ARPES study of transition metal substitutions in Sr_2IrO_4

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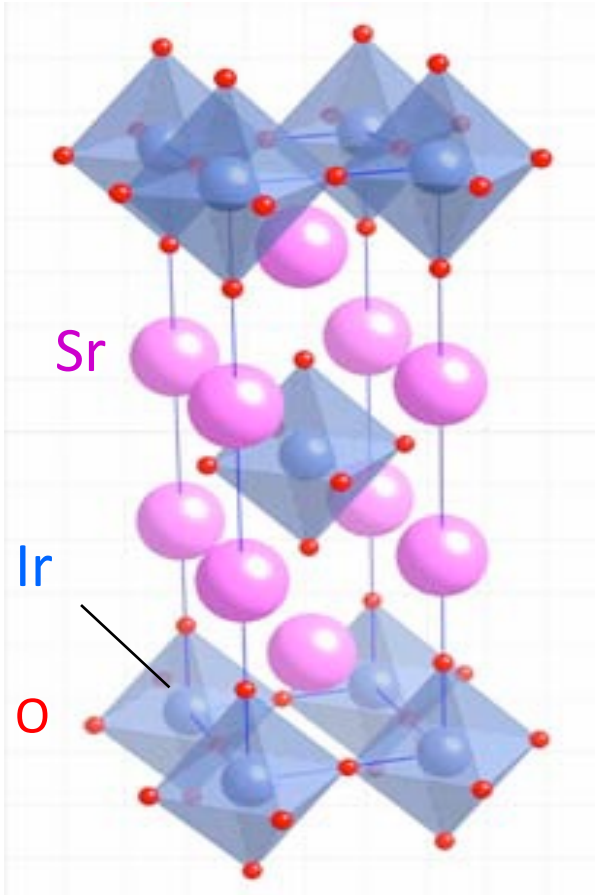
ARPES experiments : SOLEIL synchrotron, CASSIOPEE beamline

Patrick Le Fèvre, François Bertran, Julien Rault

Sample synthesis

LPS and Dorothée Colson, Anne Forget : *SPEC, CEA-Saclay, France*

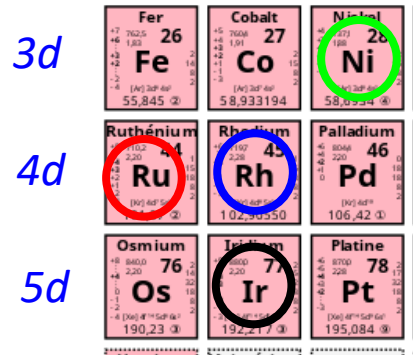
Outline



Sr_2IrO_4
(simplified structure)

- New type of Mott insulator :
  => Spin-orbit Mott insulator

- New type of correlated metals ?
  ⇒ Sr/La substitutions
  => Ir/transition metal substitutions



Ni : expected : electron doping
obtained : hole doping

Rh : expected : isovalent
obtained : hole doping

Ru : expected : hole doping
obtained : isovalent

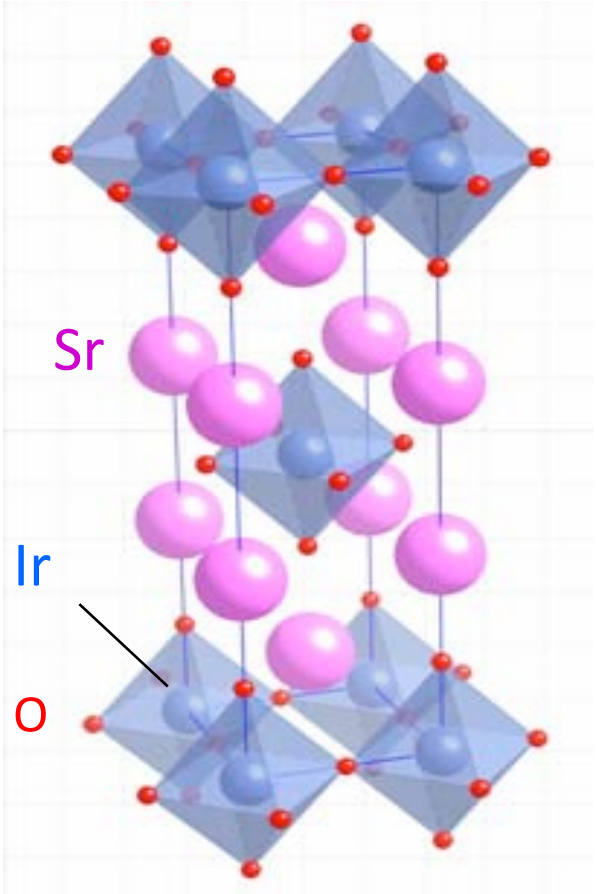
- Substitution with transition metals introduce charged defects
  => Role and origin of such defects ?

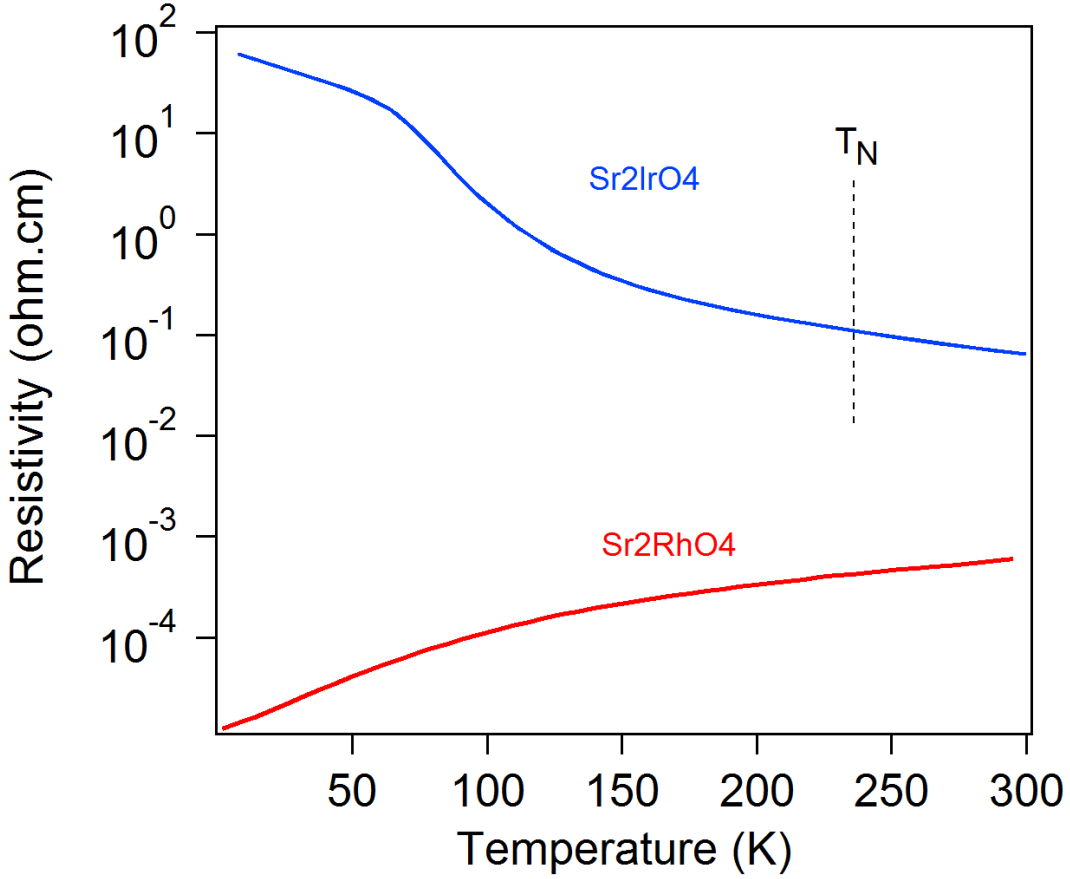
Why is Sr_2IrO_4 insulating ?

Sr_2IrO_4 $Ir^{4+} => 5d^5$

$Rh^{4+} => 4d^5$

Nagai, J. Phys Soc Japan (2010)

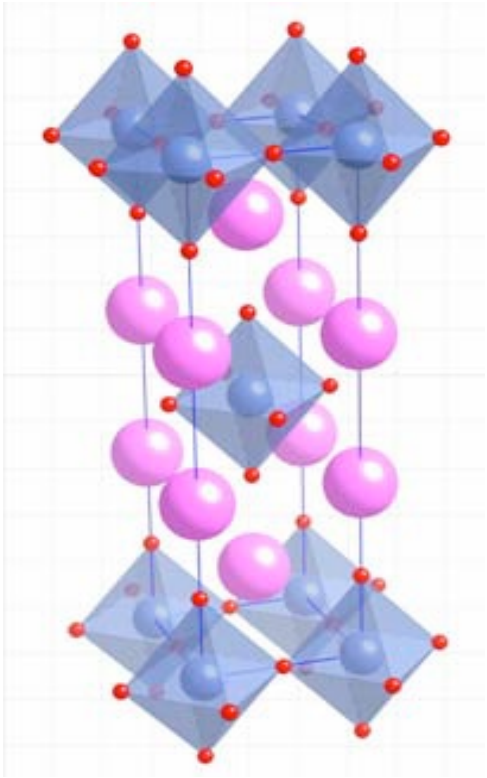




Layered perovskite : also in cuprates, manganites, ruthenates…

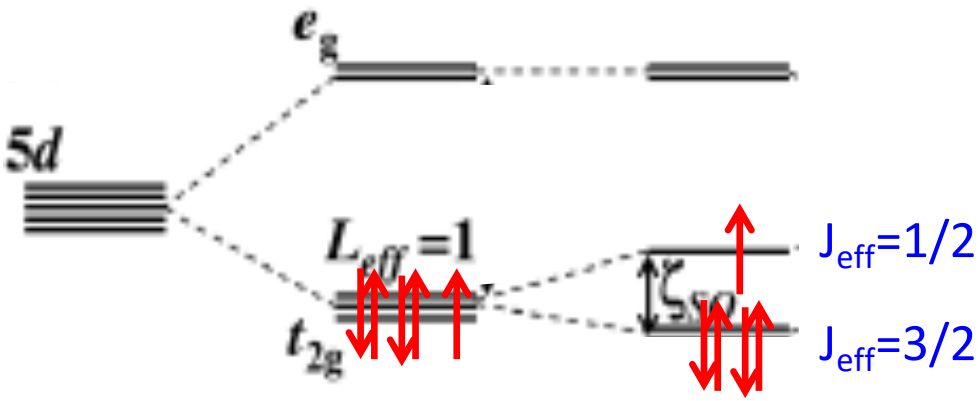
Sr_2IrO_4 : a spin-orbit Mott insulator

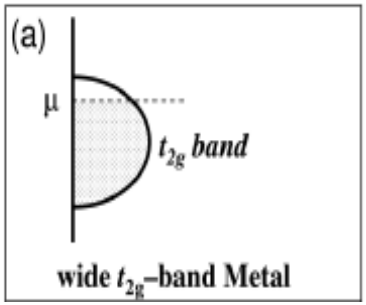
Weak correlations are expected for 5d metals. However, the strong spin-orbit splitting reshapes the band structure in a way that favors strong correlations.



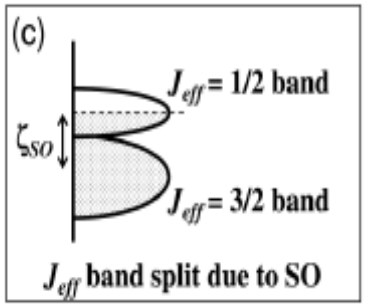
Sr_2IrO_4
(simplified structure)

B.J. Kim et al. PRL 2008

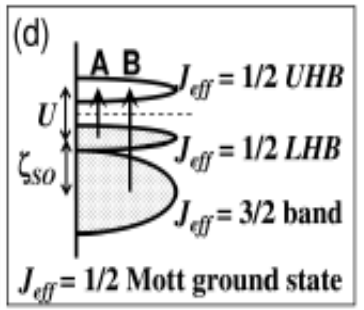




Rather weak correlations



Strong correlations

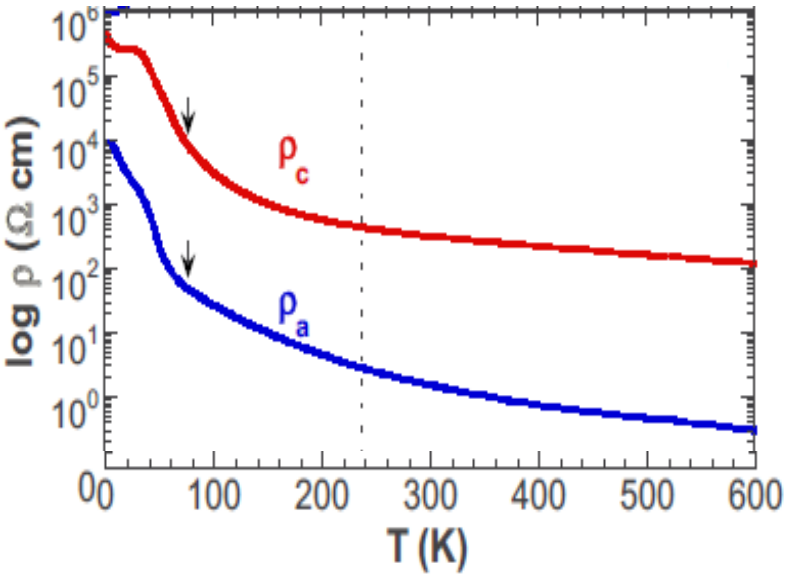


Mott insulator

Supported by DMFT calculations *C. Martins, S. Biermann et al. PRL 11*
R. Arita et al., PRL 11

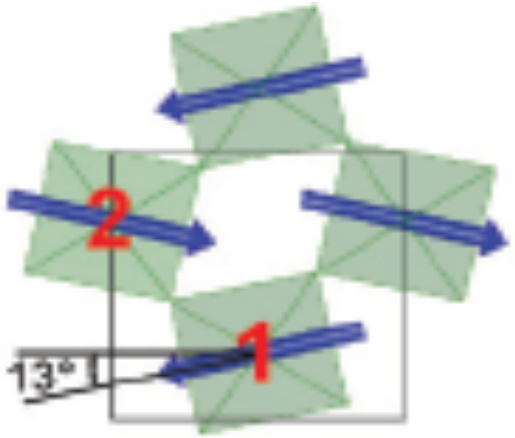
Sr_2IrO_4 : an insulator with AF transition at 240K

Resistivity => insulating below and above T_N



Chikara, G. Cao *et al.* PRB09

Magnetic order below T_N=240K



M=0,2μ_B/Ir

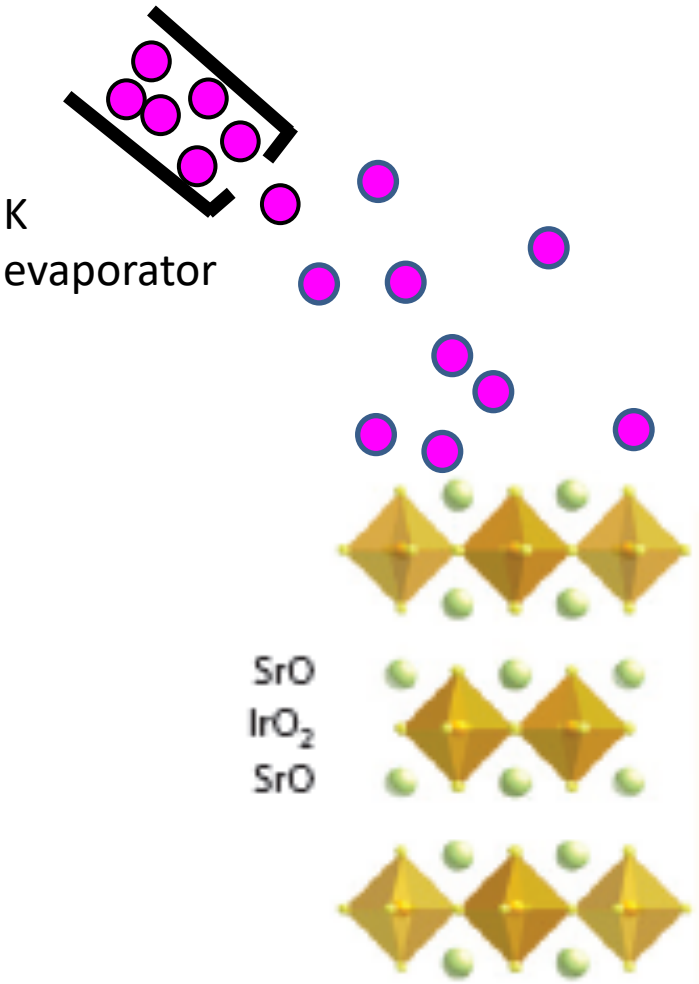
Feng Ye, PRB13
Dhital PRB 13

Mott gap ~ 0,6eV
Magnetic exchange J~60meV

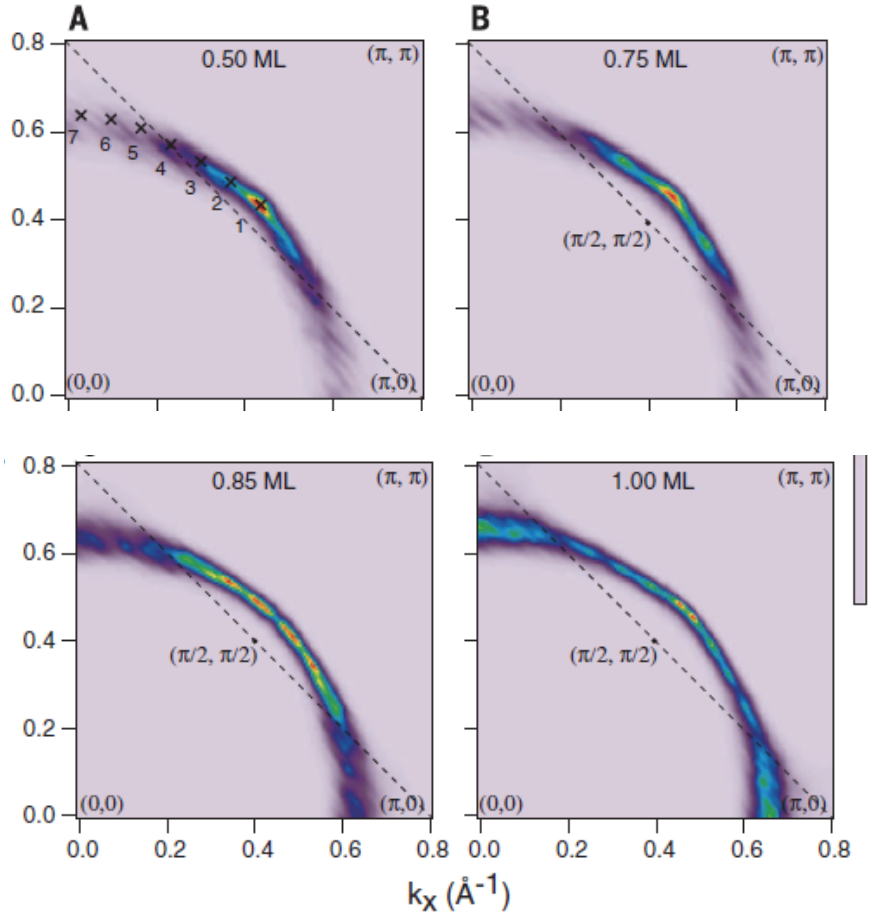
Analogous to cuprates => superconducting if doped ??

Wang, Senthil PRL 2011

Emerging metallic state ?
Is it similar to cuprates ?



Fermi Surface observed with ARPES

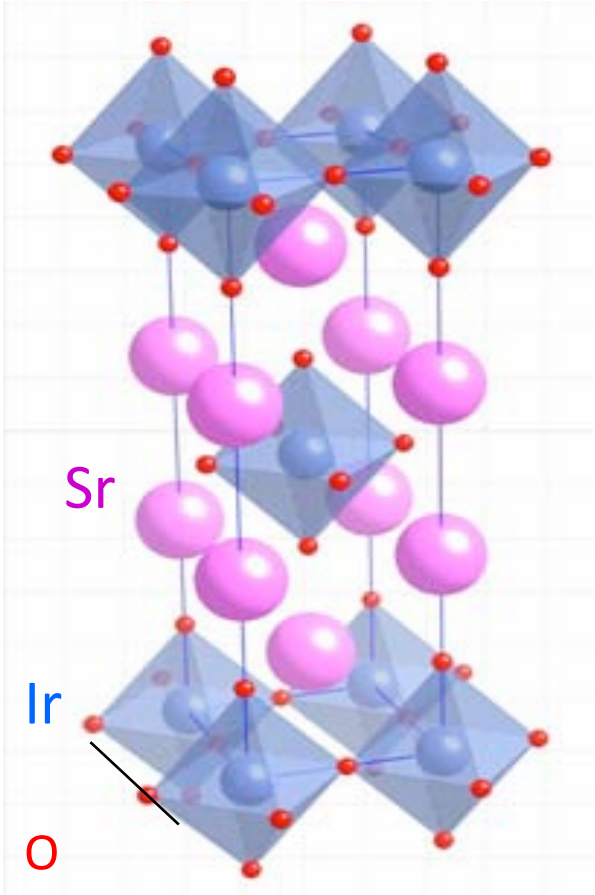


Y.K. Kim *et al.*, Science 14

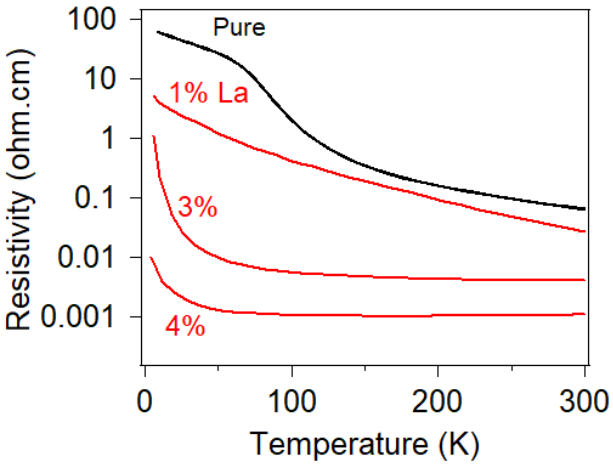
⇒ The evolution of the Fermi Surface seems to exhibit « Fermi arcs » like cuprates

⇒ A d-wave gap could open at 50K Y.K. Kim Nature Phys. 2015, Y.J. Yan PRX 15

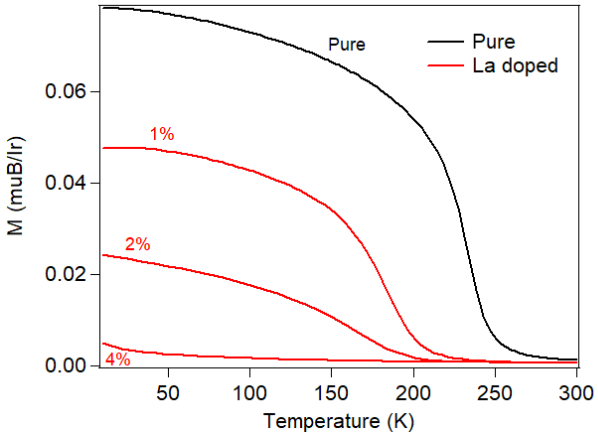
Bulk electron doping : Sr/La substitutions



Resistivity

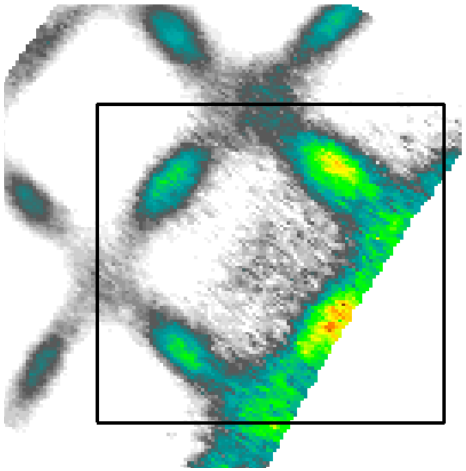


Magnetization



=> No superconductivity observed so far

« Nodal » Fermi Surface



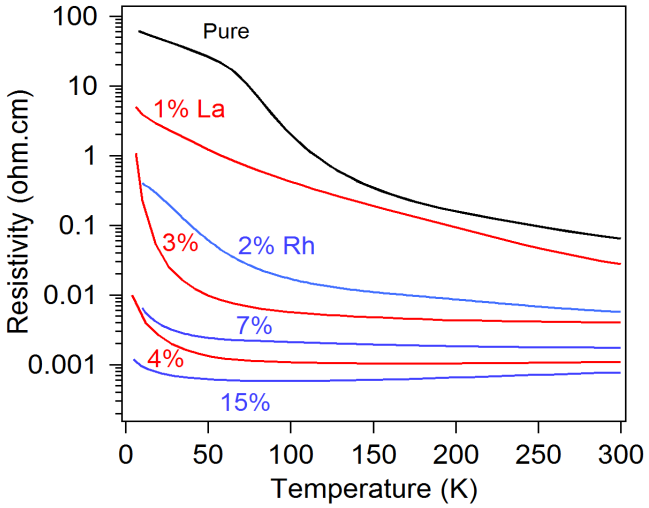
Brouet PRB 15

See also : M. Ge, G. Cao, PRB 11
X. Chen, D. Wilson, PRB 15
A. de la Torre, PRL 15…

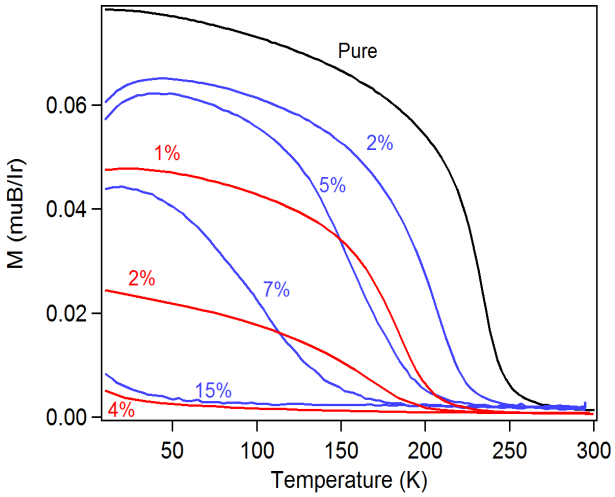
Ir-Rh substitutions

Ir-Rh substitutions also produce a weakly metallic non-magnetic state.
Its Fermi Surface is however very different with hole pockets along X points.

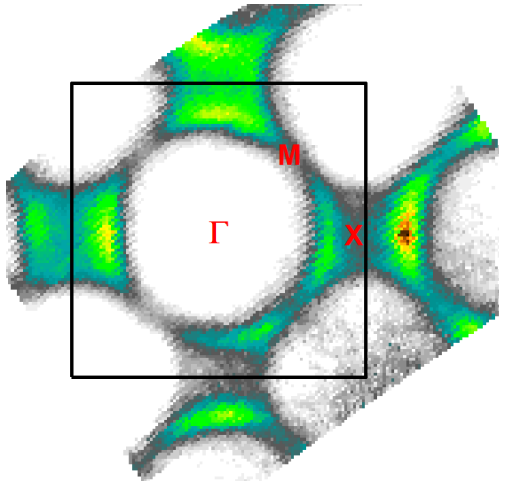
Resistivity



Magnetization (1T)

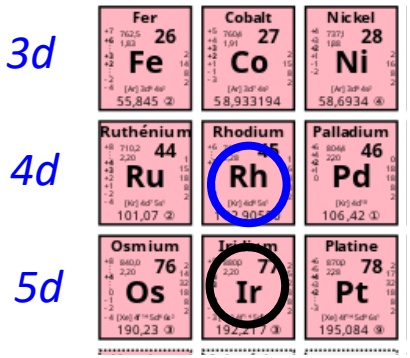


Fermi Surface 15% Rh



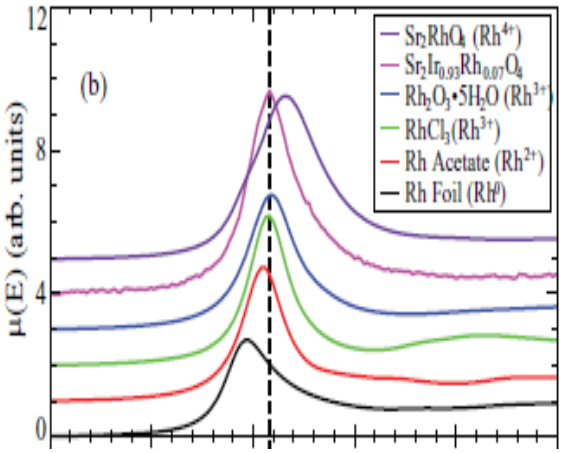
V. Brouet PRB 15

What is the effect of Rh ?

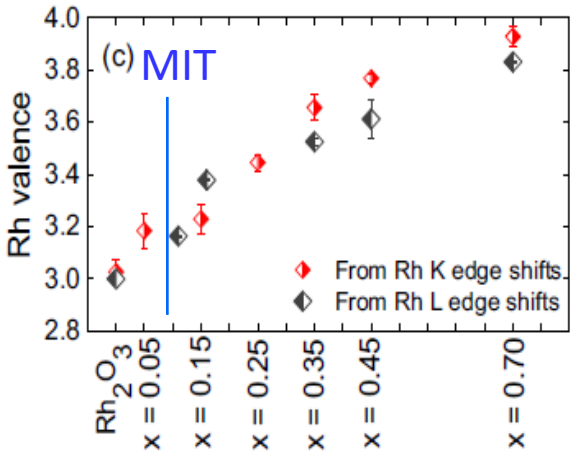


Rh : expected : isovalent, reducing SOC ?
obtained : hole doping

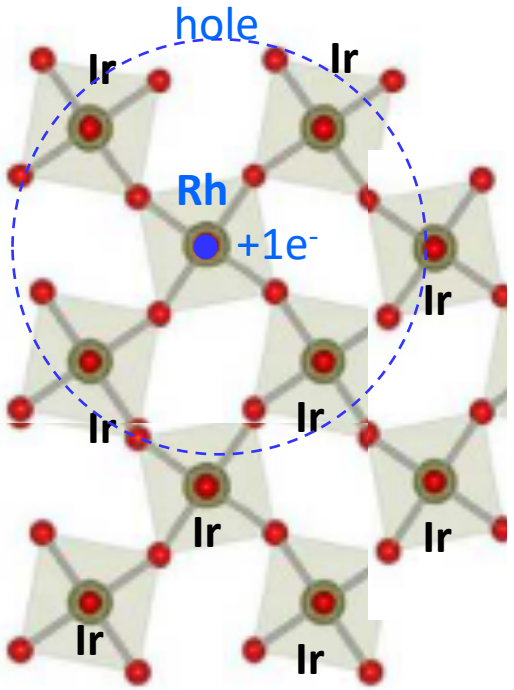
In X-ray absorption Rh looks like Rh^{3+}



Clancy PRB14



ChikaraPRB17

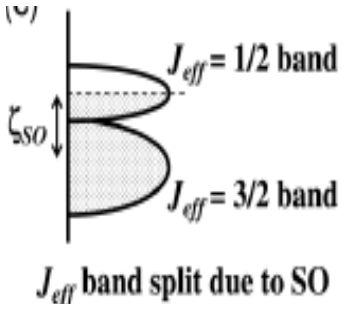


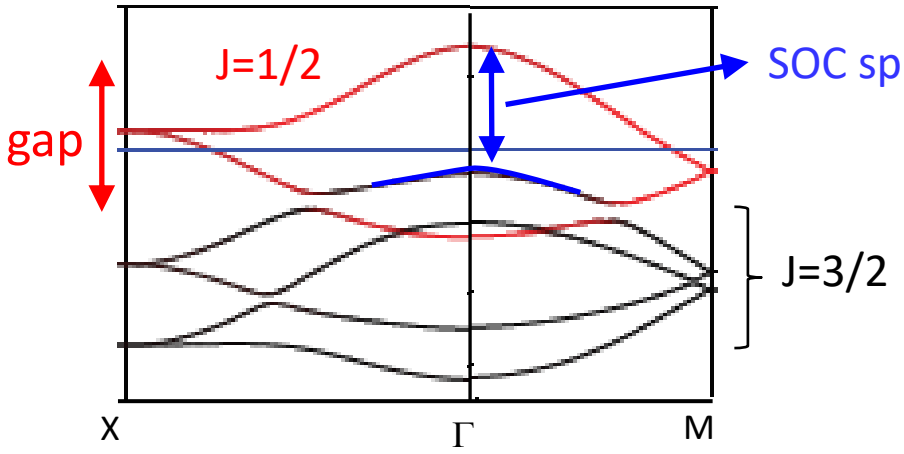
Hole doping confirmed by ARPES : Cao, Dessau NatCom 16, Louat PRB 18

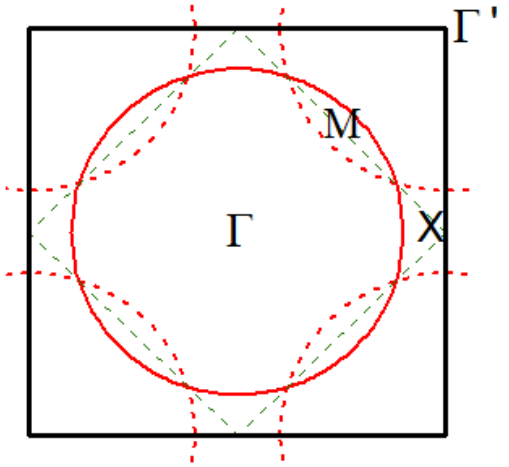
ARPES view of the electronic structure

The case of Sr_2IrO_4

Band structure

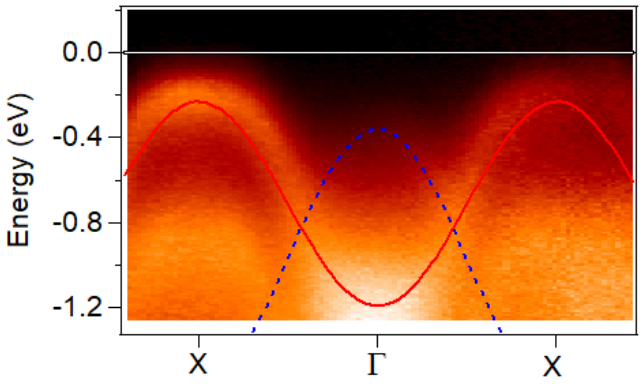


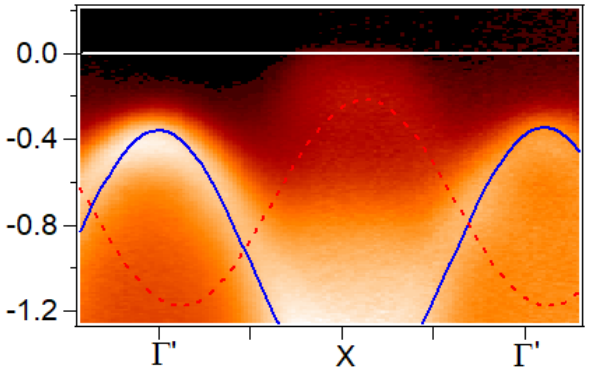


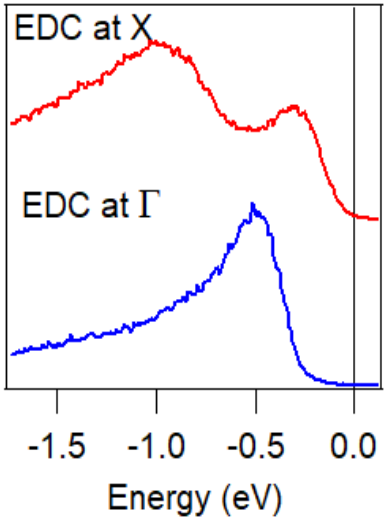


TB model by *Carter PRB13*

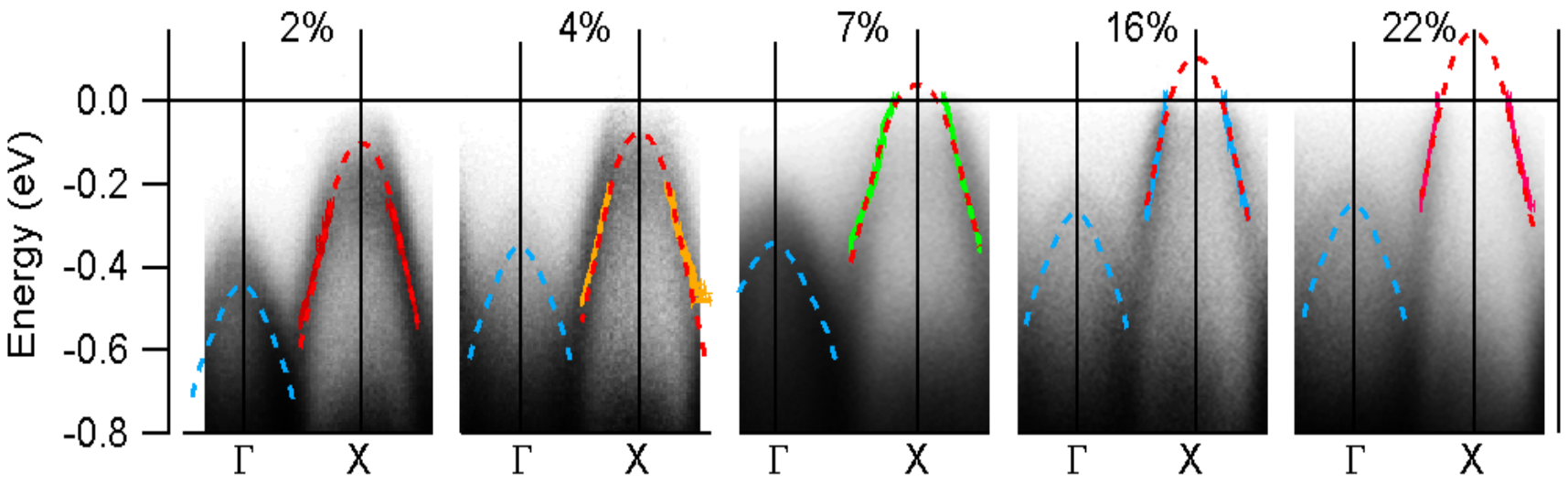
Dispersion measured by ARPES

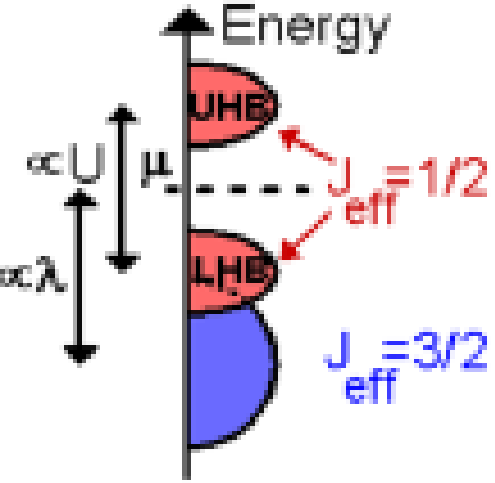




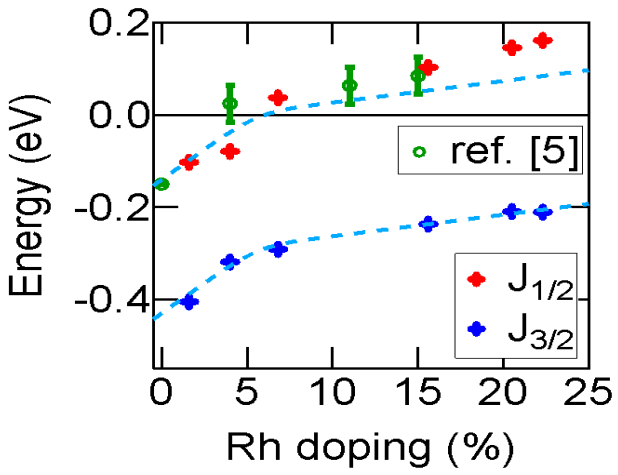


ARPES view of Rh doping

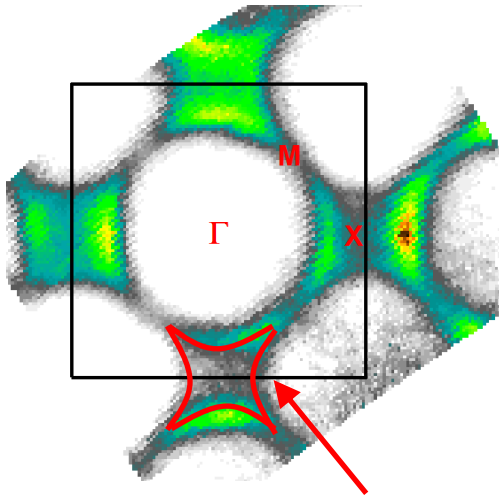




Position of bands *vs* doping



Fermi Surface - 15% Rh



A. Louat, V. Brouet PRB18

Doping with different transition metals

Rh doping

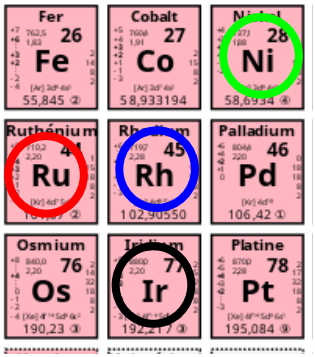
Ru doping

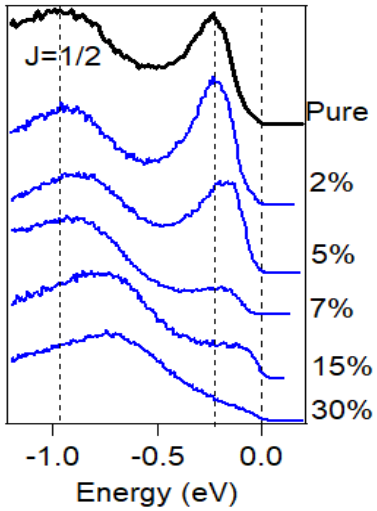
Ni doping

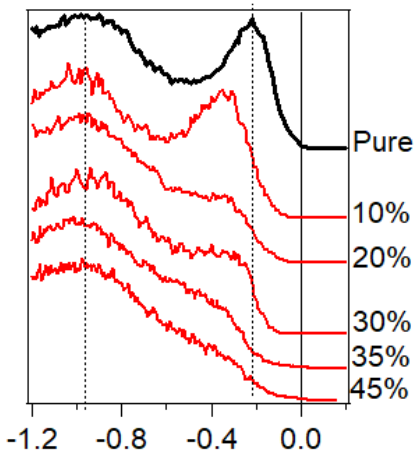
3d

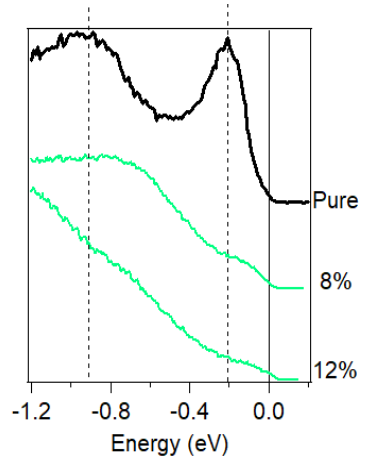
4d

5d

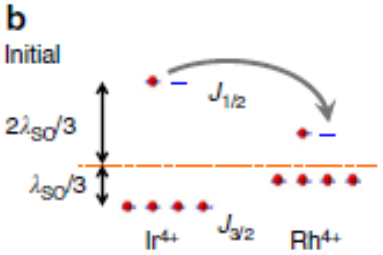




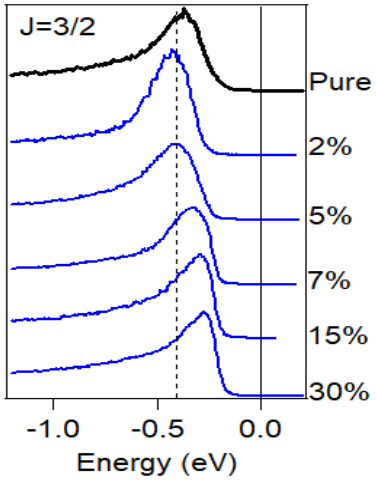


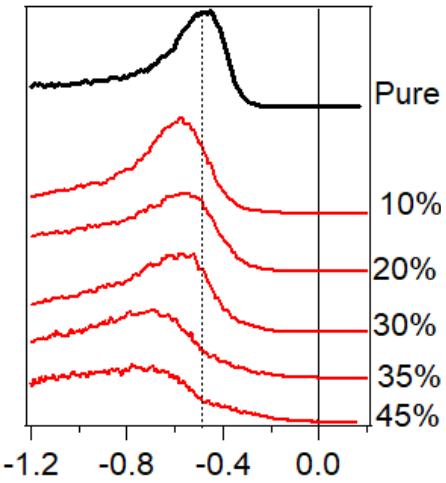


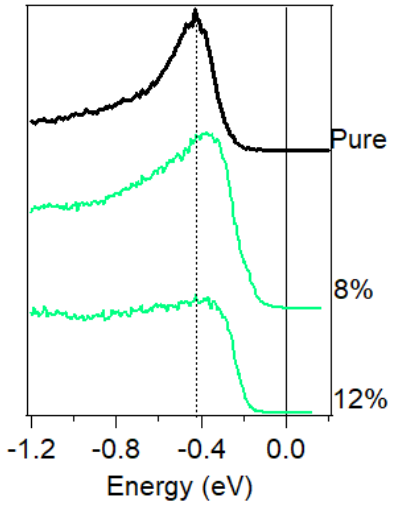
Smaller value of λ favors $Rh^{3+}=d^6$?



Y. Cao, D. Dessau et al., Nature Com. 16





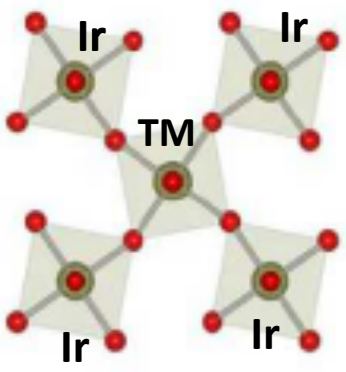


Expected :	No doping	Hole doping	Electron doping
Observed :	Hole doping	No doping	Hole doping

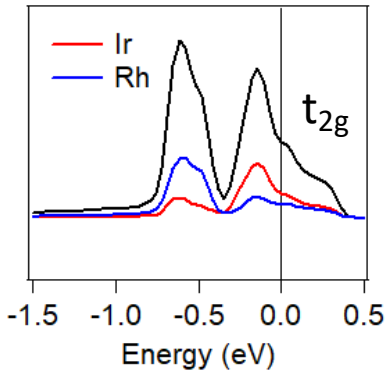
Calculation for $Sr_2(IrM)_{0.5}O_4$

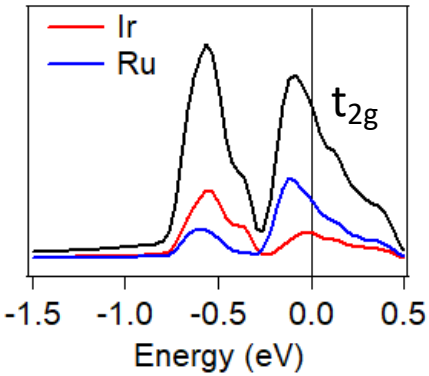
Calculation for an ordered structure with 50% Ir and 50% TM (Rh, Ru or Ni). No SOC included.

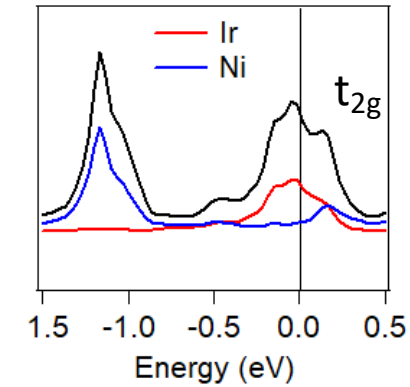
NB : more detailed calculation for supercell : *Liu, Franchini PRB 16*



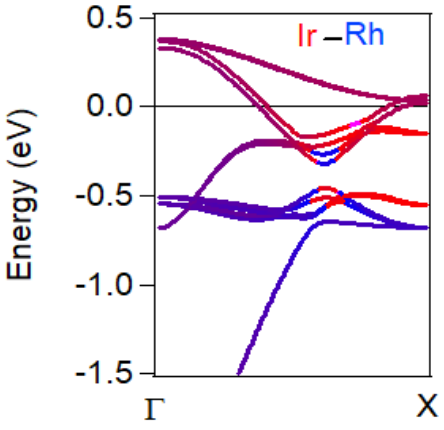
Calculated density of states (no SOC)

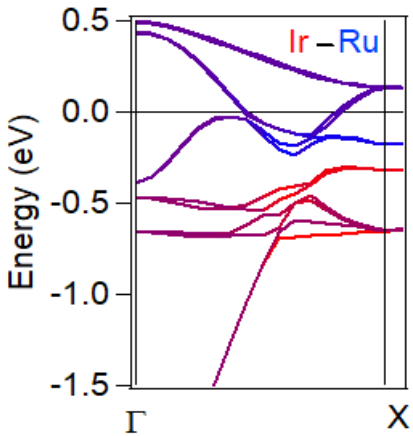


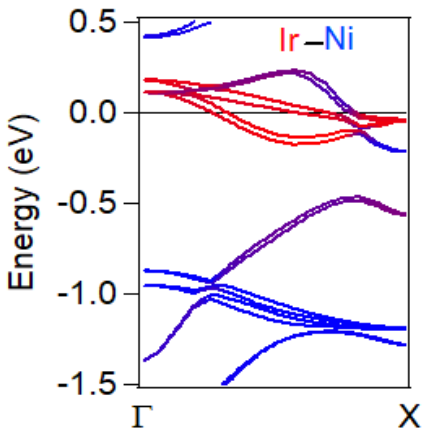




Calculated band structure along ΓX (no SOC)







Calculation for $Sr_2(IrM)_{0.5}O_4$

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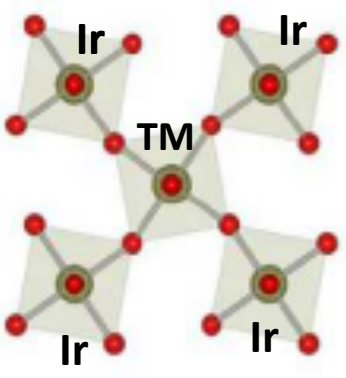
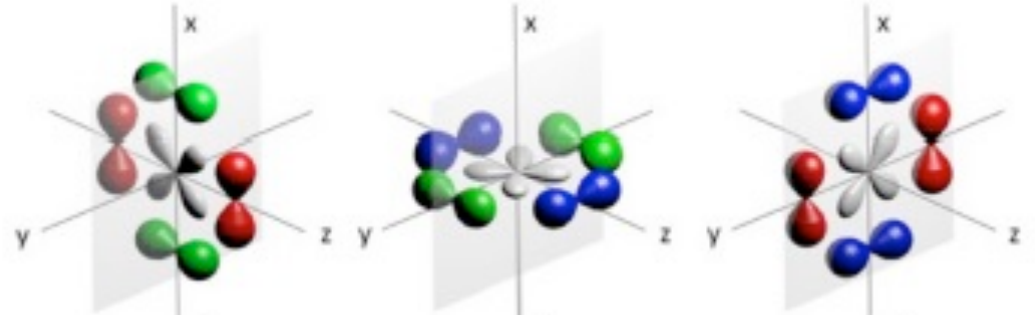
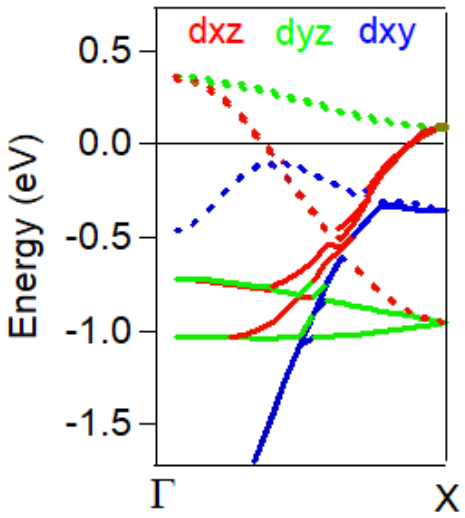
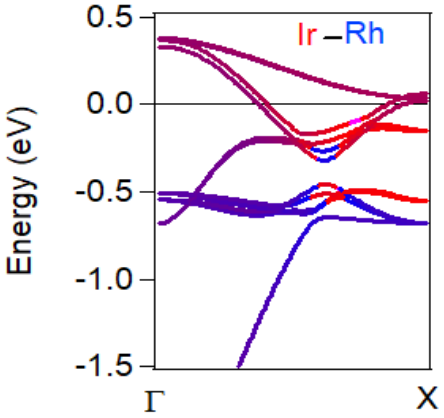


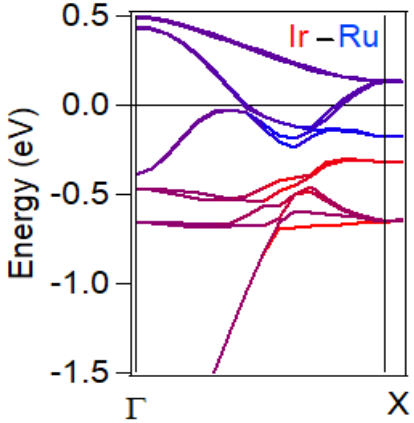


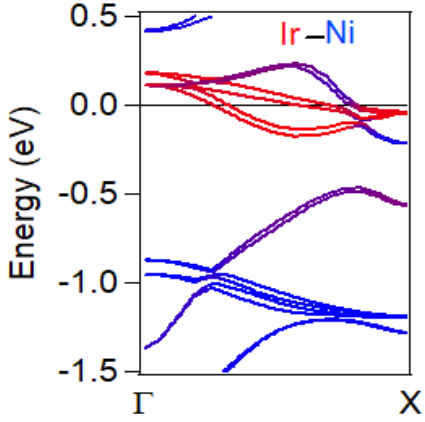
Sr_2IrO_4



Calculated band structure along ΓX (no SOC)

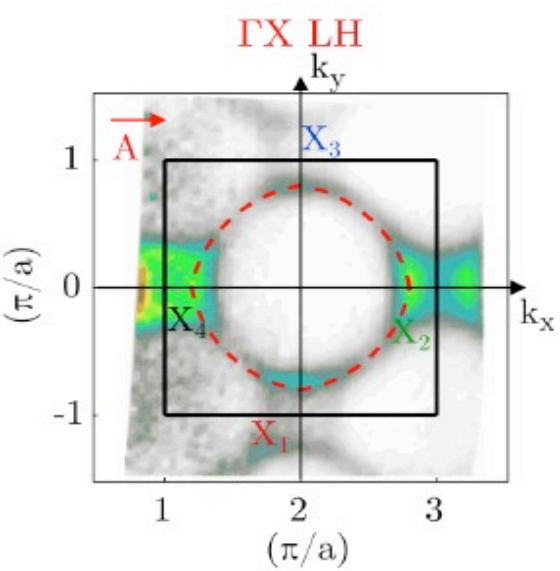


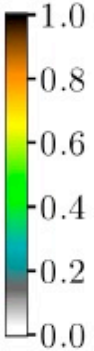




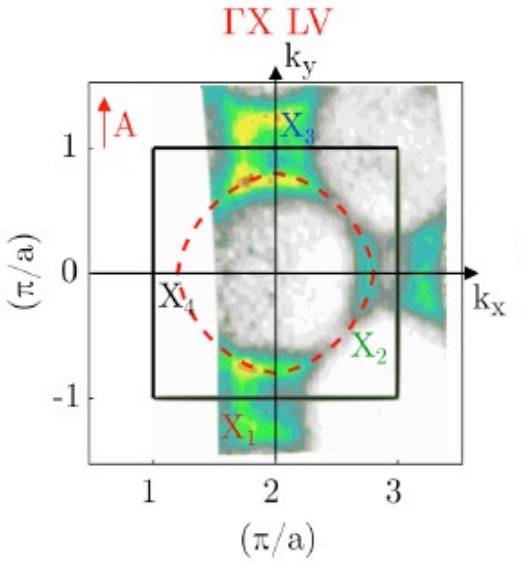
Direct study of orbital character at X point

Mostly dxz character





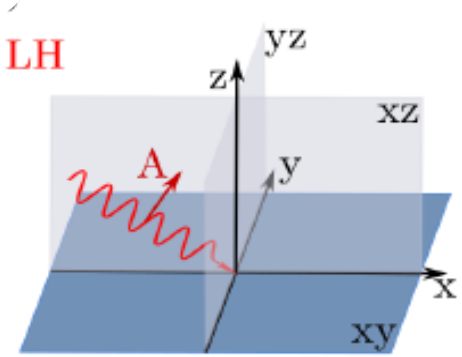
Mostly dyz character

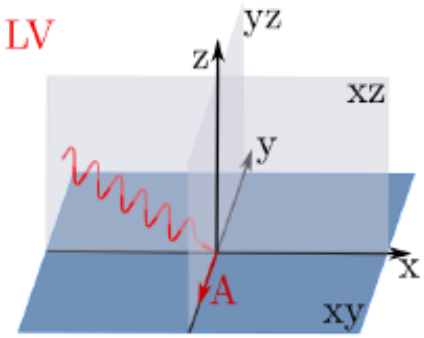


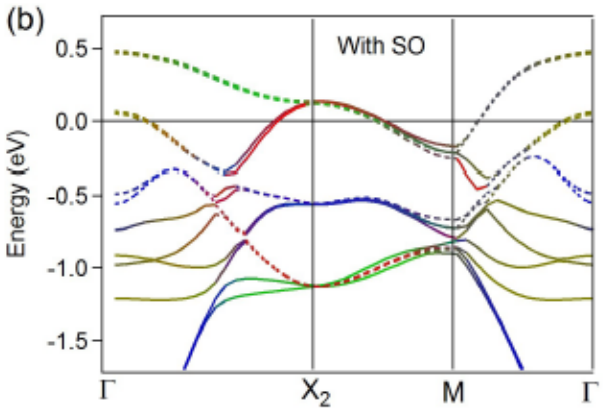
Even for a J=1/2 band, there is a strong k-dependence of orbital character.

It is strongly dxz or dyz at X.

A. Louat, V. Brouet PRB 19



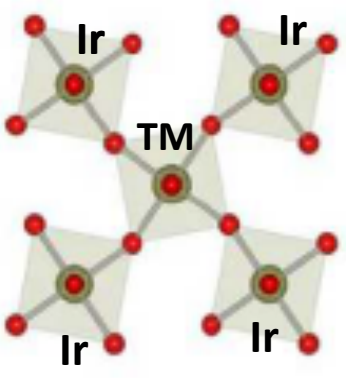




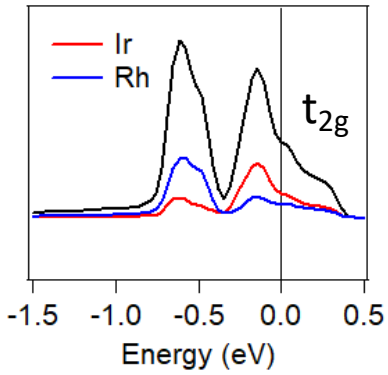
Calculation for $Sr_2(IrM)_{0.5}O_4$

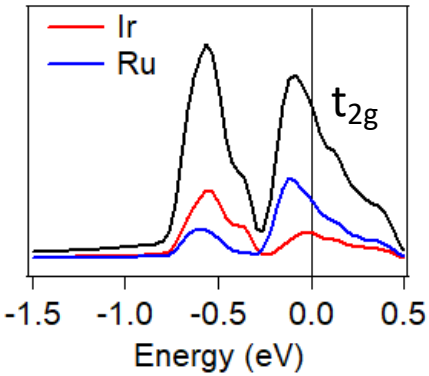
Calculation for an ordered structure with 50% Ir and 50% TM (Rh, Ru or Ni). No SOC included.

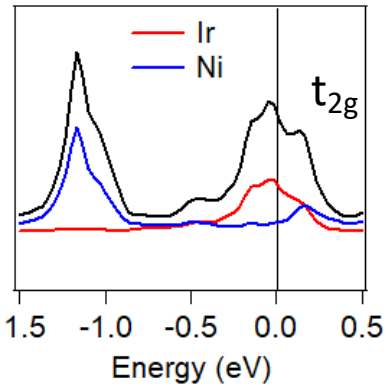
NB : more detailed calculation for supercell : *Liu, Franchini PRB 16*



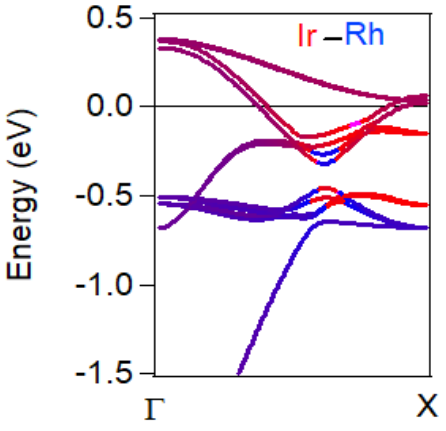
Calculated density of states (no SOC)

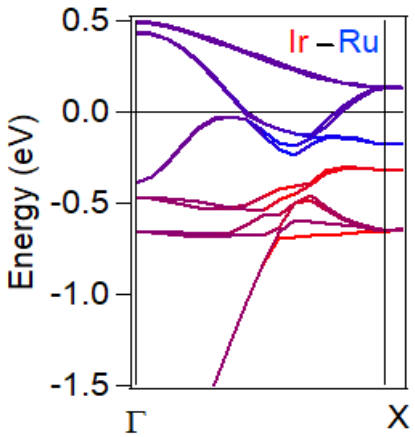


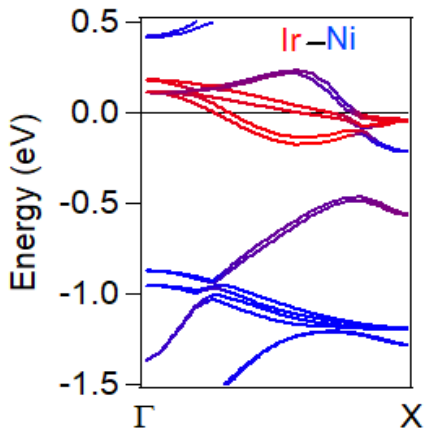




Calculated band structure along ΓX (no SOC)

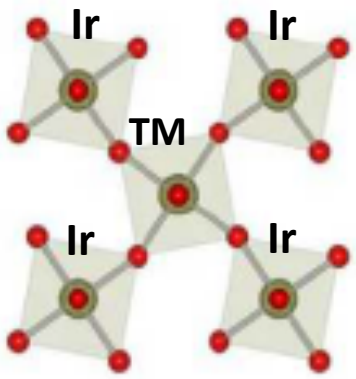




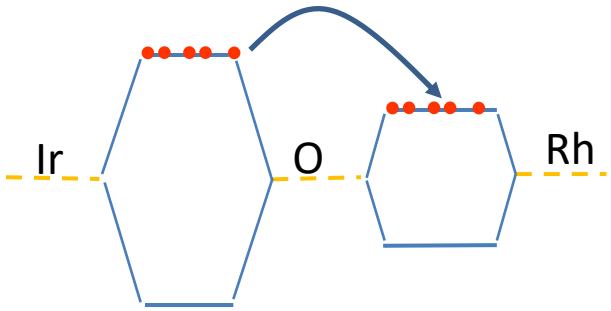


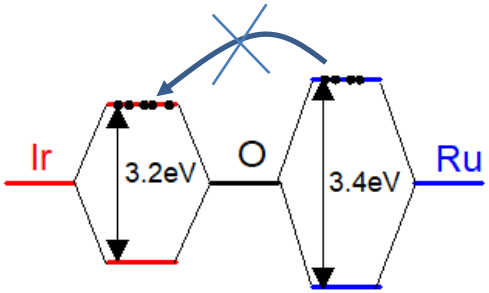
Hybridization with oxygen

Why is TM and Ir content so different at top and bottom of the band ?

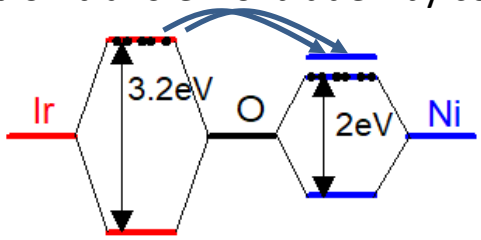


Smaller hybridization
with oxygen favors $Rh^{3+}=d^6$?





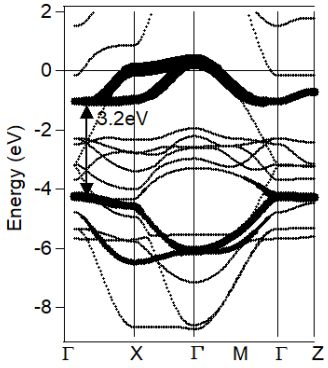
Electron transfer forbidden by correlations



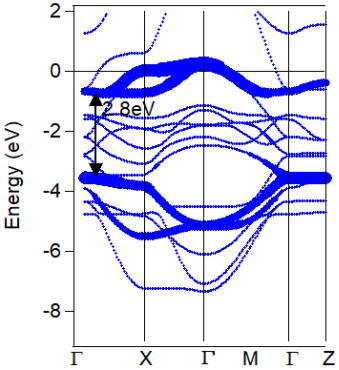
Electron transfer possible to eg

Hybridization for each atom estimated from calculation in $Sr_2\,TM\,O_4$

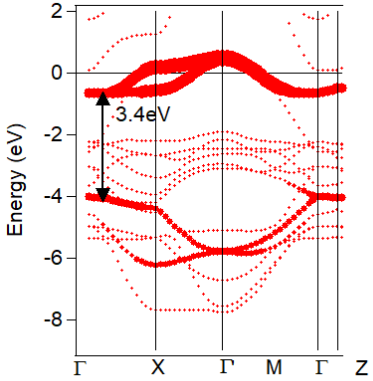
Sr_2IrO_4 : 3.2eV



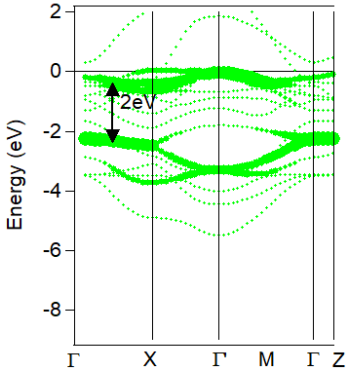
Sr_2RhO_4 : 2,8eV



Sr_2RuO_4 : 3,4eV

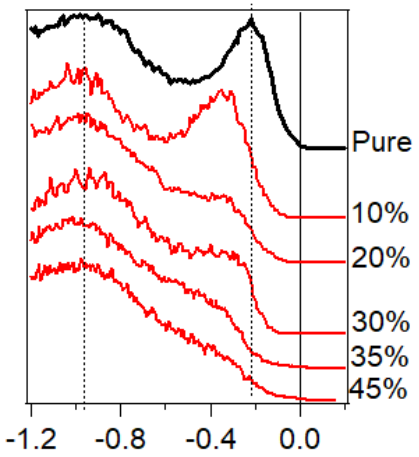


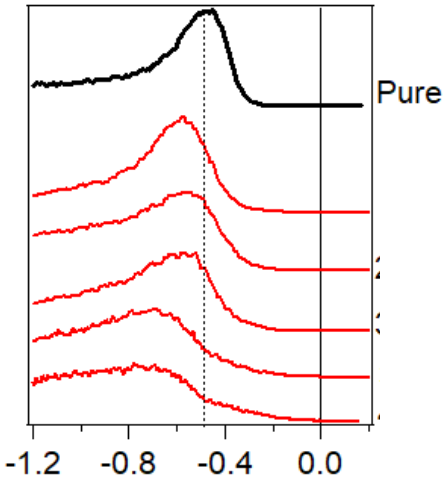
Sr_2NiO_4 : 2eV



Ir-Ru substitutions

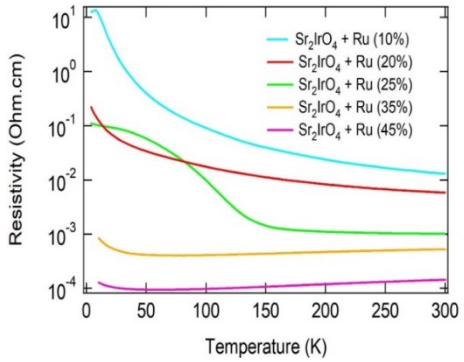
Ru doping



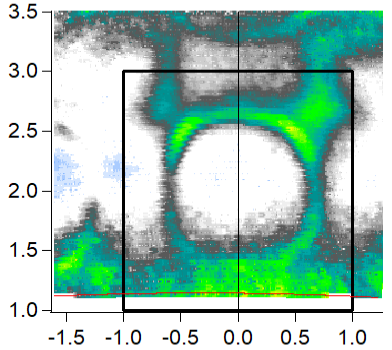


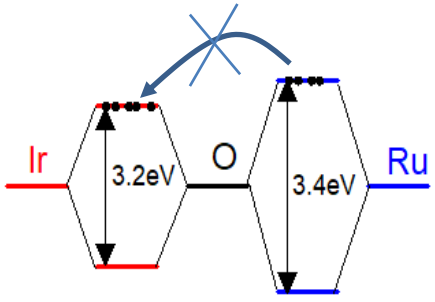
Even though the bands do not seem to move to the Fermi level, there is a insulator to metal transition.

Resistivity

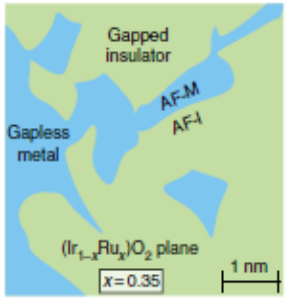


Fermi Surface (45% Ru)





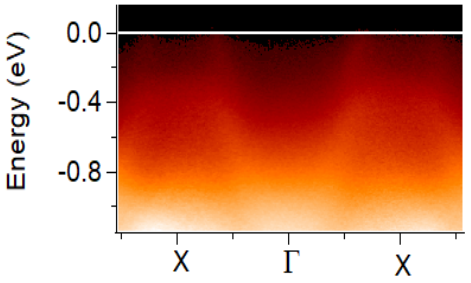
Electron transfer forbidden by correlations



Dhital NatCom14

$Sr_3Ir_2O_7$

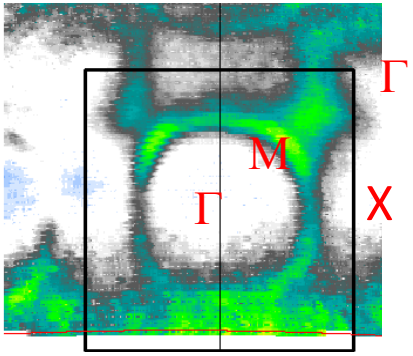
Dispersion (35% Ru)



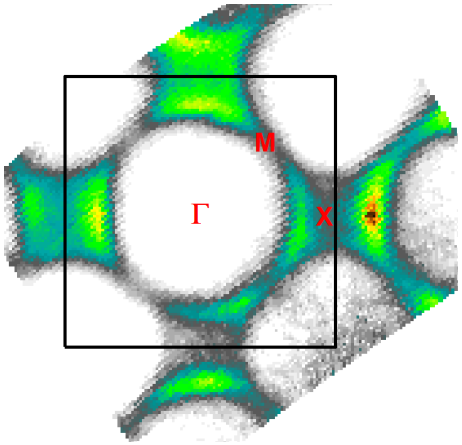
=> Coexistence of metallic and insulating regions

Fermi Surface comparaison

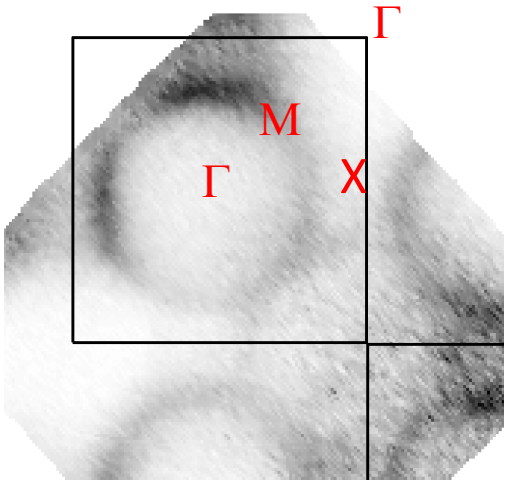
45% Ru



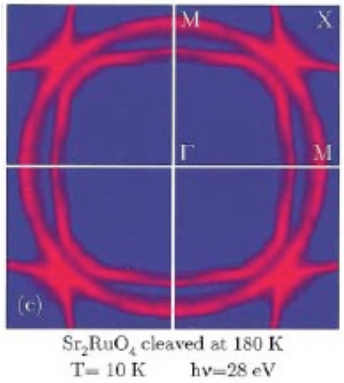
15% Rh

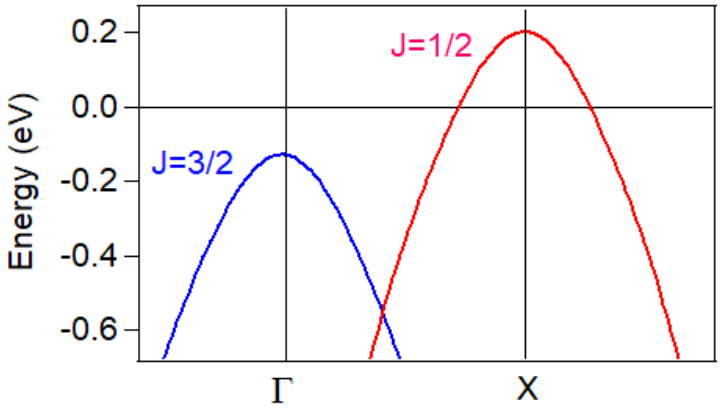


12% Ni



Sr_2RuO_4

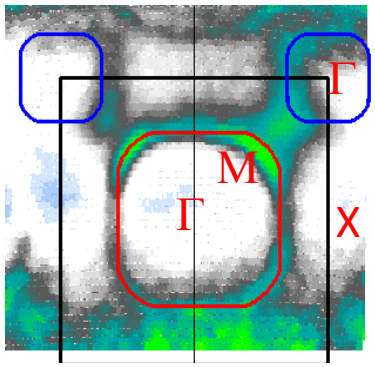




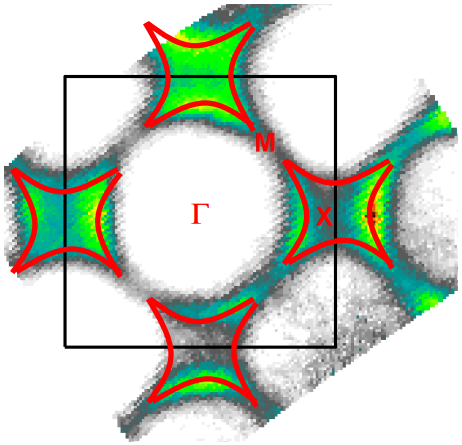
Damascelli PRL2000

Fermi Surface comparaison

45% Ru

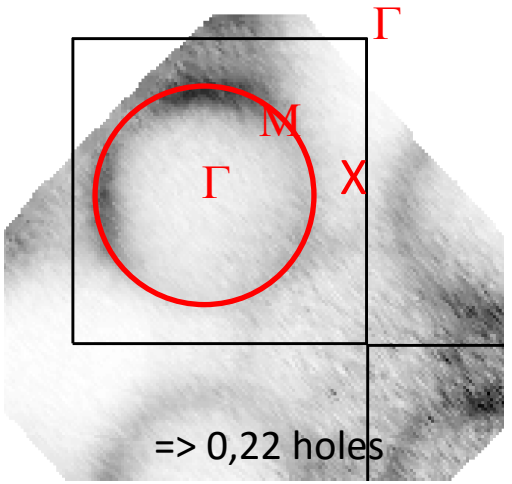


15% Rh



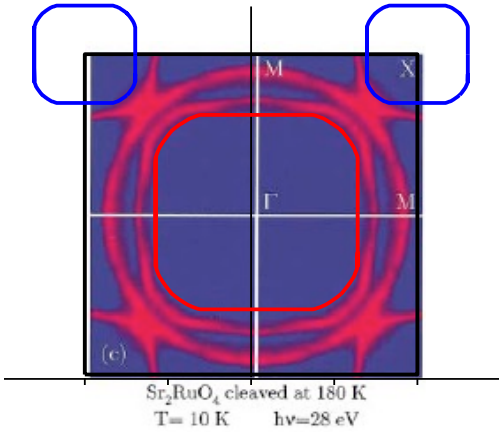
=> 0,15 holes

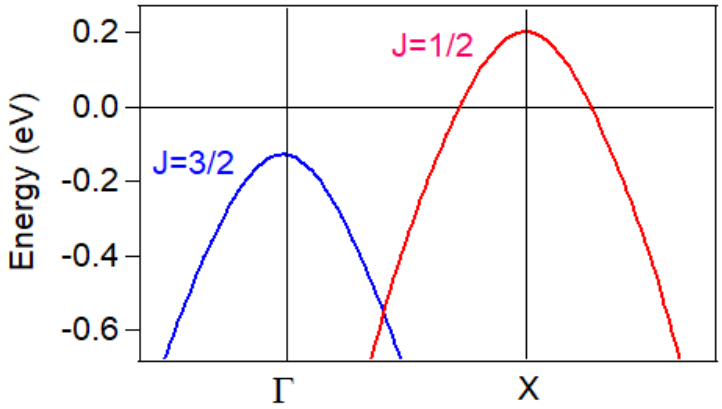
12% Ni



=> 0,22 holes

Sr_2RuO_4





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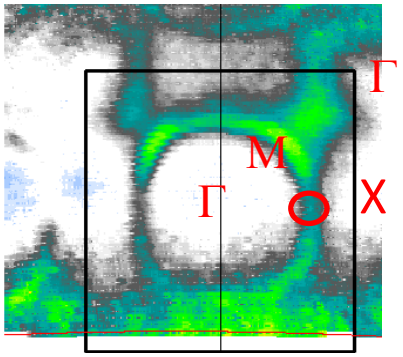
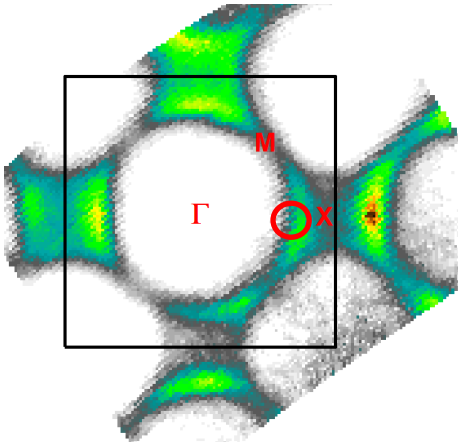
Different types of metals

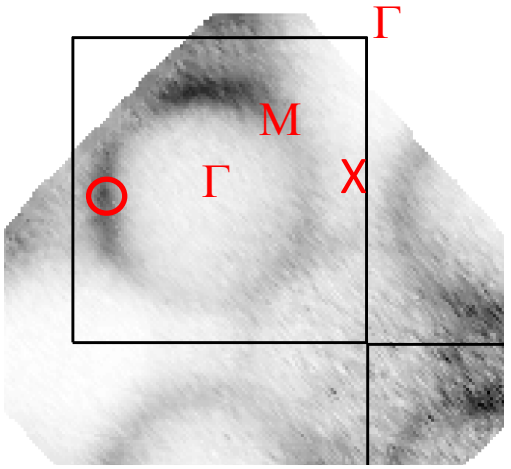
45% Ru

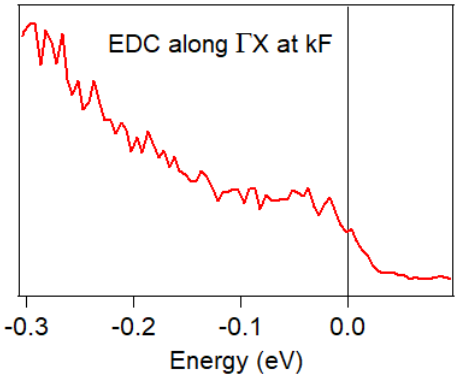


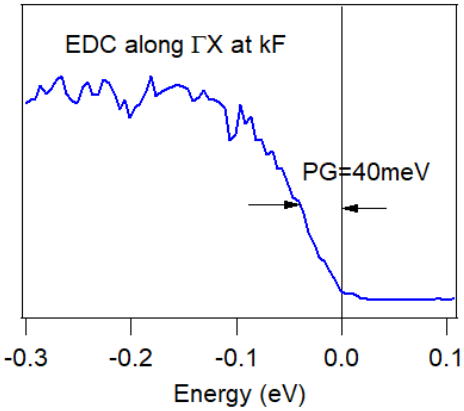
15% Rh

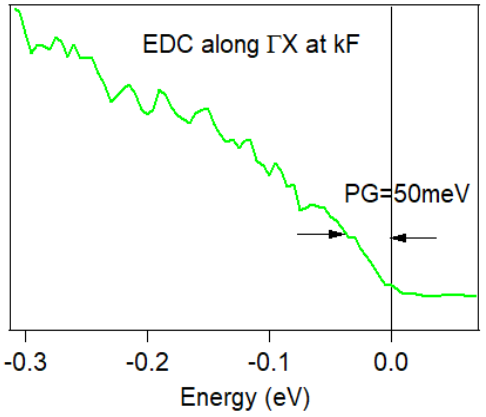


12% Ni









No gap

---------- Pseudogaps ----------

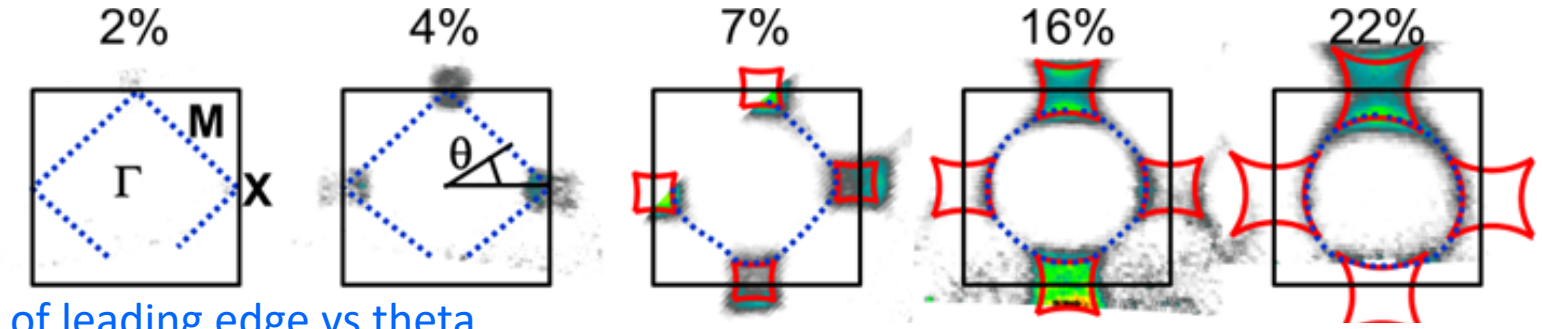
Pseudogap with Rh doping

The pseudogap is found on the entire Fermi Surface

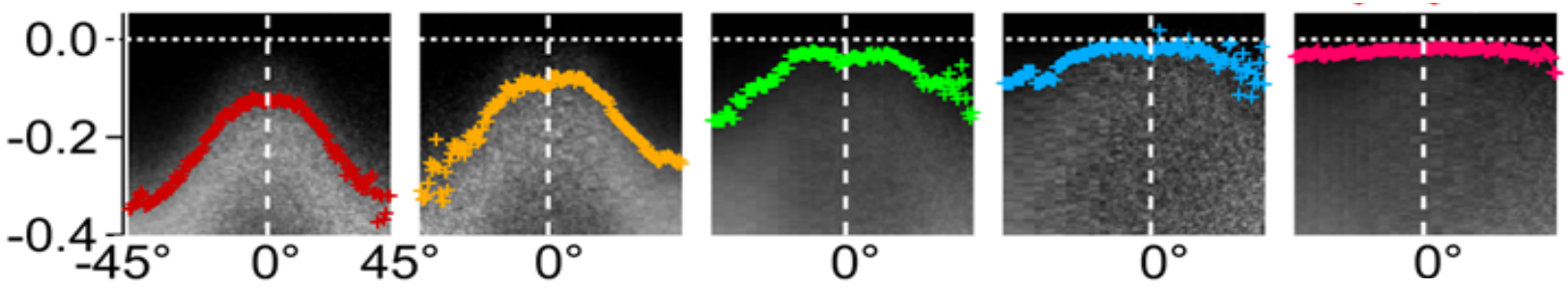
A. Louat, V. Brouet et al., PRB 18

A. Louat, V. Brouet et al., PRB19

Fermi Surface

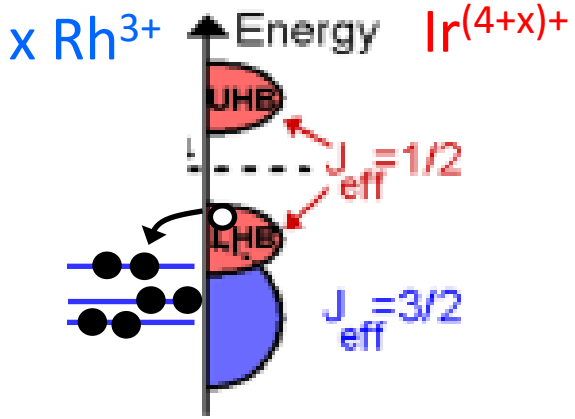


Position of leading edge vs theta



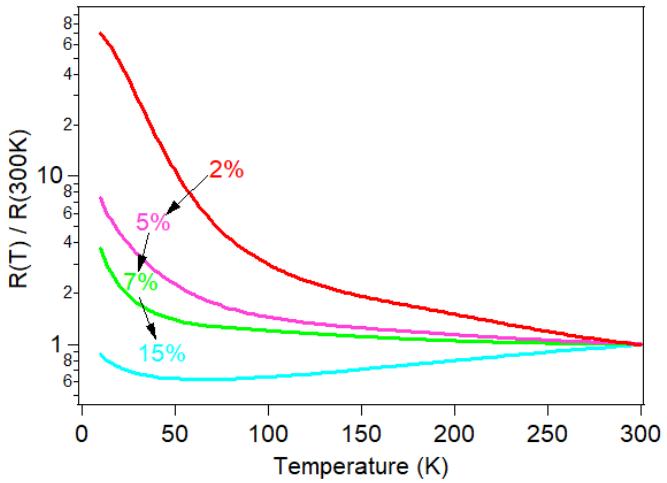
Why is it impossible to get a good metal ? There is as much « disorder » in the case of Ru doping…

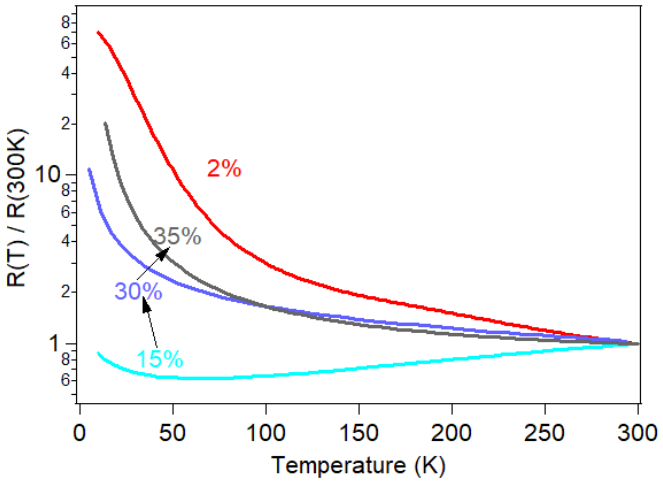
The transfer of holes and size of the gap are certainly correlated



A correlated and disordered metal

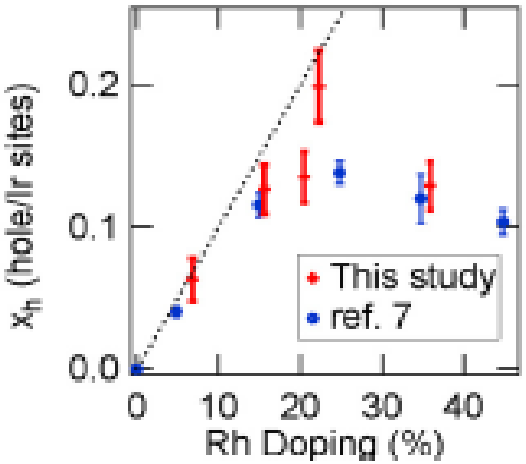
Transport



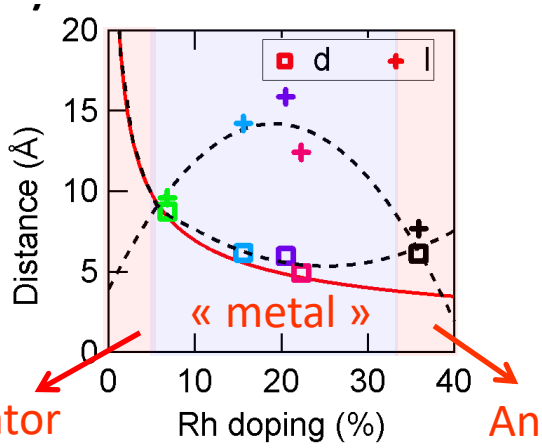


See also : T.F. Qi, G. Cao et al., PRB 12

Number of holes as a function of Rh doping



Distance between holes vs mean free path (from ARPES MDC)



Mott insulator

Anderson insulator

A. Louat, V. Brouet et al., PRB 18

Conclusions

Substitution of Ir with transition metals induce insulator to metal transition of different types

- For Rh and Ni, the metallic state is characterized by the absence of quasiparticles and a **pseudogap** on the entire Fermi Surface
- For Ru, metallicity probably develops in Ru rich domains. There is no pseudogap

Understanding how iridates react to perturbations introduced by transition metals may be important to understand iridates in general.

- Rh and Ni doping triggers the formation of local charged defects
- Ru does not transfer its hole to Ir

$\Rightarrow$ The different hybridization of these transition metals may be the key to this behavior

$\Rightarrow$ By analogy, oxygen defects may play a big role

Thanks to financial support from French National Agency for research
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