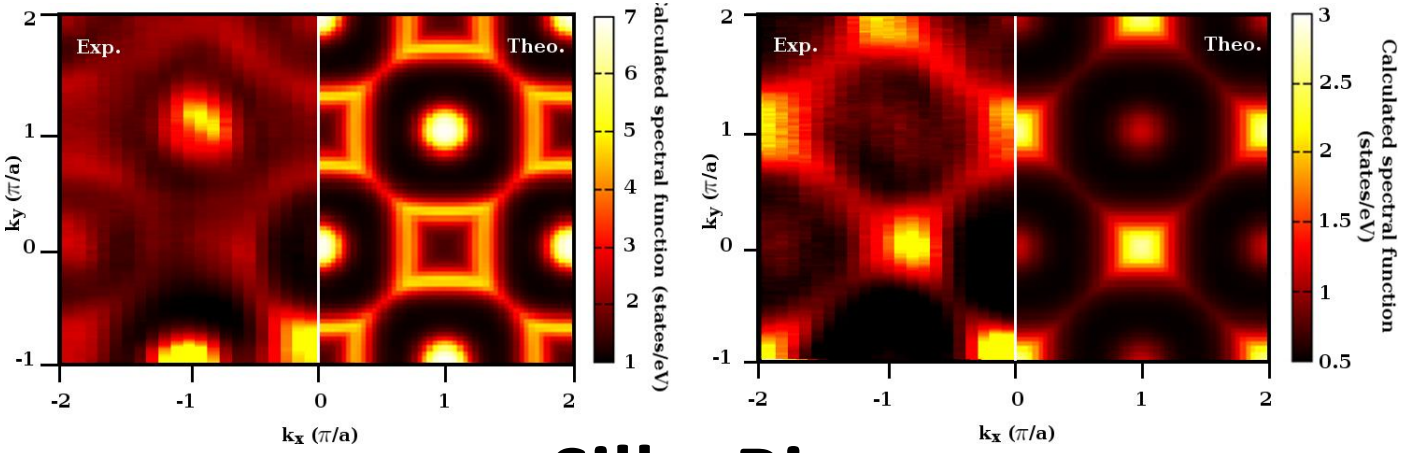
The Quantum Many-Body Problem from a Dynamical Mean Field Theory Perspective

– or –

Spectral Properties of Correlated Materials from First Principles



Phys. Rev. Mat. 2, 032001(R) (2018)

Silke Biermann

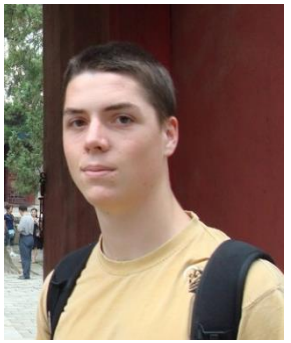
Centre de Physique Théorique,

Ecole Polytechnique, Institut Polytechnique de Paris, France

Cyril Martins
(now: MdC at Toulouse U.)

"Electronic Structure Theory" at CPHT:

Correlated materials from first principles



A. van Roekeghem
(now: permanent researcher at CEA Grenoble)

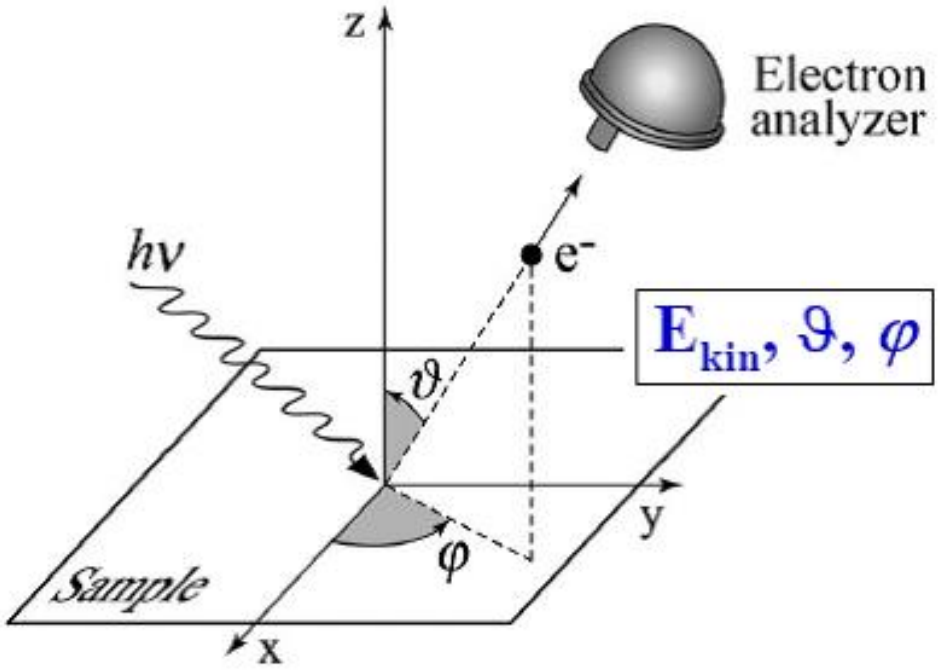
S. Backes, J. Steinbauer, **Benjamin Lenz**, A. Galler, S. Bhandary, S. Panda, M. Turtulici

(now MdC in Jussieu)

Also: A. Subedi, L. Pourovskii

Spectral properties …

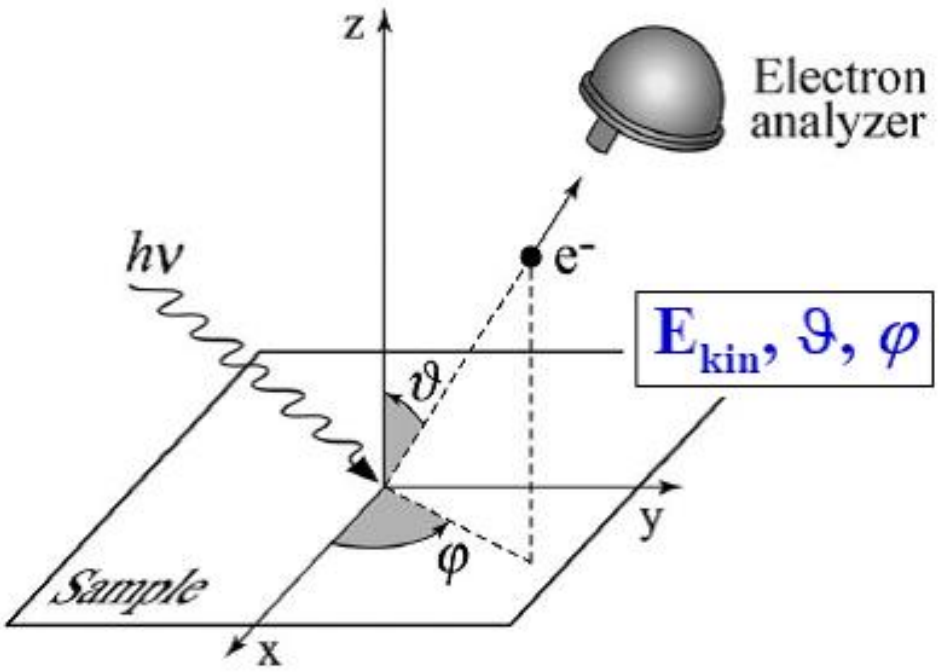
Cf Pina's Talk !



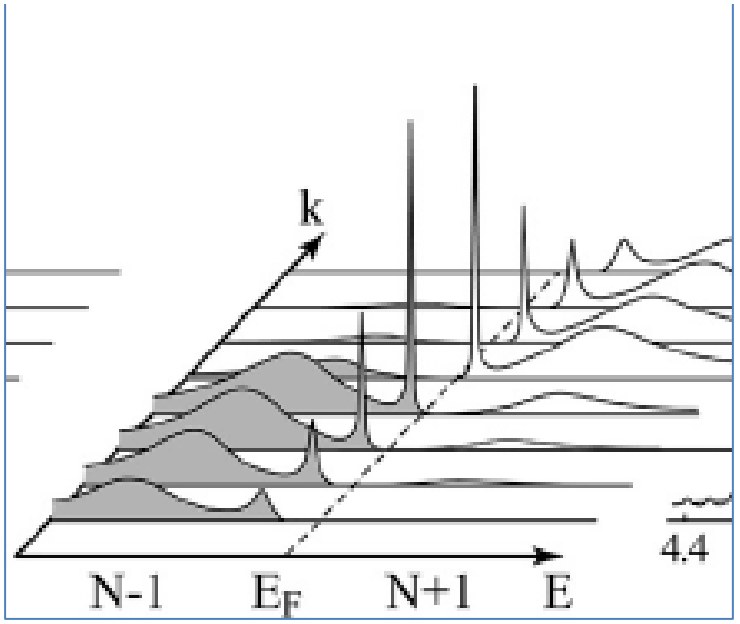
Can we calculate spectral functions from first principles ?

Intuitive definition:

Spectral function: A(k,ω) describes electron addition and removal processes.
=> Photoemission and inverse photoemission



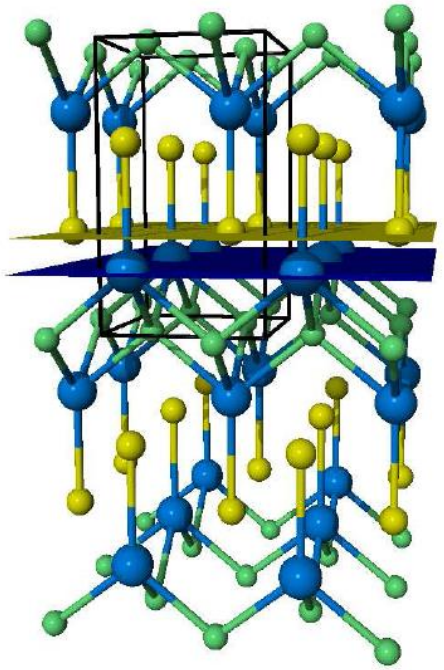
Electron addition/removal spectra



NB. Formal definition in language of Green's functions: $A(k,\omega) = -\frac{1}{\pi} G(k,\omega)$

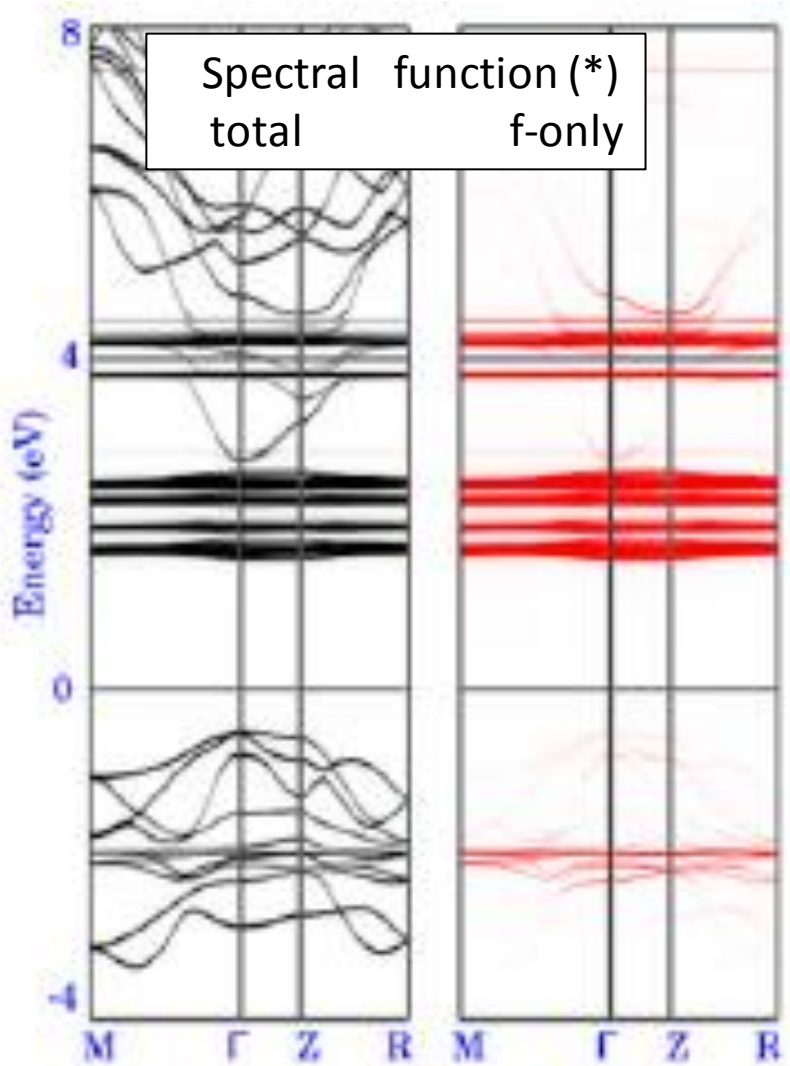
Disclaimer: neglect matrix element effects.

Example: CeSF – an f-electron pigment



Ce: $4f^1$ configuration, paramagnetic

Example :
CeSF – an f-electron pigment
(cf. Rhodia's Neolor series)



f-electrons in CeFS

- remain "atomic-like", i.e. localized on the atomic sites
- do no form bands
- no Bloch states …

=> "Mott insulator"

- Spectral feature: multiplets …

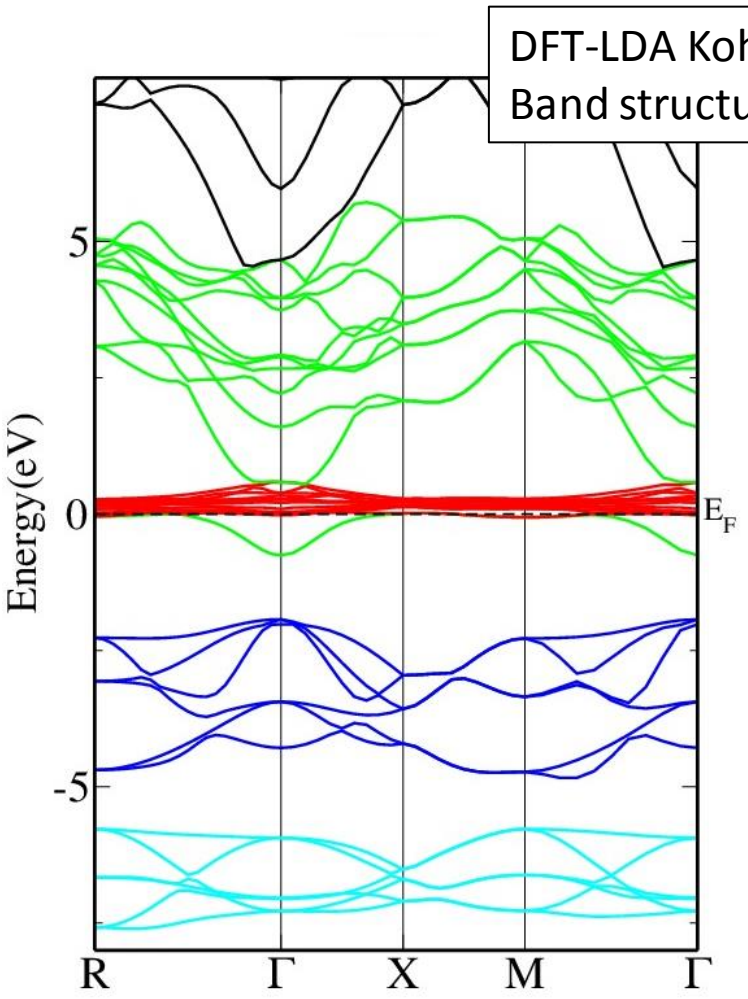
Ce: $4f^1$ configuration, paramagnetic

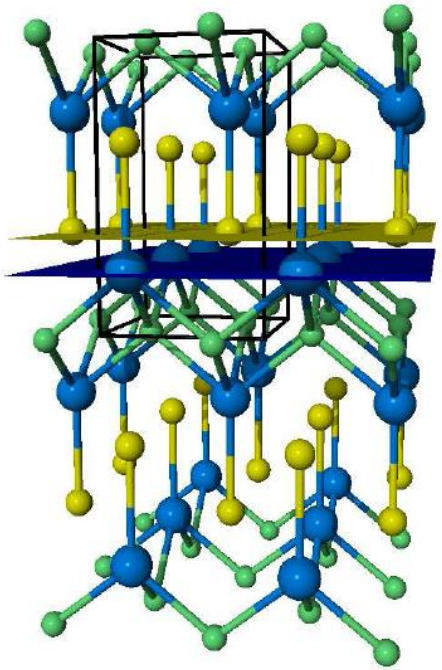
Calculated colour of CeSF:

Tomczak, Pourosvkii, Vaugier, Georges, Biermann, PNAS (2013)

(*) calculated from Density Functional + Dynamical Mean Field Theory

Example: CeSF – an f-electron pigment





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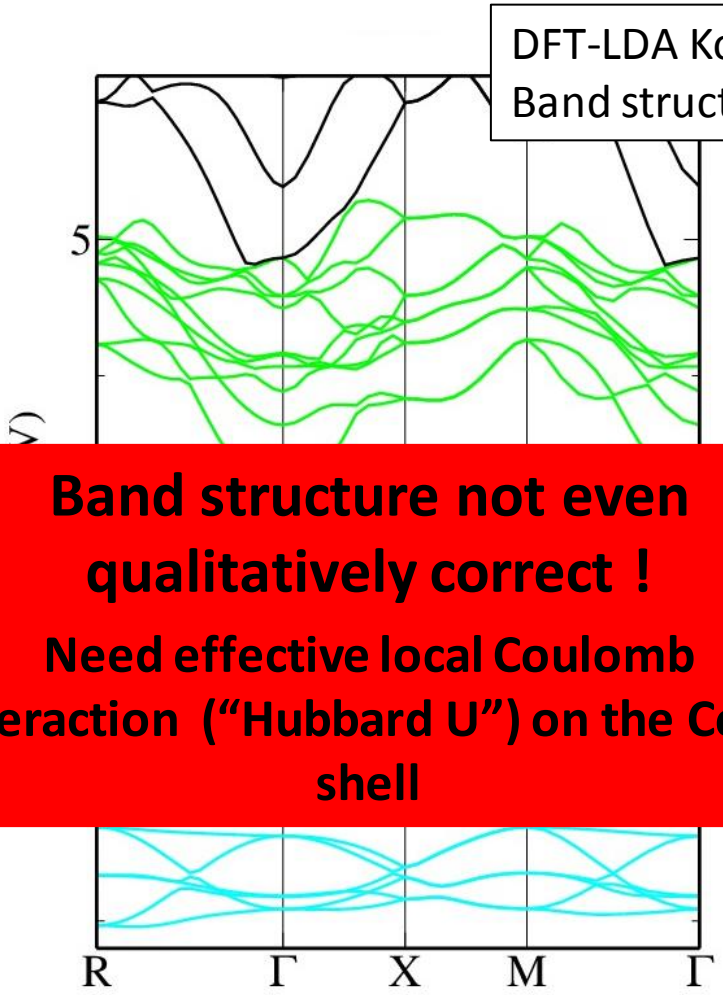
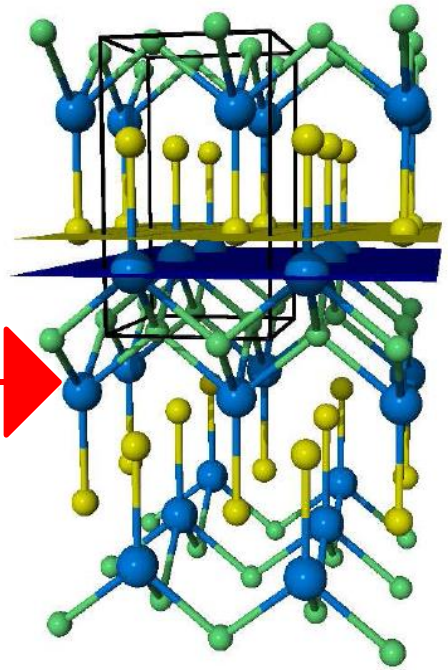
Example: CeSF – an f-electron pigment



Band structure not even qualitatively correct !

Need effective local Coulomb interaction ("Hubbard U") on the Ce 4f shell

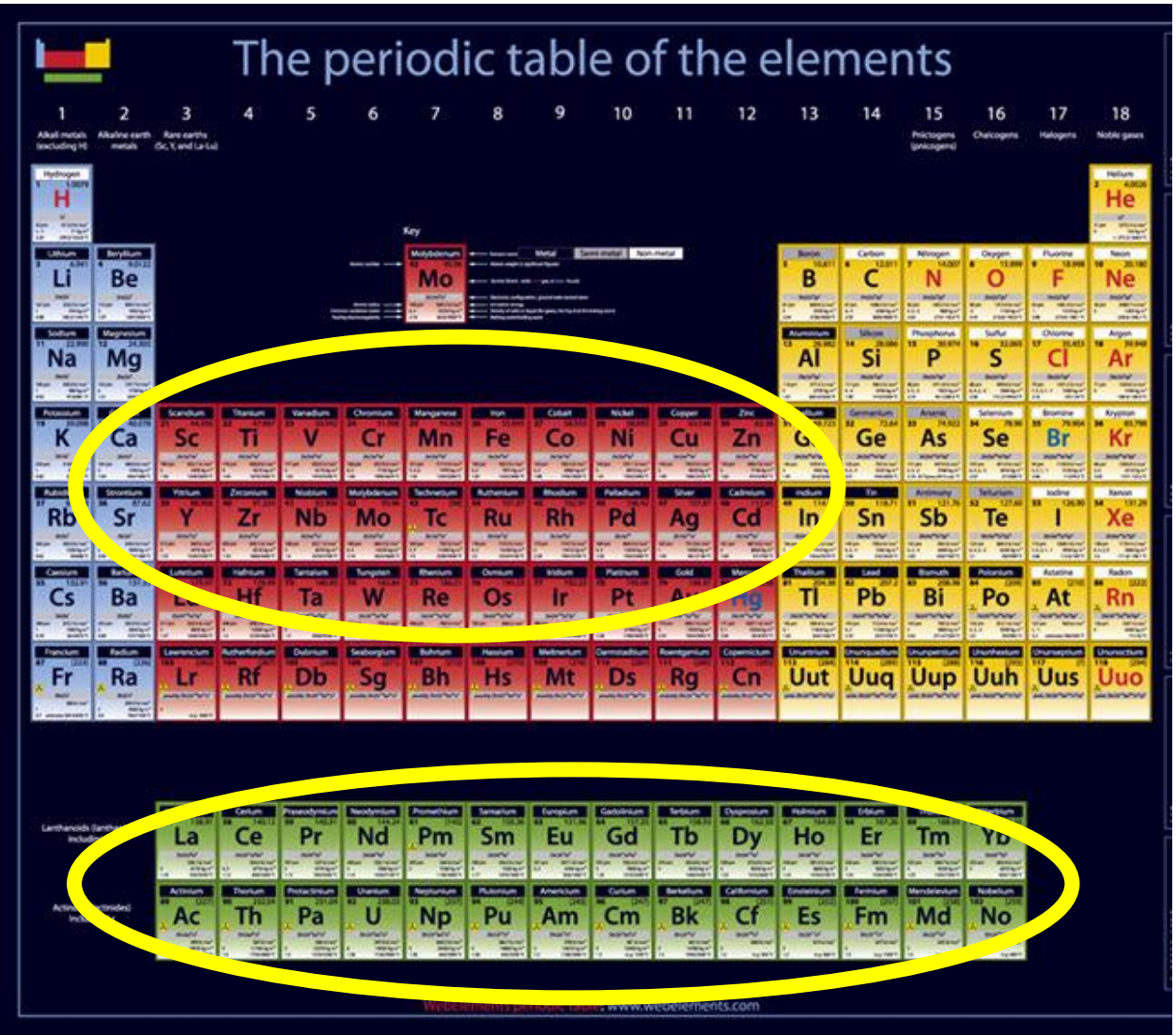


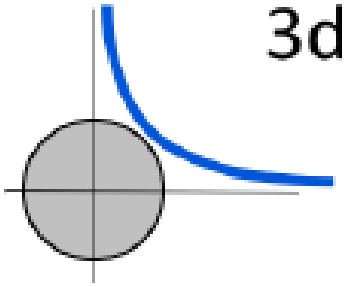
Ce: $4f^1$ configuration, paramagnetic

Tomczak, Pourosvkii, Vaugier, Georges, Biermann, PNAS (2013)

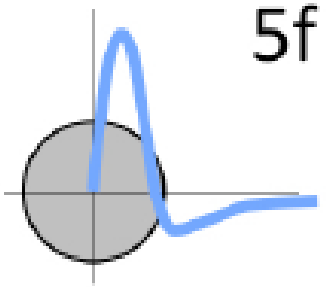
Correlated materials …

Examples: transition metal or rare-earth compounds (partially filled d- or f-shells), low-d systems …

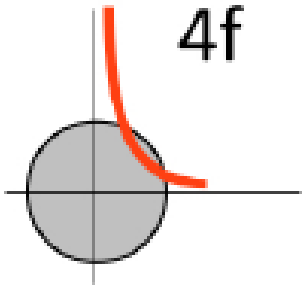




transition



actinide

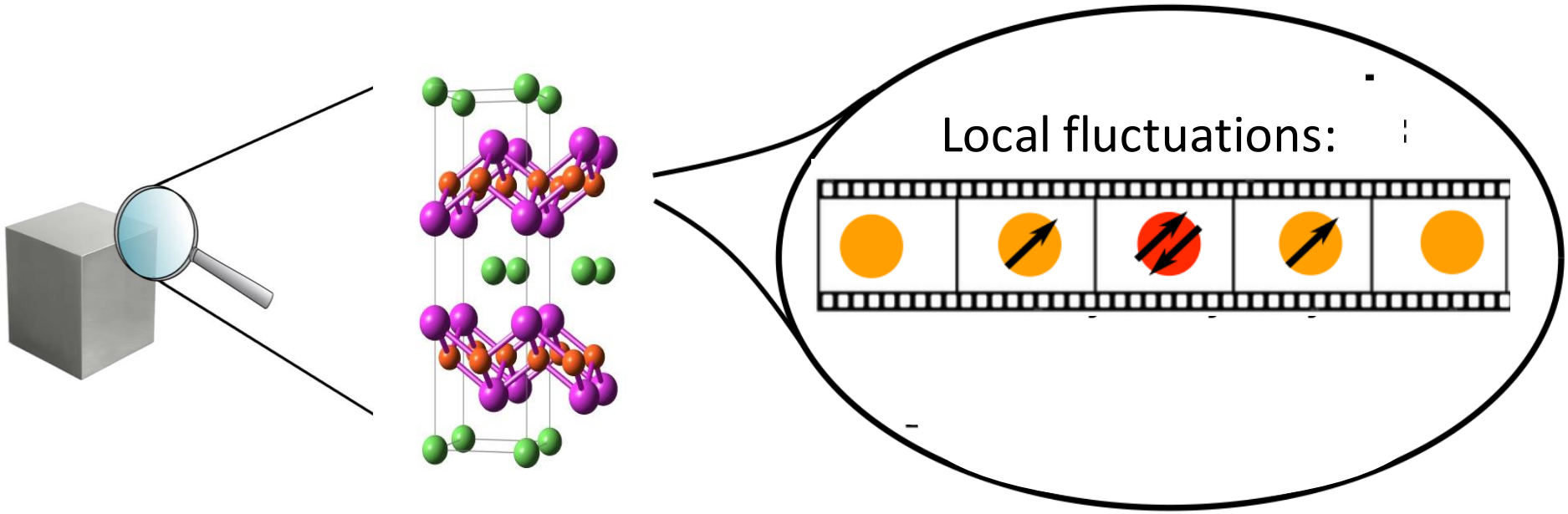


rare earth

Radial wav e function (n,l) has n-l -1 nodes

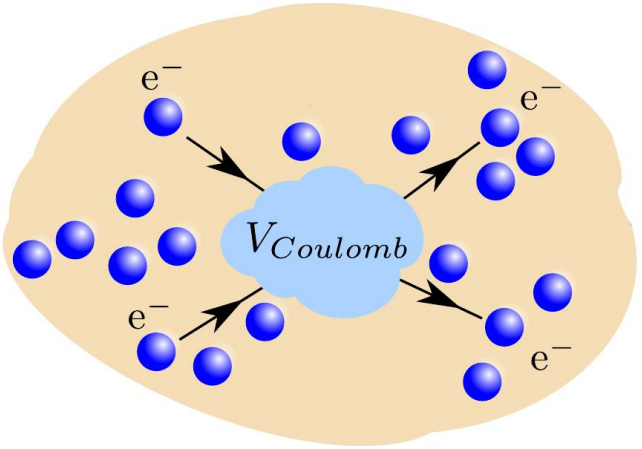
=> Spatial extension of 3d and 4f small.

Most challenging regime is in between the two (band and atomic) limits !



Correlated electron materials:

Coulomb interactions invalidate a simple single-particle picture



Correlated materials

… display collective behavior of the electrons (beyond single-particle picture), emergent properties

… are characterized by various competing energy scales, competing instabilities

… are extremely sensitive with respect to external parameters

… need theoretical treatment beyond band theory

First Principles Calculations for Correlated Materials

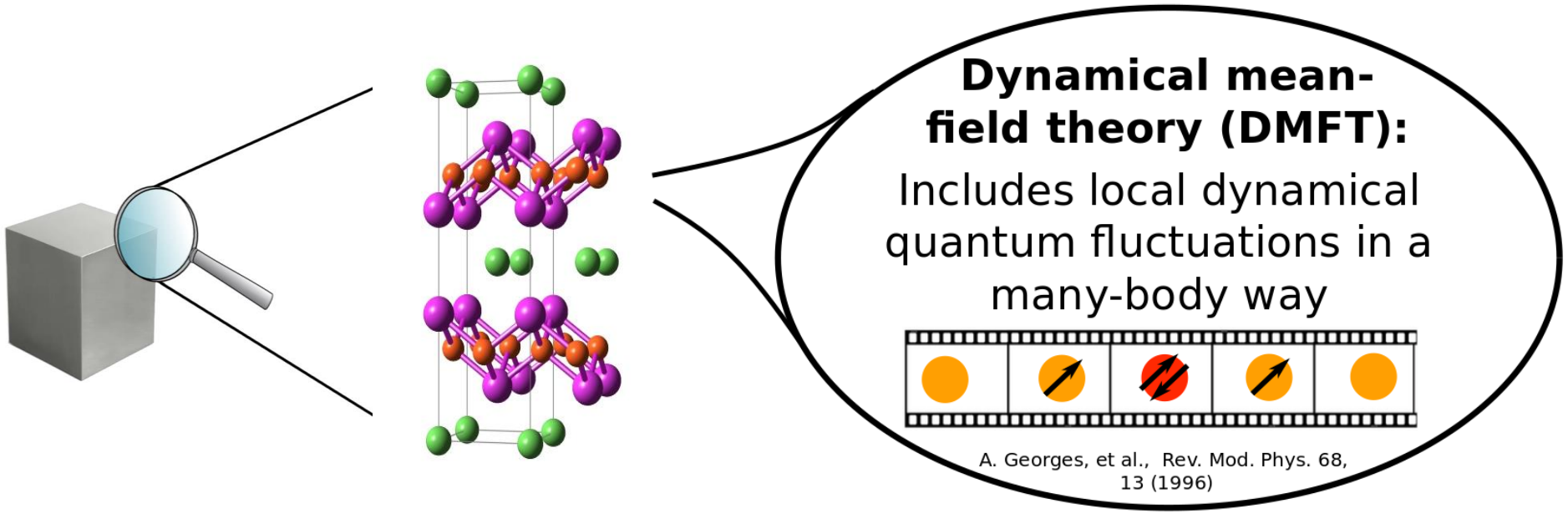
Calculate properties of **materials with strong electronic Coulomb correlations**

- ground- and excited state properties (spectra, optics, correlation functions …)
- beyond the single-particle picture ("beyond mean field")
- finite temperatures
- from *first principles*, i.e. without adjustable parameters

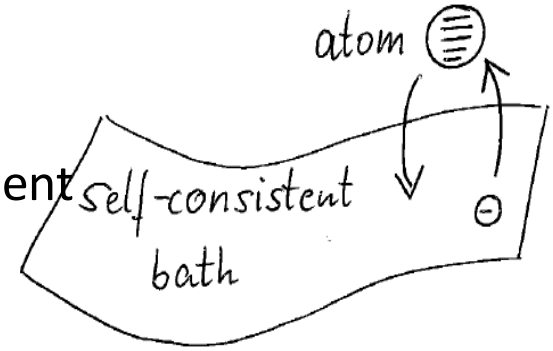
$\Rightarrow$Strategy: combine techniques from **many-body theory** & **first principles electronic structure theories**

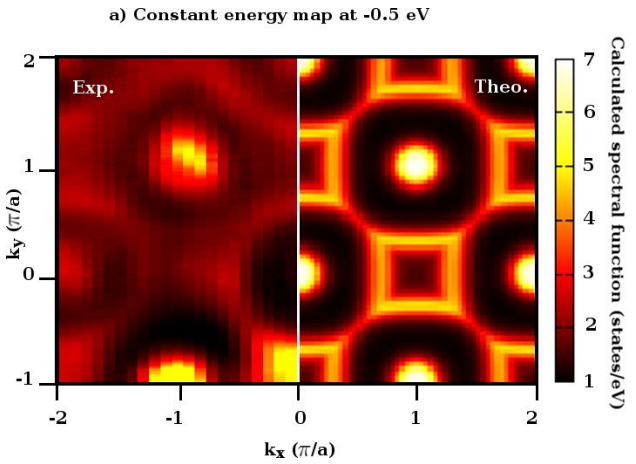
Here: **Dynamical Mean Field Theory (DMFT)**

Dynamical Mean Field Theory



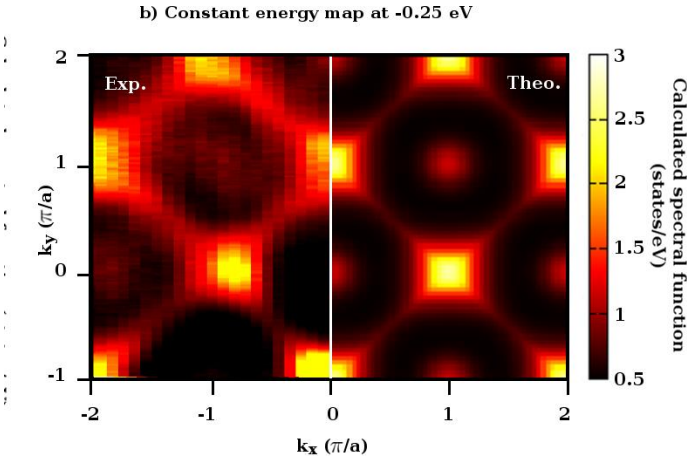
More technically speaking:
DMFT approximates dynamical orbital- and momentum-dependent many-body self-energy by a momentum-independent self-energy, calculated from a self-consistent effective local quantum impurity problem





2nd example:

Sr2IrO4



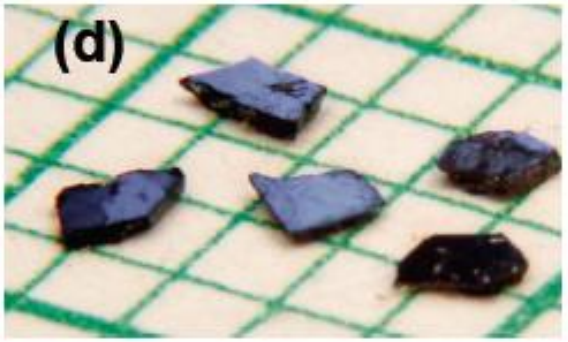
- Combined Density Functional Theory + Dynamical Mean Field Theory for Sr2IrO4 and Sr2RhO4
- Non-local correlations in Sr2IrO4
- Spectral properties of doped Sr2IrO4 – a simple picture for the pseudo-gap

Recent literature (not exhaustive) ….

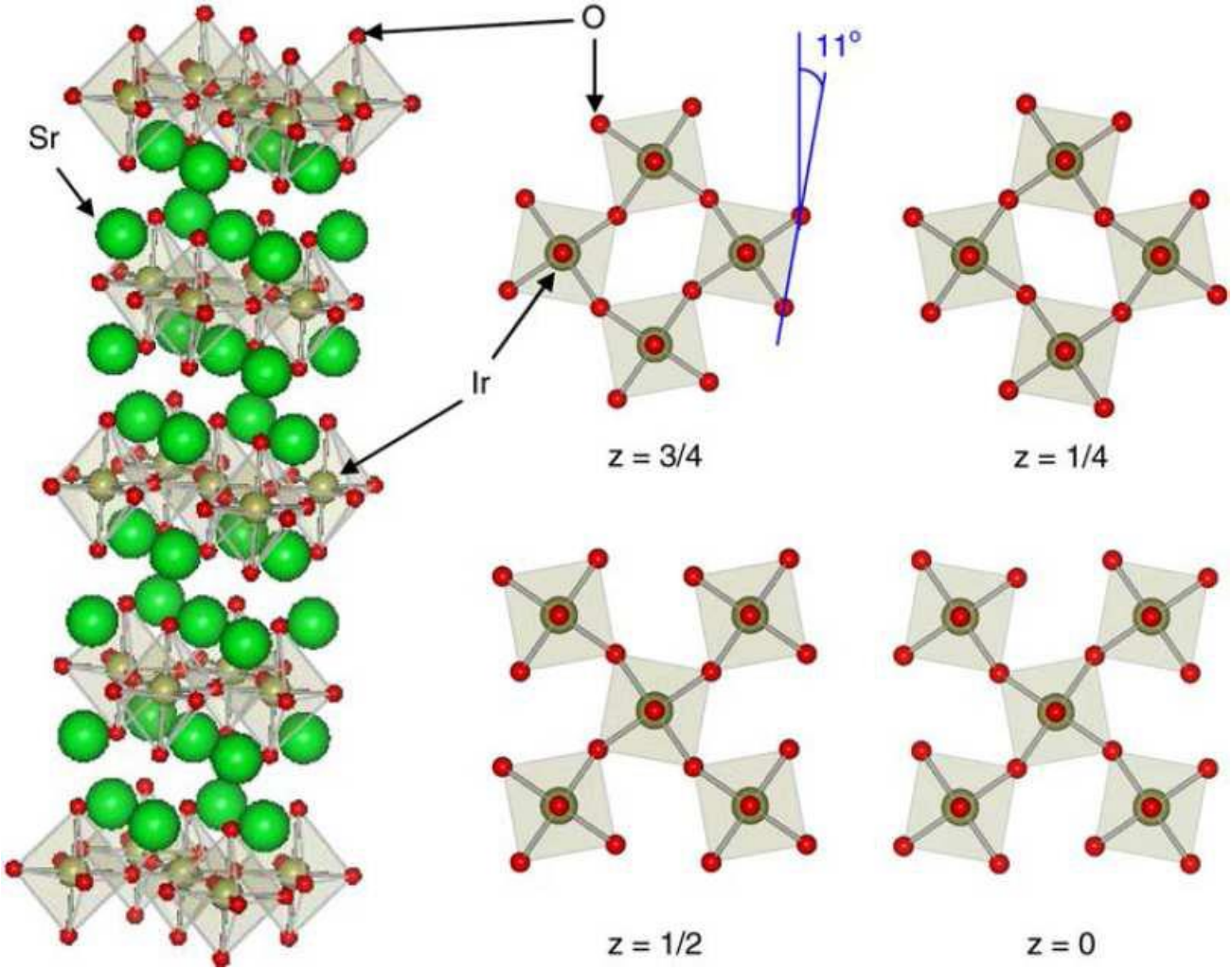
- Kim et al, Cao et al.,
- Moon et al
- Korneta et al.,
- Nichols et al.
- Li et al.,
- Wang et al,
- Lu et al.
- Moretti et al.,
- Lovesey et al.
- Yamasaki et al.
- Boseggia et al.
- Calder et al.
- Ronnow et al.
- Fujiyama et al.
- Zocco et al.
- Jackeli and Khaliullin
- Carter and Kee
- Kim, Daghofer et al.
- Paerschke et al.
- Arita et al.,
- Watanabe et al.
- Zhang et al
- Scheiderer et al.
- Hampel et al.,
- Proepper et al.
- Agrestini et al.
- Cheng et al.
- Chikara et al.
- Chatterje et al.
- Moutenet et al.
- Chen et al.
- Zhao et al.
- Cao et al.
- Lu et al.
- Wang et al.
- Guevara et al.
- ……..

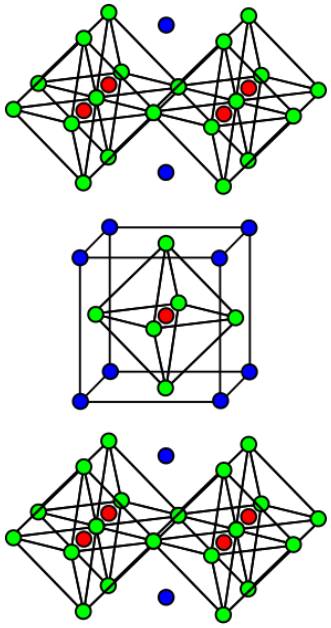
Sr2IrO4: K2NiF4-structure

(cf. La2CuO4, Sr2(Rh/Ru)O4)



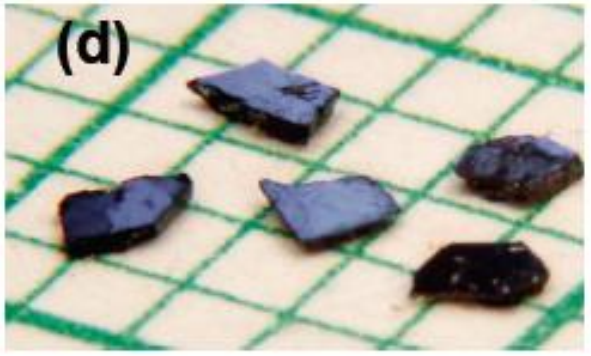
(Qi et al, PRB '12)



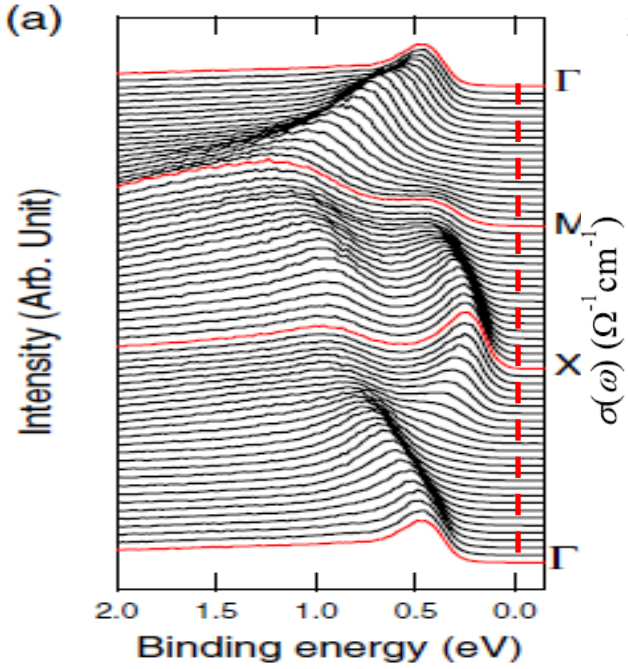


Ideal double perovskite structure

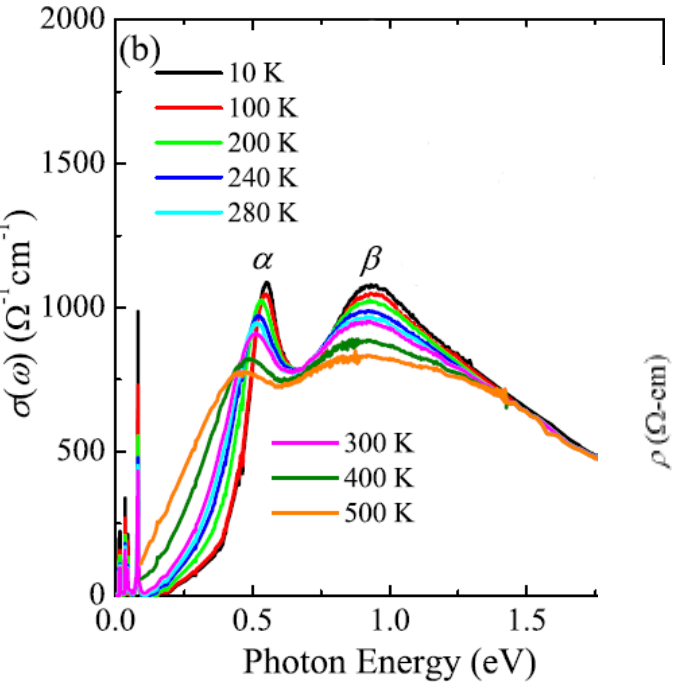
Sr2IrO4: ARPES, Optics, Transport …



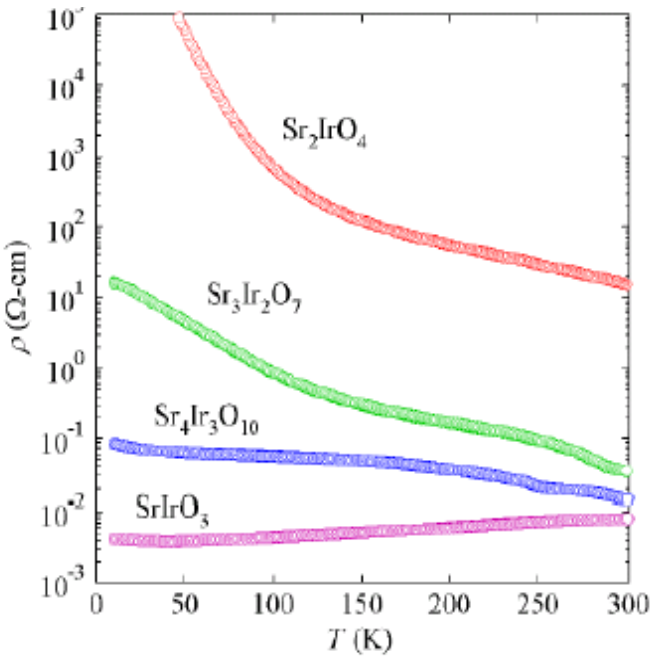
Qi et al, Phys. Rev. B 86, 125105 (2012)



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



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

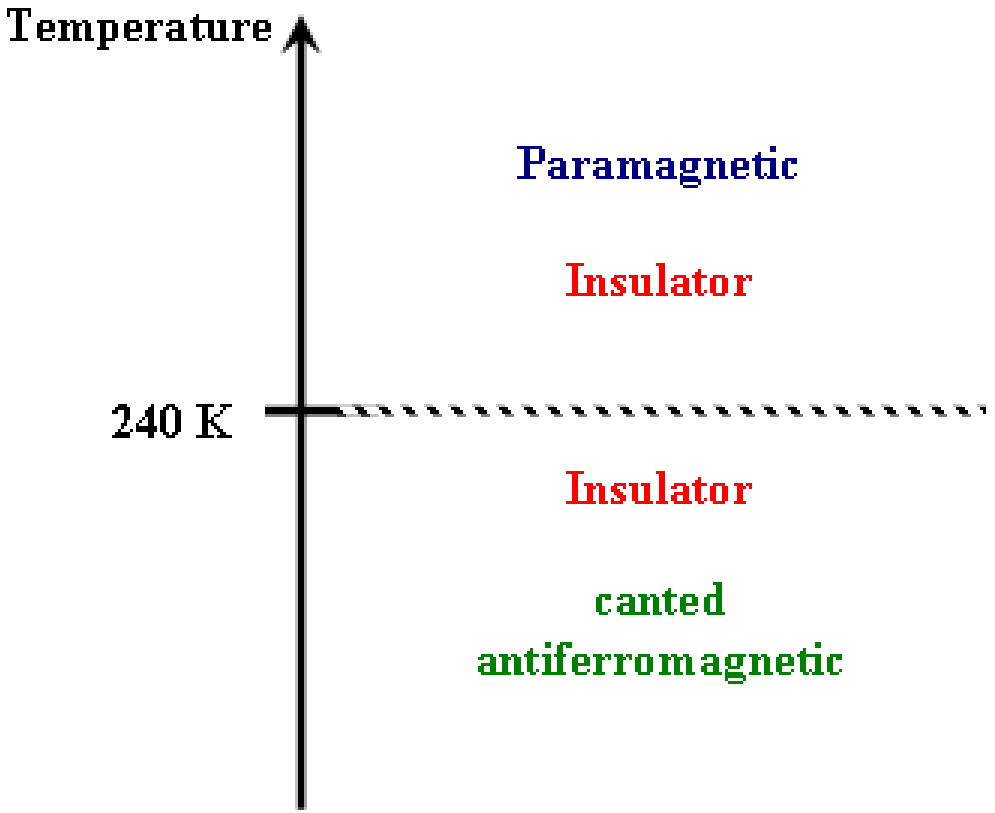


J. Matsuno H. Takagi et al, PRL (2015)

⇒ insulating behavior at all measured temperatures (up to ~ 500K). Strongly T-dependent gap.

⇒ Electronic configuration: 5d5 => not a band insulator.

Sr2IrO4: Magnetism



← **Our focus !**

Below 240K:

Canted AFM

Weak ferromagnetic moment in ab-plane

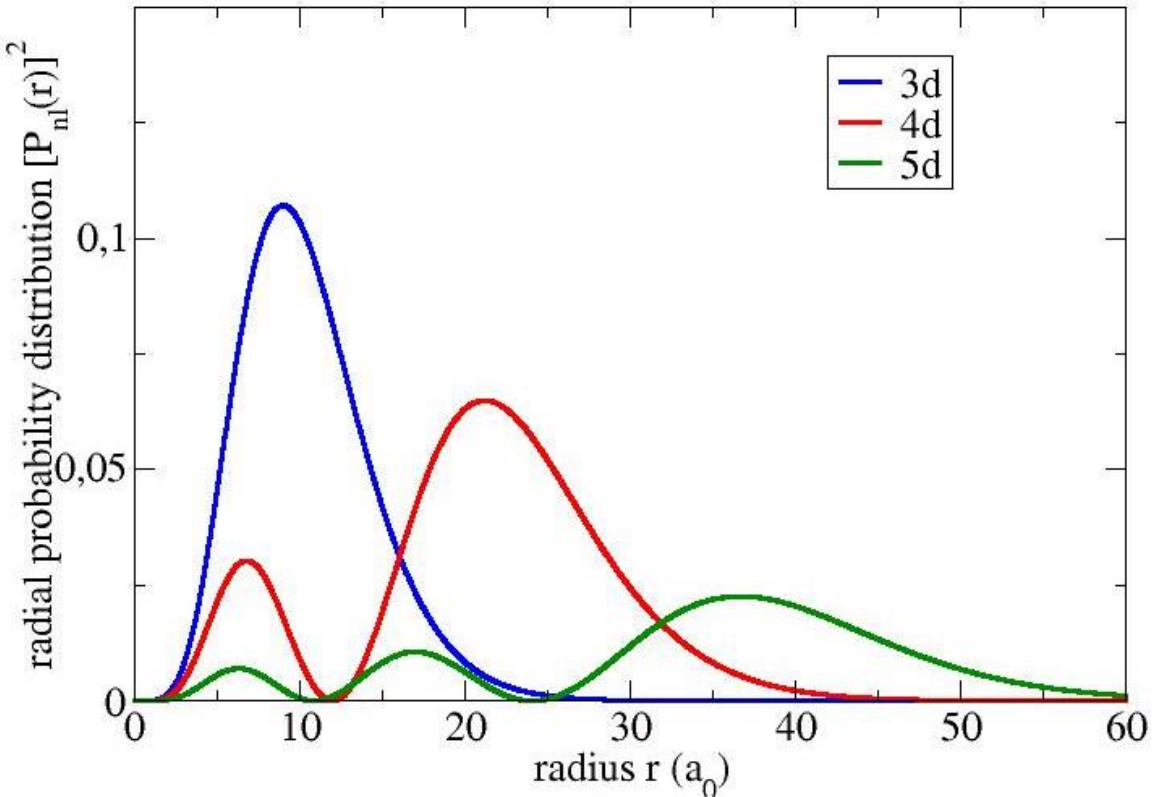
From 3d to 4d to 5d …

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

3d

4d

5d

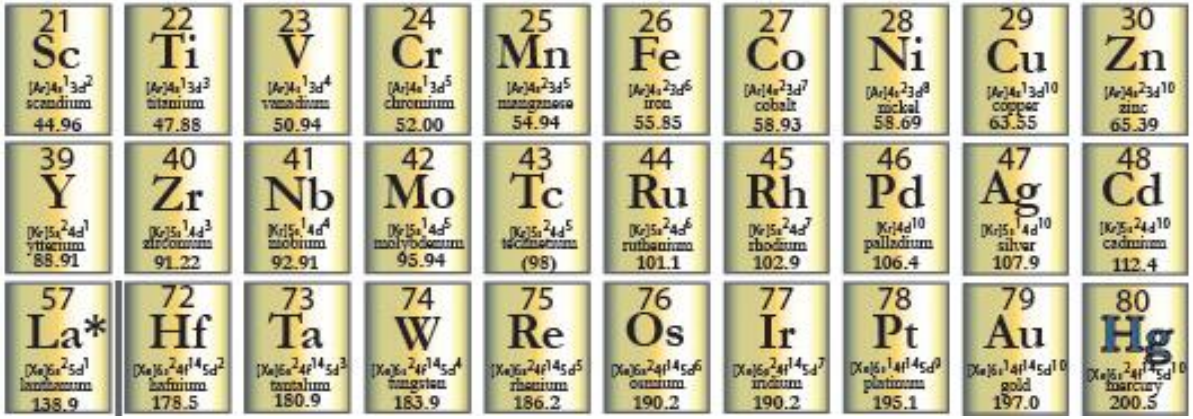


Orbital extension increases

⇒Weaker correlations

⇒Unless ……..

From 3d to 4d to 5d …



3d

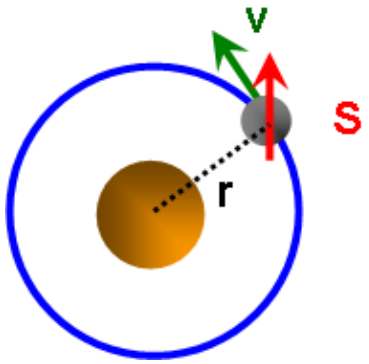
4d

5d

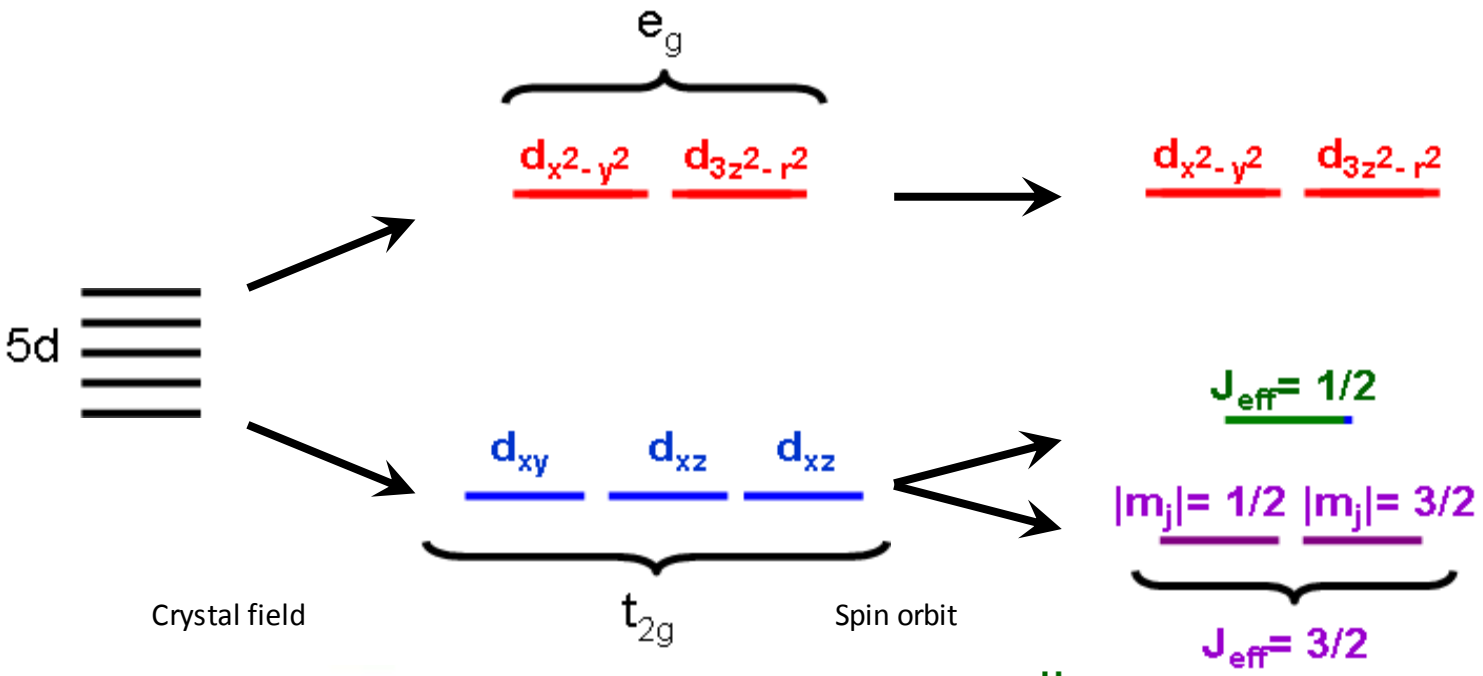
Cobalt (Z= 27)	ζ_{SO} ~ 0.06 eV
Rhodium (Z= 45)	ζ_{SO} ~ 0.19 eV
Iridium (Z= 77)	ζ_{SO} ~ 0.4 eV

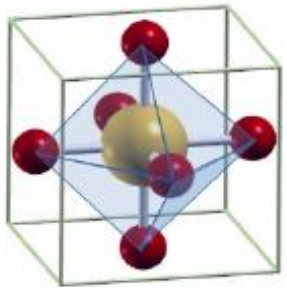
Atomic number increases

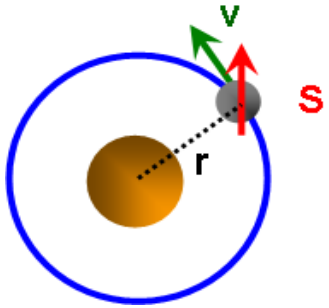
$\Rightarrow$ stronger spin-orbit coupling



Crystal field and spin-orbit: Ir in octahedral symmetry



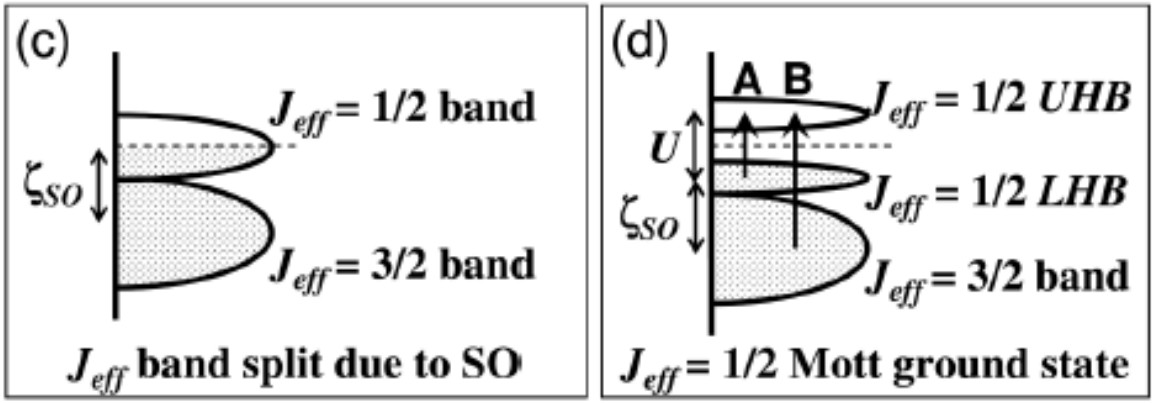




PRL 101, 076402 (2008) PHYSICAL REVIEW LETTERS week ending 15 AUGUST 2008

Novel $J_{eff} = 1/2$ Mott State Induced by Relativistic Spin-Orbit Coupling in Sr_2IrO_4

B. J. Kim,[1] Hosub Jin,[1] S. J. Moon,[2] J.-Y. Kim,[3] B.-G. Park,[4] C. S. Leem,[5] Jaejun Yu,[1] T. W. Noh,[2] C. Kim,[5] S.-J. Oh,[1] J.-H. Park,[3,4,*] V. Durairaj,[6] G. Cao,[6] and E. Rotenberg[7]



Phase-Sensitive Observation of a Spin-Orbital Mott State in Sr_2IrO_4

B. J. Kim,[1,2*] H. Ohsumi,[3] T. Komesu,[3] S. Sakai,[3,4] T. Morita,[3,5] H. Takagi,[1,2*] T. Arima[3,6]

RIXS => “Jeff=1/2” state

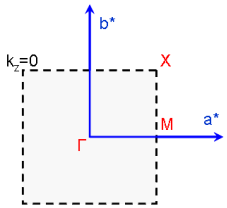
DFT-LDA

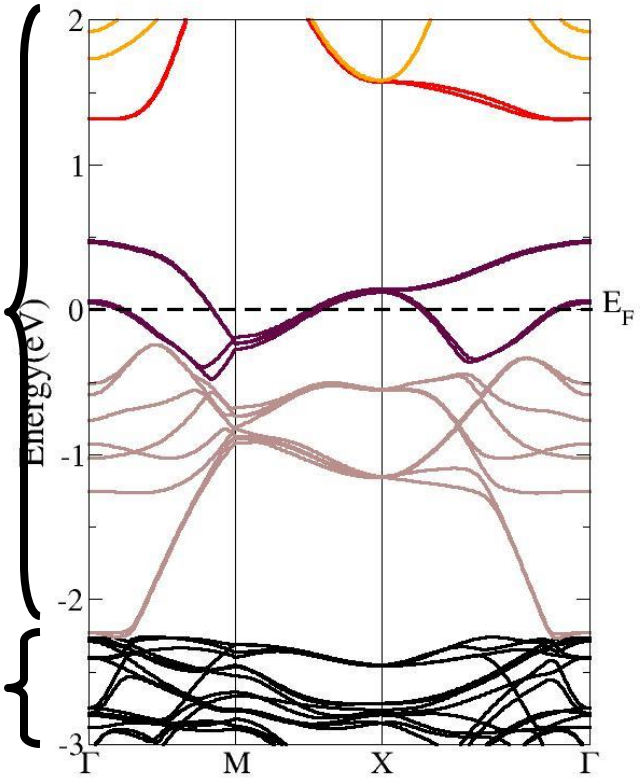
DFT part

Solving
Kohn-Sham equations

$$\left[-\frac{\hbar^2}{2m_0}\nabla^2 + V_{KS}(\mathbf{r})\right]\psi_{\mathbf{k}}(\mathbf{r}) = \varepsilon_{n\mathbf{k}}\psi_{\mathbf{k}}(\mathbf{r})$$

within the Local Density Approximation (LDA)





Ir 5d

O 2p

e_g

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

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

Used as a starting point for LDA+DMFT calculations.

Martins et al, Phys. Rev. Lett. 107, 266404 (2011)
(see also: Arita et al, Phys. Rev. Lett. 108, 086403 (2012), Zhang et al., Phys Rev Lett. 2013, Moutenet et al, Phys Rev B 2018)

Projection on the the j_{eff} = 1/2 and 3/2 orbitals

Martins et al., Phys. Rev. Lett. 107, 266404 (2011)
J. Phys. Cond.. Matt. (2017).

DFT part

Solving
Kohn-Sham equations

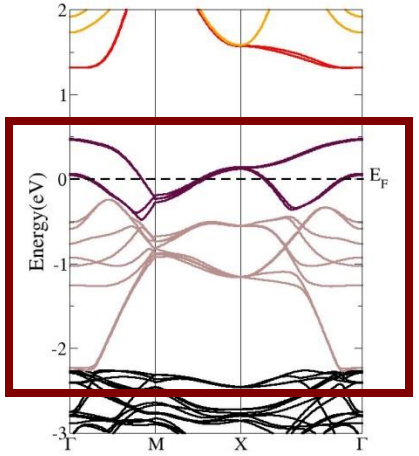
Interfacing

Constructing projection
operators on
the correlated states

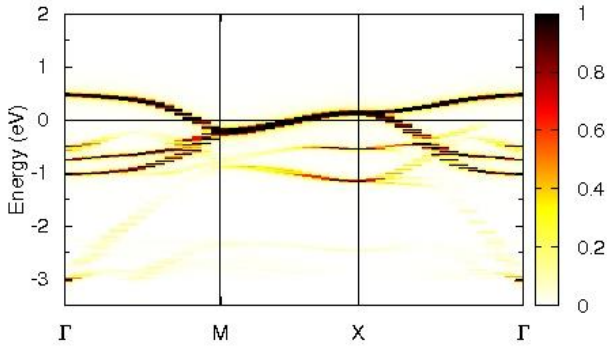
$$\hat{P}^{\alpha}(\mathbf{k}) = \sum_{m \in \mathcal{C}} |w^{\alpha}_{\mathbf{k}m}\rangle\langle w^{\alpha}_{\mathbf{k}m}|.$$

Wannier-like orbitals obtained
with a projection scheme

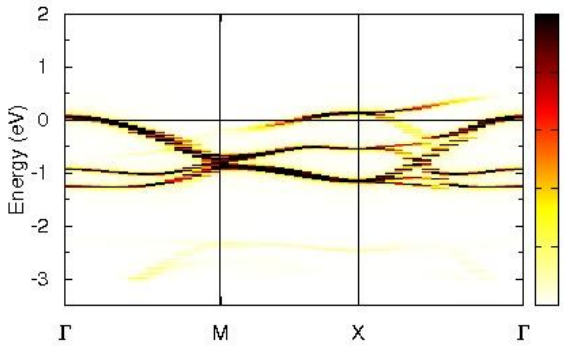
Aichhorn et al, Phys Rev B 80, 085101 (2009)



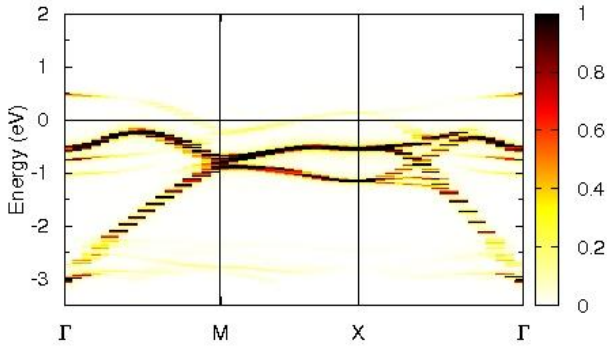
The j_{eff}=1/2 state is almost half-filled with a bandwidth of ~ 1.5 eV
the j_{eff}=3/2 states are almost completely filled.



j_{eff}=1/2



j_{eff}=3/2 $|m_j|$=3/2



j_{eff}=3/2 $|m_j|$=1/2

Reduction of Effective Degeneracy by correlations

At the level of the DFT band structure, Sr2IrO4 is not (yet) a single-orbital system:

	Sr_2IrO_4		
Wannier orbitals	$\left\|\frac{1}{2},\pm\frac{1}{2}\right\rangle$	$\left\|\frac{3}{2},\pm\frac{1}{2}\right\rangle$	$\left\|\frac{3}{2},\pm\frac{3}{2}\right\rangle$
occupation (LDA + SO)	1.16	1.98	1.84
charge (LDA + SO+DMFT)	1.02	2.00	1.98

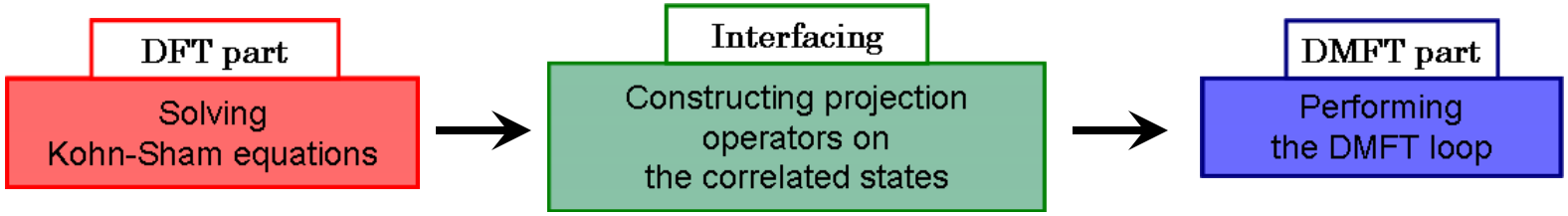
PRL 107, 266404 (2011) PHYSICAL REVIEW LETTERS week ending 23 DECEMBER 2011

Reduced Effective Spin-Orbital Degeneracy and Spin-Orbital Ordering in Paramagnetic Transition-Metal Oxides: Sr_2IrO_4 versus Sr_2RhO_4

Cyril Martins,[1,2] Markus Aichhorn,[3,1] Loïg Vaugier,[1] and Silke Biermann[1,2]

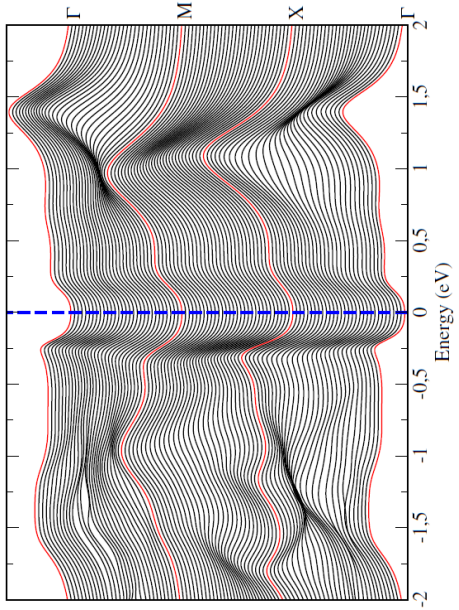
Sr_2IrO_4 a spin-orbital ordered Mott insulator

Martins et al., Phys. Rev. Lett. 107, 266404 (2011)



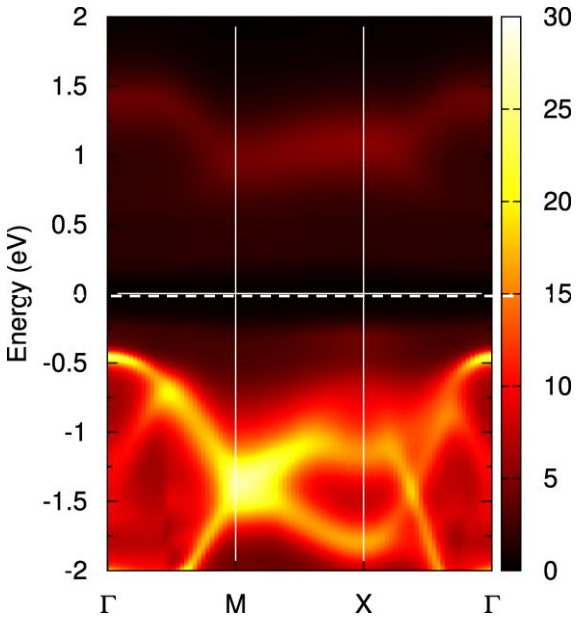
An insulating paramagnetic state with a gap of about 0.2 – 0.3 eV is obtained. (U = 2.2 eV and J = 0.3 eV)

Jeff=1/2 spectral function



Orbitally-resolved spectral density for the jeff=1/2

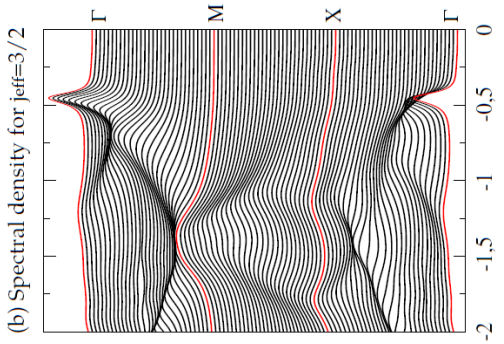
Total spectral function



Calculated spectral density

Jeff=3/2 spectral function

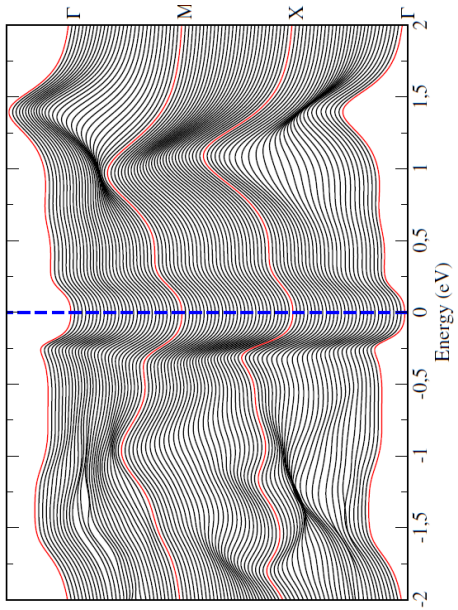
Orbitally-resolved spectral density for the jeff=3/2



Spectral function of Sr_2IrO_4 at 300 K: LDA+DMFT vs. Experiment (Perfetti et al.)

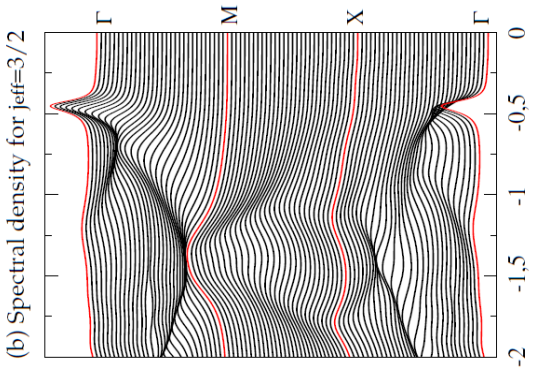
Martins et al., Phys. Rev. Lett. 107, 266404 (2011)

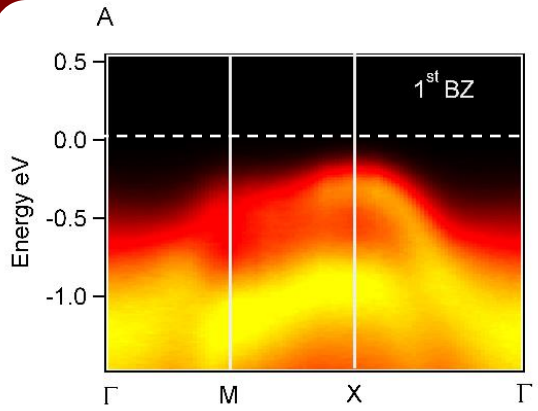
Orbitally-resolved spectral density for the jeff=1/2



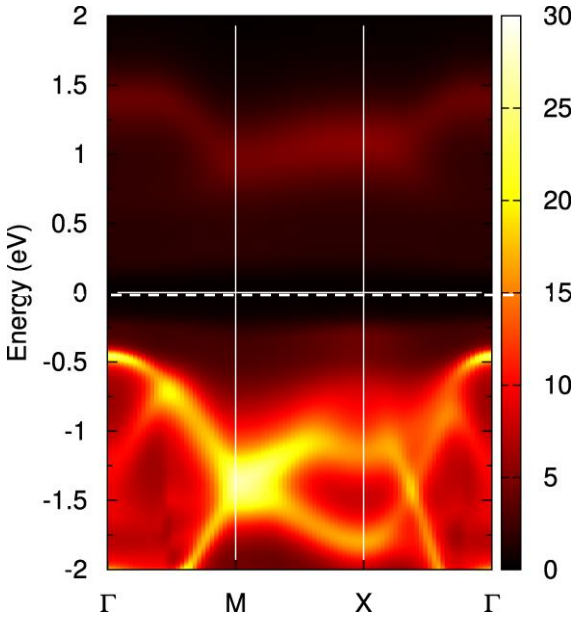
While the j_{eff}=3/2 orbitals are well described, the k-dispersion of the j_{eff}=1/2 is missing.

Orbitally-resolved spectral density for the jeff=3/2

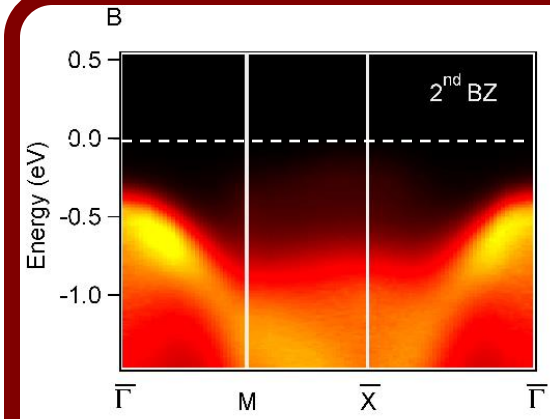




Photoemission spectrum at 300 K In the 1st Brillouin Zone



Calculated spectral density

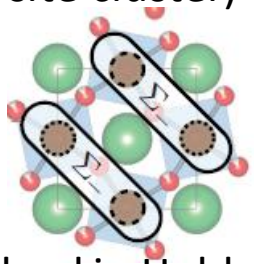


Photoemission spectrum at 300 K In the 2nd Brillouin Zone

Spectral function of Sr_2IrO_4 at 300 K: DFT + cluster-DMFT vs. Experiment

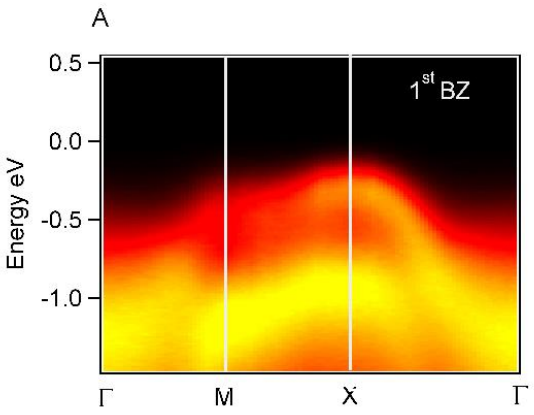
Martins et al., Phys. Rev. Lett. 107, 266404 (2011)

Cluster –DMFT
(2-site cluster)

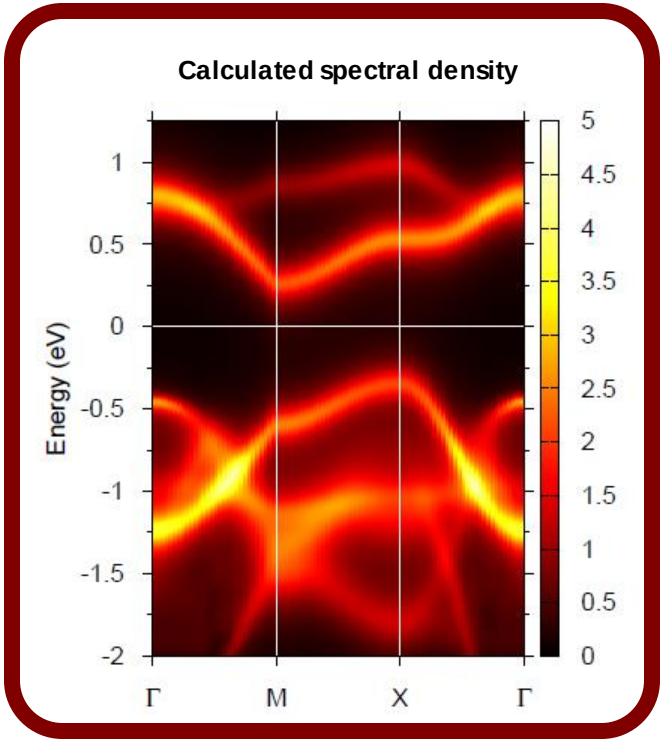


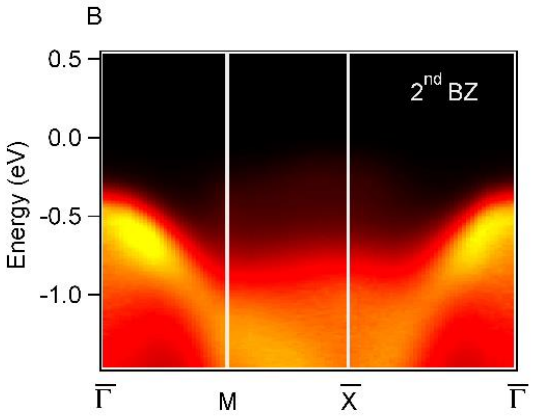
solved in Hubbard i

By taking into account inter-site (non-local) correlations, excellent agreement with experiments



Photoemission spectrum at 300 K In the 1st Brillouin Zone





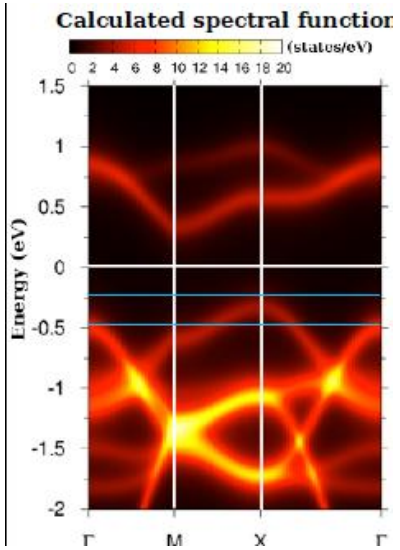
Photoemission spectrum at 300 K In the 2nd Brillouin Zone

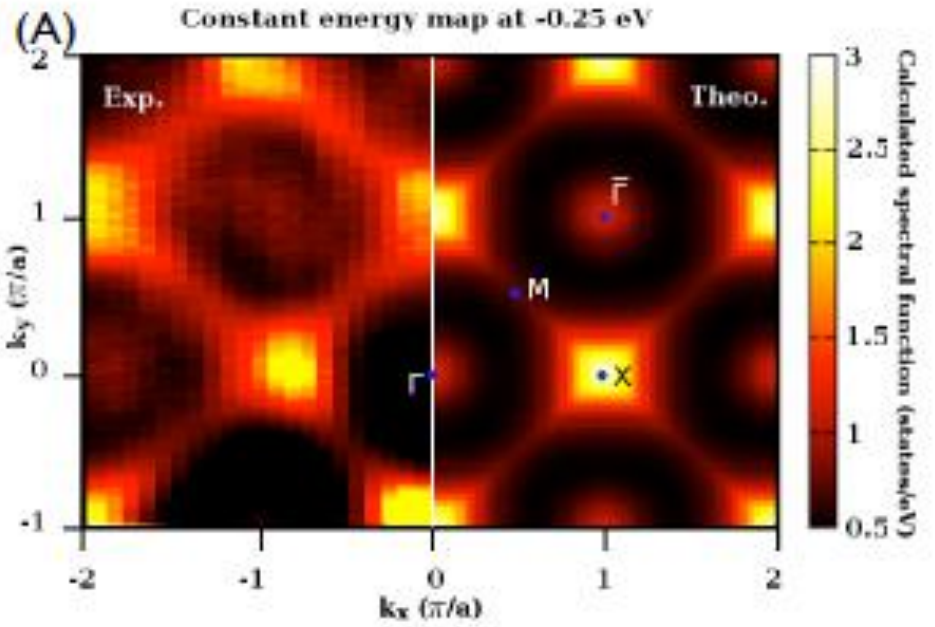
ARPES by L. Perfetti et al.

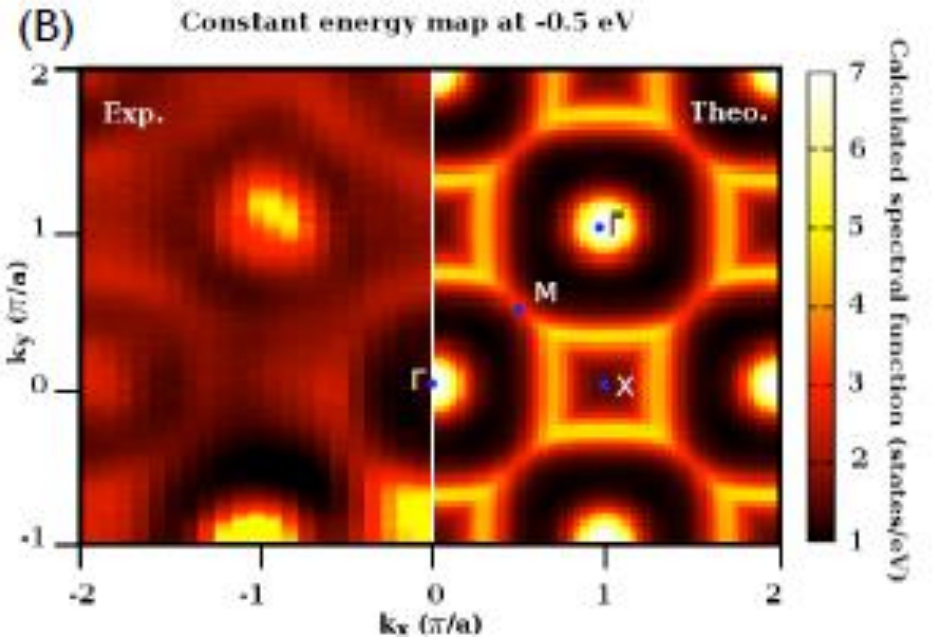
Theory: Keep 3/2 states from DFT+DMFT; Cluster-Hubbard I for ½ state

C. Martins, B. Lenz, V. Brouet, F. Bertran, L. Perfetti, S.B. PRMat. Rapid 2018

Sr2IrO4 -- Constant energy maps: experiment vs. theory







C. Martins, B. Lenz, V. Brouet, F. Bertran, L. Perfetti, S.B. PRMat. Rapid (2018)

Doped case: Sr2IrO4 + 10% electron doping

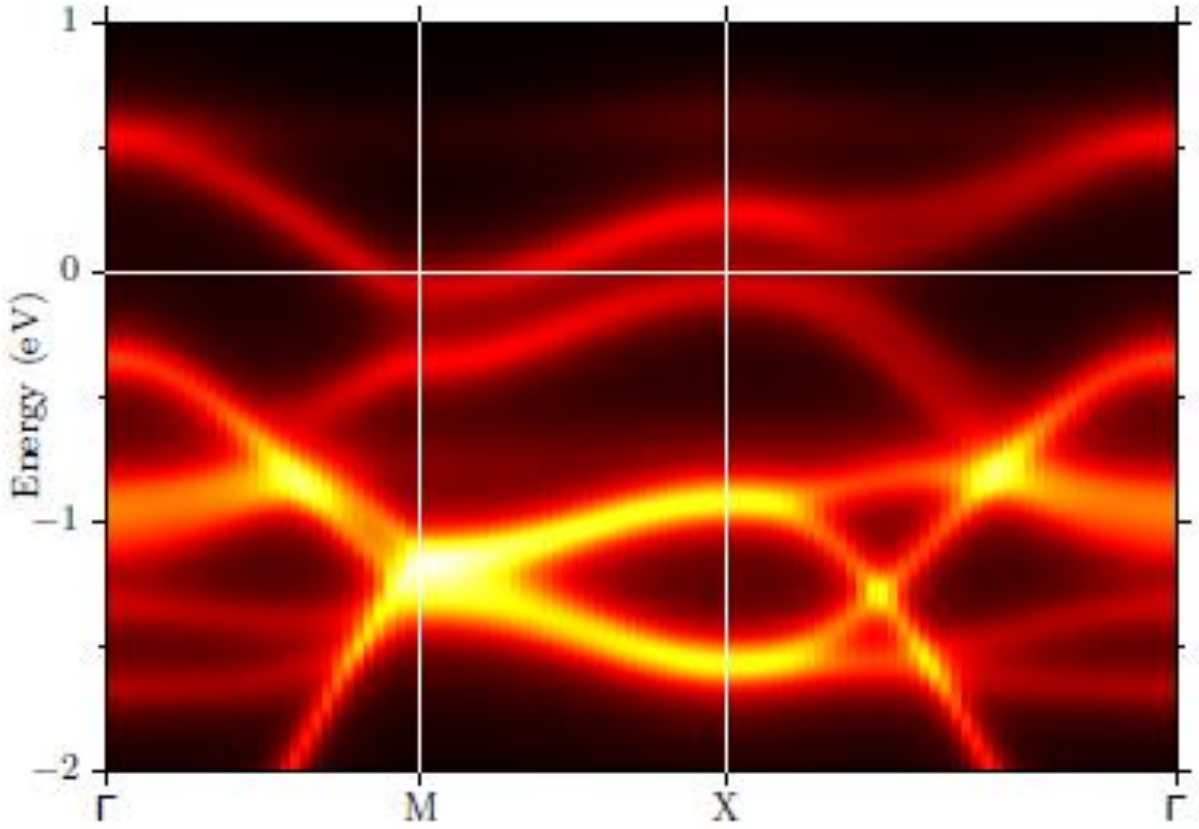


FIG. 3. Calculated momentum-resolved spectral function $A(\mathbf{k}, \omega + i\eta)$ of 10% electron-doped Sr_2IrO_4 along the $\Gamma - M - X - \Gamma$ path.

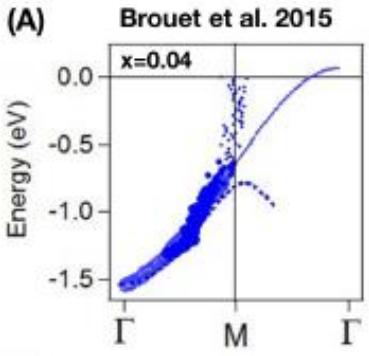
However: U is reduced by metallic screening!

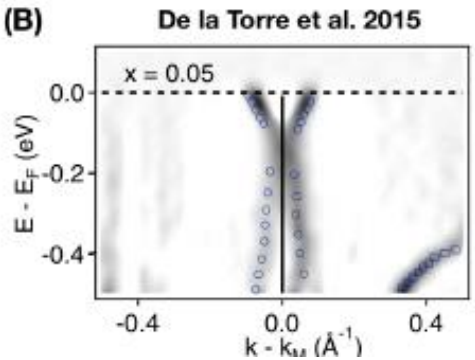
In practice:

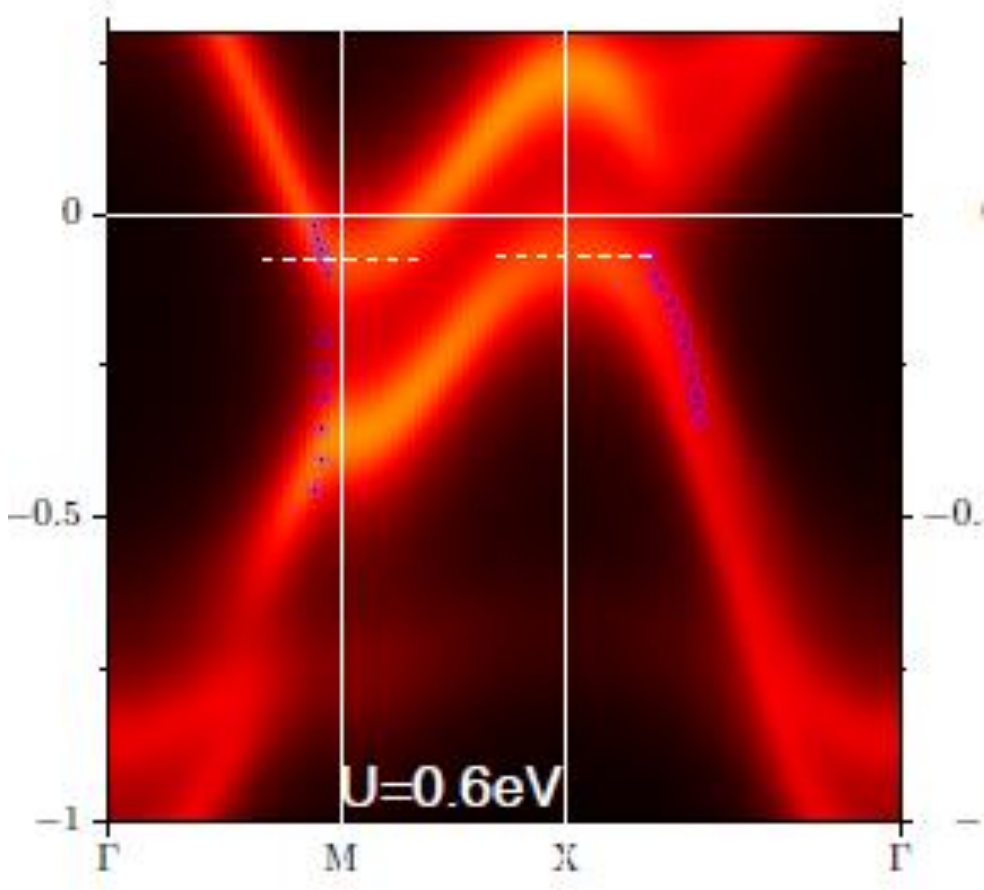
"VCA"-like solver (no bath sites)

C. Martins, B. Lenz, V. Brouet, F. Bertran, L. Perfetti, S.B. PRMat. Rapid 2018

Doped case: Comparison to ARPES

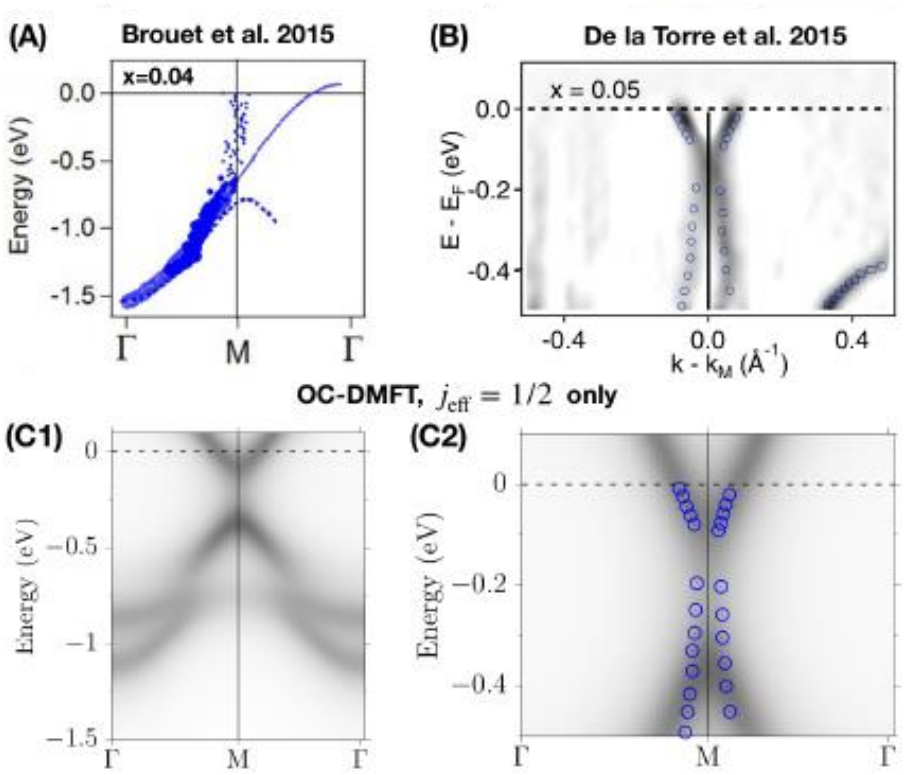


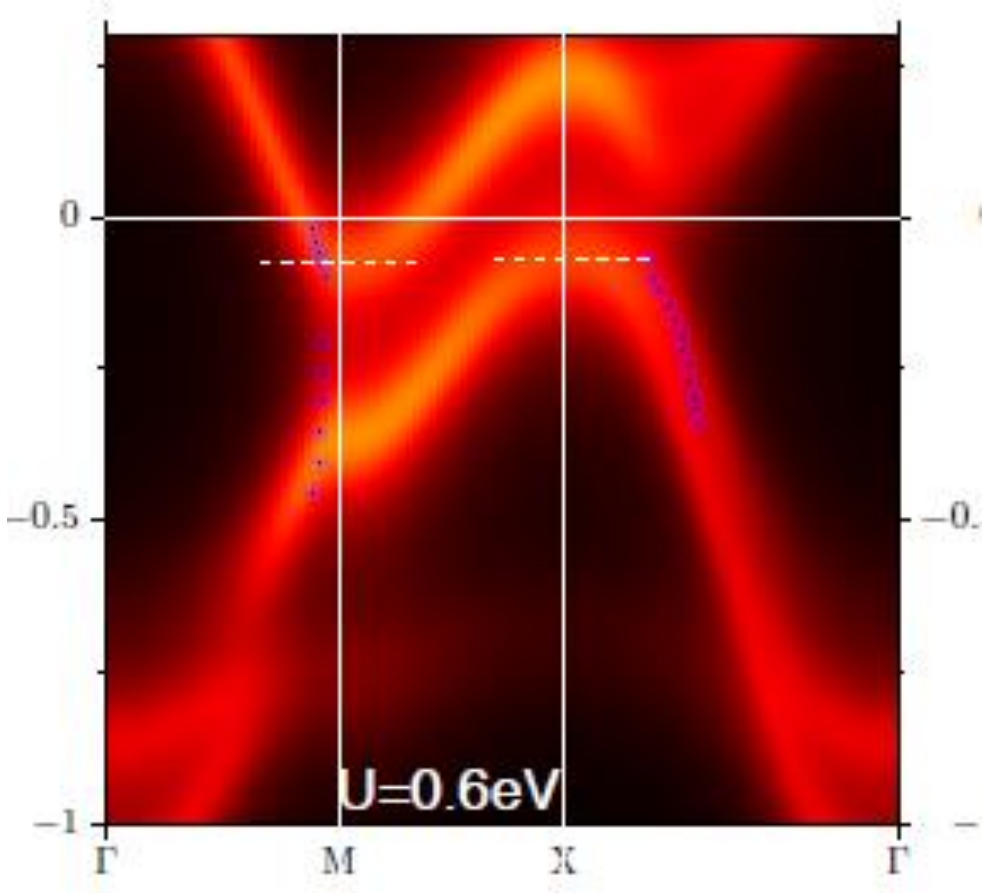




Blue-dashed: ARPES from De la Torre et al. 2015

Doped case: Comparison to ARPES





Blue-dashed: ARPES from De la Torre et al. 2015

Doped case:
Sr2IrO4 + 10% electron doping

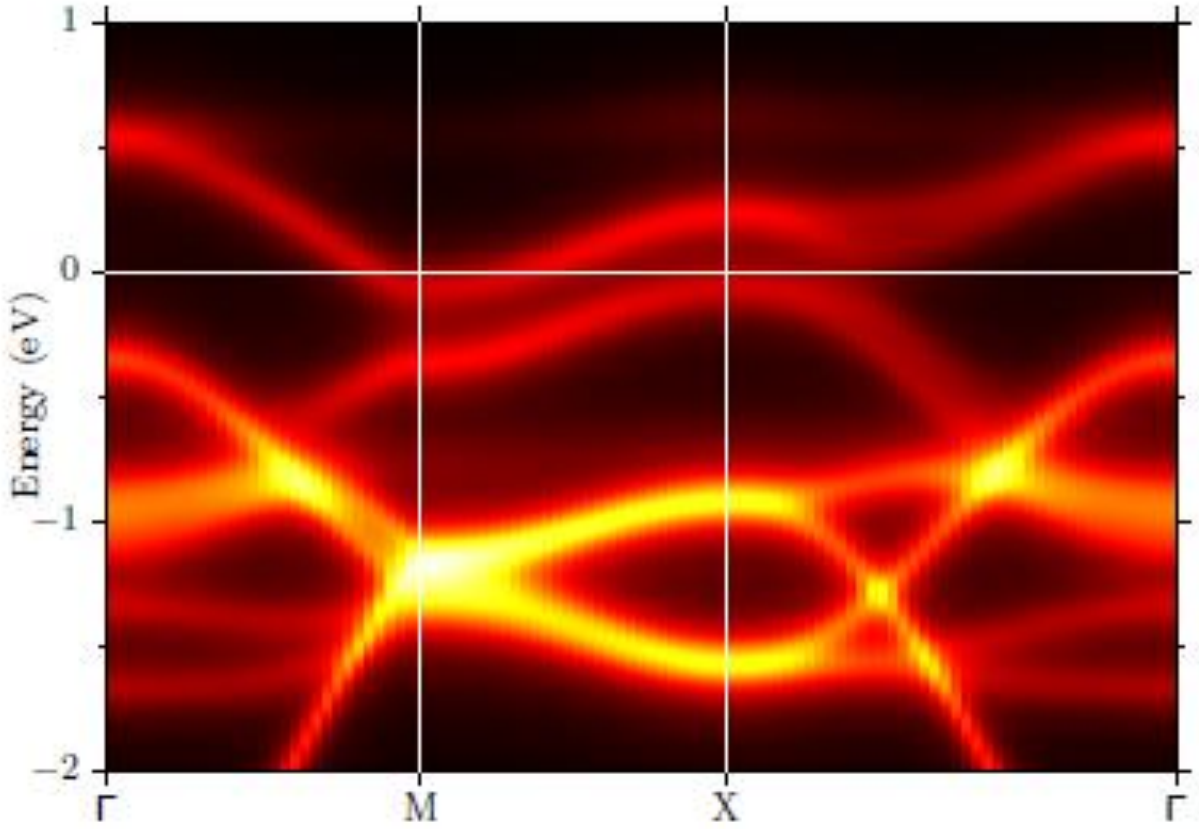
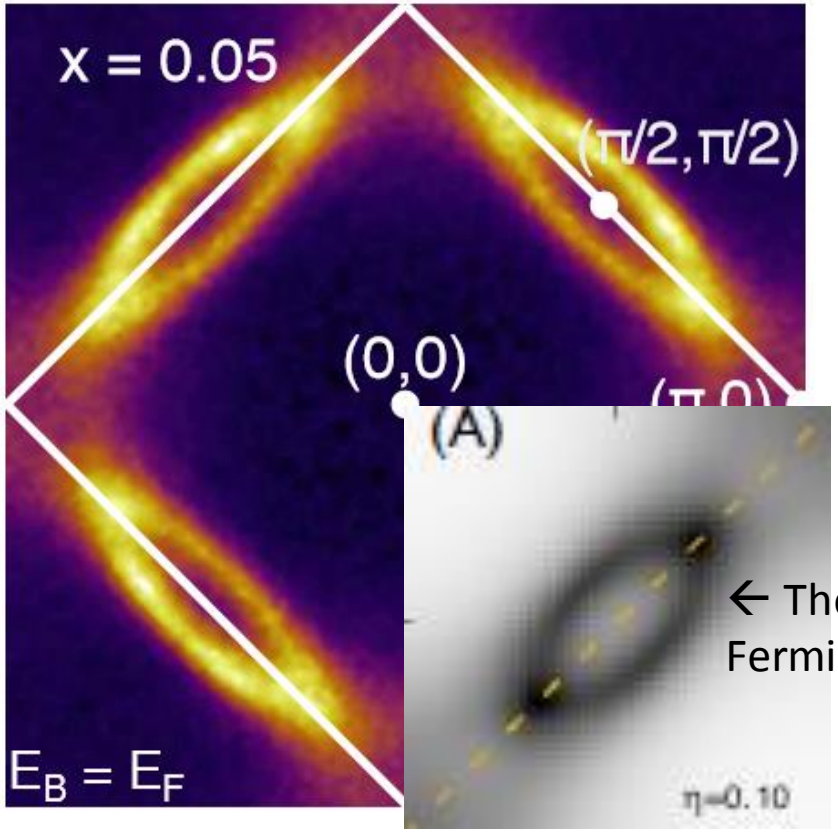


FIG. 3. Calculated momentum-resolved spectral function $A(\mathbf{k}, \omega + i\eta)$ of 10% electron-doped Sr_2IrO_4 along the $\Gamma - M - X - \Gamma$ path.

C. Martins, B. Lenz, V. Brouet, F. Bertran, L. Perfetti, S.B. PRMat. Rapid 2018

Fermi surface of (Sr,La)2IrO4:



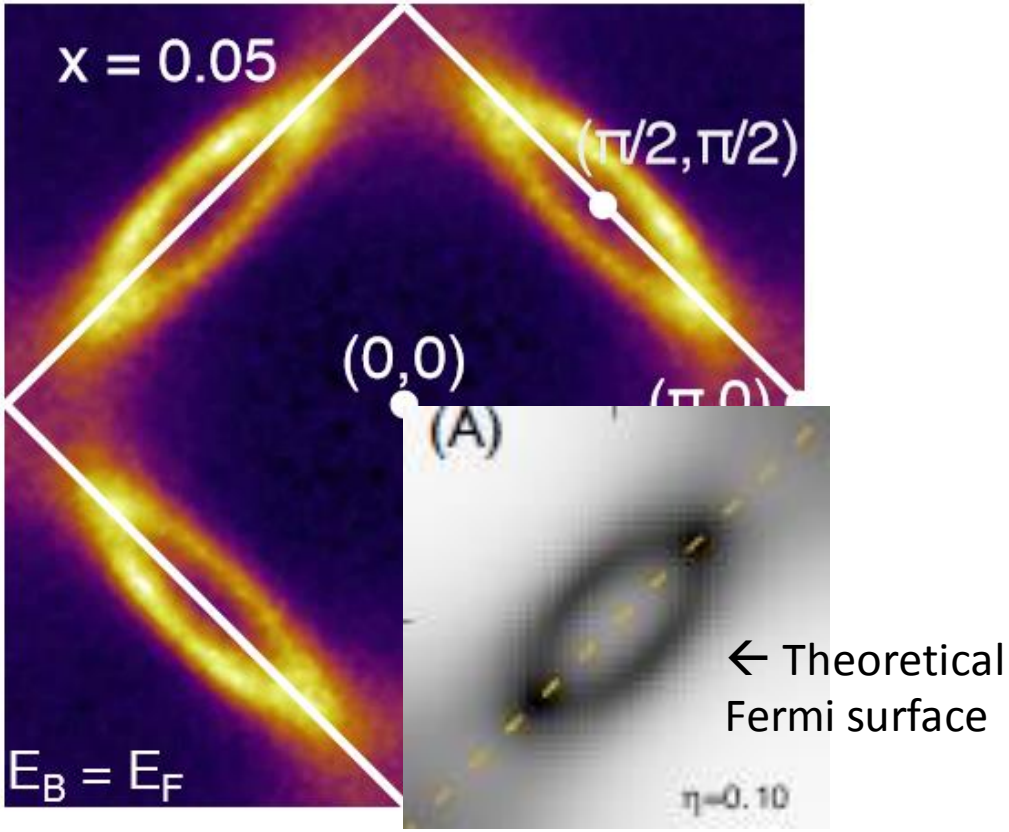
← Theoretical Fermi surface

(Martins, Lenz, Perfetti, Brouet, Biermann, Phys. Rev. Mat. Rapid (2018))

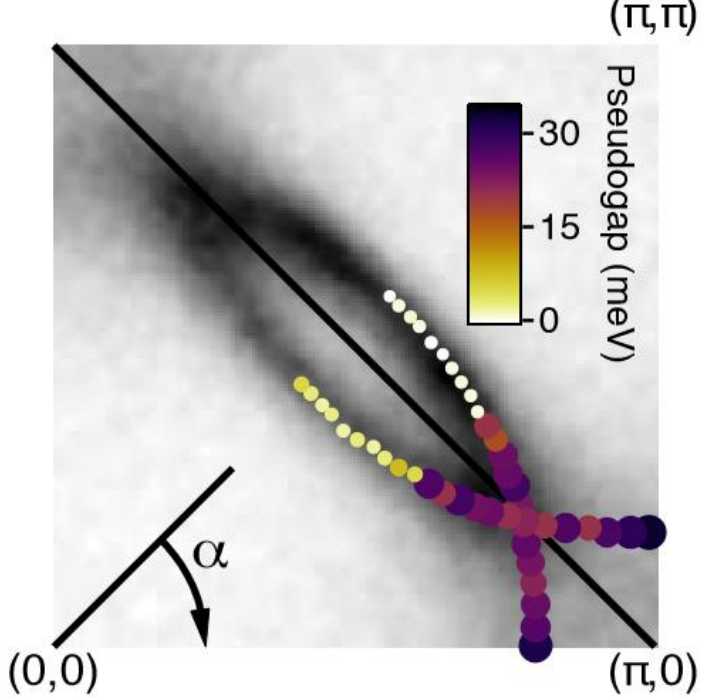
De la Torre *et al*. PRL 2015

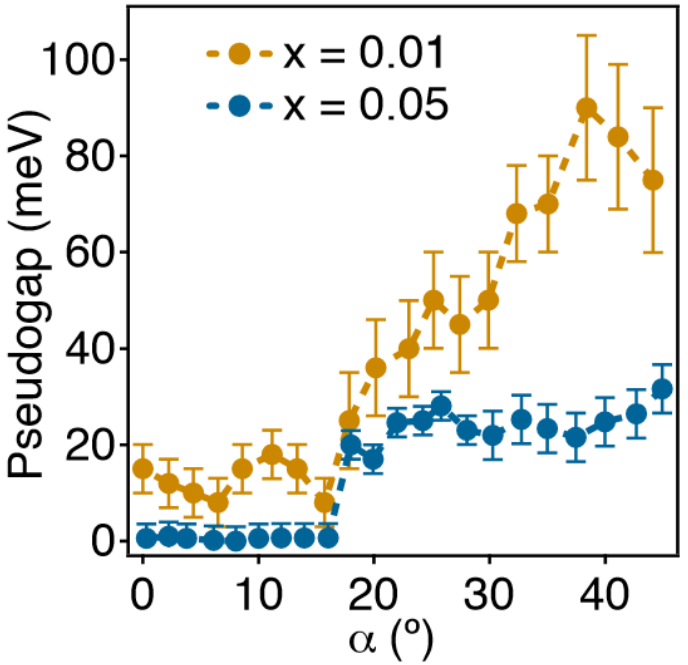
See also: Moutenet et al for a QMC study of the FS within a model for Sr2IrO4

Fermi surface of (Sr,La)2IrO4:



← Theoretical Fermi surface



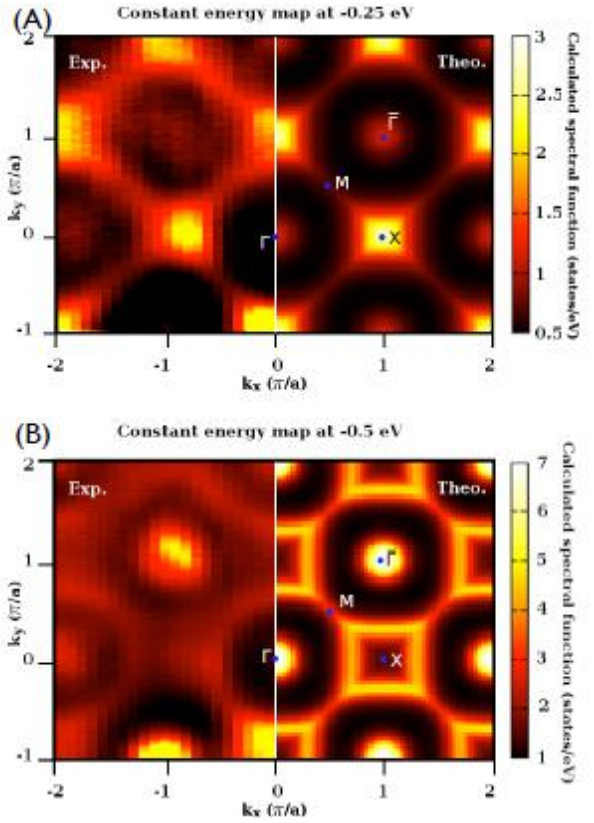


De la Torre *et al*. PRL 2015

Summary

”First principles” calculations of spectral properties of iridates: Sr2IrO4

- Reduced effective degeneracy
- Role of non-local correlations: antiferromagnetic fluctuations crucial (note: not a statement on range!)
- Doped case: good agreement with ARPES!
- Pseudogap-like features a result of several effects: antiferro fluctuations, filled band coming close …
- “OC-DMFT” a promising cluster scheme



Martins et al., PRL 2011, JPCM 2016, PRM (R) 2018

Collaborators and Funding

Sr2IrO4/Sr2RhO4 (PRL 2011 and JPCM 2017)	**C. Martins,** M. Aichhorn, L. Vaugier, S. Biermann
Sr2IrO4 vs. Ba2IrO4	**C. Martins,** B. Lenz, L. Perfetti, S. Moser
Non-local effects in Sr2IrO4 (Phys. Rev. Mat. Rapid 2018)	**B. Lenz, C. Martins,** L. Perfetti, V. Brouet

Many useful discussions: Y. Sidis, P. Bourges, J.Jeong

Thank you for your attention !