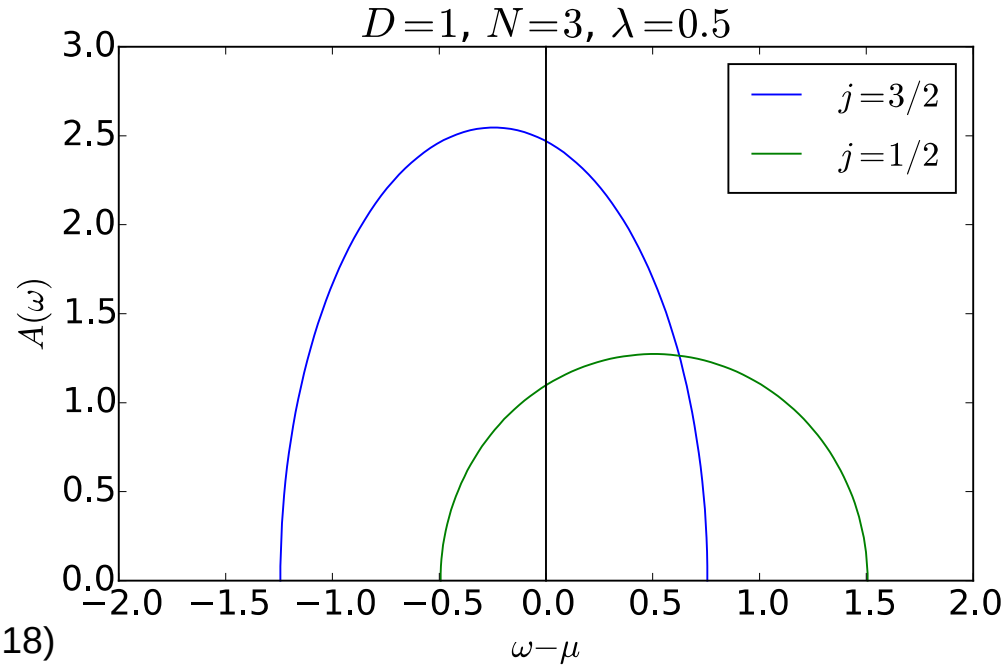
SOC Hamiltonian

$$H_{SO} = \lambda \mathbf{L}_{t2g} \cdot \mathbf{S}$$

Hamiltonian diagonal in *j*-basis

Weak sign problem in CTHYB

Bethe lattice



R. Triebl, … M. Aichhorn, PRB 98, 205128 (2018)

Atomic limit: the *j*-basis

Diagonalization leads to *j*-basis (only *j* = ½ terms):

$$|j=\frac{1}{2},m_j=-\frac{1}{2}\rangle = \frac{1}{\sqrt{3}}|d^{\downarrow}_{xy}\rangle + \frac{i}{\sqrt{3}}|d^{\uparrow}_{xz}\rangle - \frac{1}{\sqrt{3}}|d^{\uparrow}_{yz}\rangle$$

$$|j=\frac{1}{2},m_j=\frac{1}{2}\rangle = \frac{1}{\sqrt{3}}|d^{\uparrow}_{xy}\rangle + \frac{i}{\sqrt{3}}|d^{\downarrow}_{xz}\rangle + \frac{1}{\sqrt{3}}|d^{\downarrow}_{yz}\rangle$$

Advantage: Single-particle part of hamiltonian is diagonal

Disadvantage: Interaction part becomes involved

Pre-factors of interaction terms:

	one orbital	opp. spin	aligned spin
Cubic, crystal field	*U*	*U* – 2 *J*	*U* – 3 *J*
j-basis	*U* – 4/3 *J*	*U* – 7/3 *J*	*U* – 7/3 *J*

Atomic limit and the Crystal-Field Analogue

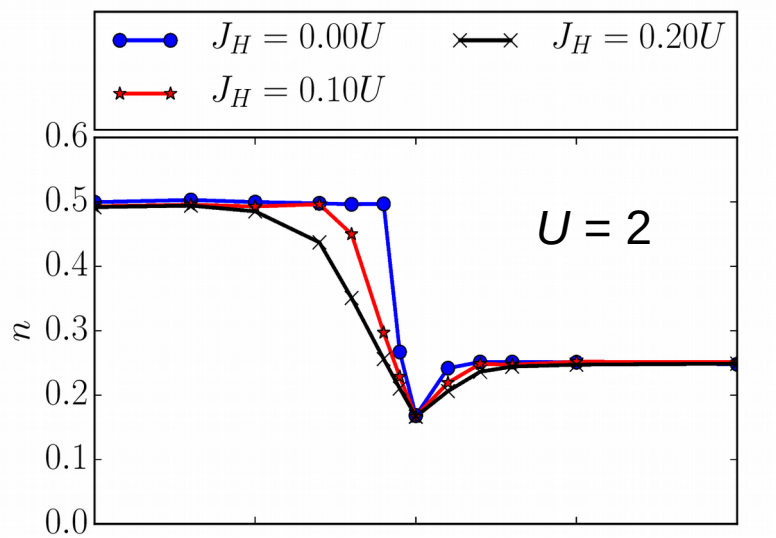
Limit of large SOC / Crystal field:

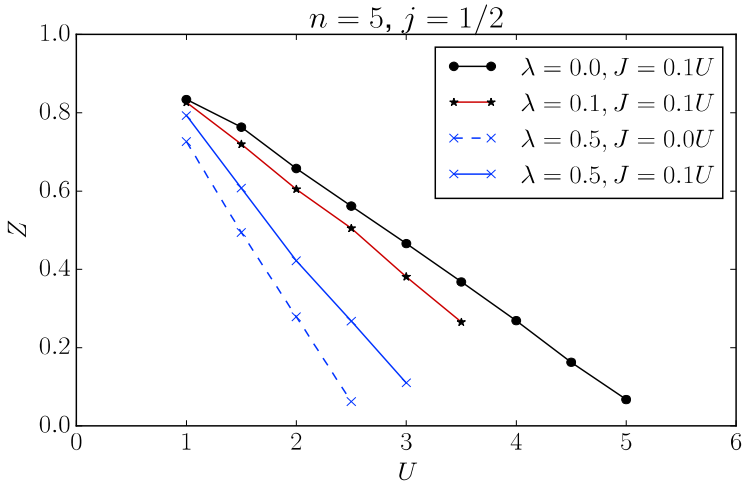
N	SOC	Crystal field
1	$U - 7/3\ J$	$U - 3\ J$
2	$U - J$	$U + J$
3	$U - 7/3\ J$	$U - 3\ J$
4	$U - 3\ J + 3/2\ \lambda$	$U - 5\ J + 3/2\ \Delta$
5	$U - 4/3\ J$	U

Qualitative differences for $N = 2$ and $N = 5$!

To what extent is this difference reflected in a true calculation?

The easy cases: 1 electron or 1 hole





Main effect:

Reduction of number of orbitals

Large SOC limit:

$N = 1$: One electron in two bands

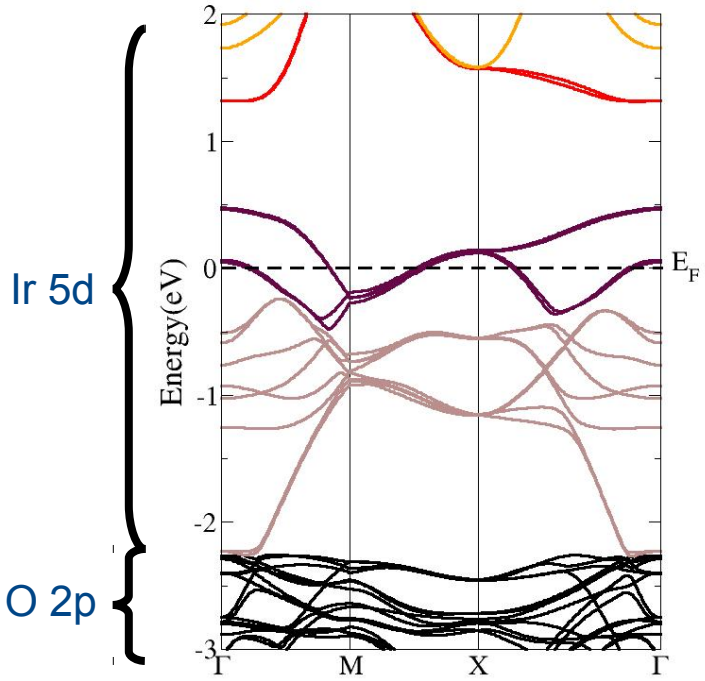
$N = 5$: One electron in one band

Hunds coupling prevents polarization

Sizable Effect for moderate SOC strength!

Depends on J due to transformation of hamiltonian!

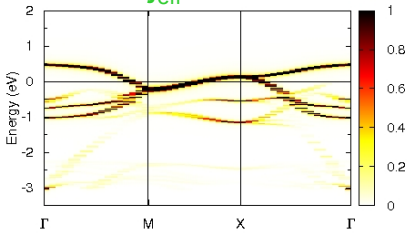
1 hole: Iridate Sr_2IrO_4



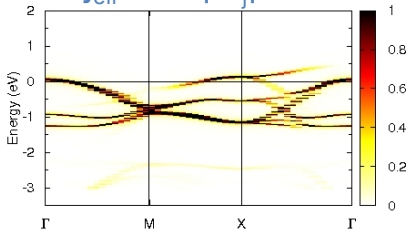
e_g

j_{eff} = 1/2
j_{eff} = 3/2

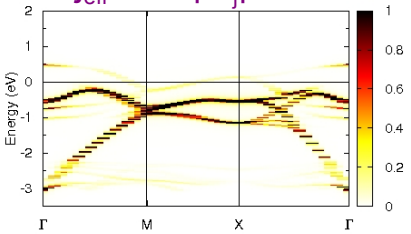
4 bands at Fermi level:
metallic Kohn-Sham band structure



***n* = 1.14**



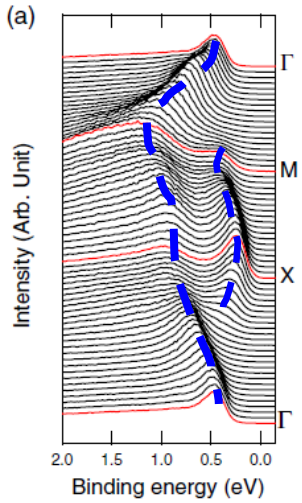
***n* = 1.96**



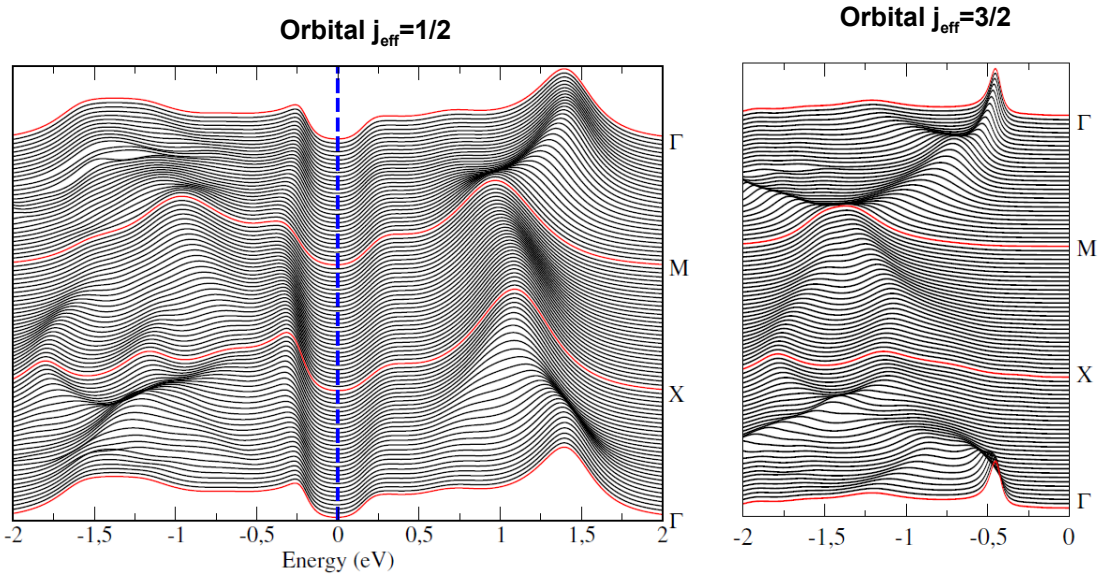
***n* = 2.00**

Experimental spectrum

Kim *et al*, PRL 101, 076402 (2009)



DMFT, orbitally resolved spectral density



C. Martins, M. Aichhorn, et al., PRL 107, 266404 (2012)

Sr_2IrO_4 is a j_{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 due to:

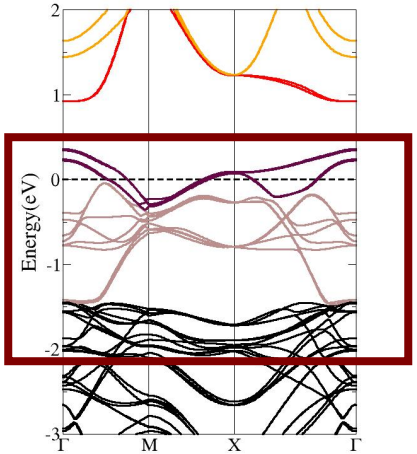
- Spin orbit coupling ($\zeta_{SO} \sim 0.4$ eV)
- the structural distortions (rotation of 11° of the IrO_6 octahedra)

Iridate vs. Rhodate: Effect of spin-orbital polarization

Sr_2RhO_4 has identical crystal structure as Sr_2IrO_4

But smaller SO coupling 0.2

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



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

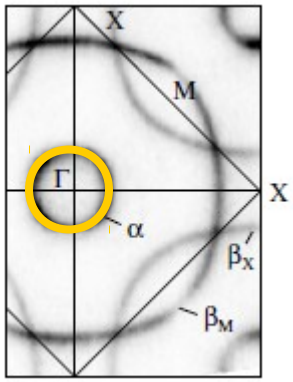
Sr_2RhO_4 is a "three-quarter-filled two-bands" system

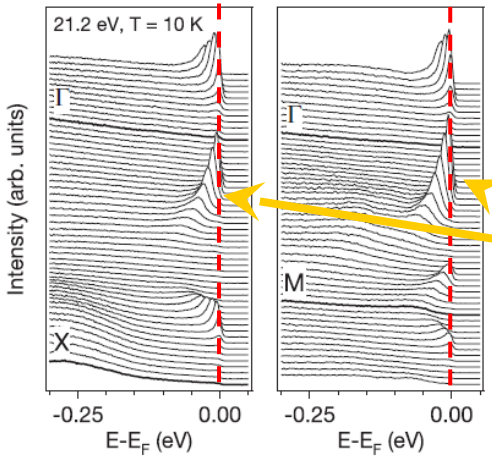
Sr_2RhO_4 is a METAL!

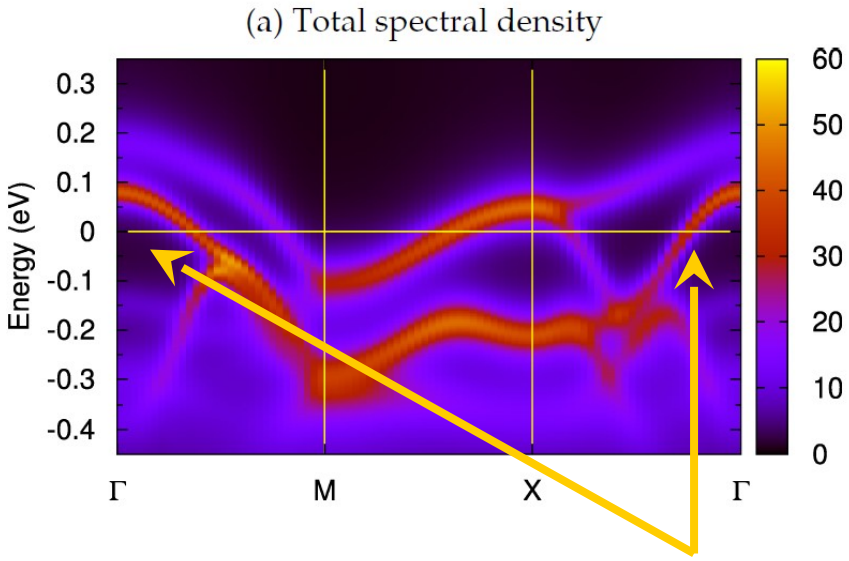
Iridate vs. Rhodate: Effect of spin-orbital polarization

Sr_2RhO_4 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 : 0.17 Å^{-1}

α pocket : 0.18-19 Å^{-1}

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

C. Martins, M. Aichhorn et al., JPCM 2017
PsiK highlight of the month, Nov 2016

Iridate vs. Rhodate: Effect of spin-orbital polarization

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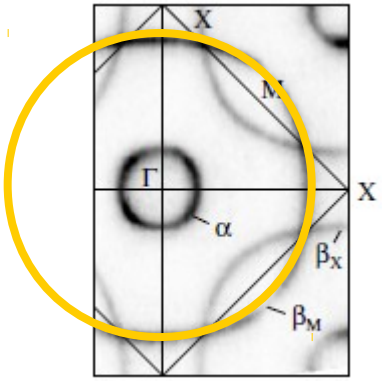
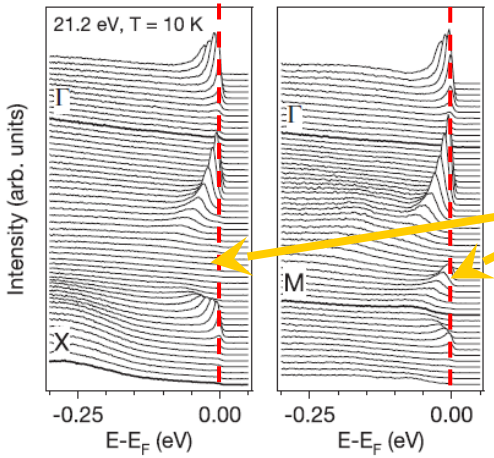
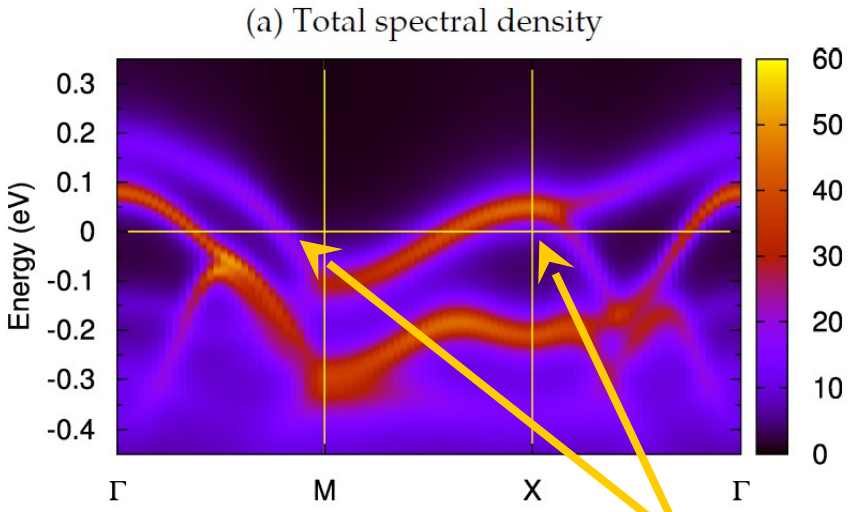






β_M and β_X pocket : 0.66 Å^{-1}

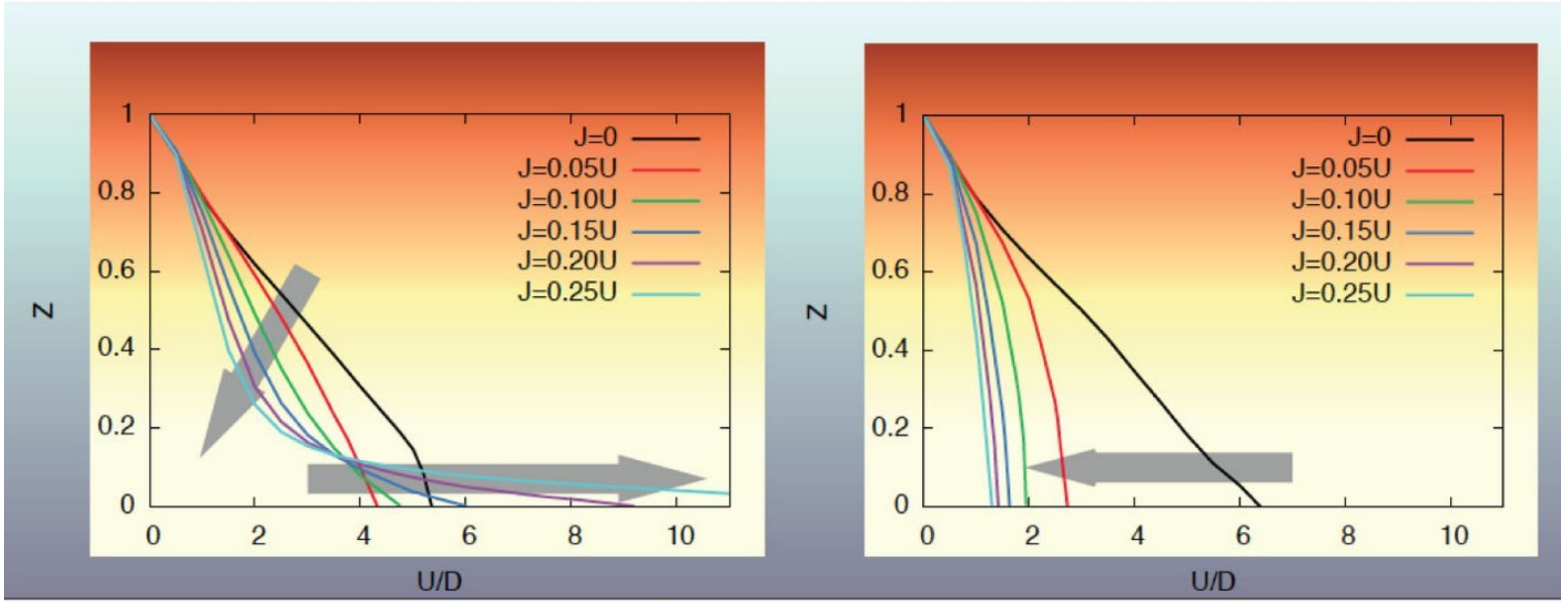
β_M and β_X pocket : 0.65-0.69 Å^{-1}

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

C. Martins, M. Aichhorn et al., JPCM 2017
PsiK highlight of the month, Nov 2016

2 electrons in 3 bands: Hund's metals

L. de Medici *et al.*, PRL 107, 256401 (2011)



Off half filling

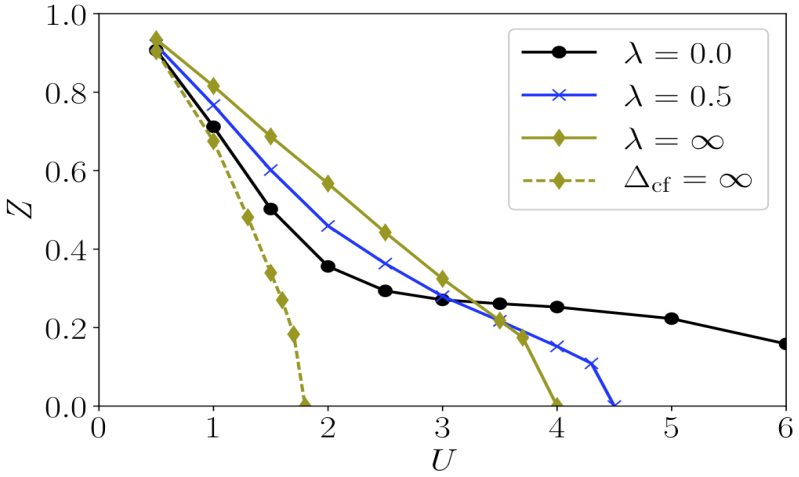
Half filling

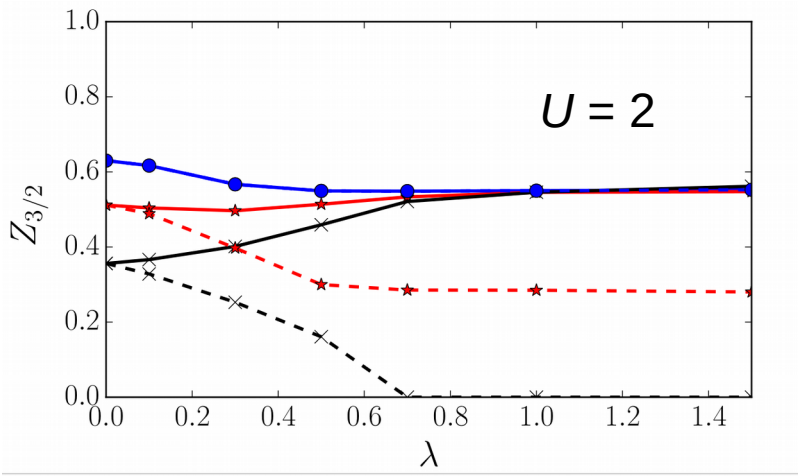
Two antagonistic effects: Lowering coherence scale

Strong influence on metal/insulator transition

How does this change with spin orbit?

2 electrons in 3 bands: Hund's metals



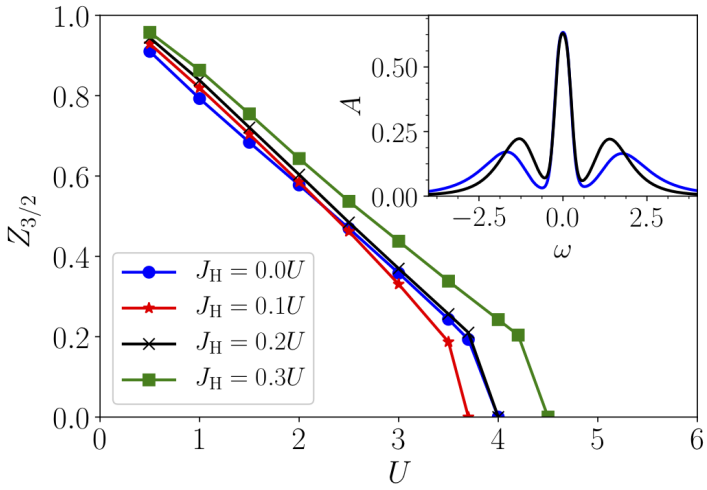


Hunds metal “tail” disappears

System becomes half filled

BUT: Large SOC: weak dependence on J!
Crystal field very different!

Atomic Hamiltonian:
after transformation very different Hunds rule!



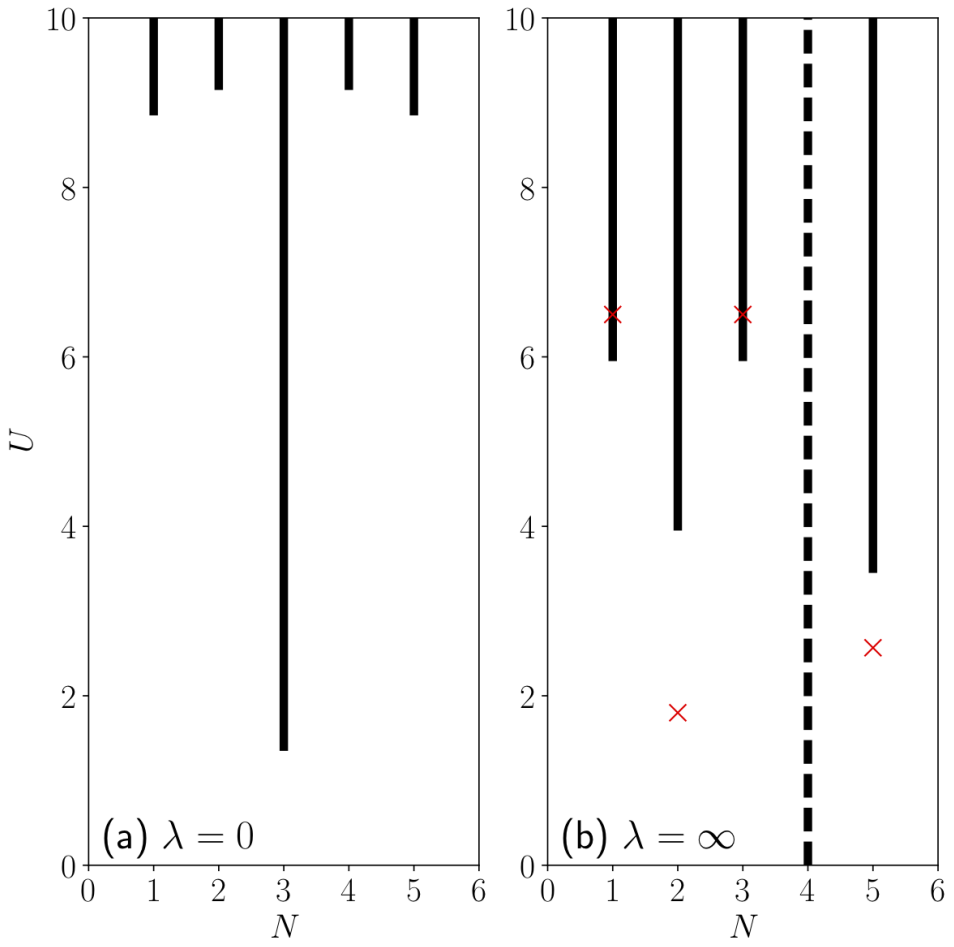
2 electrons in 3 bands: Hund's metals

Without SOC:

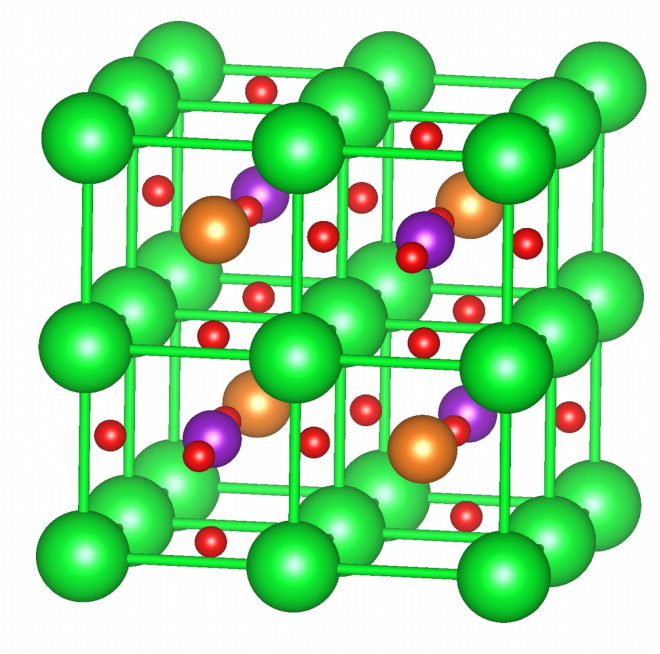
- Insulator at N=3
- N=2 as doped Mott insulator
- Hunds tail

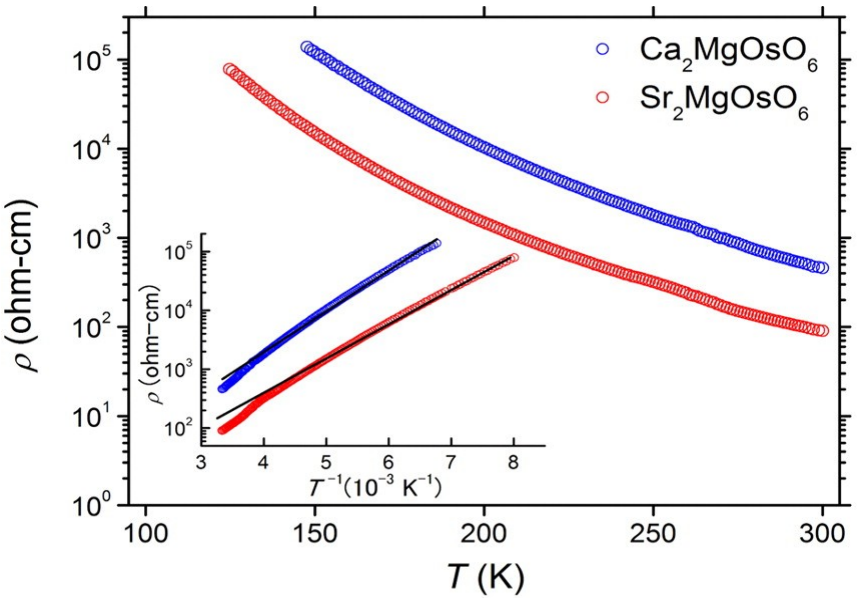
With SOC:

- Insulator at N=2
- No Mott state close to metal
- No Hunds tail



2 electrons in 3 bands: Osmates Sr_2MgOsO_6





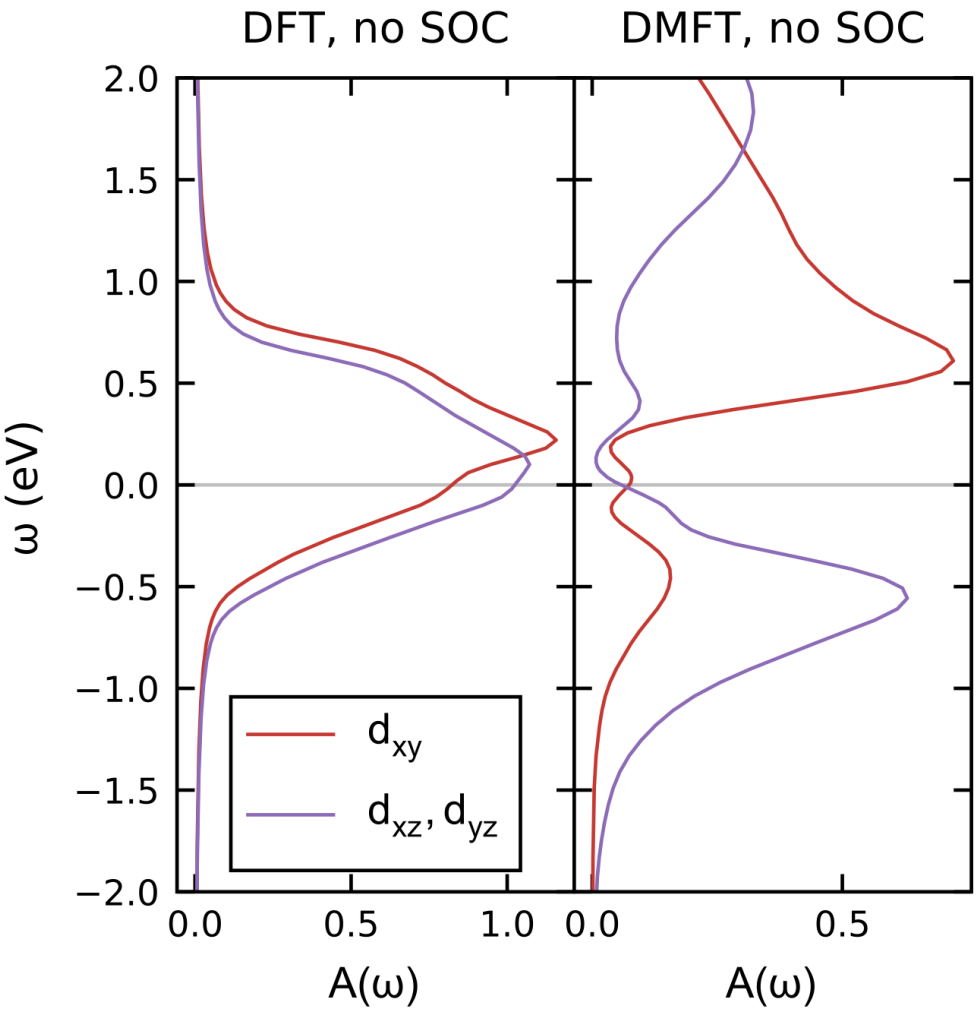
Y. Yuan et al., Inorg Chem. 54, 3422 (2015)

Double perovskite

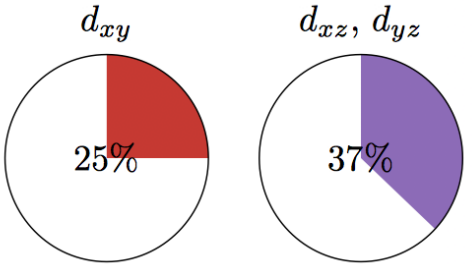
Slightly distorted cages

“These double-perovskite oxide are likely to be Mott insulators”

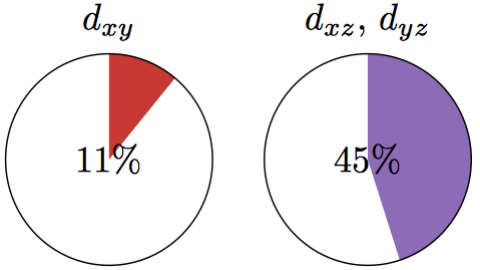
Osmate Sr_2MgOsO_6 without SOC



DFT:

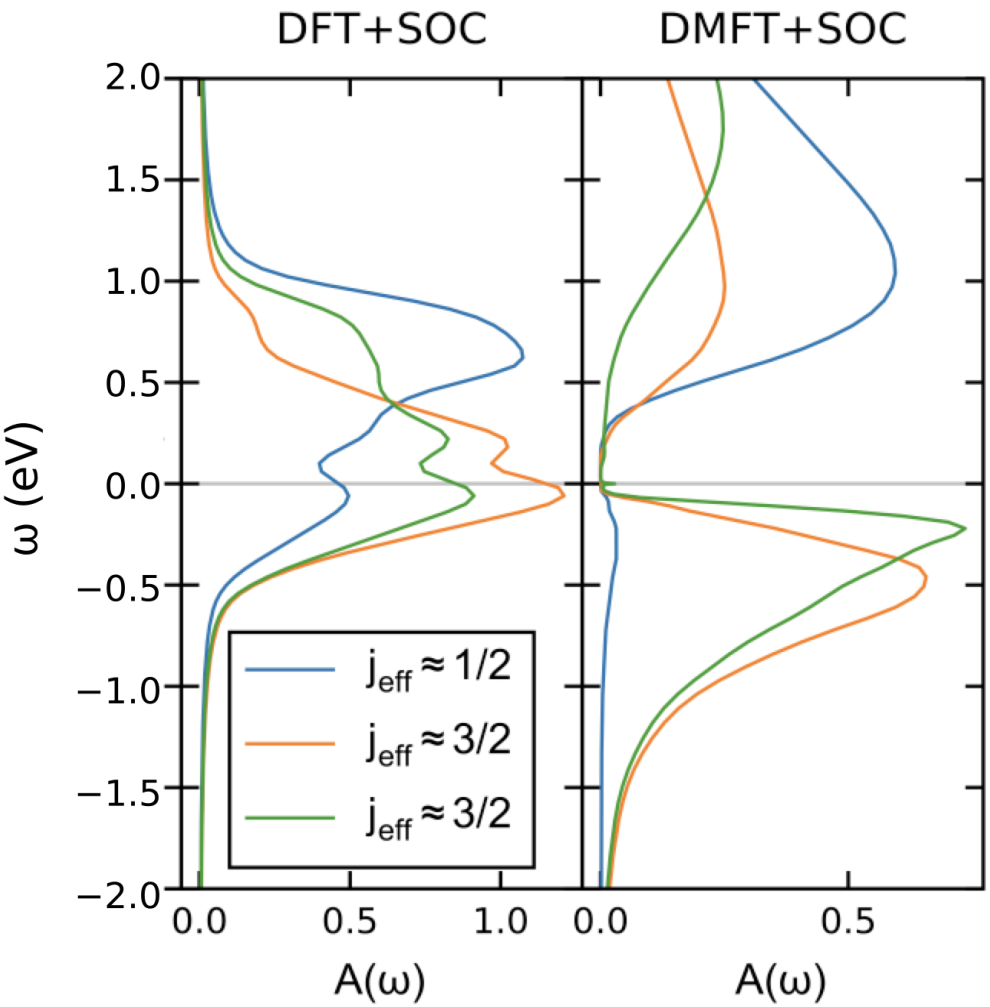


DMFT: bad metal

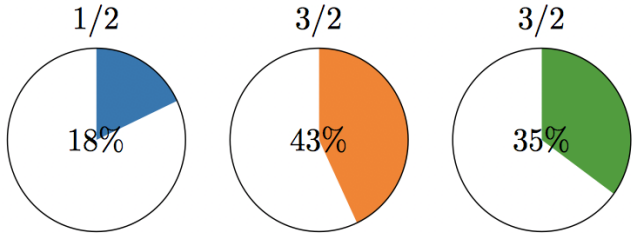


G. Kraberger and M. Aichhorn, in prep.

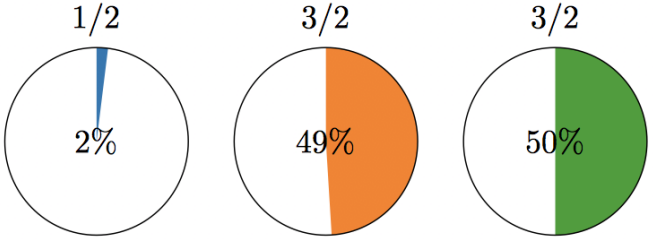
Osmate Sr_2MgOsO_6 with SOC



DFT: j-basis



DMFT: polarised insulator



The self energy and effective SOC

$$\Sigma_a = \frac{2}{3}\Sigma_{\frac{3}{2}} + \frac{1}{3}\Sigma_{\frac{1}{2}}$$

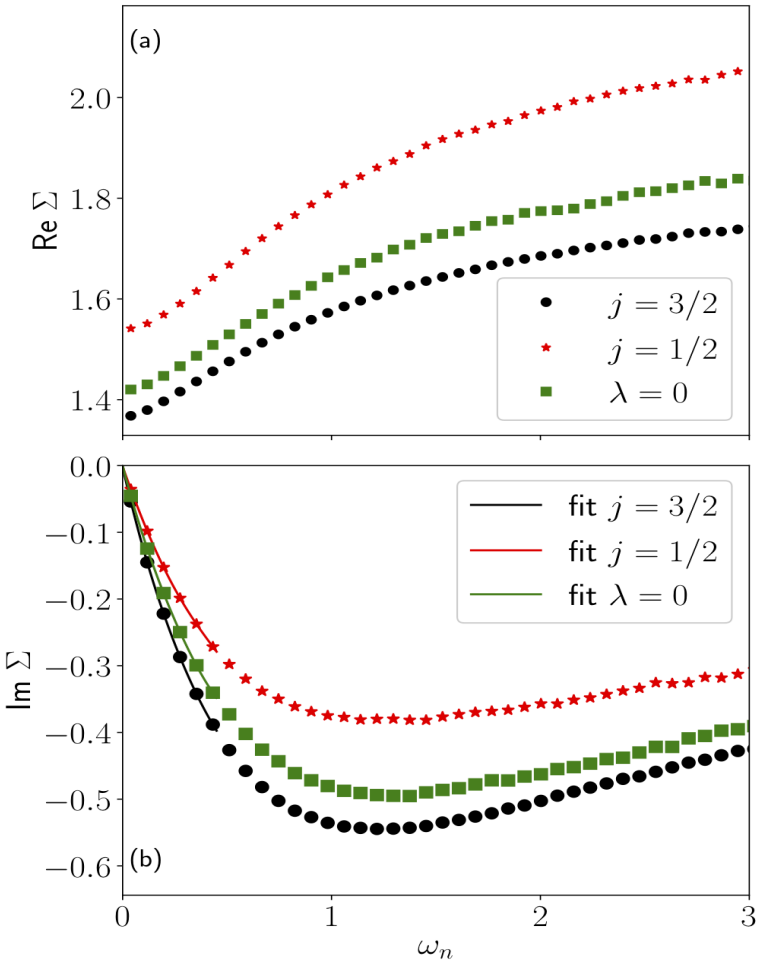
$$\Sigma_d = \Sigma_{\frac{1}{2}} - \Sigma_{\frac{3}{2}}$$

$$\Rightarrow \quad \Sigma = \Sigma_a \mathbb{I} + \frac{2}{3}\Sigma_d \mathbf{l}\cdot\mathbf{s}$$

Eff. Spin-Orbit Coupling:

$$\lambda_{\text{eff}} = \lambda + \frac{2}{3}\,\text{Re}\Sigma_d(i\omega_n \to 0)$$

$$\lambda_{\text{eff}} > \lambda$$



Strong Correlations and Spin-Orbit Coupling

Spin-Orbit coupling important for heavy atoms

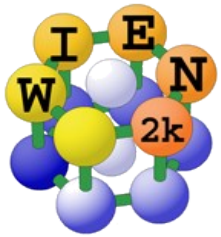
In model calculations:

- Effect strongest for N=5, strong reduced for N=3 and N=2
- Effective crystal field picture has to be used with care

Iridates: Spin-Orbit assisted Insulator

- Reduced Spin-Orbital Degeneracy, effective one-band model

Osmates Double Pervoskites: Similar effect, but N=2



triqs

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Strong Correlations and Spin-Orbit Coupling

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