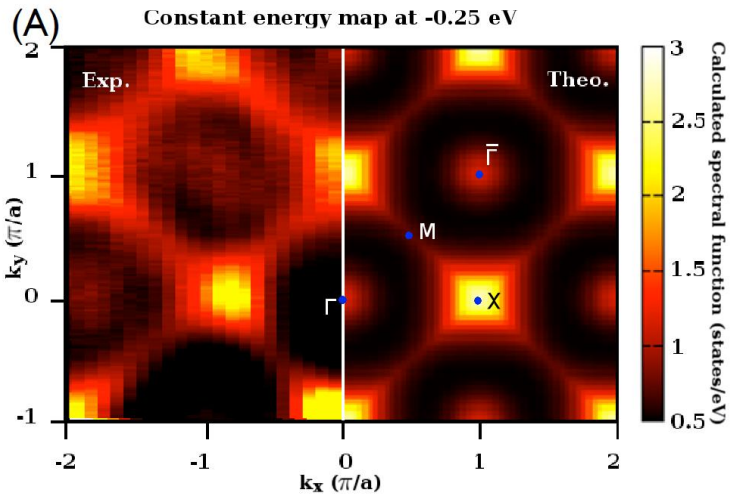
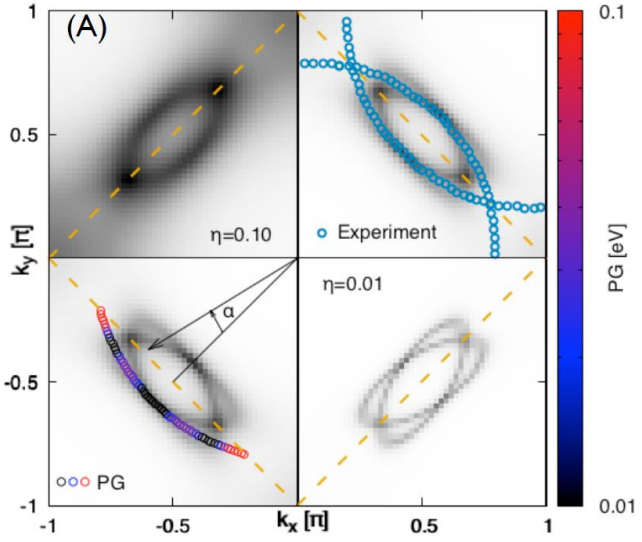
GDR NBODY General Meeting 2022

Toulouse, January 12th 2022

Spectral functions of doped and pure Sr_2IrO_4 : theory versus experiment





Cyril Martins

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In collaboration with

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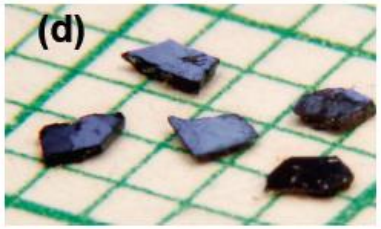
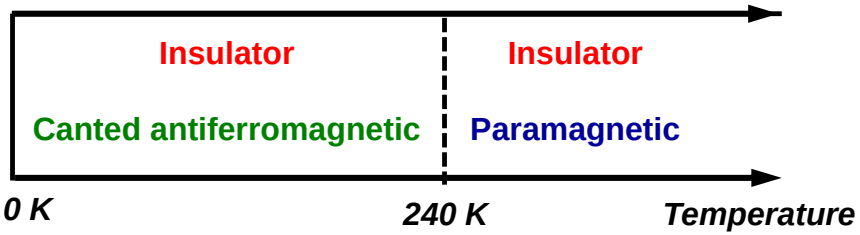
Sr_2IrO_4 – a 5d transition metal oxide



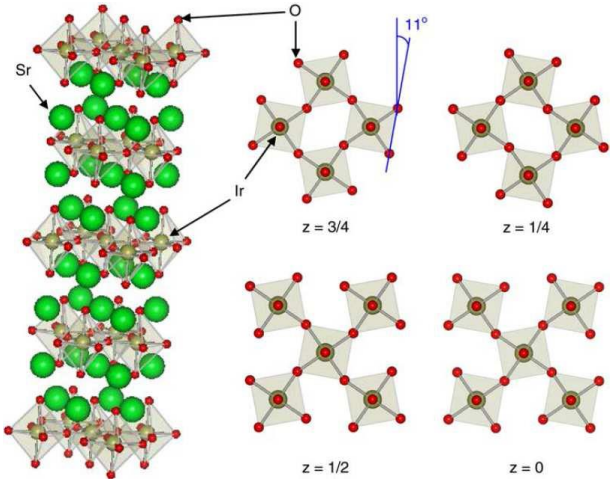
Qi et al, Phys. Rev. B 86, 125105 (2012)

An insulator at all temperatures with a magnetic transition at 240 K



A structure similar to La_2CuO_4 or Sr_2RuO_4

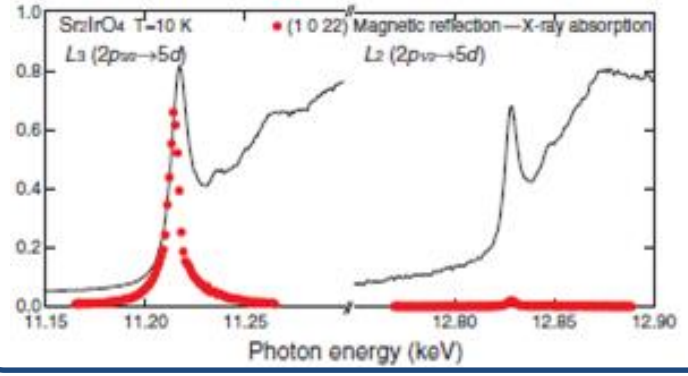
IrO_6 octahedra are rotated around the z axis by about 11° .



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

Experimental evidence of a spin-orbit driven physics

Kim et al, Science 323, 1329 (2009)



No L2-edge observed in resonant X-ray scattering

Observation of a gap at all temperatures in :

Transport – resistivity :	*Qi et al, PRB 86, 125105 (2012)*
Optical spectra :	*Moon et al, PRB 80, 195110 (2009)*
STM :	*Li et al., Sci. Rep. 3, 3073 (2013)*
Photoemission :	*Kim et al., PRL 101, 076402 (2008)*

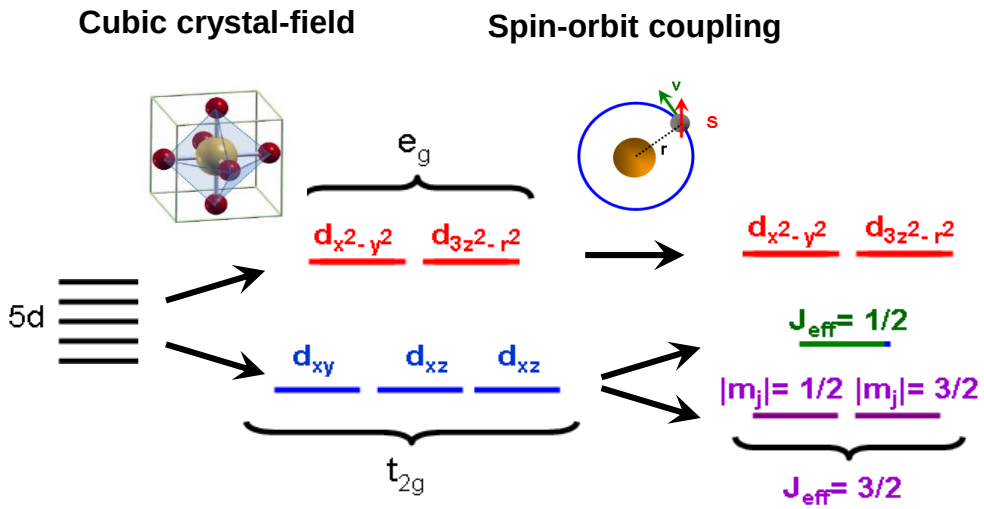
Sr_2IrO_4 – a spin-orbit driven Mott insulator above 240 K

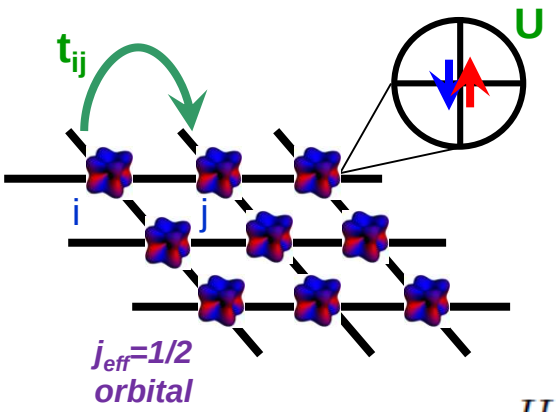
The j_{eff}=1/2 picture for Sr_2IrO_4

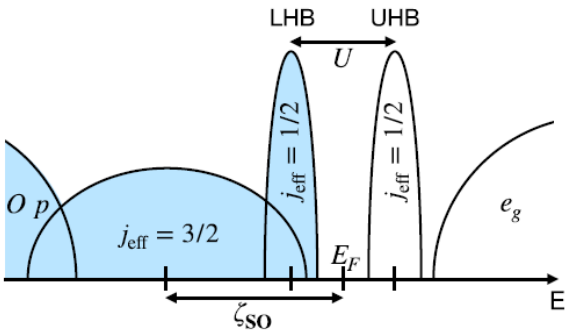
Each Ir atom accommodates 5 electrons in its 5d orbitals.

one half-filled J_{eff}=1/2 state and two completely filled J_{eff}=3/2 states

because of the crystal field and the spin-orbit coupling,







$$H = \sum_{\langle ij \rangle \sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

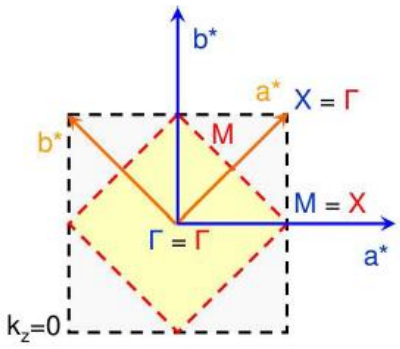
Sr_2IrO_4 is a j_{eff}=1/2 Mott insulator

described by a single-orbital Hubbard model on a 2d square lattice

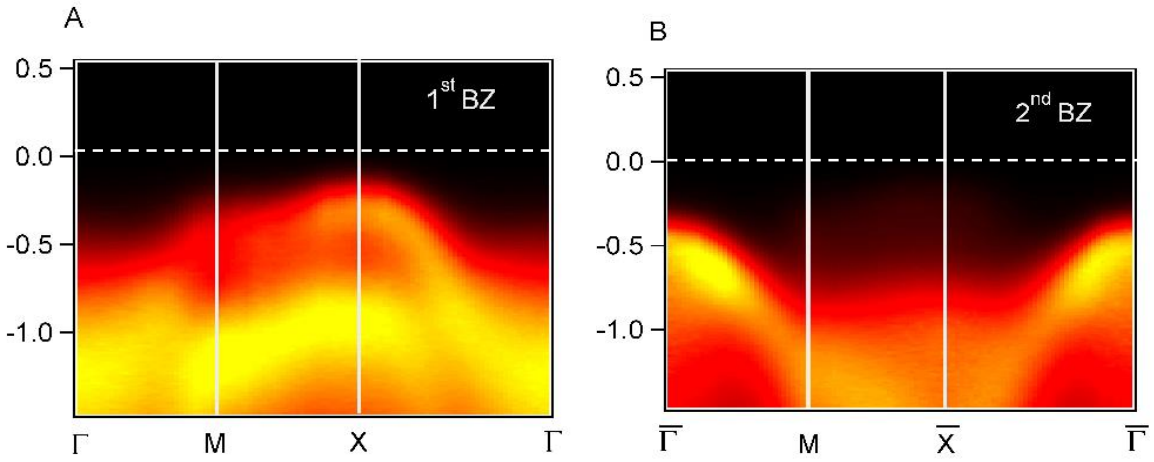
Kim et al., Phys. Rev. Lett. 101, 076402 (2008)
Martins et al., Phys. Rev. Lett. 107, 266404 (2011)

Sr_2IrO_4 – a spin-orbit driven Mott insulator above 240 K

Confirmation of the Mott insulating phase
by angle-resolved photoemission spectroscopy (ARPES) in the paramagnetic phase



First and second Brillouin Zone



Measured ARPES spectra in the first and second Brillouin Zone at 300 K

As in the antiferromagnetic phase :

- The lowest energy excitation disperse up to -0.25 eV at X point.
- Observation of strong matrix element effects.

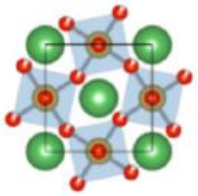
Martins et al., Phys. Rev. Mat. 2, 032001(R) (2018)

Previous ARPES in the antiferromagnetic phase

Kim et al., PRL 101, 076402 (2008)
Yamasaki et al., PRB 89, 121111(R) (2014)
Liu et al., Sci. Rep. 5, 13036 (2015)
Wang et al., PRB 87, 245109 (2013)
Kawasaki et al., PRB 94, 121104(R) (2016)

Sr_2IrO_4 vs. high-T_C superconducting cuprates

Sr_2IrO_4 vs. La_2CuO_4



- **A similar crystal structure**
- **An insulating state described a single-orbital Hubbard model**

F. Wang & T. Senthil, Phys. Rev. Lett. 106, 136402 (2011)

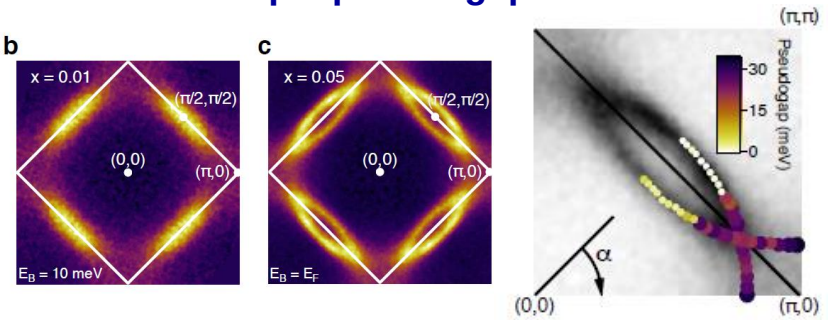
- **An antiferromagnetic state** (at low temperature) **driven by superexchange constants of similar order of magnitude.**

$$J \approx 0,1 \text{ eV}$$

Fujiyama et al., Phys. Rev. Lett. 108, 247212 (2012)

In electron-doped Sr_2IrO_4

- **A metallic paramagnetic state**
- **Observation of lens-like Fermi contours**
- **An anisotropic pseudogap**



Constant energy maps of the spectral density of $(Sr_{1-x}La_x)_2IrO4$

de la Torre et al., Phys. Rev. Lett. 115, 176402 (2015)

BUT STRONG DISCREPANCIES TOO…

- **The key-role of the spin-orbit coupling in Sr_2IrO_4**
- **No superconducting phase ever observed** (up to now…)

Electron doping achieved by :

- ***<u>La substitution :</u>***
  - *Ge et al., PRB 84, 100402 (2011)*
  - *Batisti et al., PRB 95, 235141 (2017)*
- ***<u>Deposit on a K surface layer :</u>***
  - *Kim et al., Science 345, 187 (2014)*
  - *Kim et al., Nat. Phys. 12, 37 (2016)*
- ***<u>Oxygen depletion :</u>***
  - *Korneta et al., PRB 82, 115117 (2010)*

Outlines

NON-LOCAL CORRELATIONS IN SR_2IRO_4 : INTRODUCING ORIENTED CLUSTER-DMFT

Necessity to take into account non-local correlations in Sr_2IrO_4

The oriented cluster DMFT (OC-DMFT) in a nutshell

SPECTRAL FUNCTIONS : OC-DMFT VS. EXPERIMENTS

Spectral function and energy cuts of Sr_2IrO_4

Fermi arcs and pseudogap in electron-doped Sr_2IrO_4

NEW INSIGHTS FROM EXPERIMENTS : THE END OF THE J_{EFF}=1/2 MODEL ?

Symmetries of the « bands » from polarized ARPES

Magnetization density distribution from polarized neutron diffraction

Spectral functions of doped and pure Sr_2IrO_4 : theory versus experiment

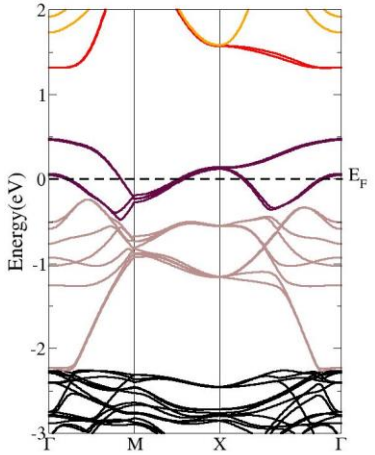
Non-local correlations in Sr_2IrO_4 : introducing Oriented Cluster-DMFT

Necessity to take into account non-local correlations in Sr_2IrO_4

Performing first-principles calculations for Sr_2IrO_4 within LDA+DMFT

DFT part

Solving
Kohn-Sham equations



Interfacing part

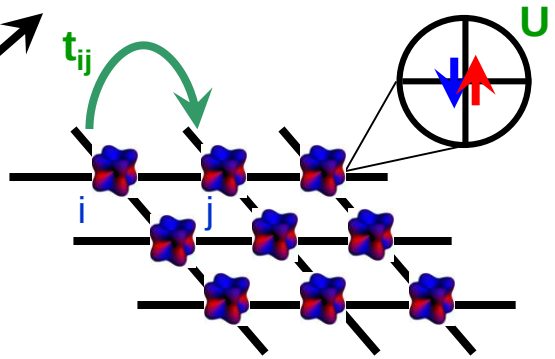
Constructing
Wannier orbitals based
on the j_{eff}=1/2 orbitals

DMFT part

Solving
the Hubbard model
Within DMFT

C-RPA part

Evaluating
on-site interaction
Hubbard U



To evaluate « consistently » local interaction Hubbard U term,
use of the constrained Random Phase Approximation (c-RPA)

About LDA+DMFT

Lichtenstein & Katsnelson, Phys Rev B 57, 6884 (1998)
Anisimov et al, J. Phys. Cond Mat. 9, 7359 (1997)

About constrained-RPA

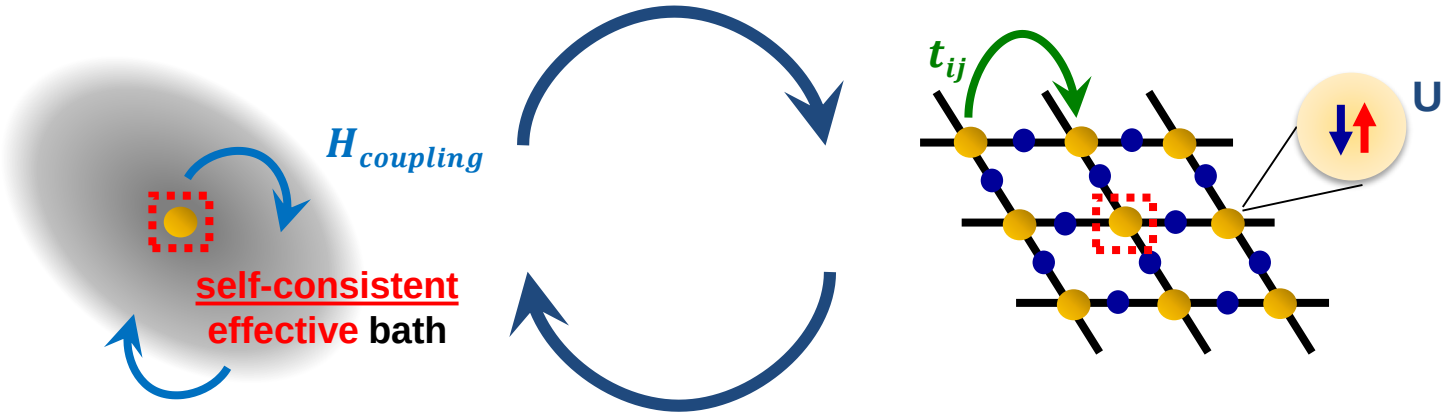
Aryasetiawan et al., Phys. Rev. B 70, 195104 (2004)
Vaugier et al., Phys. Rev. B 86, 165105 (2012)

Necessity to take into account non-local correlations in Sr_2IrO_4

DMFT is an extension to quantum many body systems of classical mean-field theory.

THE SELF-CONSISTENT CYCLE IN DMFT

$$\Sigma_{ii}(\omega) \equiv \Sigma_{imp}(\omega) \quad \text{and} \quad \Sigma_{ij}(\omega) \equiv 0$$



$$G_{imp}(\omega) \equiv G_{loc}(\omega) = \sum_{\boldsymbol{k}} G(\boldsymbol{k}, \omega)$$

DMFT is based on the Green function formalism.

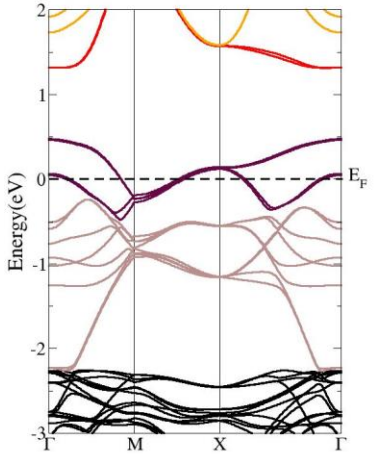
Local quantum fluctuations are taken into account.
Temperature effects can be taken into account too.

Necessity to take into account non-local correlations in Sr_2IrO_4

Performing first-principles calculations for Sr_2IrO_4 within LDA+DMFT

DFT part

Solving
Kohn-Sham equations



Interfacing part

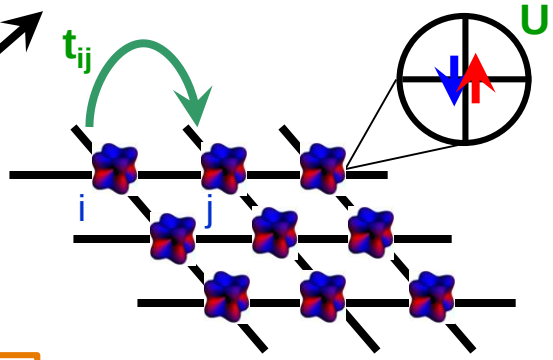
Constructing
Wannier orbitals based
on the j_{eff}=1/2 orbitals

DMFT part

Solving
the Hubbard model
Within DMFT

C-RPA part

Evaluating
On-site and Inter-site
interaction



On-site interaction $U = 2.2$ eV
Inter-site interaction $V = 0.9$ eV

To evaluate « consistently » **local and non-local** interaction,
use of the constrained Random Phase Approximation (c-RPA)

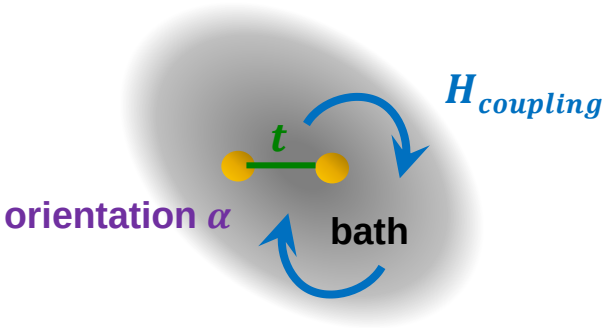
Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)

The oriented cluster DMFT (OC-DMFT) in a nutshell

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)

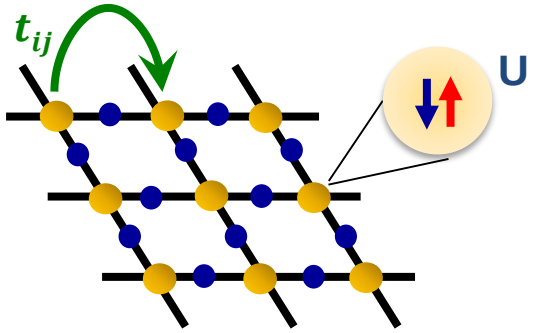
OC-DMFT extends cluster-DMFT
to oriented clusters as representative entities in a solid.

AUXILIARY PROBLEM



An oriented two-site cluster coupled to a bath
(along x or y, for a 2d square lattice)

« REAL » LATTICE PROBLEM



$$H = \sum_{\langle ij \rangle \sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

Hubbard model

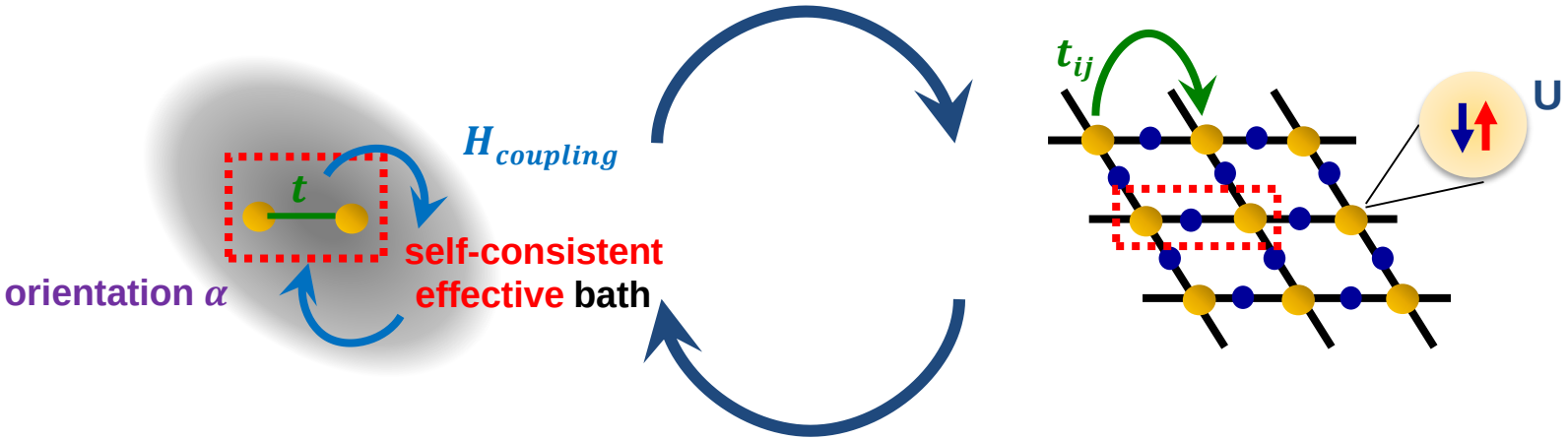
Hubbard, Proc Roy Soc Lond A, 276 (1964)

The oriented cluster DMFT (OC-DMFT) in a nutshell

Self-consistency condition

Translational symmetry and point group symmetry are restrored.

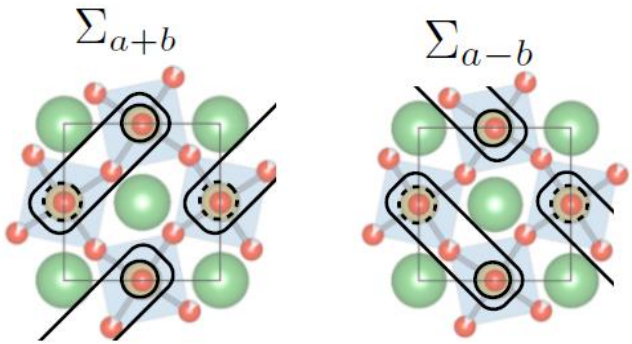
$$G_{loc}(\omega) = \sum_{\mathbf{k},\alpha} G_\alpha(\mathbf{k},\omega) \qquad G_\alpha(\mathbf{k},\omega) = [\omega + \mu - H(\mathbf{k}) - \Sigma^\alpha_{dimer}(\omega)]^{-1}$$

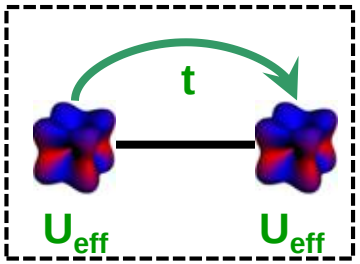


Mapping the « real » problem onto the auxiliary one

The local quantum fluctuations of the lattice
and the short-range antiferromagnetic fluctuations are included.

The oriented cluster DMFT (OC-DMFT) in a nutshell



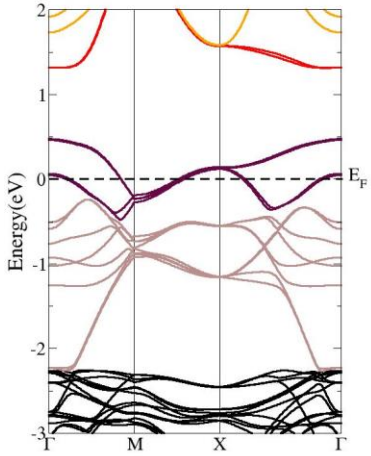


We used two Ir-Ir dimers oriented along $a+b$ and $a-b$

an effective Hubbard interaction $U_{\mathrm{eff}} = U - V$ for a dimer

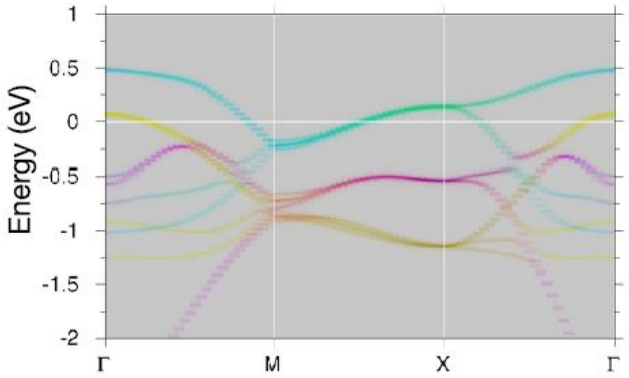
DFT part

Solving Kohn-Sham equations



Interfacing part

Constructing Wannier orbitals based on the j_{eff}=1/2 orbitals



OC-DMFT part

Adding local and non-local self-energies (Hubbard I type)

Wannier projected j_{eff}=1/2 bands in turquoise

$$\left|j_{\mathrm{eff}} = \frac{1}{2}, \pm\frac{1}{2}\right\rangle = \frac{1}{\sqrt{3}}(|xy, \pm\sigma\rangle \pm |yz, \mp\sigma\rangle + i|xz, \mp\sigma\rangle)$$

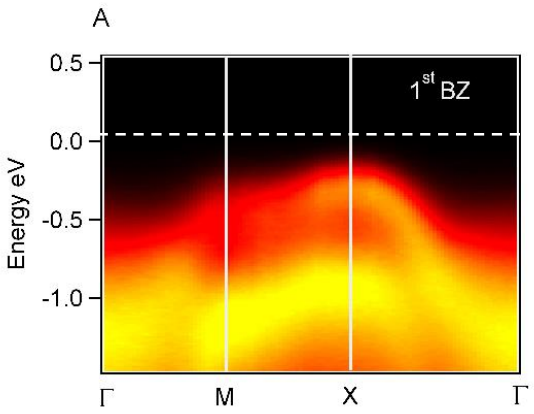
Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)

Spectral functions of doped and pure Sr_2IrO_4 : theory versus experiment

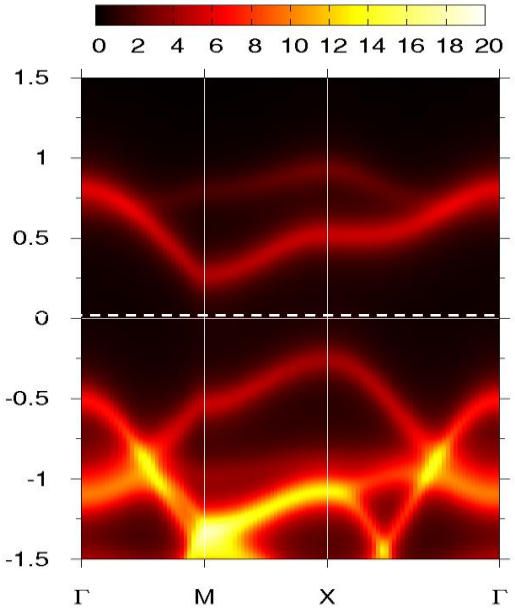
Spectral functions : OC-DMFT vs. Experiments

Spectral function and energy cuts of Sr_2IrO_4

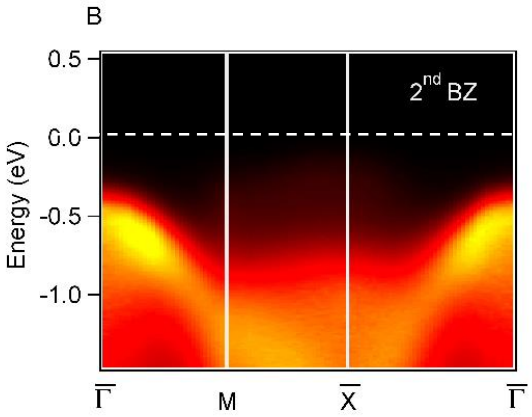
Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)



Measured ARPES spectrum in the first Brillouin Zone at 300 K



Calculated spectral function within OC-DMFT
(no matrix elements included)



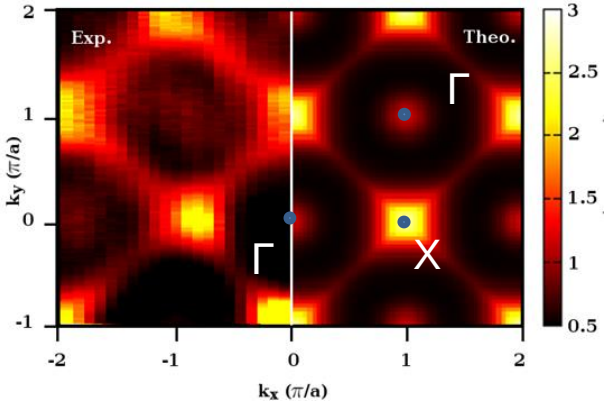
Measured ARPES spectrum in the second Brillouin Zone at 300 K

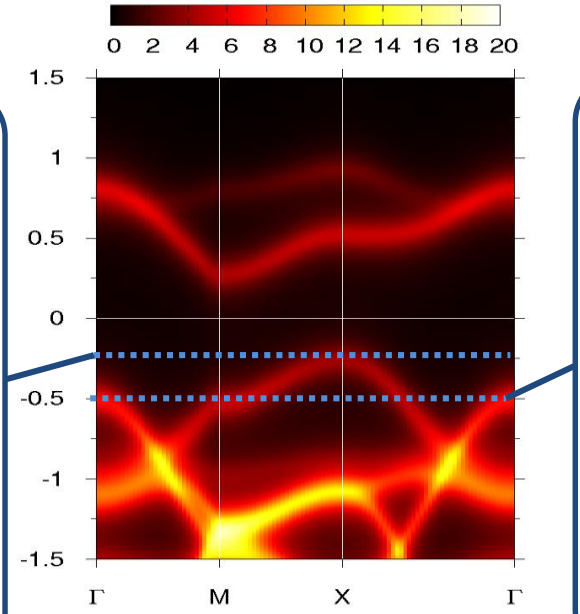
AN IMPRESSIVELY GOOD AGREEMENT BETWEEN THEORY AND EXPERIMENT !!

Spectral function and energy cuts of Sr_2IrO_4

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)

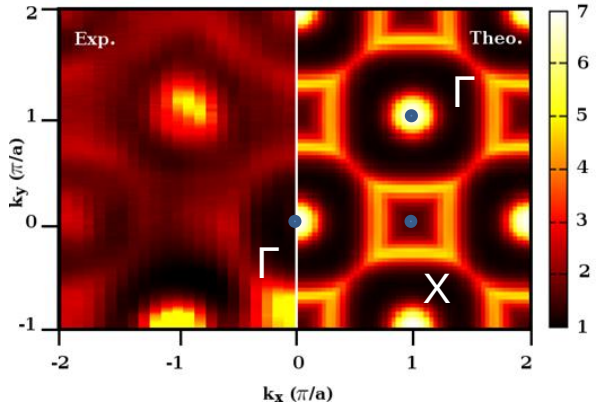
Constant energy map of the spectral density at -0.25 eV





Calculated spectral function within OC-DMFT
(no matrix elements included)

Constant energy map of the spectral density at -0.5 eV



AN IMPRESSIVELY GOOD AGREEMENT BETWEEN THEORY AND EXPERIMENT !!

Spectral function and energy cuts of Sr_2IrO_4

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
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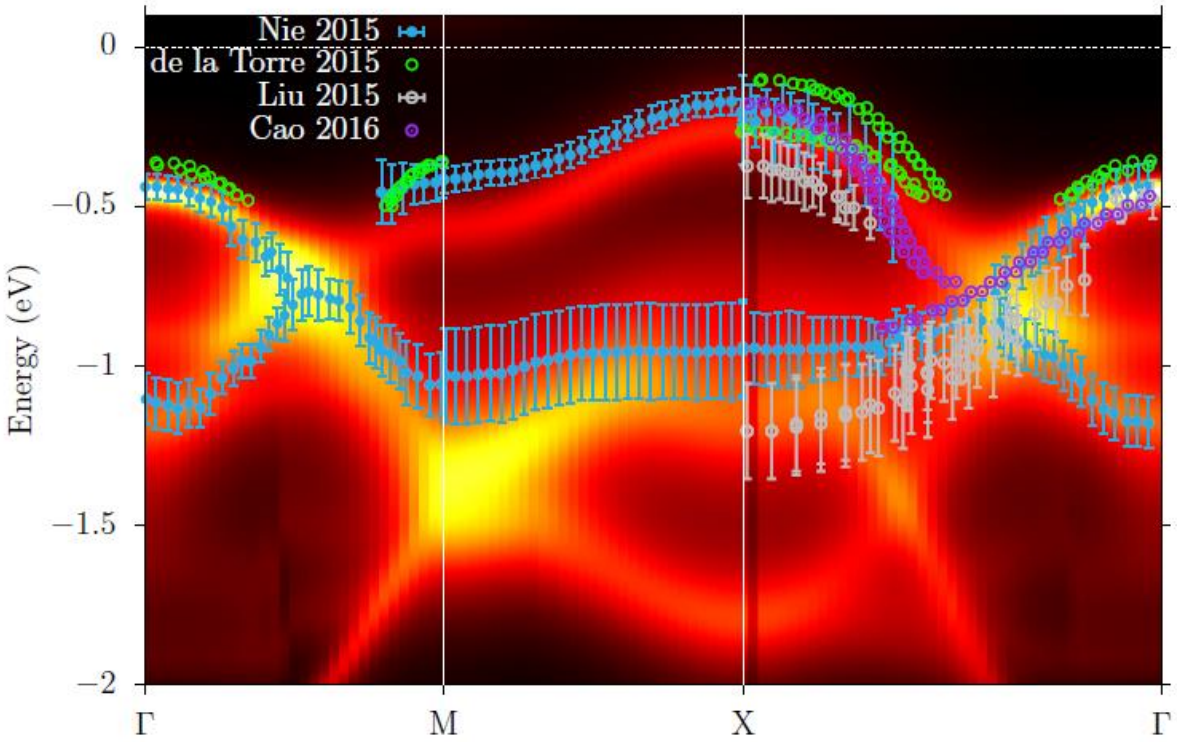


AN IMPRESSIVELY GOOD AGREEMENT BETWEEN THEORY AND EXPERIMENT

even in the antiferromagnetic phase !!

Fermi arcs and pseudogap in electron-doped Sr_2IrO_4

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)

For electron-doped Sr_2IrO_4

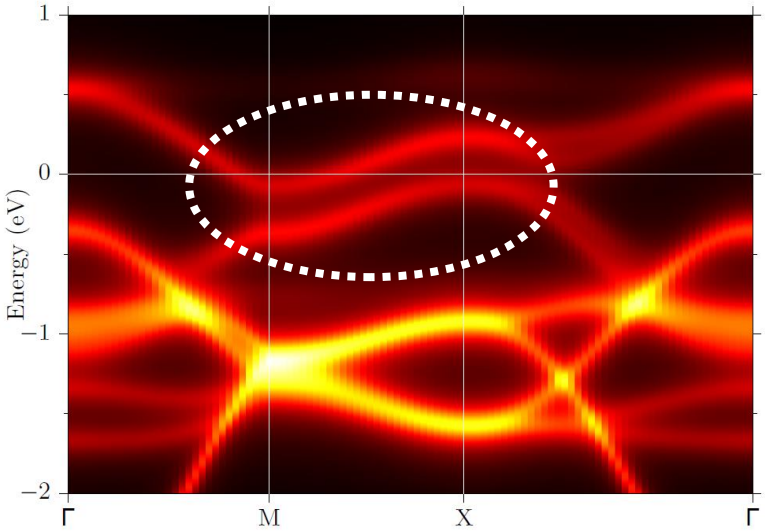
AN OVERALL GOOD AGREEMENT with ARPES spectra of $(Sr_{1-x}La_x)_2IrO4$

Fermi pockets around M from the j_{eff}=1/2 upper branch

The j_{eff}=1/2 lower branch close to the Fermi level at X.

Here, we use
an effective Hubbard interaction
as an adjustable parameter.
because the electronic screening
is further enhanced in a metal

Calculated spectral function of 10% electron-doped Sr_2IrO_4 within OC-DMFT



U_{eff} = 0.6 eV

Fermi arcs and pseudogap in electron-doped Sr_2IrO_4

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)

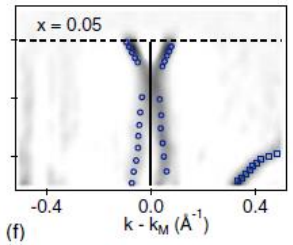
For electron-doped Sr_2IrO_4

AN OVERALL GOOD AGREEMENT with ARPES spectra of $(Sr_{1-x}La_x)_2IrO4$

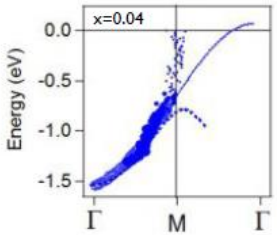
Fermi pockets around M from the j_{eff}=1/2 upper branch

The j_{eff}=1/2 lower branch close to the Fermi level at X.

No Dirac point at M at -0.1 eV

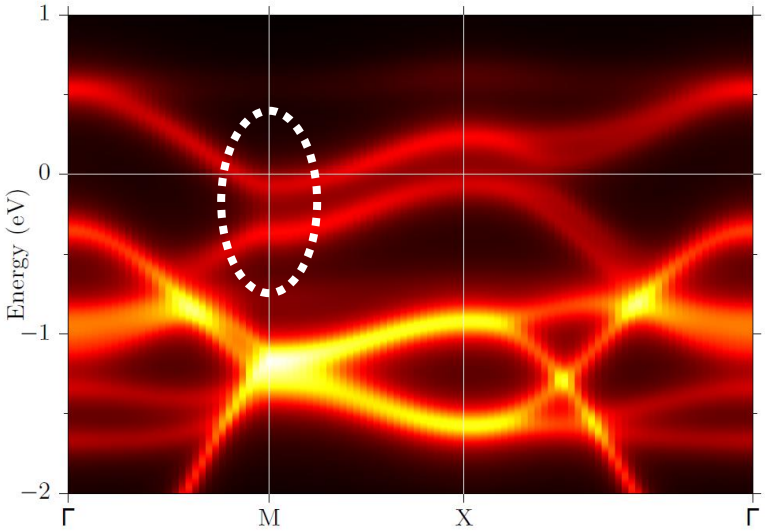


de la Torre et al., PRL 115, 176402 (2015)



Brouet et al., PRB 92, 081117 (2015)

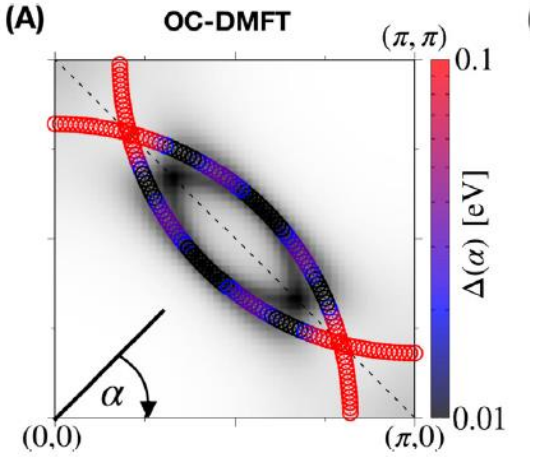
Calculated spectral function of 10% electron-doped Sr_2IrO_4 within OC-DMFT



U_{eff} = 0.6 eV

Fermi arcs and pseudogap in electron-doped Sr_2IrO_4

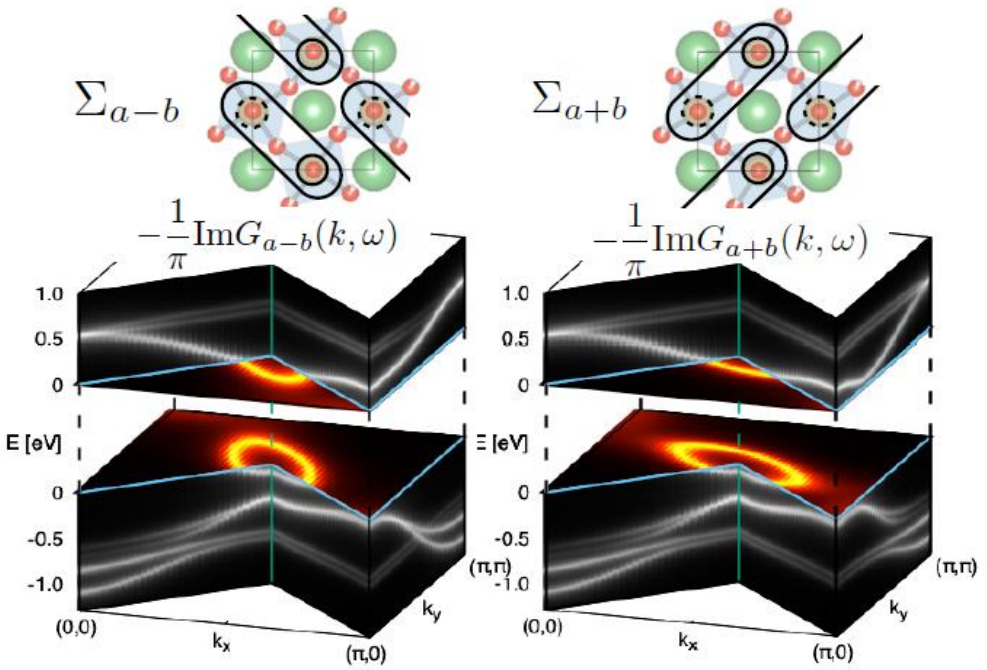
Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)



With increased resolution η :
Two-lense nature of pocket visible

With experimentally (reduced) resolution η :
Only one Fermi pocket

Its tip corresponds to the region between the pockets.



In the average configuration,
2 canted pockets around M :

Each oriented dimer leads to a Fermi pocket tilted by ~15°

Fermi arcs and pseudogap in electron-doped Sr_2IrO_4

Martins et al., Phys. Rev. Materials 2, 032001(R) (2018)
Lenz et al., *J. Phys.: Condens. Matter* 31 293001 (2019)

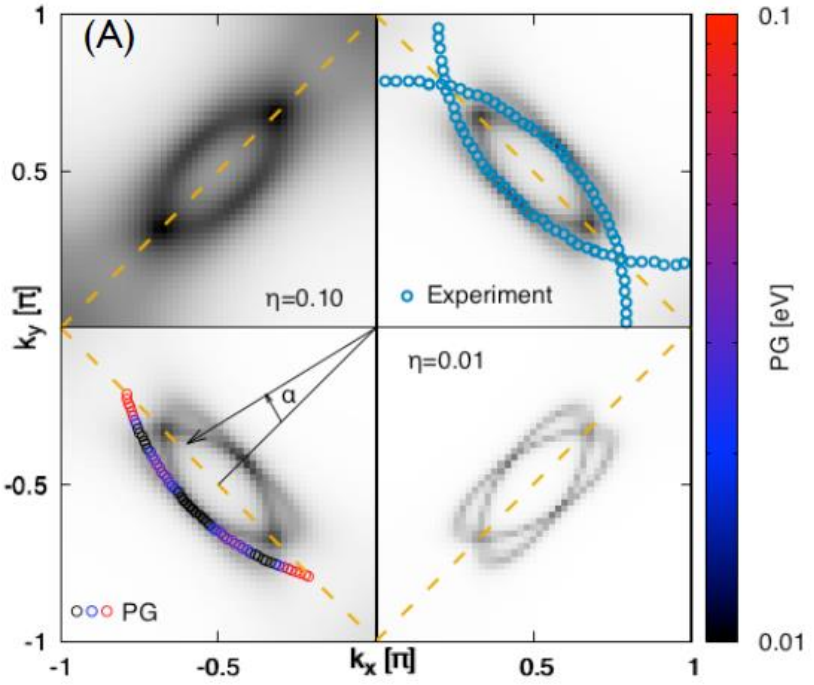
Size and shape of lens-shaped Fermi pockets around M

in **VERY GOOD AGREEMENT** with experiments

A PSEUDOGAP for $4° < \alpha < 15°$ with artificially increased resolution η

Additional pseudogap-like feature emerges for $21° < \alpha < 26°$ at sufficiently large temperature,

Fermi surface of 10% electron-doped Sr_2IrO_4



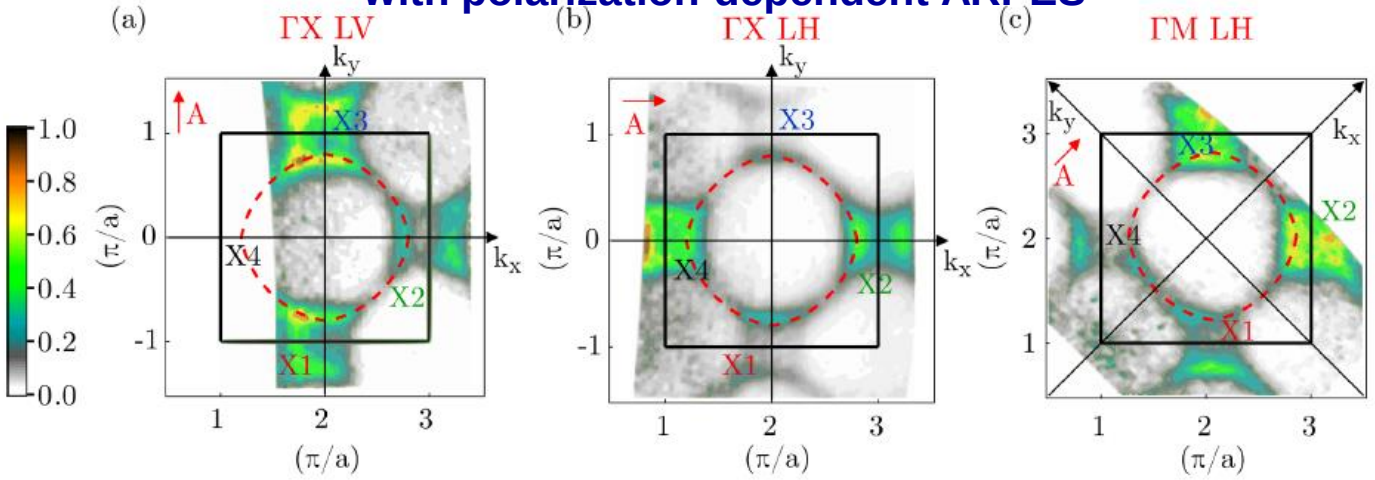
Spectral functions of doped and pure Sr_2IrO_4 : theory versus experiment

New insights from experiments :
The end of the J_{eff}=1/2 model ?

Symmetries of the « bands » from polarized ARPES

A. Louat et al., Phys. Rev. B 100, 205135 (2019)

Highlighting orbital characters of the spectral function with polarization-dependent ARPES



d_{yz} character | d_{xz} character | d_{xy} character

Fermi surface for 15% Rh doped Sr_2IrO_4 at 50 K measured by ARPES using different polarization

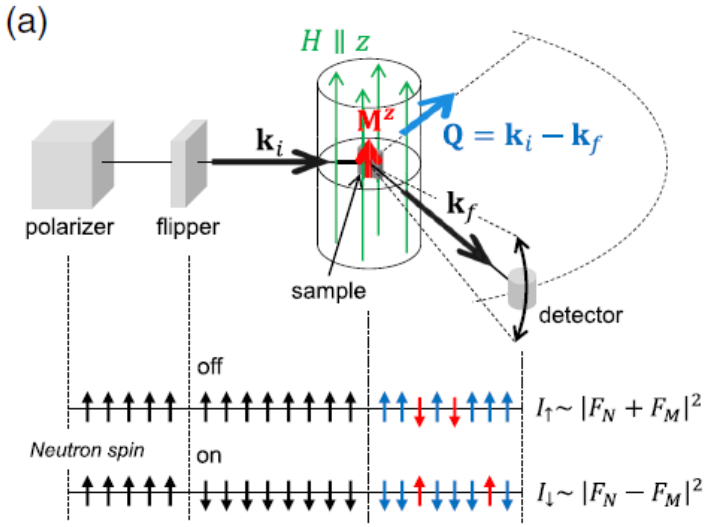
Contrary to the j_{eff}=1/2 model,

a well-defined and k-dependent orbital character in terms of cubic harmonics.

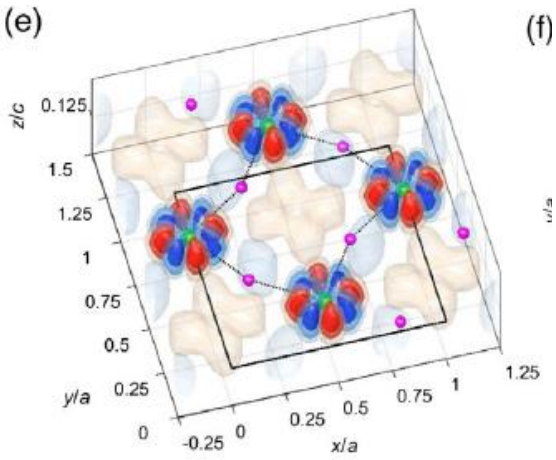
Electron and spin density distribution from polarized neutron diffraction

J. Jeong et al., Phys. Rev. Lett 125, 097202 (2020)

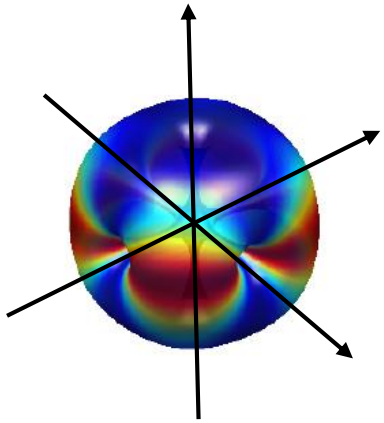
Probing the electron and spin density distribution with polarized neutron diffraction



Experimental setup for a polarized neutron diffraction experiment (B = 5 T ; T = 4 K)



Magnetization density distribution reconstructed from multipole expansion refinement



Electron and spin density distribution for a j_{eff}=1/2 orbital

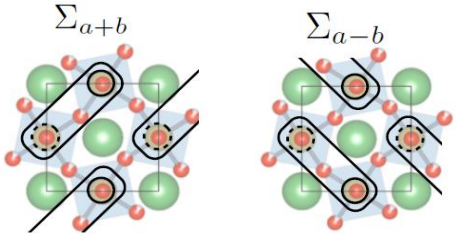
Contrary to the j_{eff}=1/2 model,
an anisotropic and aspherical magnetization density distribution (closer to a d_{xy} orbital).

Role of non-local correlations in pure and electron-doped strontium iridate (Sr_2IrO_4)

Conclusion & Outlooks

Conclusions and outlooks

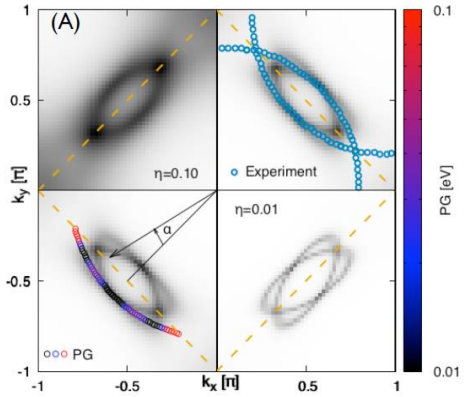
The oriented-cluster DMFT



- **Preserve the local point-group symmetry of the system**
- **Allows to take into account short-range antiferromagnetic fluctuations**

For pure and electron-doped Sr_2IrO_4

- **Spectral functions within OCDMFT in very good agreement with ARPES.**
- **The pseudogap is a direct consequence of the non-local antiferromagnetic fluctuations.**



BUT

The most recent experimental data

seem to jeopardize the jeff=1/2 picture even in pure Sr_2IrO_4 !

Thank you for your attention