Electronic Structure of Iron Pnictides from First Principles
–
a Dynamical Mean Field Perspective

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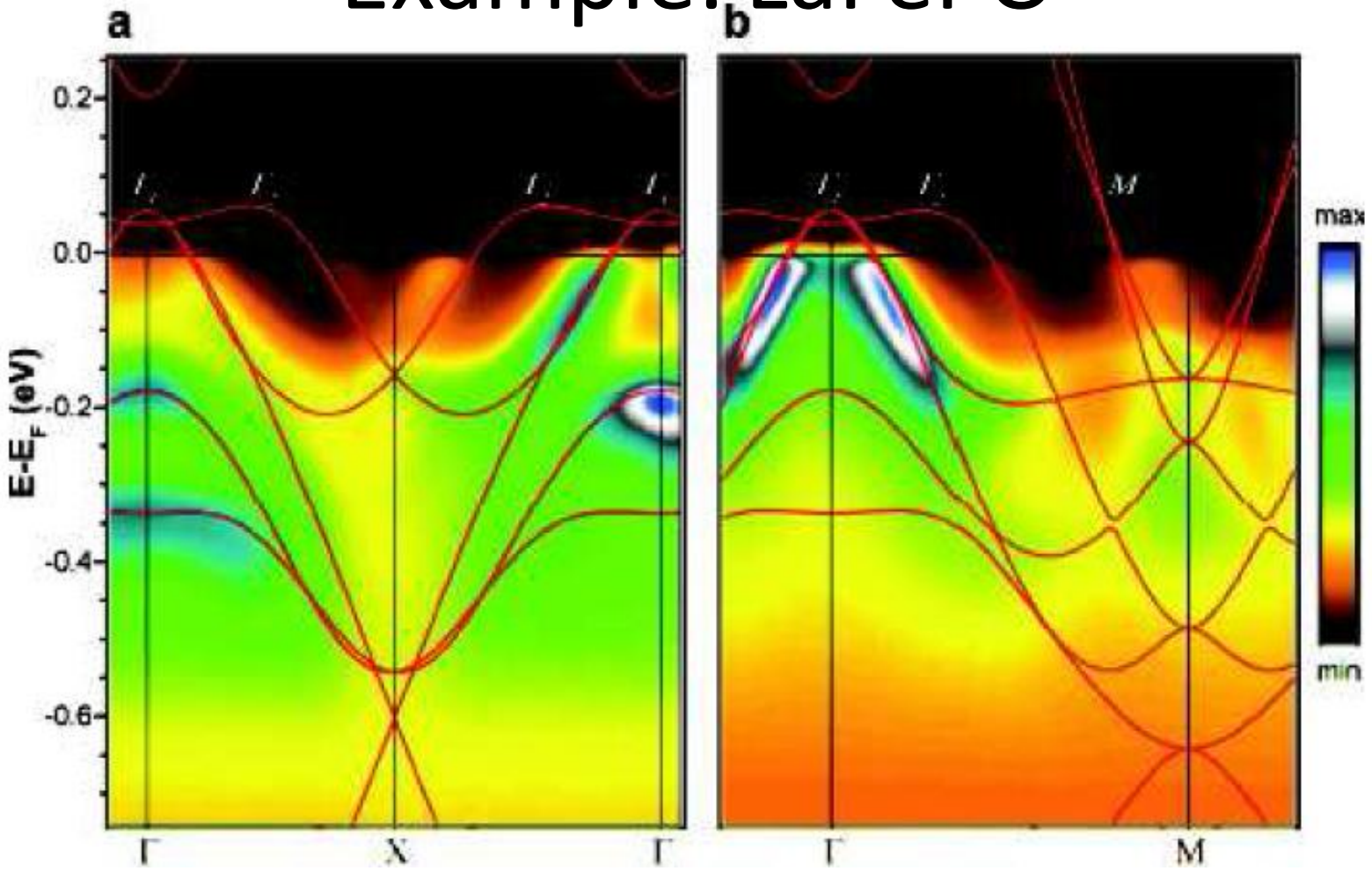
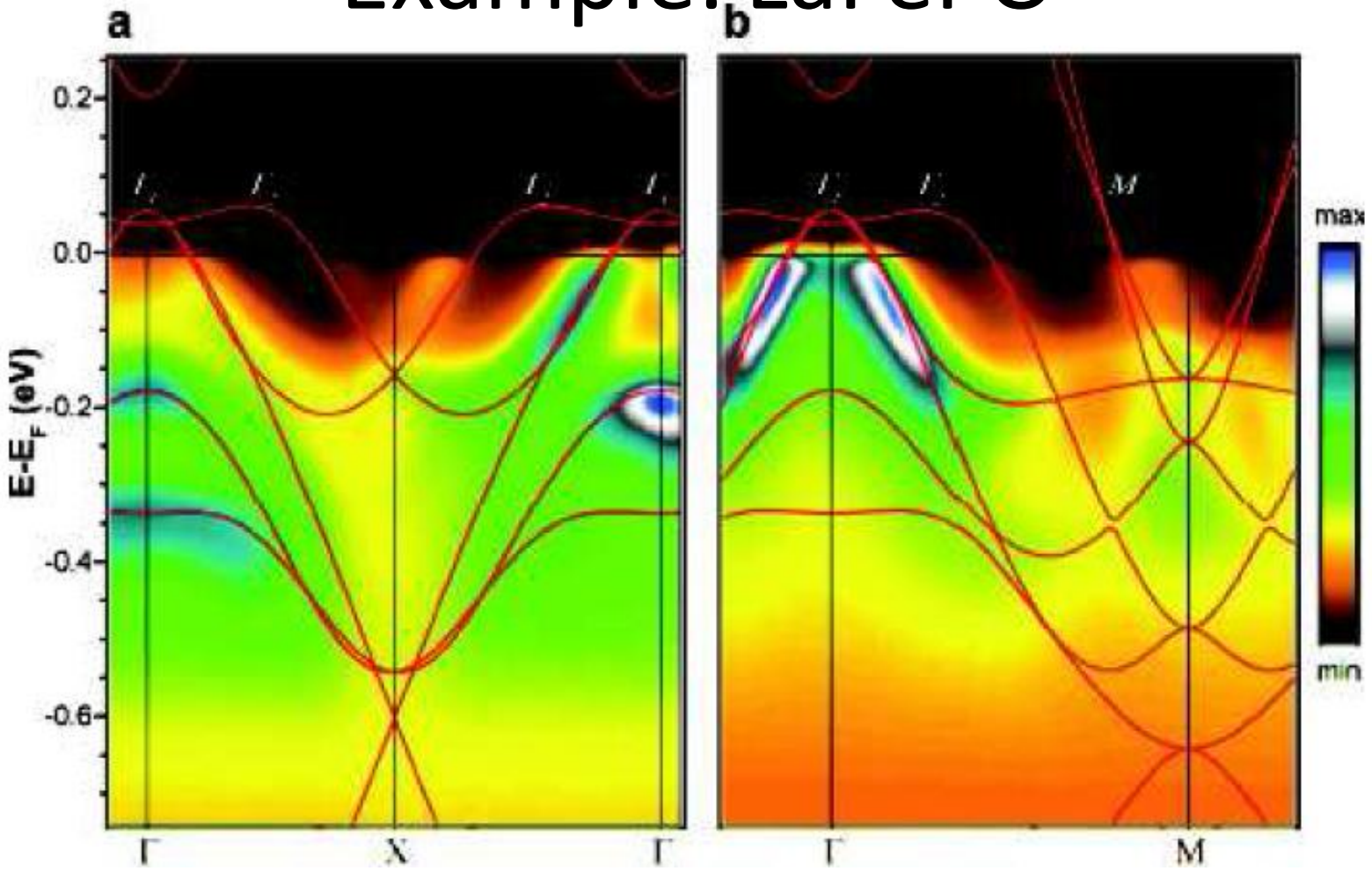
Ecole Polytechnique, Palaiseau, France

Example: LaFePO



Fe-d states: photoemission versus band structure
(Lu et al., 2008)

Example: LaFePO



"…after shifting the calculated bands up by 0.11 eV and then renormalizing by a factor 2.2 …."

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**

Outline

- Combined density functional theory and dynamical mean field theory ("DFT+DMFT")
- Quick Example: ceria
- Iron pnictides within DFT+DMFT: LaFeAsO, FeSe, BaFe2As2, BaCo2As2
- Doping-dependence of coherence in Ba122
- What about (Hubbard) U?
- Beyond "DFT+DMFT": Screened Exchange+DMFT
- Conclusions

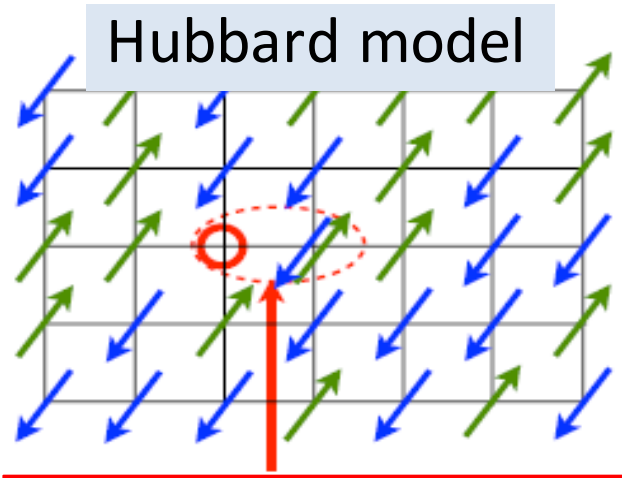
Disclaimer: focus on work in Paris. Other DFT+DMFT calculations on pnictides: groups of Held, Kotliar, Sangiovanni, Valenti, …

“Ab initio” modeling of materials …

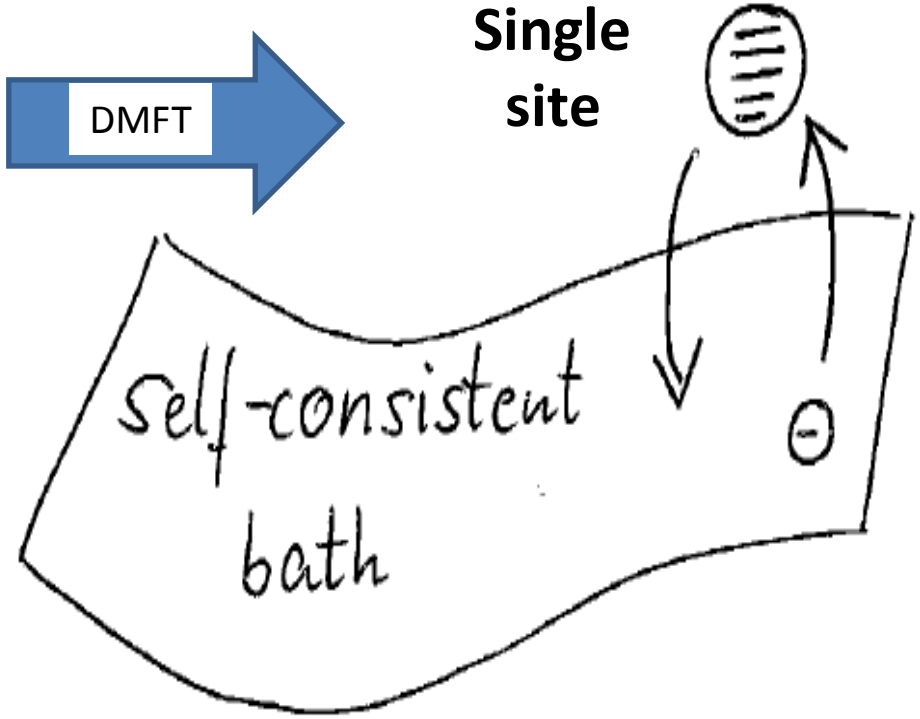
… beyond the band picture ?

Combine
ab initio techniques
with
many-body theory

Dynamical Mean Field Theory for Lattice Models



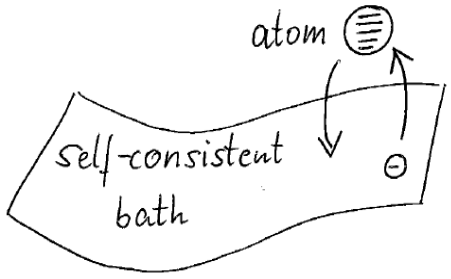
Local Coulomb interaction
= "Hubbard U"



Georges, Kotliar, Krauth, Rozenberg, Rev. Mod. Phys. 1996

DMFT loop

Anderson impurity model solver



$$\hat{\mathcal{G}}_0^{-1}$$

$$\hat{\mathcal{G}}_0 = \left(\hat{\Sigma} + \hat{G}^{-1}\right)^{-1}$$

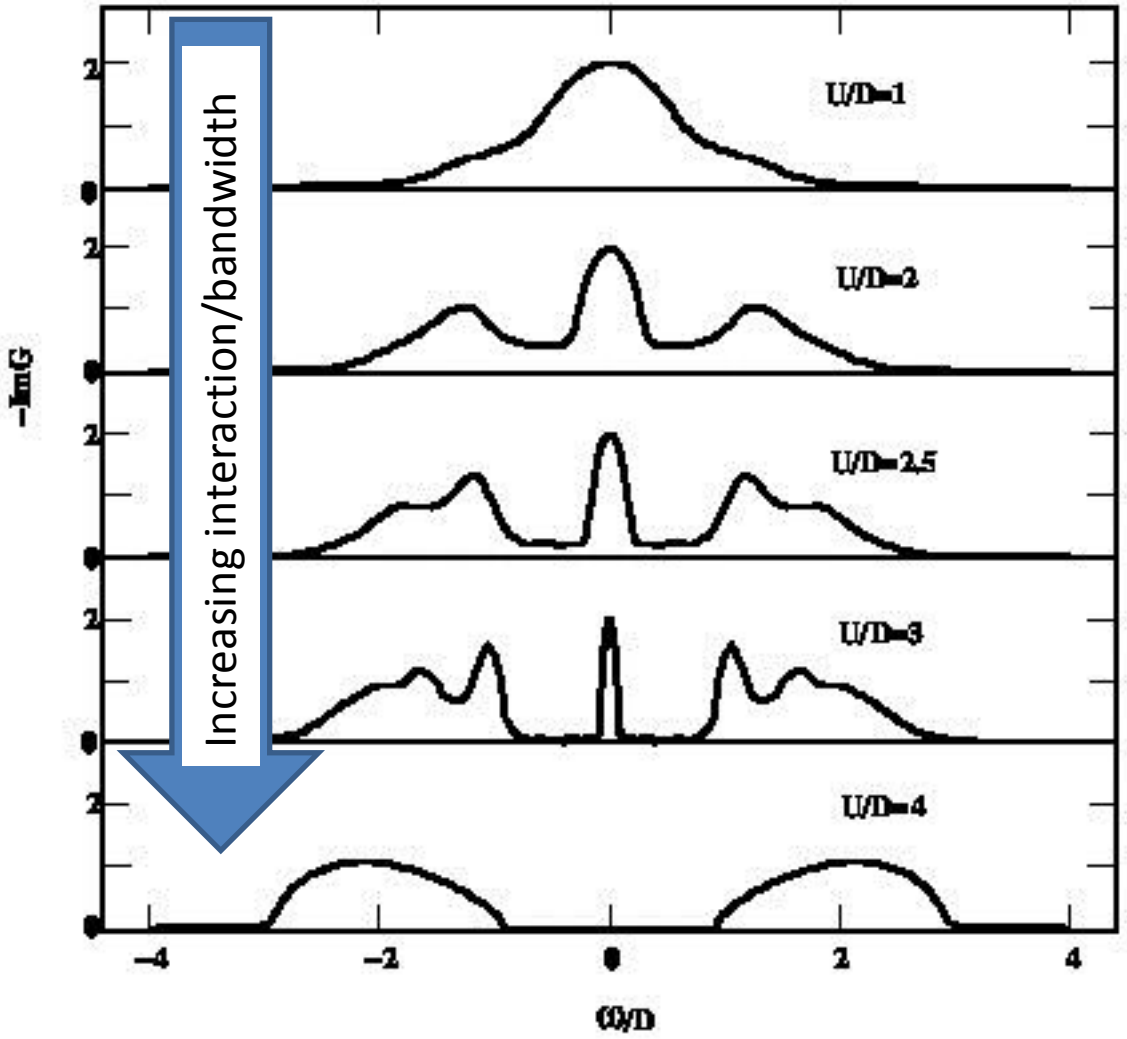
$$\hat{G}(\tau) = -\langle \hat{T} c(\tau) c^{\dagger}(0) \rangle$$

$$\hat{\Sigma} = \hat{\mathcal{G}}_0^{-1} - \hat{G}^{-1}$$

Self-consistency condition:

$$\hat{G}(\omega) = \sum_k \left(\omega + \mu - \hat{H}_o(k) - \hat{\Sigma}(\omega)\right)^{-1}$$

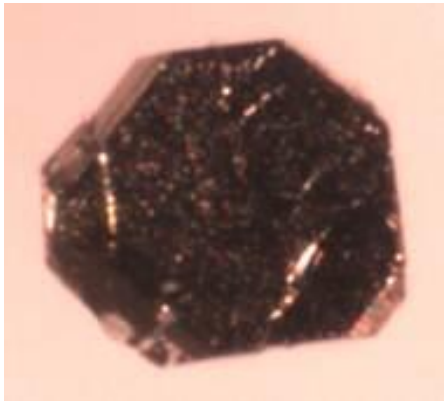
The Mott transition

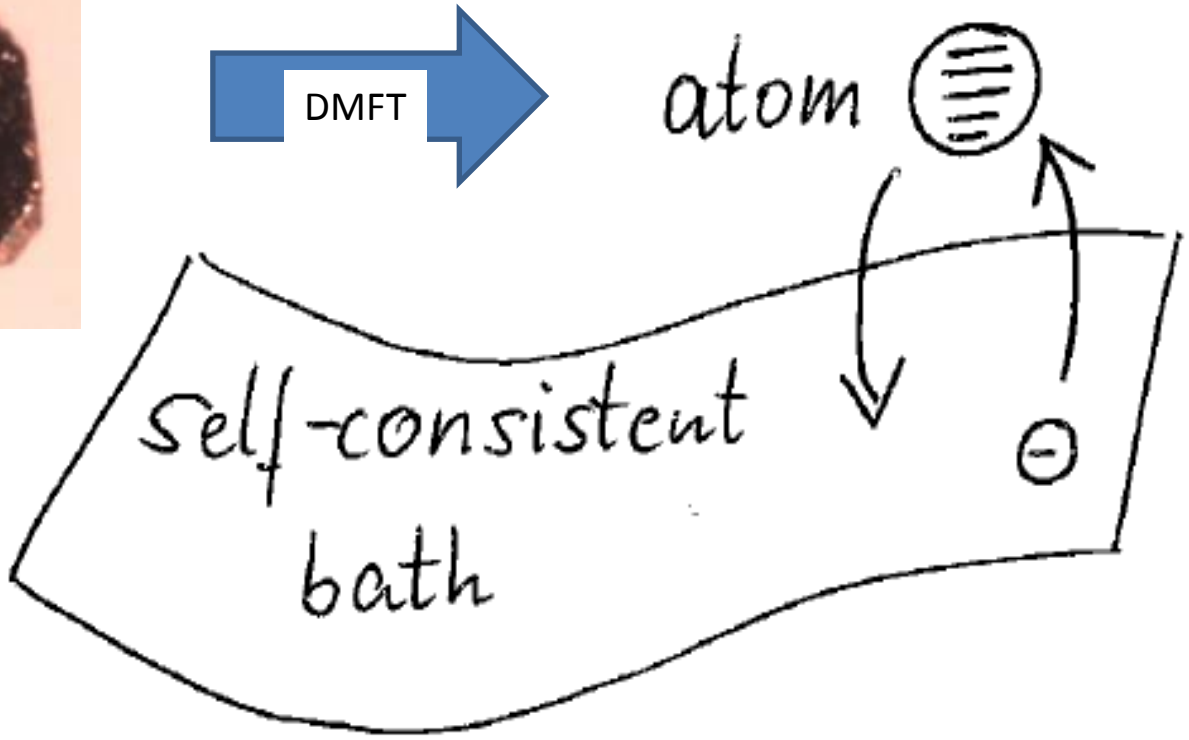


Spectral function of half-filled Hubbard model, paramagnetic phase

Zhang, Rozenberg, Kotliar, 1993
Georges, Kotliar, 1992

Dynamical mean field theory within realistic electronic structure calculations: “DFT+DMFT”





Lichtenstein, Katsnelson, 1998
Anisimov, … Kotliar, 1997

Density Functional Theory

Nobel Price in Chemistry, 1998

Interacting electron gas

Mapping

Non-interacting electrons in effective potential

such that ground state density is the same for the two systems

P. Hohenberg, W. Kohn, 1964,

W. Kohn, L. Sham, 1965

Density Functional Theory

Nobel Price in Chemistry, 1998

Interacting electron gas

Mapping

Non-interacting electrons in effective potential

such that ground state density is the same for the two systems

DFT used here as a means to generate the one-body part of a many-body Hamiltonian

DFT+DMFT – basic strategy

One-particle part of Hamiltonian from DFT-LDA

$$H = \sum_{\{im\sigma\}} (H^{LDA}_{im,i'm'} - H^{double\ counting}_{im,i'm'}) a^{+}_{im\sigma} a_{i'm'\sigma}$$

$$+ \frac{1}{2} \sum_{imm'\sigma\ \text{(correl. orb.)}} U^{i}_{mm'} n_{im\sigma} n_{im'-\sigma}$$

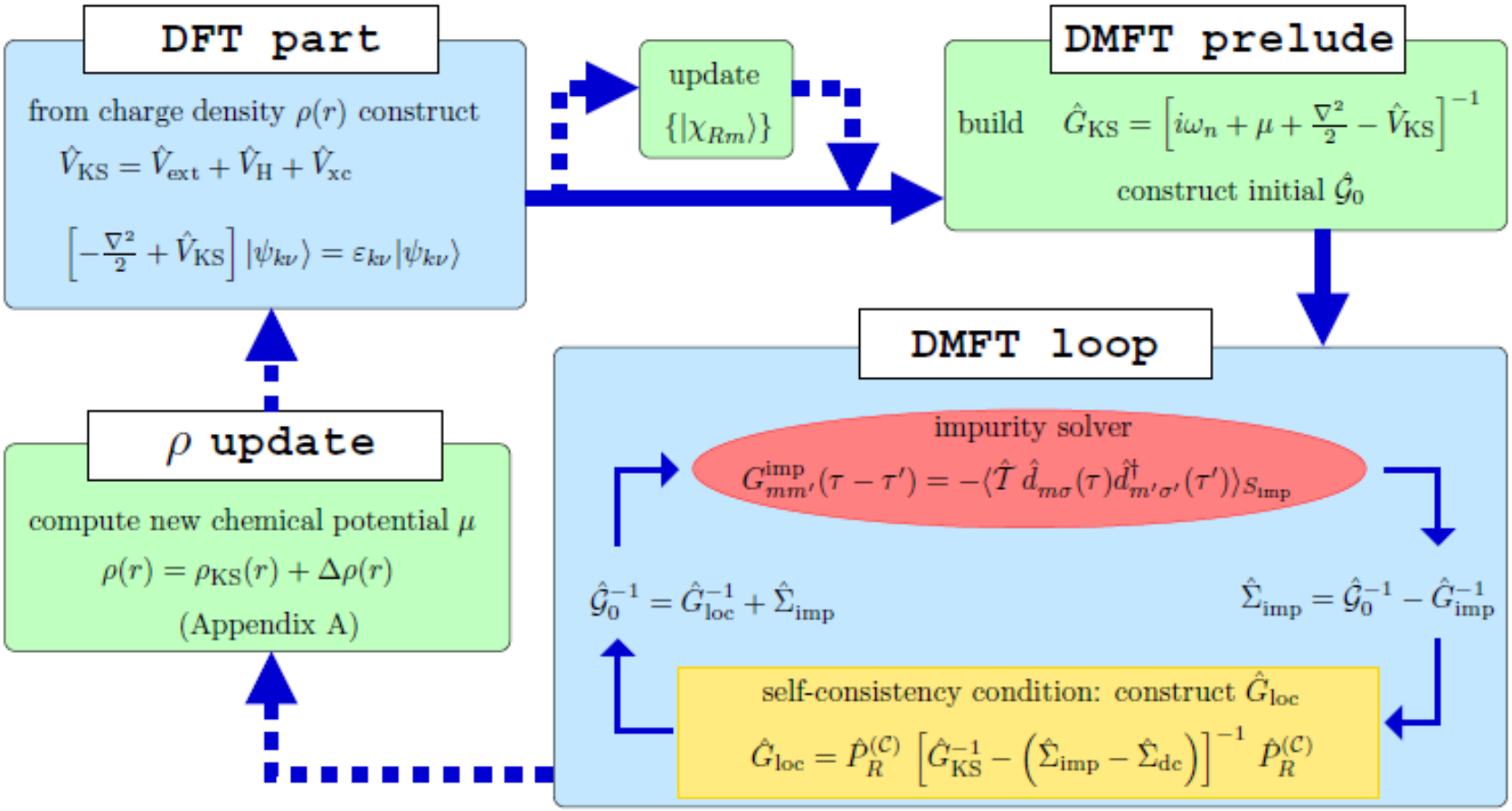
Hubbard interaction for "correlated shell"

$$+ \frac{1}{2} \sum_{im\neq m'\sigma\ \text{(correl. orb.)}} (U^{i}_{mm'} - J^{i}_{mm'}) n_{im\sigma} n_{im'\sigma}$$

Hund's rule coupling

- Solve within Dynamical Mean Field Theory (DMFT)
- => "LDA+DMFT" (Anisimov et al., Lichtenstein et al., 1997/98)

Charge-self-consistent DFT+DMFT :

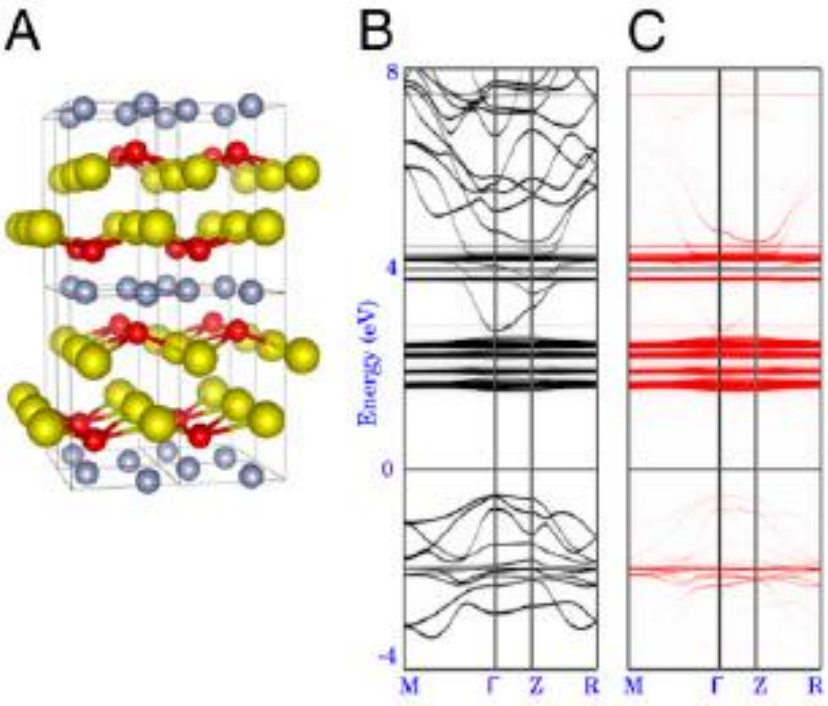


see e.g. F. Lechermann, A. Georges, A. Poteryaev, S. B., M. Posternak, A. Yamasaki, O. K. Andersen, PRB 2006

DMFT for CeSF

- Incorporates “atomic physics” of localised degrees of freedom into the itinerant band picture
- Multiplets are described correctly!

Example: spectral function of Mott insulating CeSF

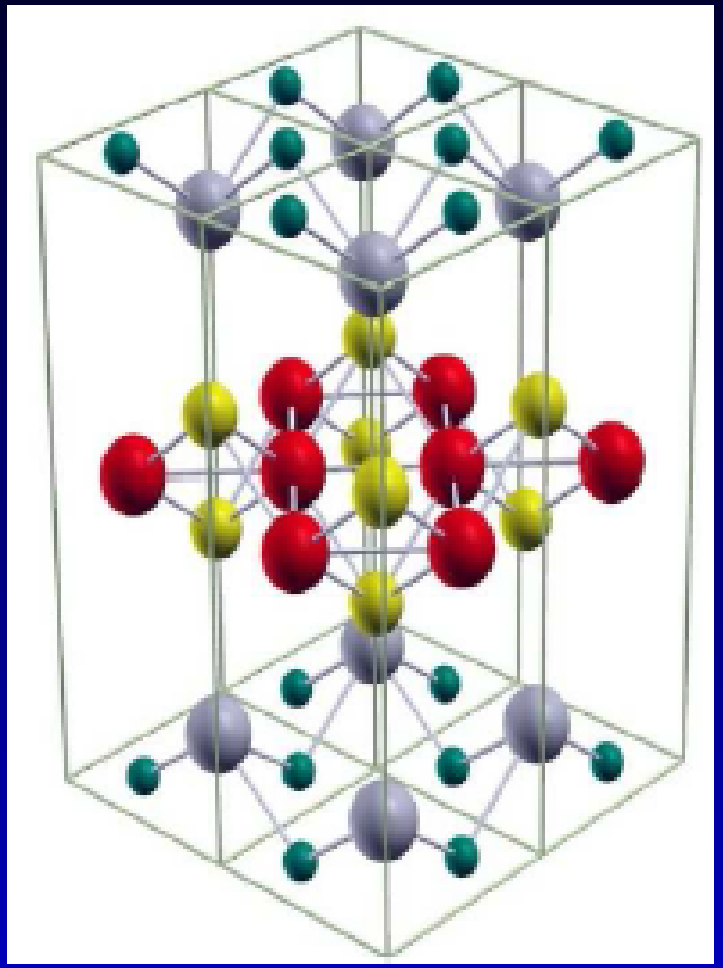


Calculated colour of CeSF:

Tomczak et al., PNAS 2013

A DMFT view on pnictides …

LaFeAsO



Fe

As

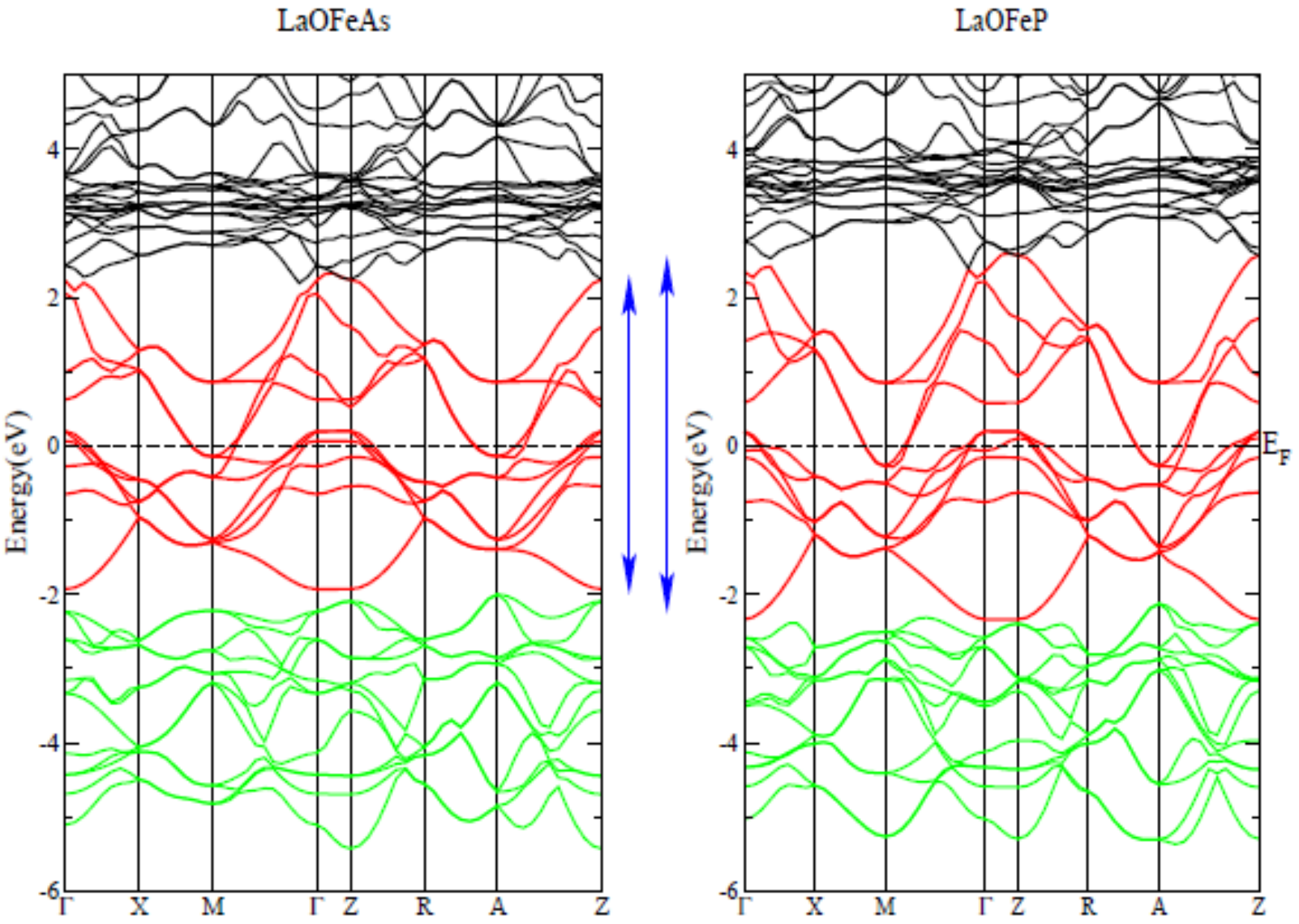
RE=La, Ce, Pr, Sm,...

O

Parent compound of new iron oxypnictide superconductors

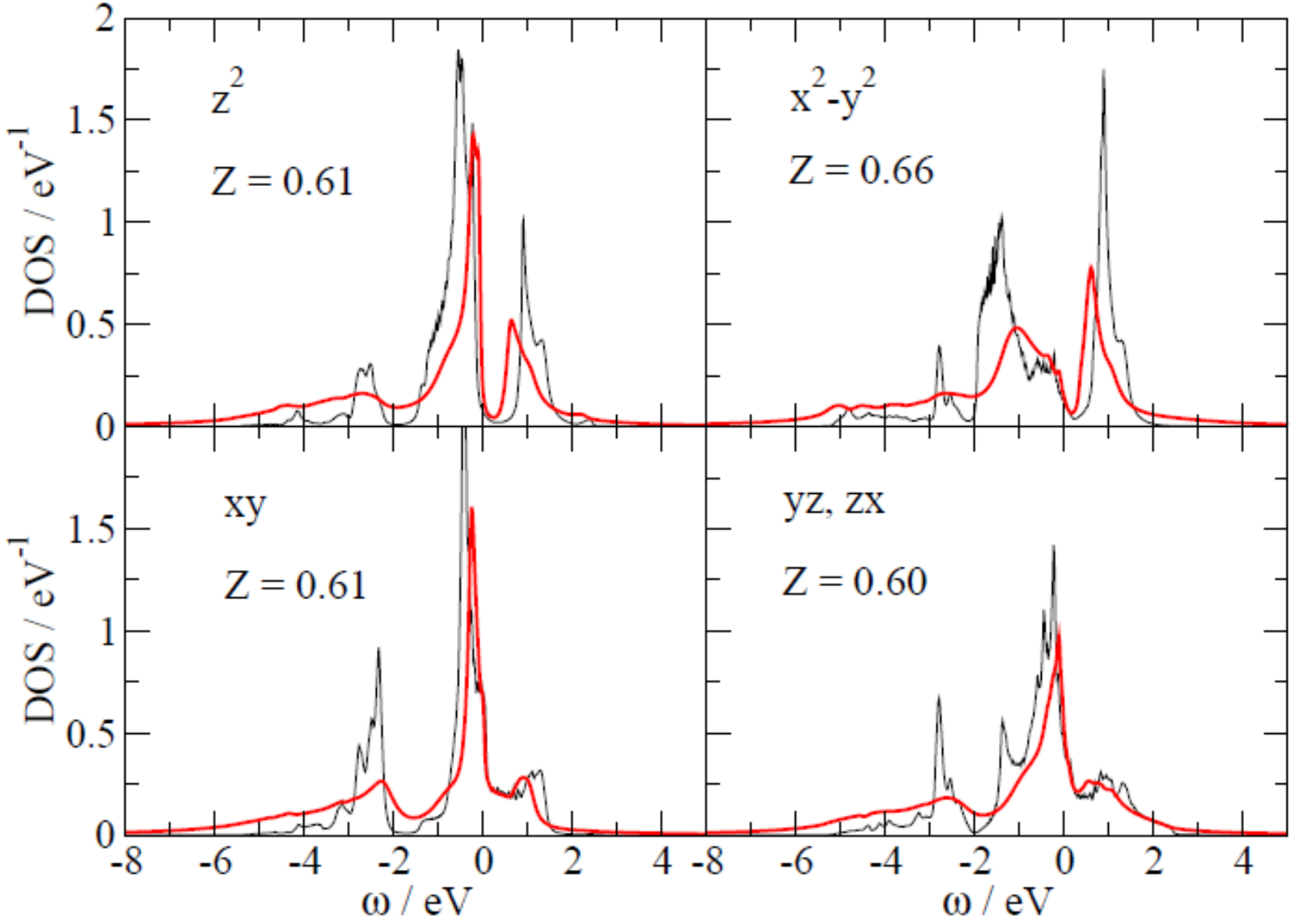
Fe-d states, hybridising with As-p and O-p bands

LDA band structure



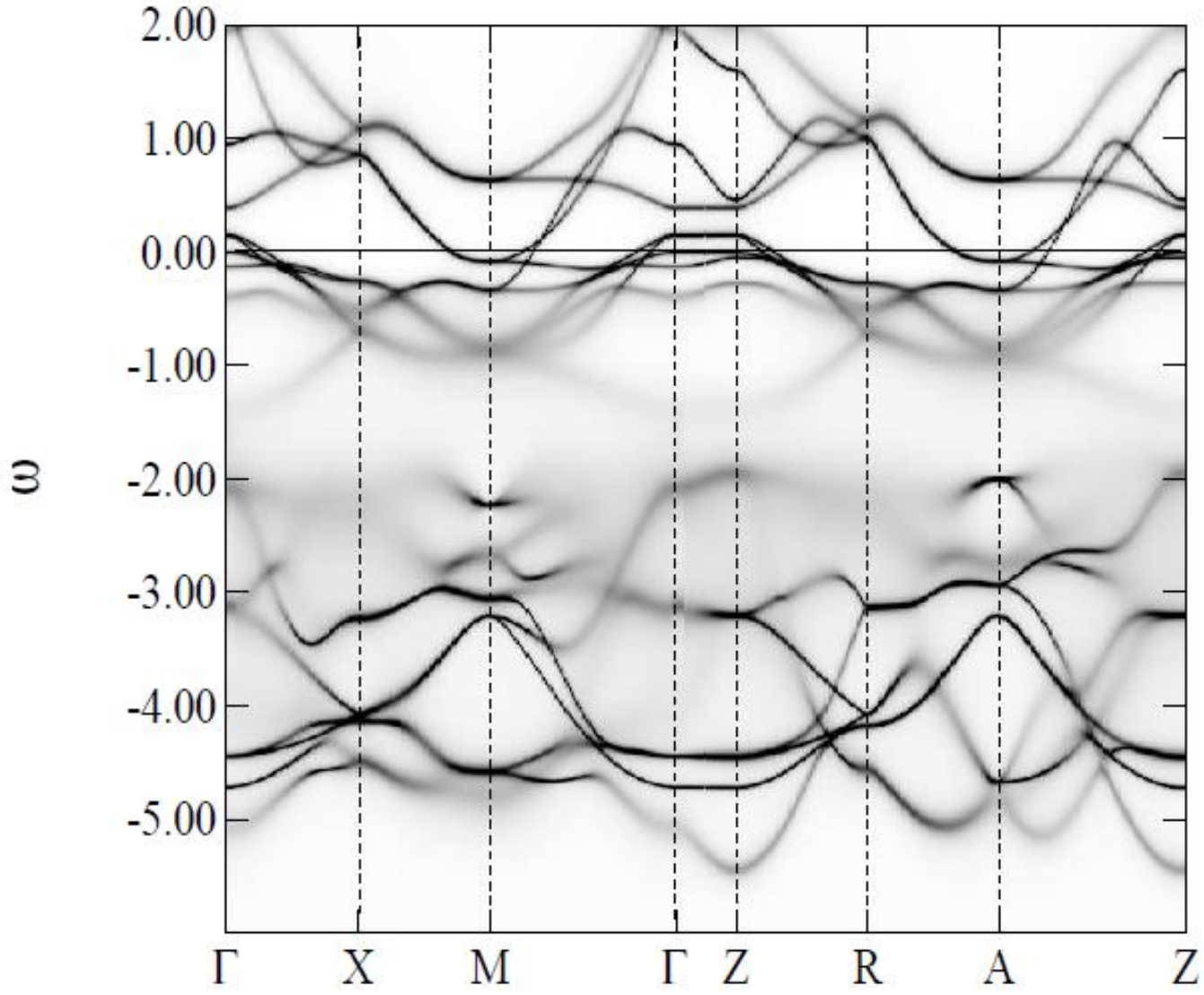
Vildosola, Pourovskii, Arita, Georges, Biermann, PRB 2008

Orbital-resolved spectral function



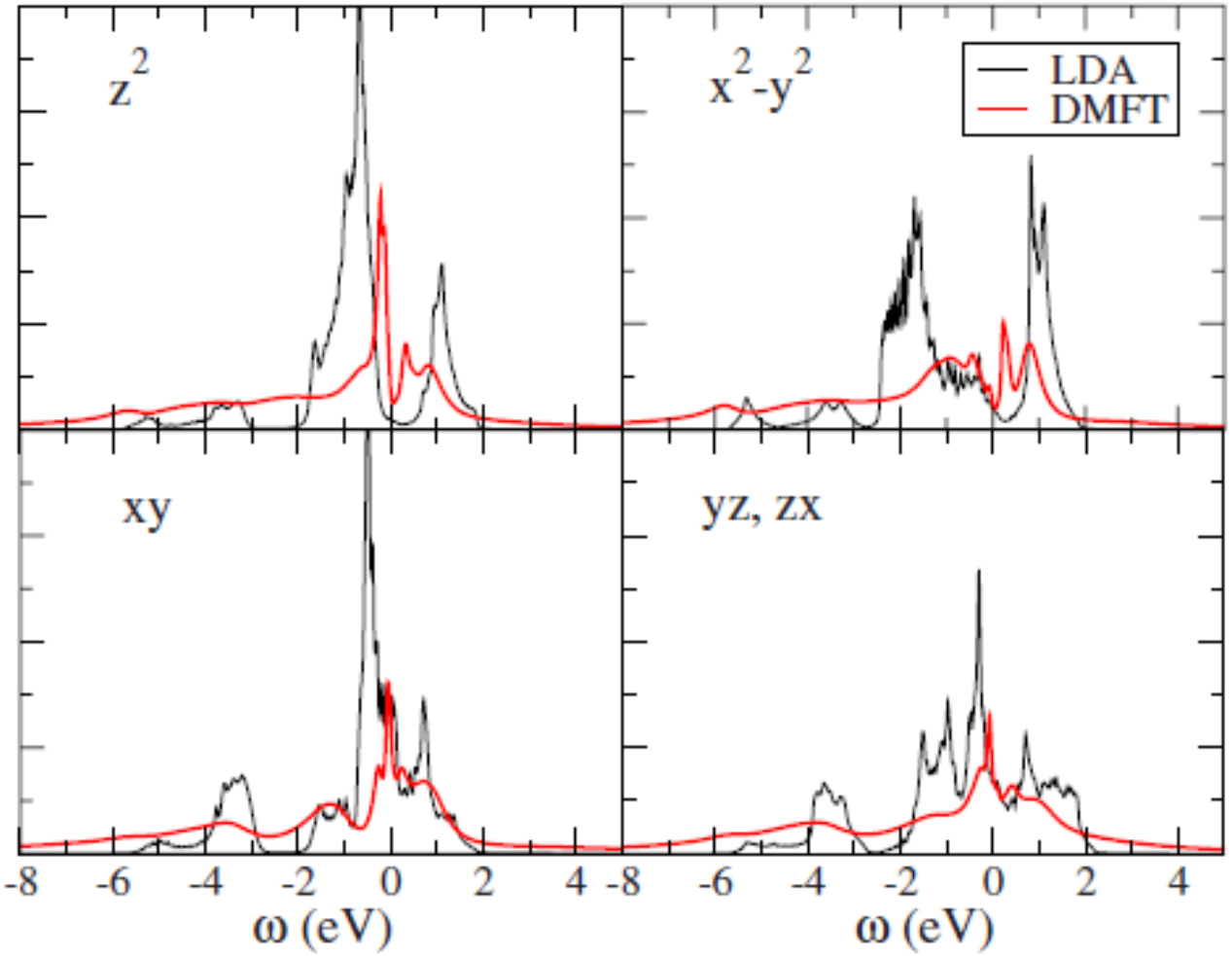
Aichhorn, Pourovskii, Vildosola,Ferrero, Parcollet, Miyake, Georges, Biermann, PRB 2009

LaFeAsO: K-resolved spectral function



Aichhorn, Pourovskii, Vildosola,Ferrero, Parcollet, Miyake, Georges, Biermann, PRB 2009

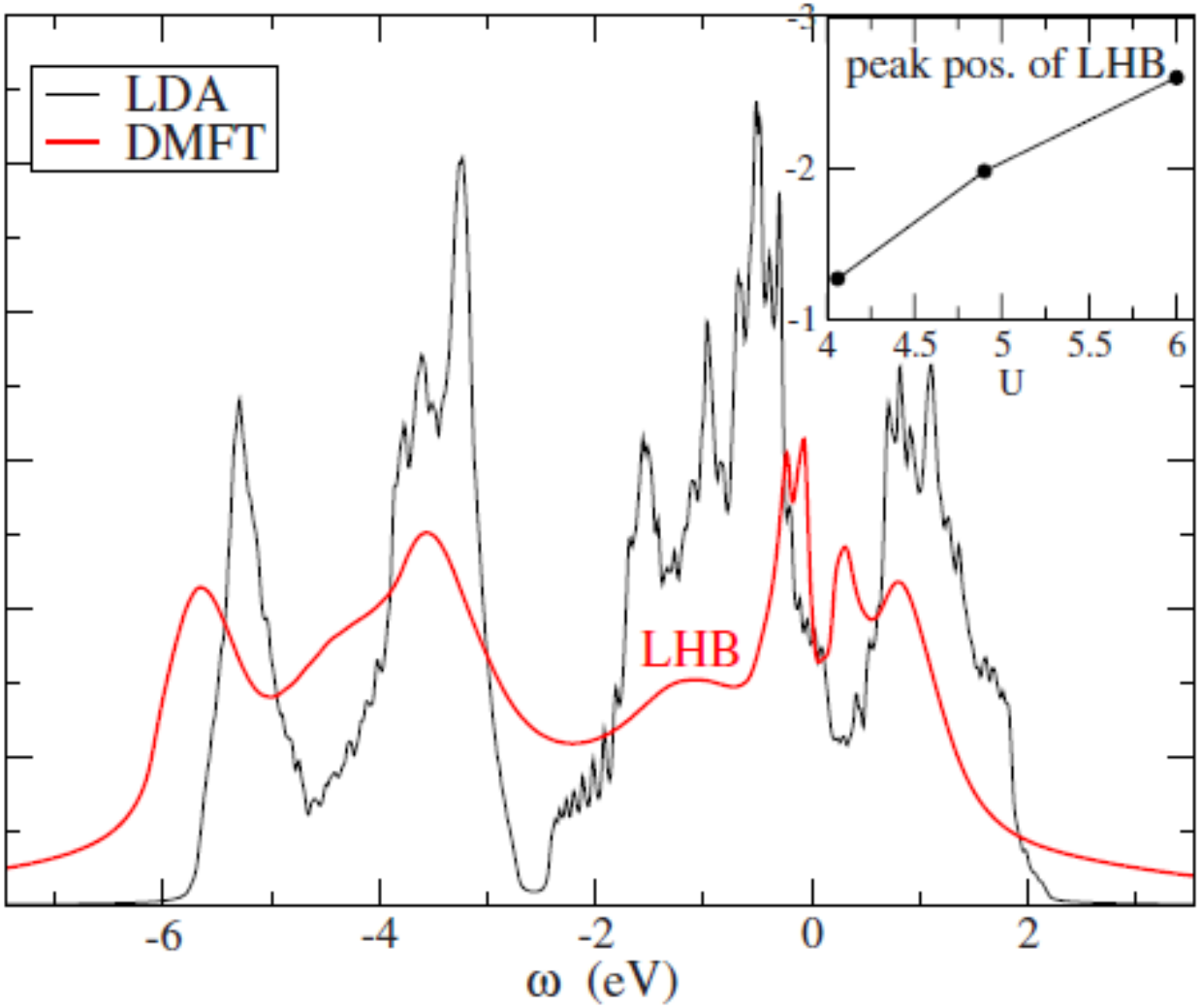
FeSe



effective masses: ~ 2 for $x^2 - y^2$, ~ 5 for xy
But: large quasi-particle (?) damping
$(-\mathrm{Im}\Sigma(i0^+)_{xy} \simeq 0.2\,\mathrm{eV}\,)$

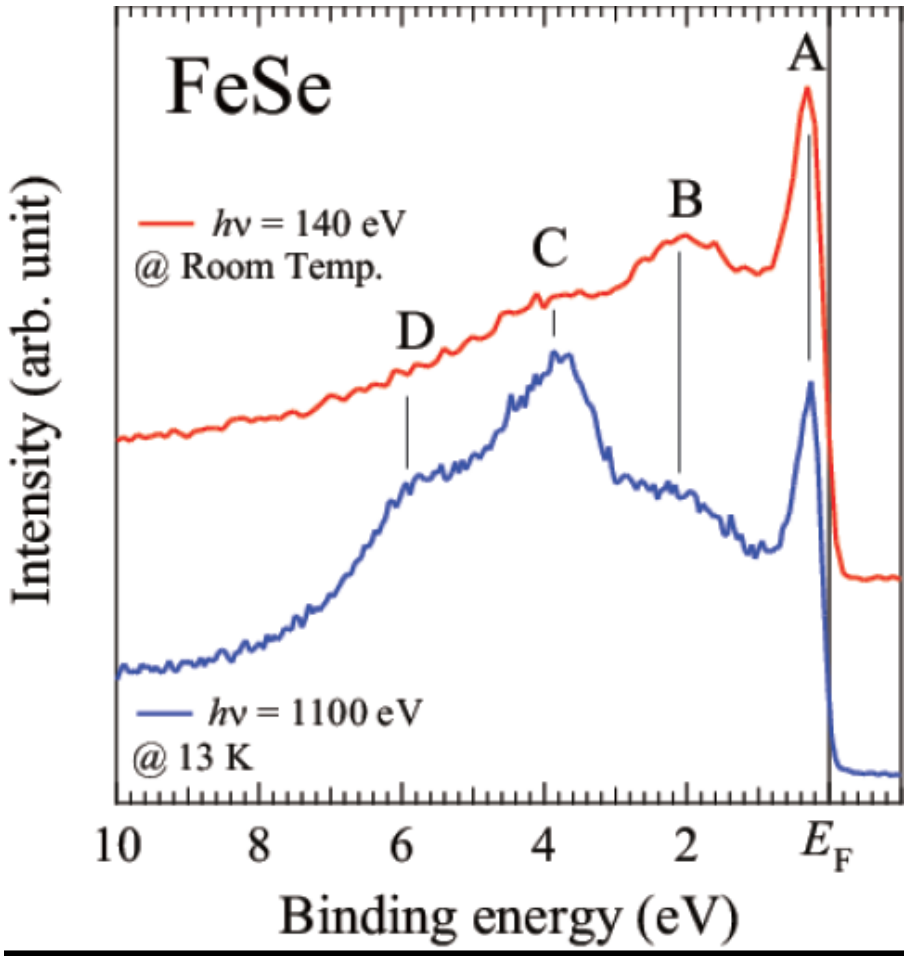
Aichhorn et al., PRB 2010

Lower Hubbard band in FeSe



Aichhorn et al., PRB 2010

Photoemission: Is B our Hubbard band?



Yoshida et al., 2009

Confirmation from PES?

PHYSICAL REVIEW B **82**, 184511 (2010)

Electron correlation in the FeSe superconductor studied by bulk-sensitive photoemission spectroscopy

A. Yamasaki,[1] Y. Matsui,[1] S. Imada,[2] K. Takase,[3] H. Azuma,[3] T. Muro,[4] Y. Kato,[4,*] A. Higashiya,[5,†] A. Sekiyama,[6] S. Suga,[6] M. Yabashi,[4] K. Tamasaku,[5] T. Ishikawa,[5] K. Terashima,[2] H. Kobori,[1] A. Sugimura,[1] N. Umeyama,[7,8] H. Sato,[9] Y. Hara,[10] N. Miyagawa,[7] and S. I. Ikeda[8]

FeSe. The self-energy correction provides the larger mass enhancement value ($Z^{-1}=3.6$) than in Fe-As superconductors and enables us to separate an incoherent part from the spectrum. These features are quite consistent with the results of recent dynamical mean-field calculations, in which the incoherent part is attributed to the lower Hubbard band.

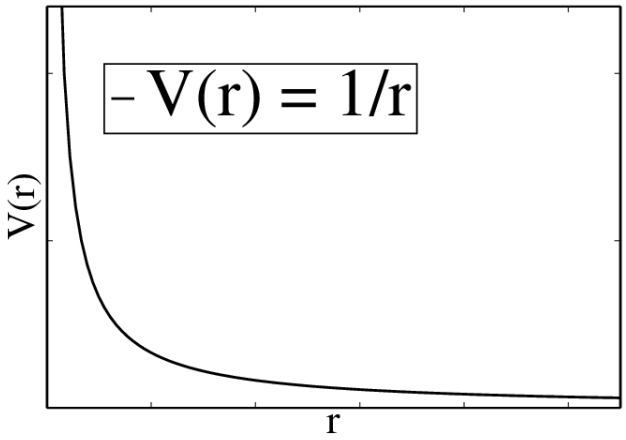
Confirmation from ARPES

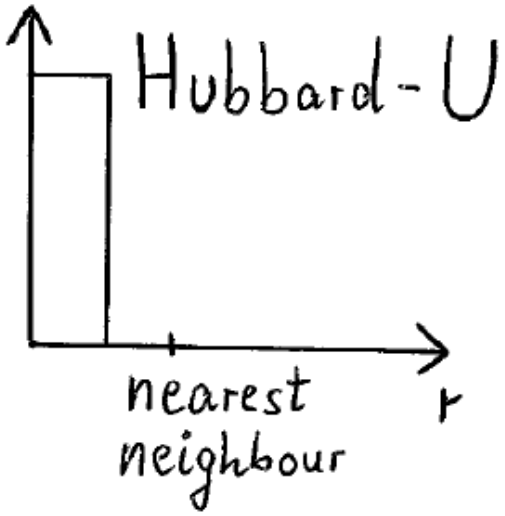
-> recent work from Dresden group

What about U?

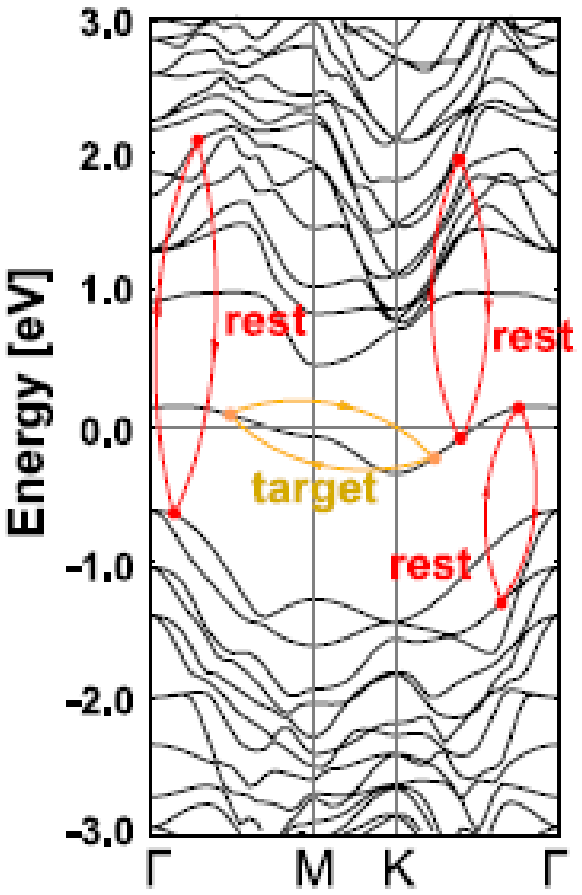
How to bridge between …

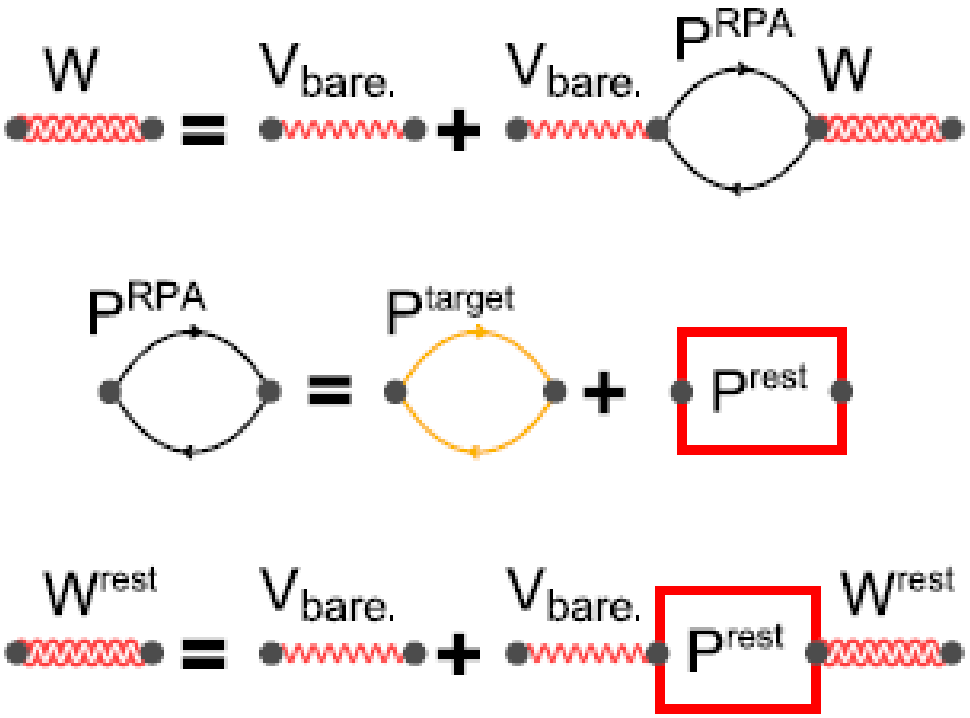
1/r Coulomb interaction & its description as “+U” ?





Constrained Random Phase Approximation





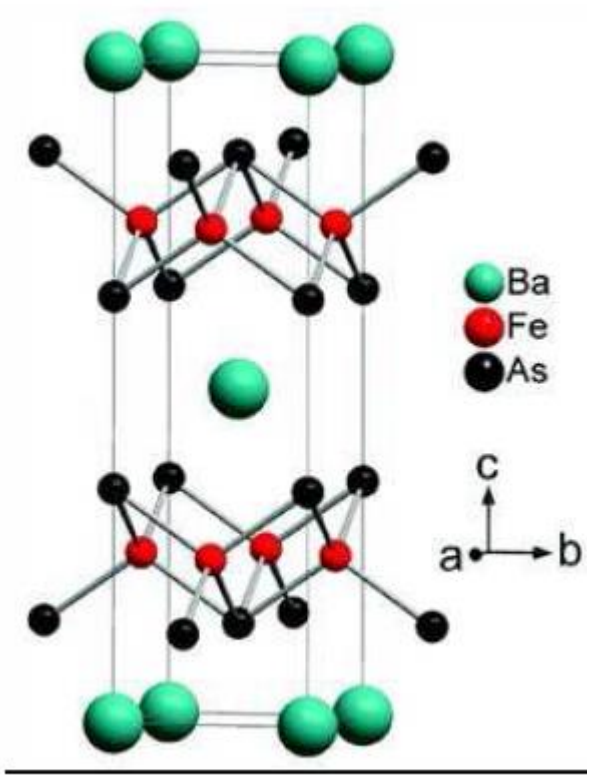
RPA ⇨ cRPA

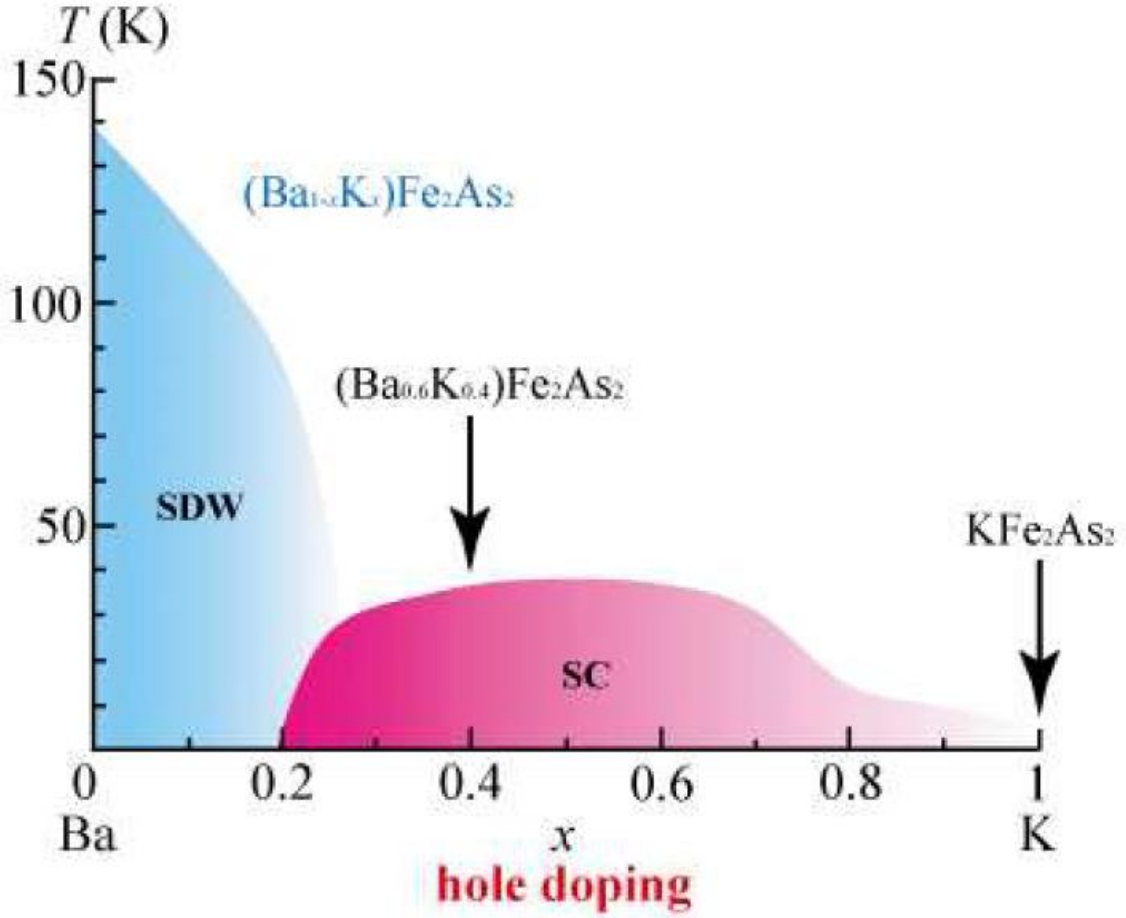
$$U^{cRPA}_{lmno} = \langle \varphi_l \varphi_m | W^{rest} | \varphi_n \varphi_o \rangle$$

Aryasetiawan, Imada, Georges, Kotliar, Biermann, Lichtenstein, PRB 2004.

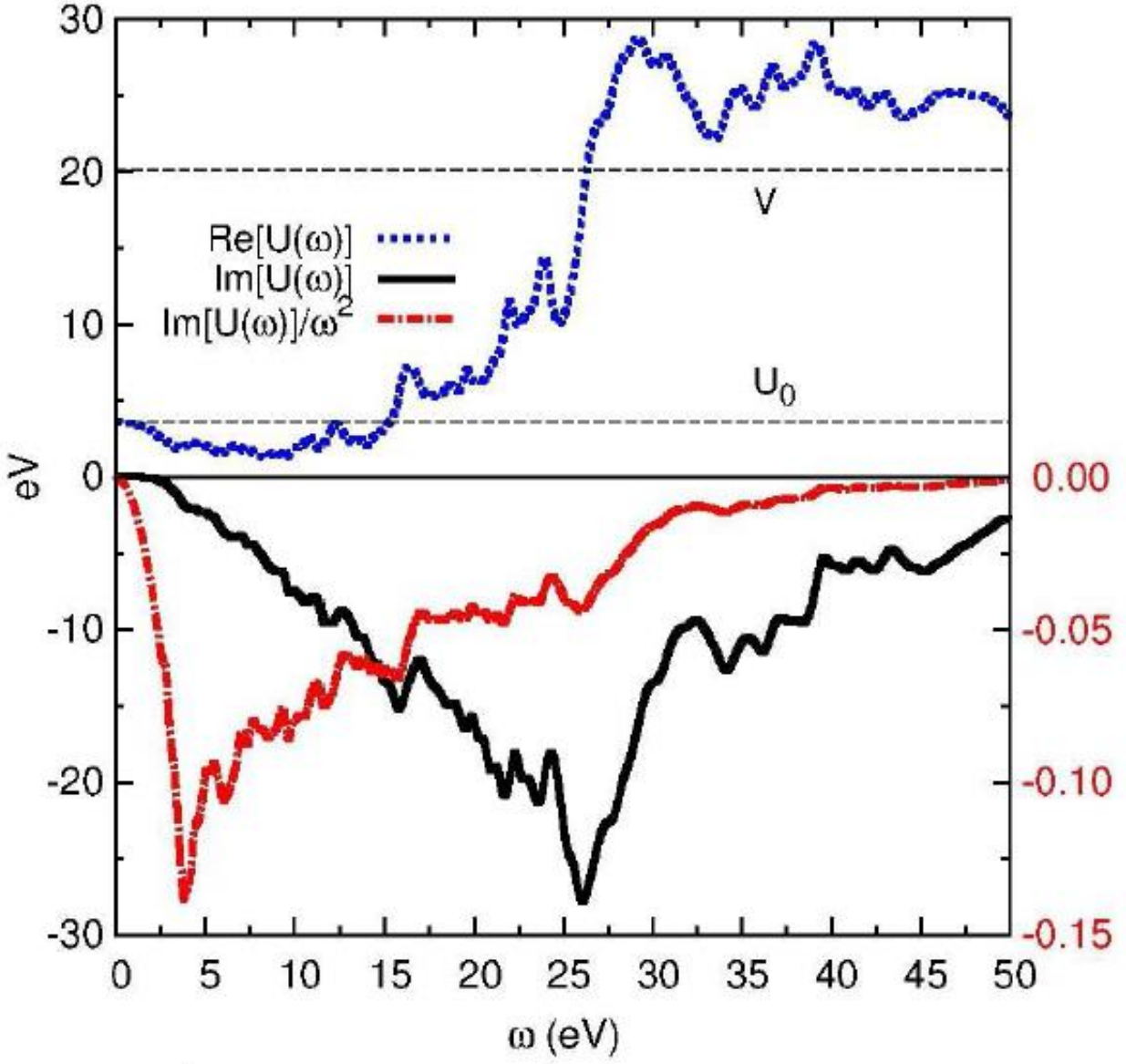
[Figure from Hansmann et al., JPCM 2013]

BaFe2As2


Ba
Fe
As
c
a
b

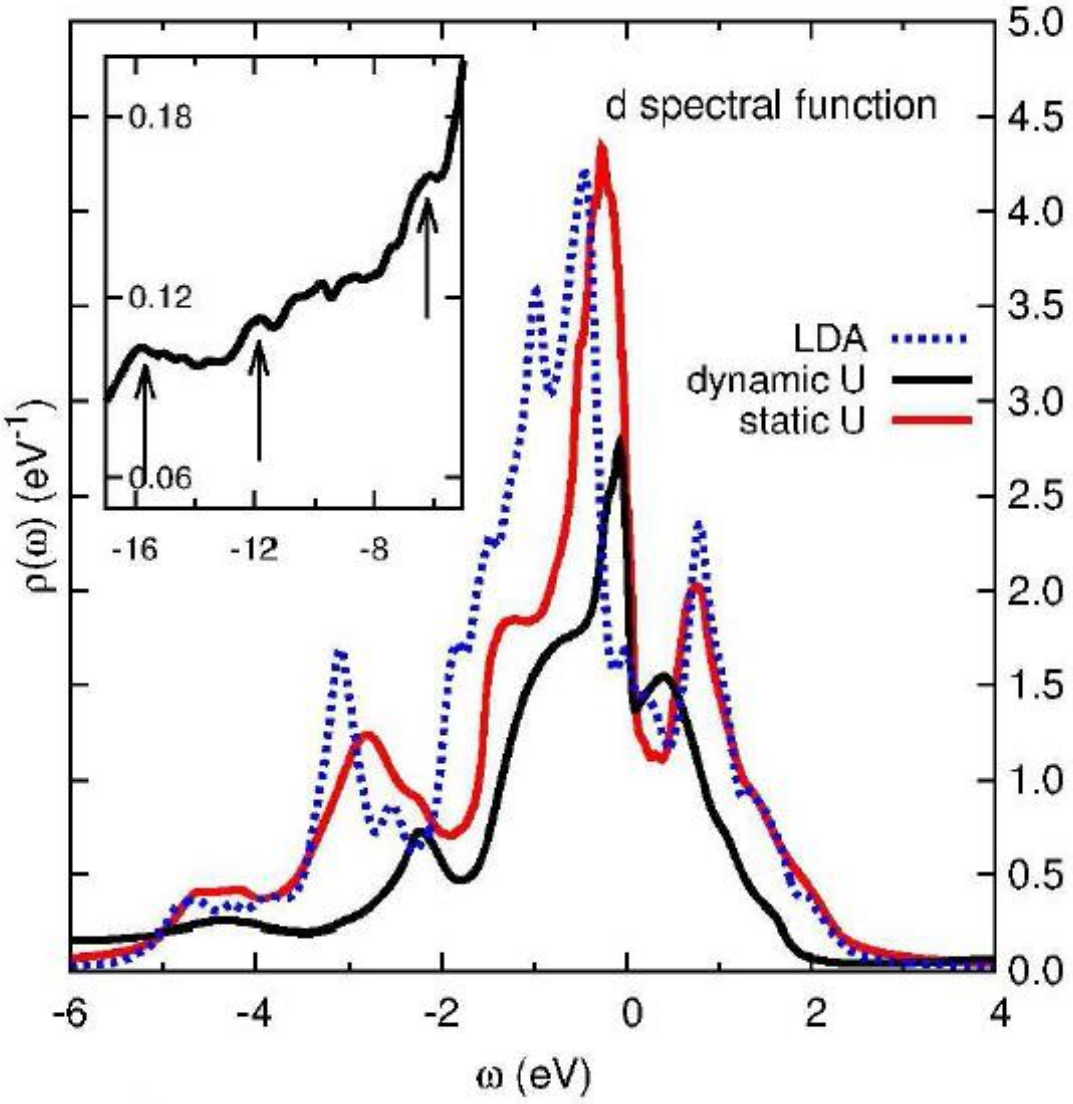

T (K)
150
100
50
(Ba1-xKx)Fe2As2
(Ba0.6K0.4)Fe2As2
SDW
SC
KFe2As2
0
Ba
0.2
0.4
0.6
0.8
1
K
x
hole doping

U(ω) for BaFe2As2



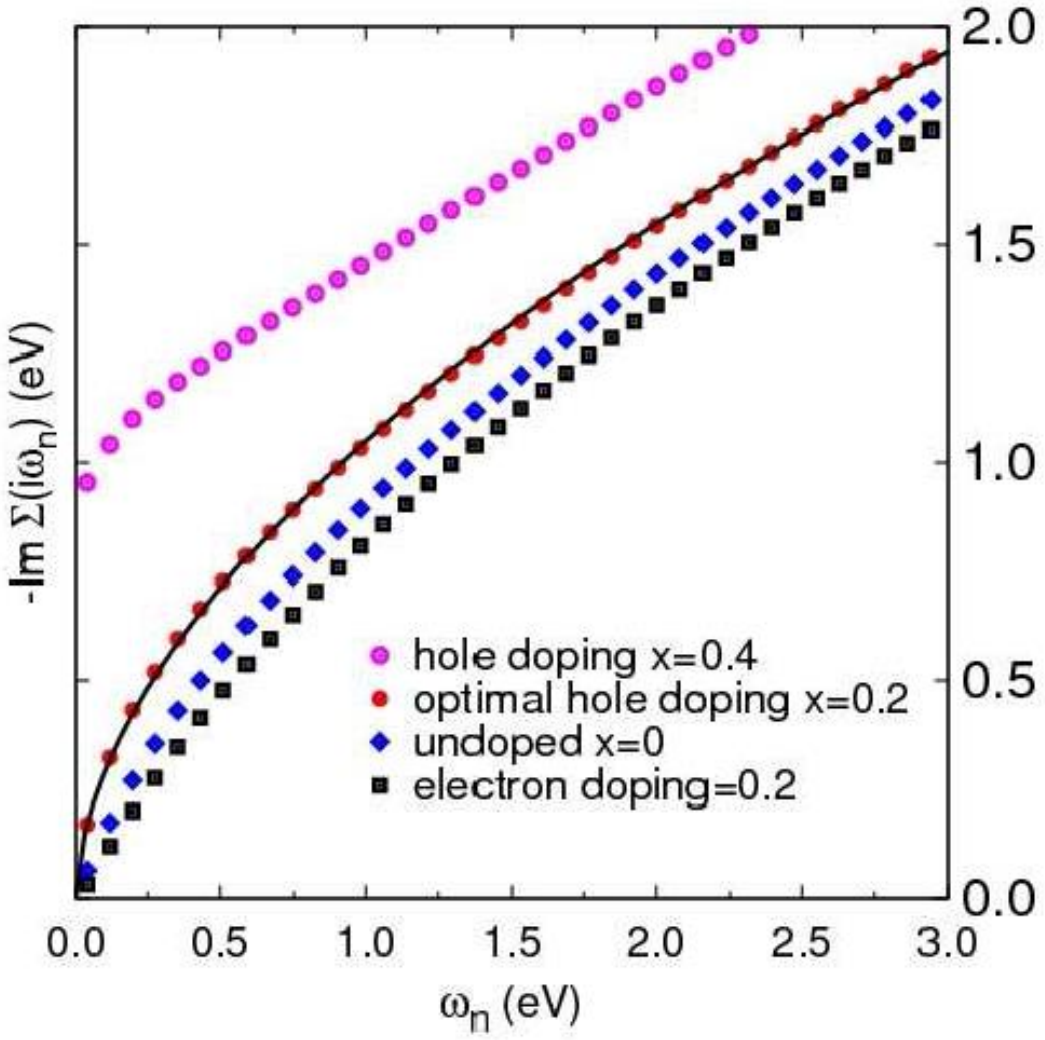
Werner, Casula, Miyake, Aryasetiawan, Millis, SB, Nature Phys. 2012

BaFe2As2



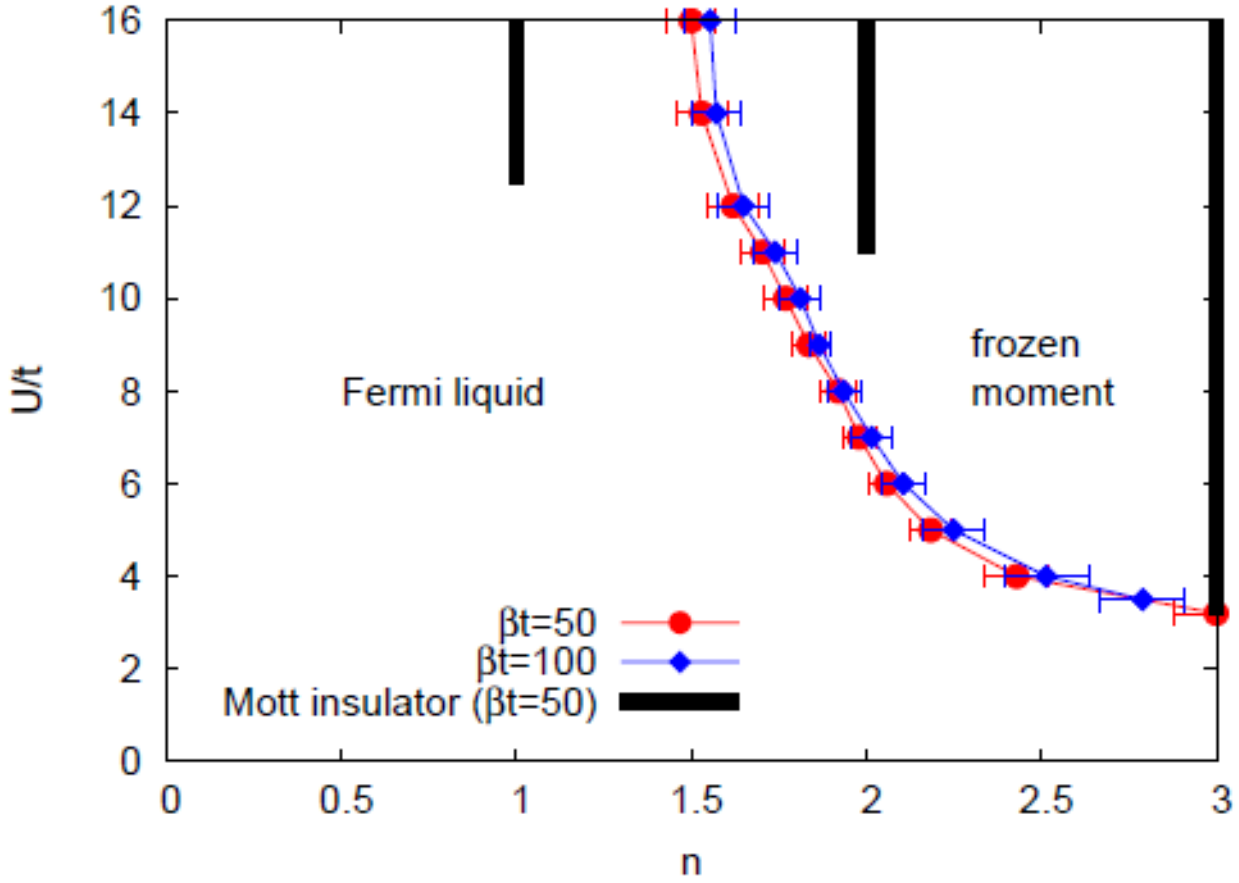
Werner, Casula, Miyake, Aryasetiawan, Millis, SB, Nature Phys. 2012

Fractional power law behavior!



Werner, Casula, Miyake, Aryasetiawan, Millis, SB, Nature Phys. 2012

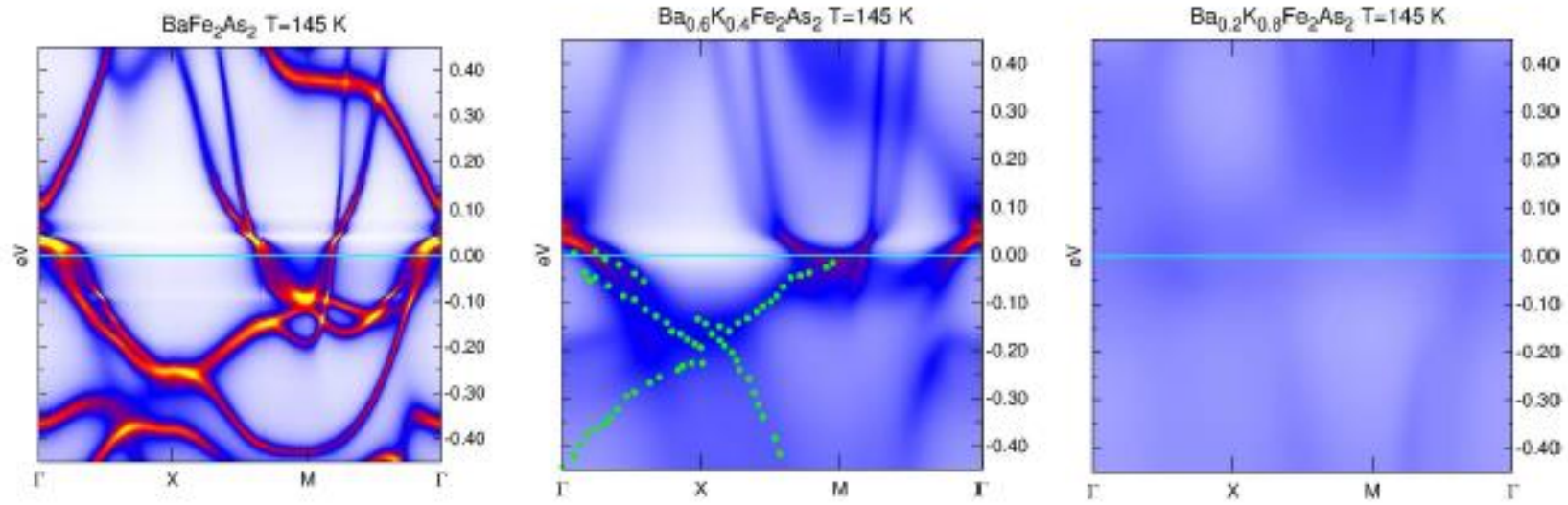
“Spin-freezing” in 3-orbital model



Werner, Millis, PRL 2008

“Hund’s metals”

- Haule, Kotliar, New J. Phys. 2008: sensitivity to Hund’s coupling
- A. Liebsch PRB 2010, L. de’ Medici and collaborators PRL 2011: effective masses doping-dependent
- NOW: coherence doping-dependent !



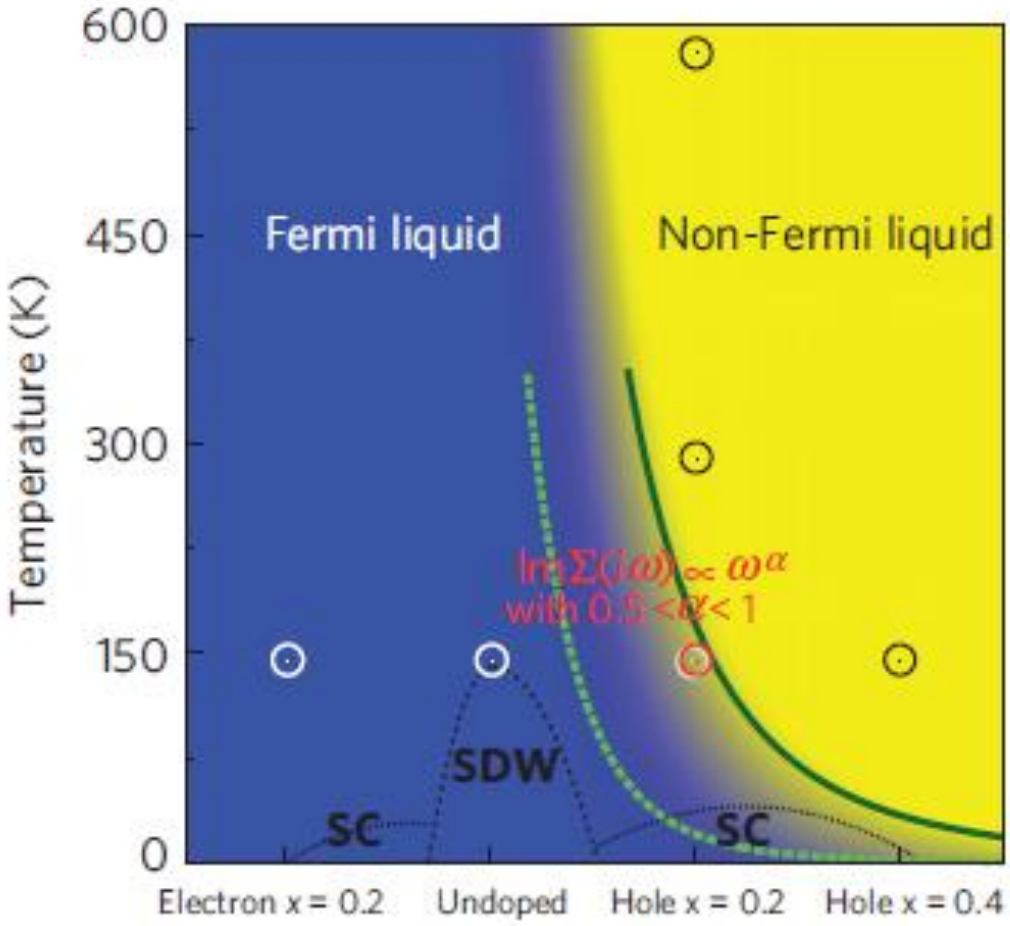
x = 0 x = 0.4 x = 0.8

Werner, Casula, Miyake, Aryasetiawan, Millis, SB, Nature Phys. 2012

Satellites and large doping and temperature dependence of electronic properties in hole-doped $BaFe_2As_2$

Philipp Werner[1,2]*, Michele Casula[3], Takashi Miyake[4,5,6], Ferdi Aryasetiawan[7,8], Andrew J. Millis[9] and Silke Biermann[5,10]

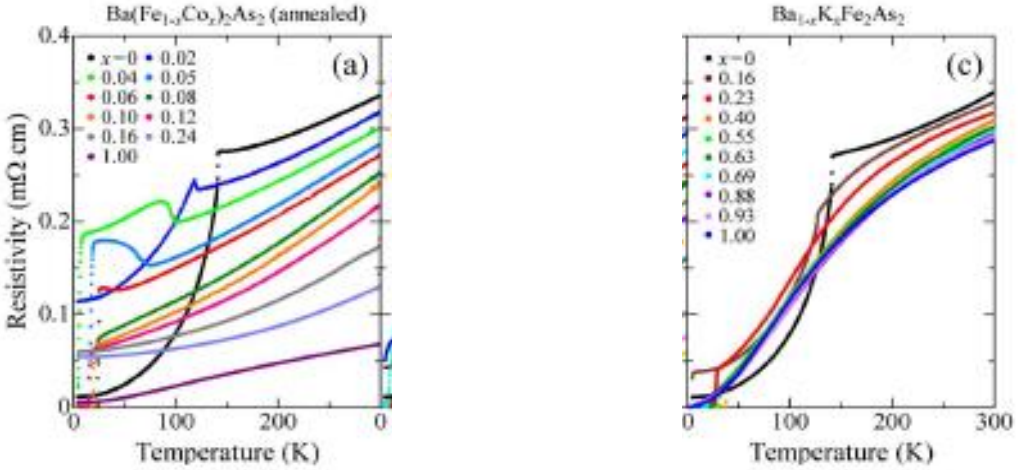
Nat. Phys. 2012



Experimental confirmation?!

Normal-state charge dynamics in doped $BaFe_2As_2$: Roles of doping and necessary ingredients for superconductivity

M. Nakajima,[1,2,3,*] S. Ishida,[1,2,3] T. Tanaka,[1,3] K. Kihou,[2,3] Y. Tomioka,[2,3] T. Saito
H. Fukazawa,[3,4] Y. Kohori,[3,4] T. Kakeshita,[1,3] A. Iyo,[2,3] T. Ito,[2,3] H. Eisaki,[2,3] an



With Co doping, the temperature dependence changes from T linear to T^2 in the overdoped non-SC region.

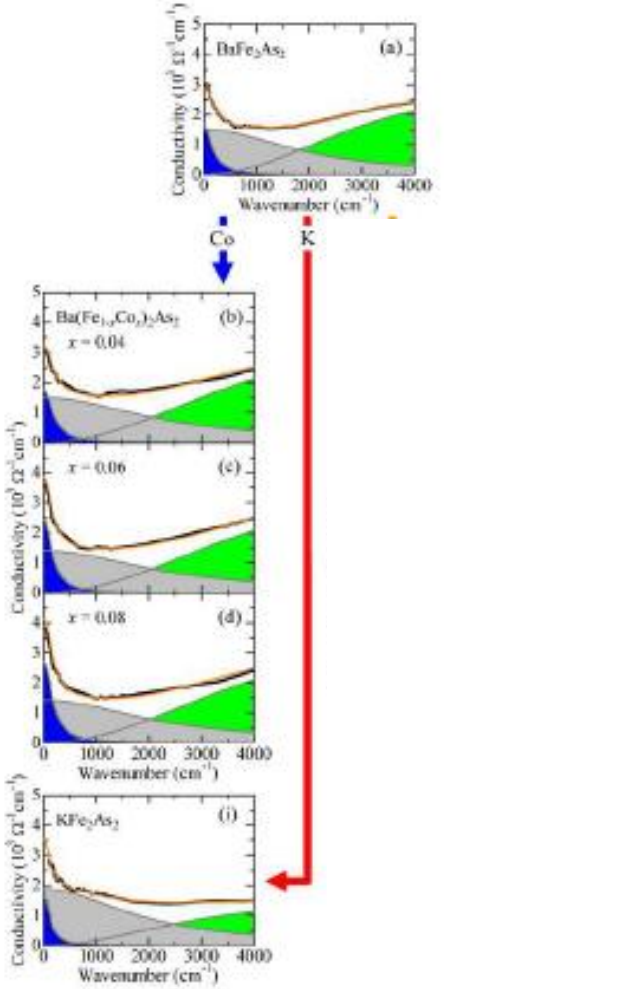
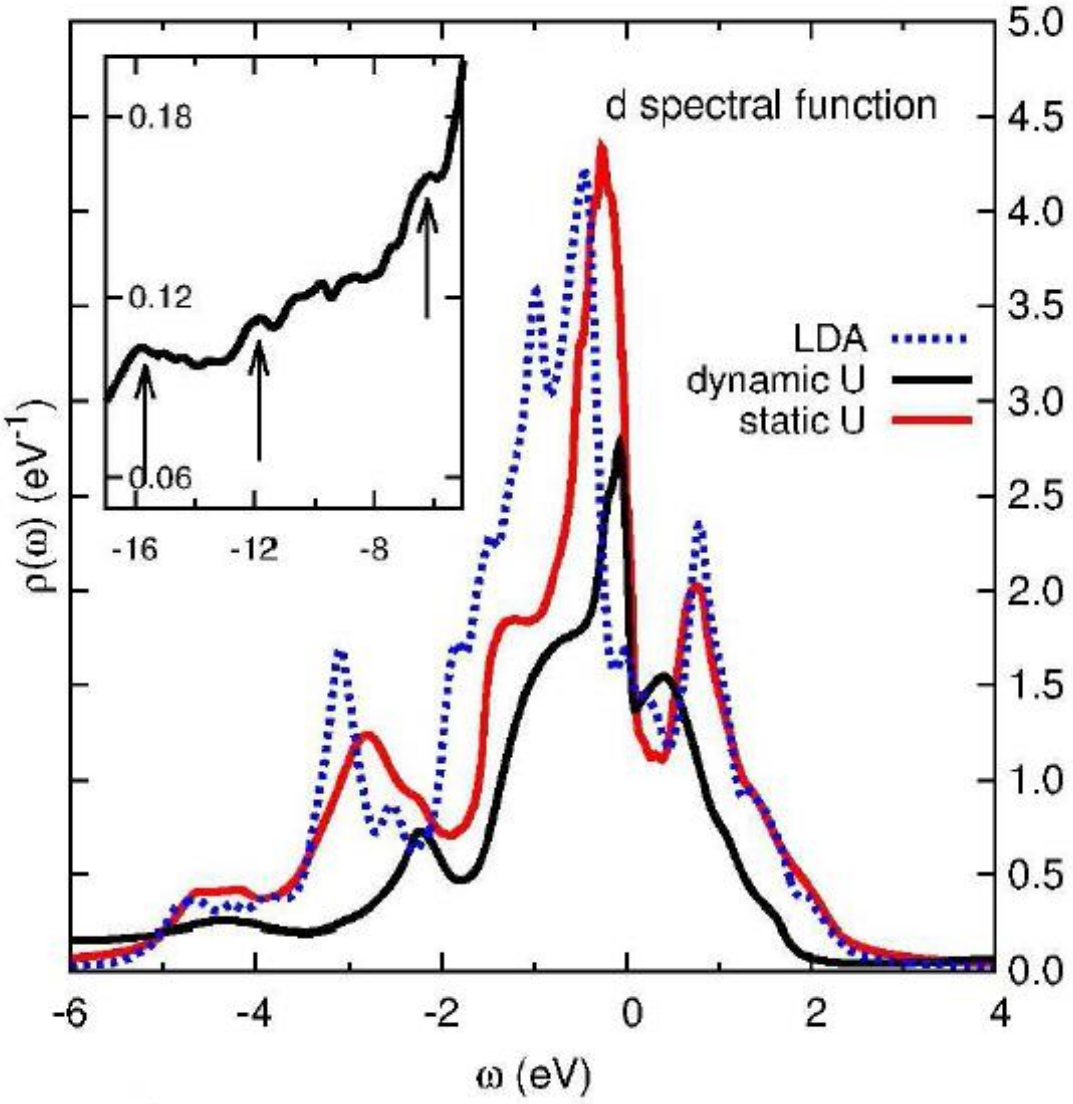


FIG. 2. Decomposition of the optical conductivity spectrum at $T = 300$ K for (a) $BaFe_2As_2$, (b–d) Co-Ba122, (e–h) P-Ba122, and (i) KFe_2As_2.

- BaFe2As2 “bad metal”
- Coherence increases with Co-doping
- K-doping does not help …

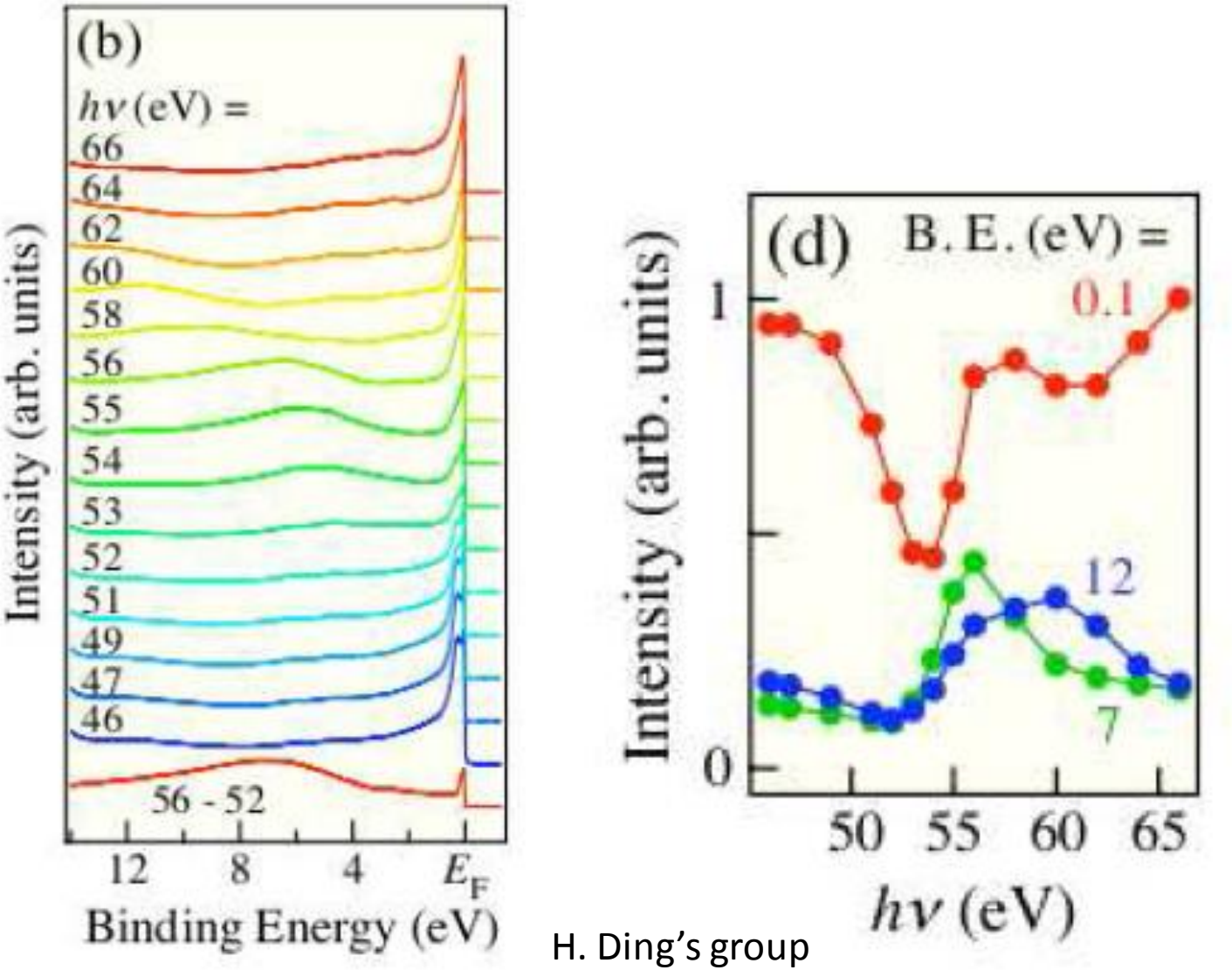
Nakajima et al, Sci. Rep. 2014

BaFe2As2



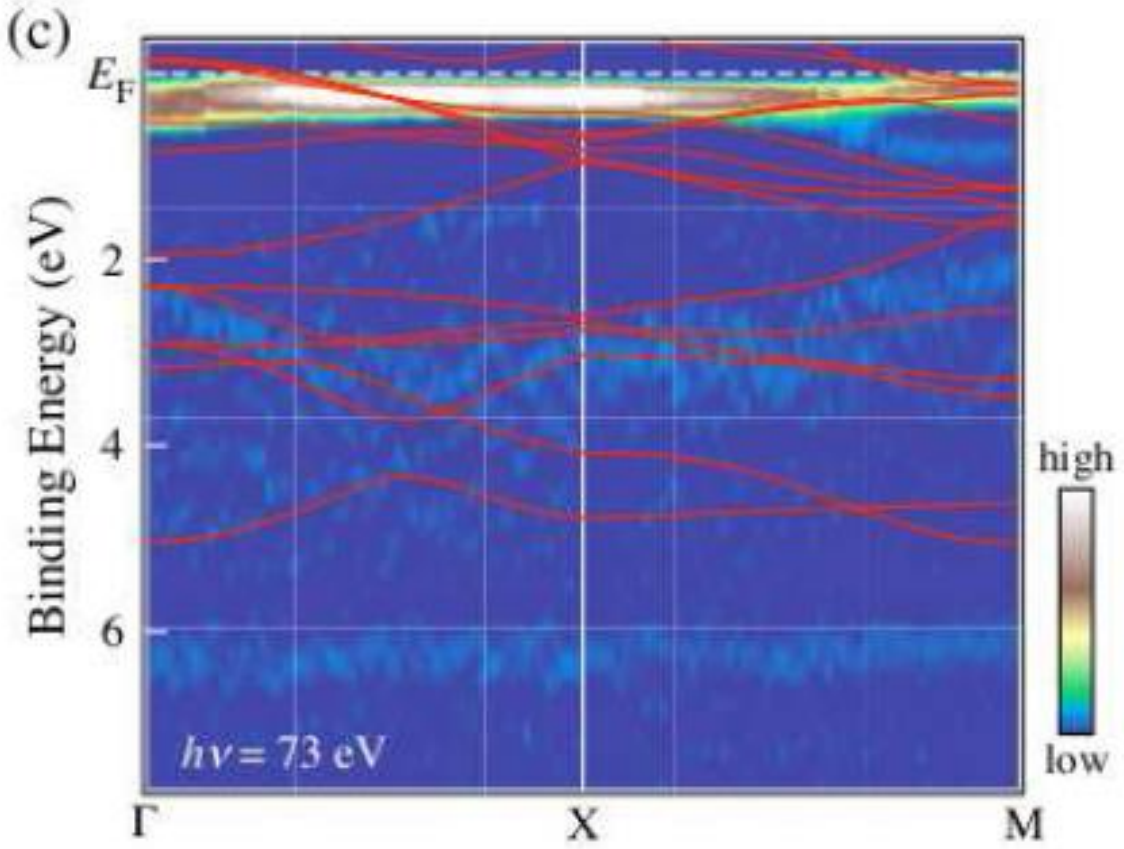
Werner, Casula, Miyake, Aryasetiawan, Millis, SB, Nature Phys. 2012

BaFe2As2 Photoemission

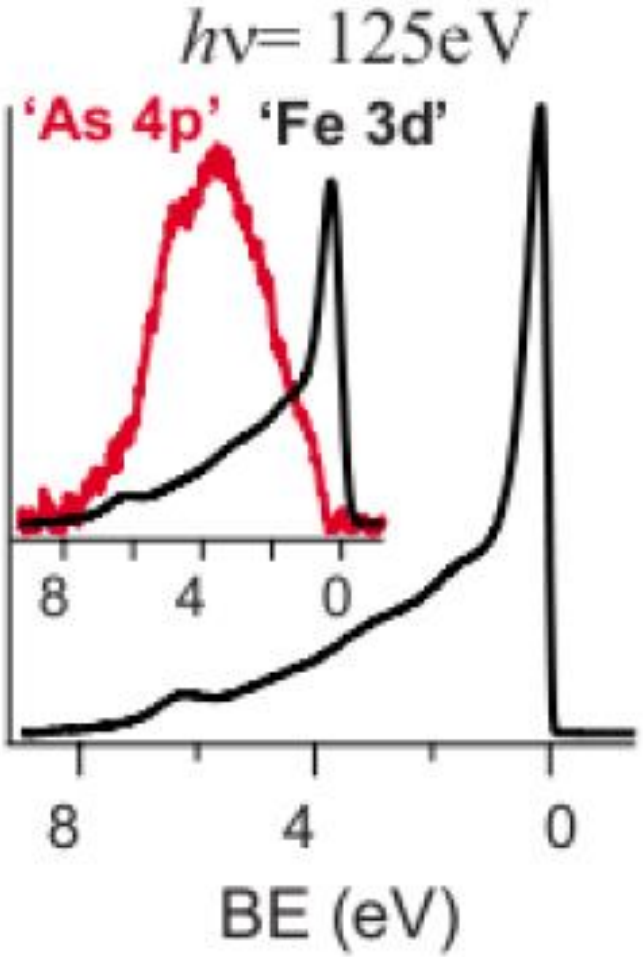


H. Ding's group

BaFe2As2 Photoemission



Ding et al



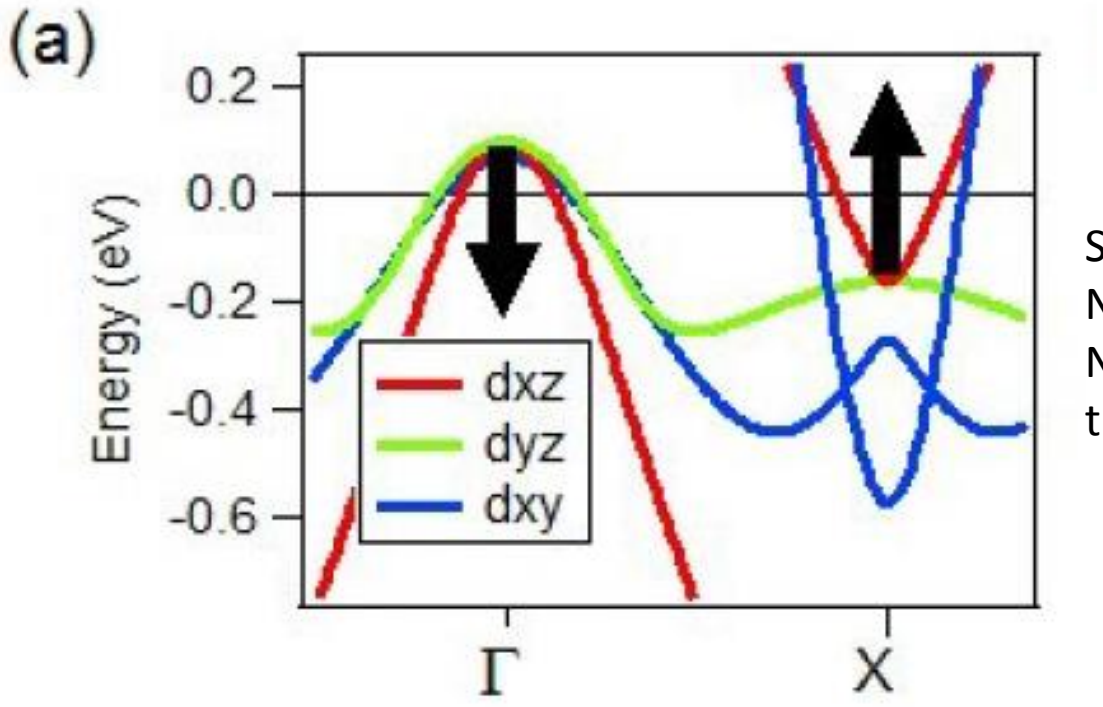
Yi et al.

What's left?

- Fermi surface and low-energy zoom?

What's left?

- Fermi surface and low-energy zoom?



Shift of pockets!
Not a renormalization effect!
Not fully in DMFT -- however:
tiny energy scales …

Ba(FeCo)2As2 – from Brouet et al., PRL 2013

What's left?

- Fermi surface and low-energy zoom?

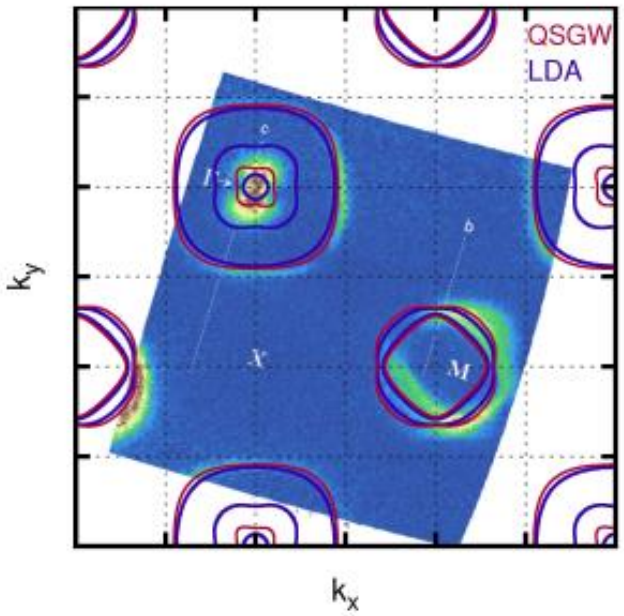


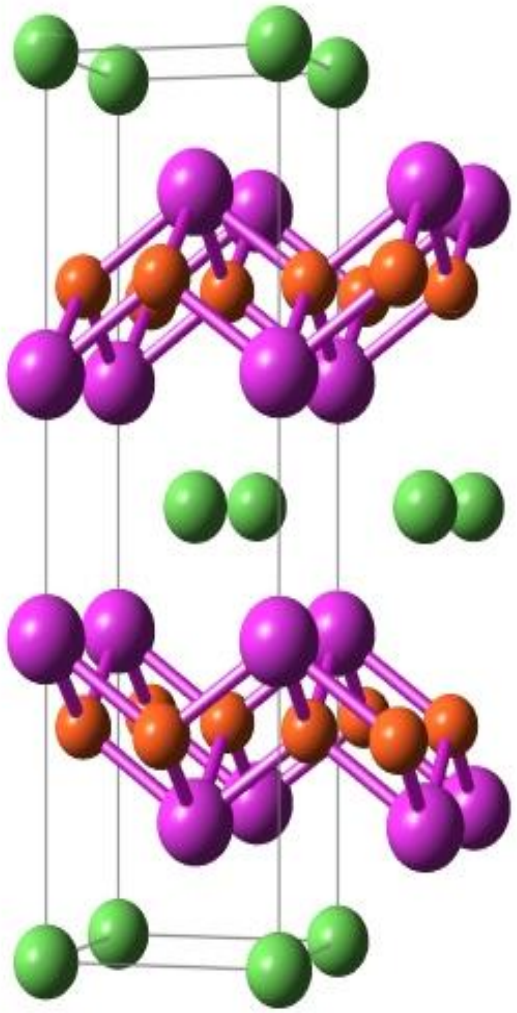
FIG. 3: **Fermi surface of LiFeAs.** $k_z = 0$–plane in the Brillouin zone for 2 Fe atoms; experimental intensity from Ref. 10. Notice how QSGW drastically shrinks the inner pockets at Γ.

Shrinking of pockets!
-- Not seen in DMFT, see work by R. Valenti's group, but contained in GW calculations

LiFeAs – from Tomczak et al., PRL 2013. (ARPES from Borisenko et al.)

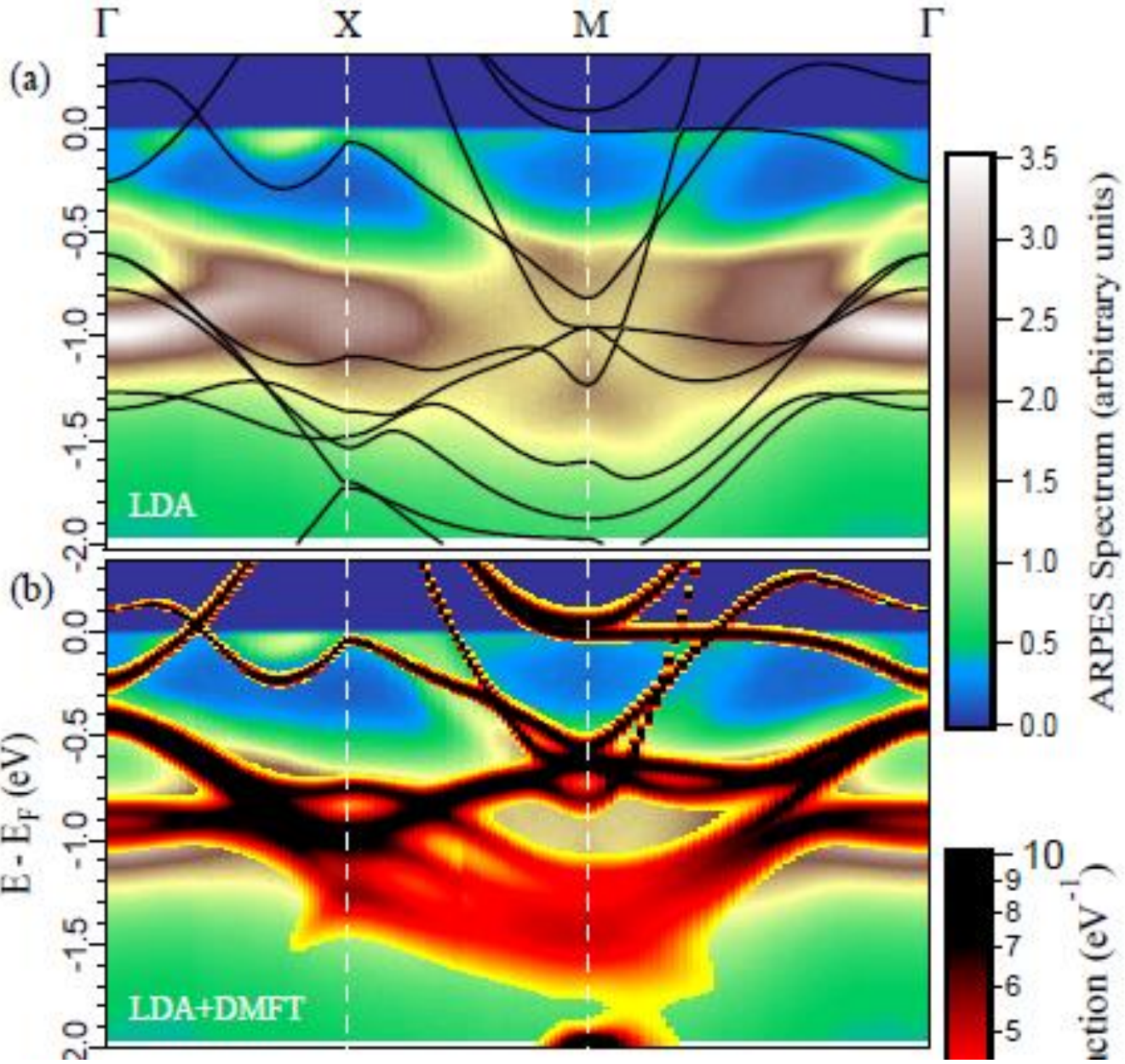
Yet another puzzle …

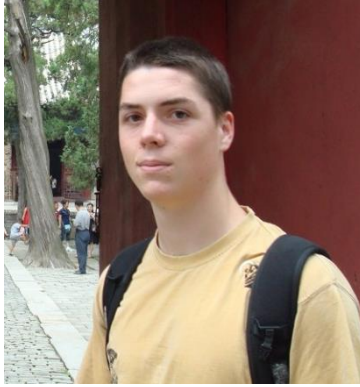
Cobalt Pnictide: BaCo2As2



Fe-d7 configuration
=> weakly correlated

Cobalt Pnictide: BaCo2As2



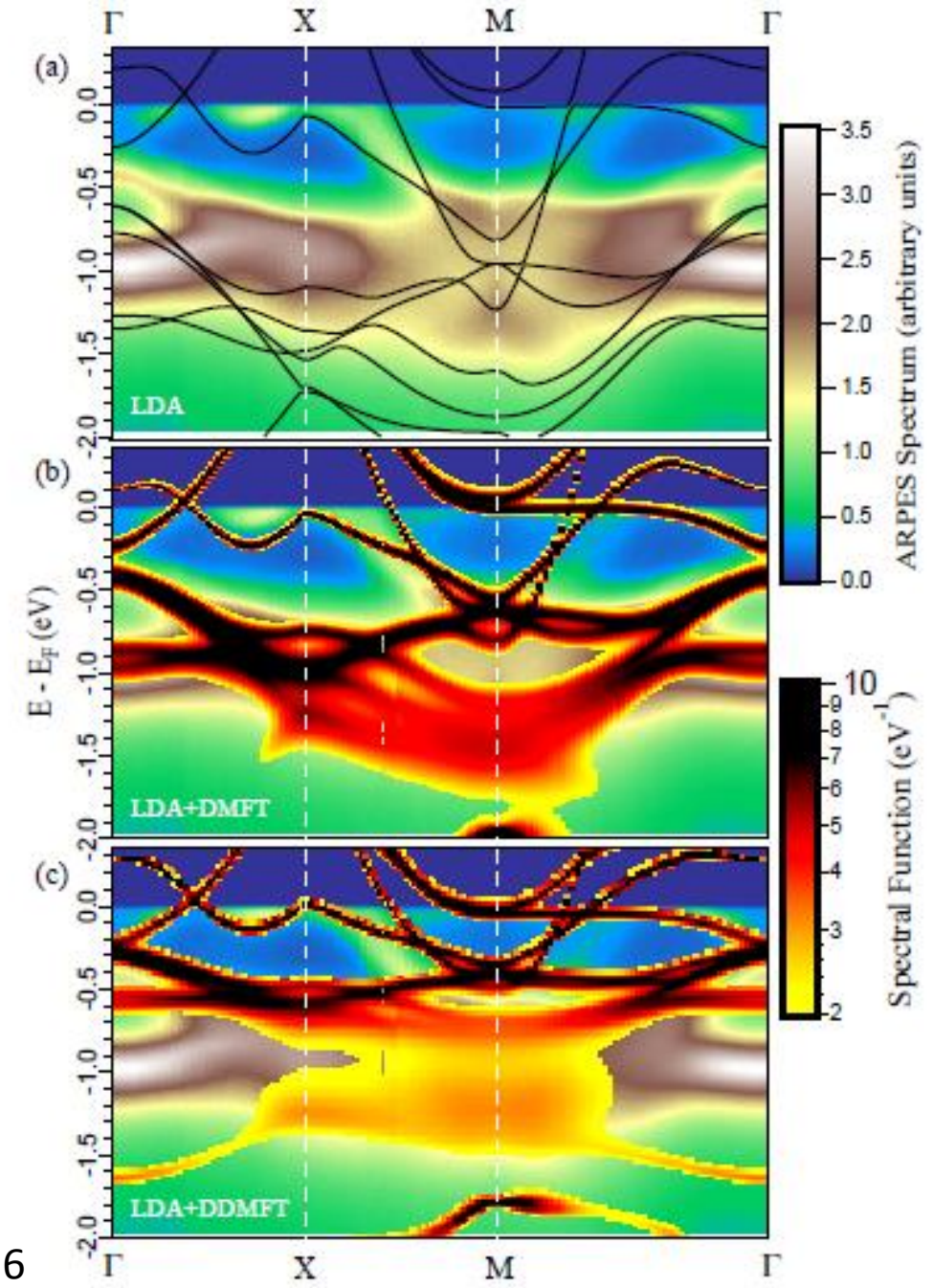


A. Van Roekeghem
IOP-CAS & Ecole Polytechnique

Xu et al., PRX (2013) &
A. van Roekeghem et al., arxiv 14083136

See also:
PES by Dhaka et al.

With dynamical effects ….



A. van Roekeghem et al., arxiv 14083136

Have we worked too much ….?

Have we worked too much ….?

No! Not enough!

Dynamical U(w) …

- … renormalizes one-particle band structure by

$$Z_B = \exp\left(1/\pi \int_0^\infty \mathrm{d}\nu \, \mathrm{Im}U_{\mathrm{ret}}(\nu)/\nu^2\right)$$

Casula, et al., PRL 2012

Dynamical U(w) …

- … renormalizes one-particle band structure
- But: which one?
- No reason to take DFT-LDA …!

Dynamical U(w) …

- … renormalizes one-particle band structure
- But: which one?
- No reason to take DFT-LDA …!
- Lesson to be learnt from “GW+DMFT” :

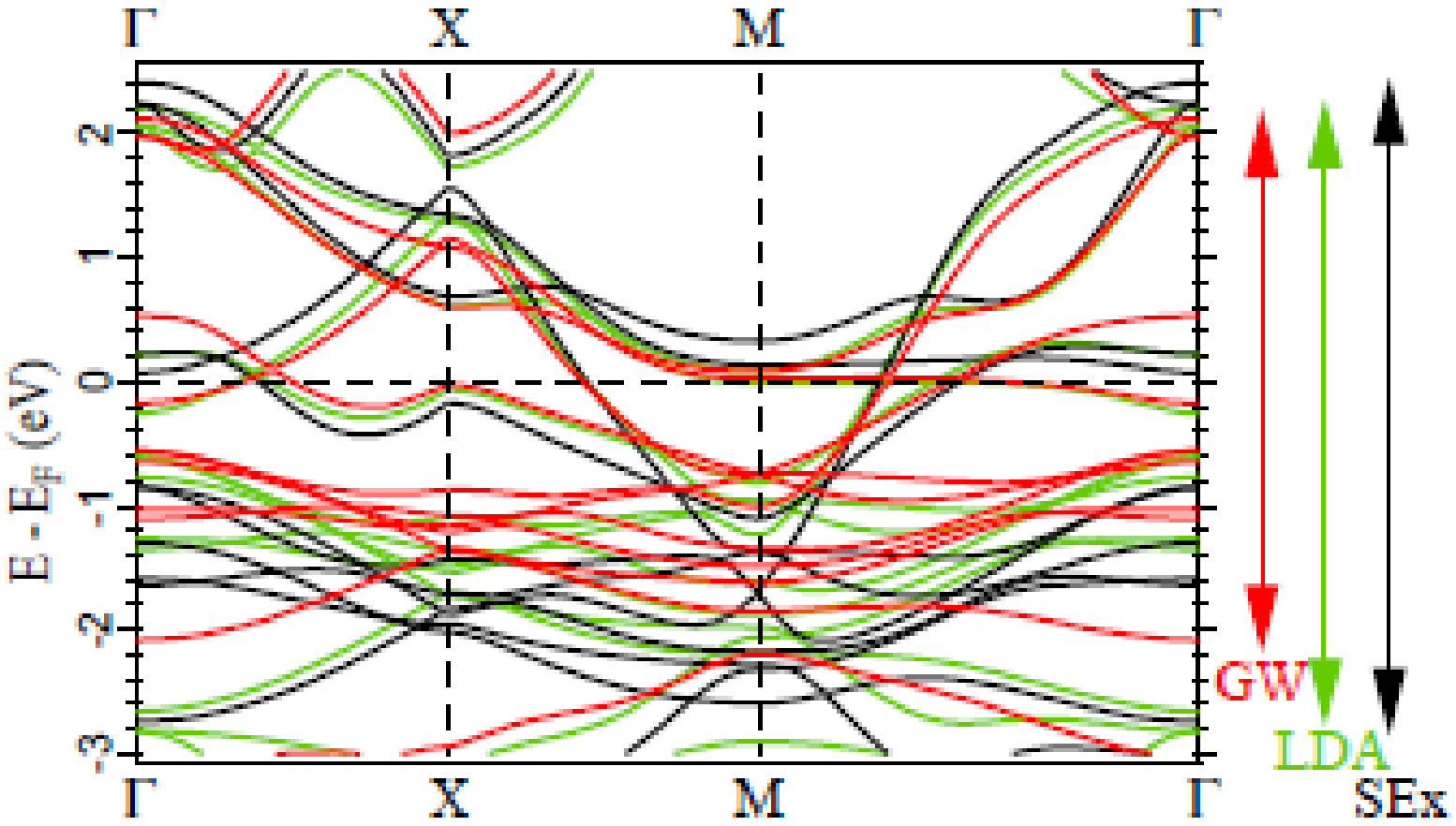
(Biermann, Aryasetiawan, Georges, PRL 2003)

DMFT on top of one-body Hamiltonian incorporating nonlocal effects of many-body perturbation theory (GW approximation)

(Tomczak et al.: “nonlocal GW” to good approximation static!)

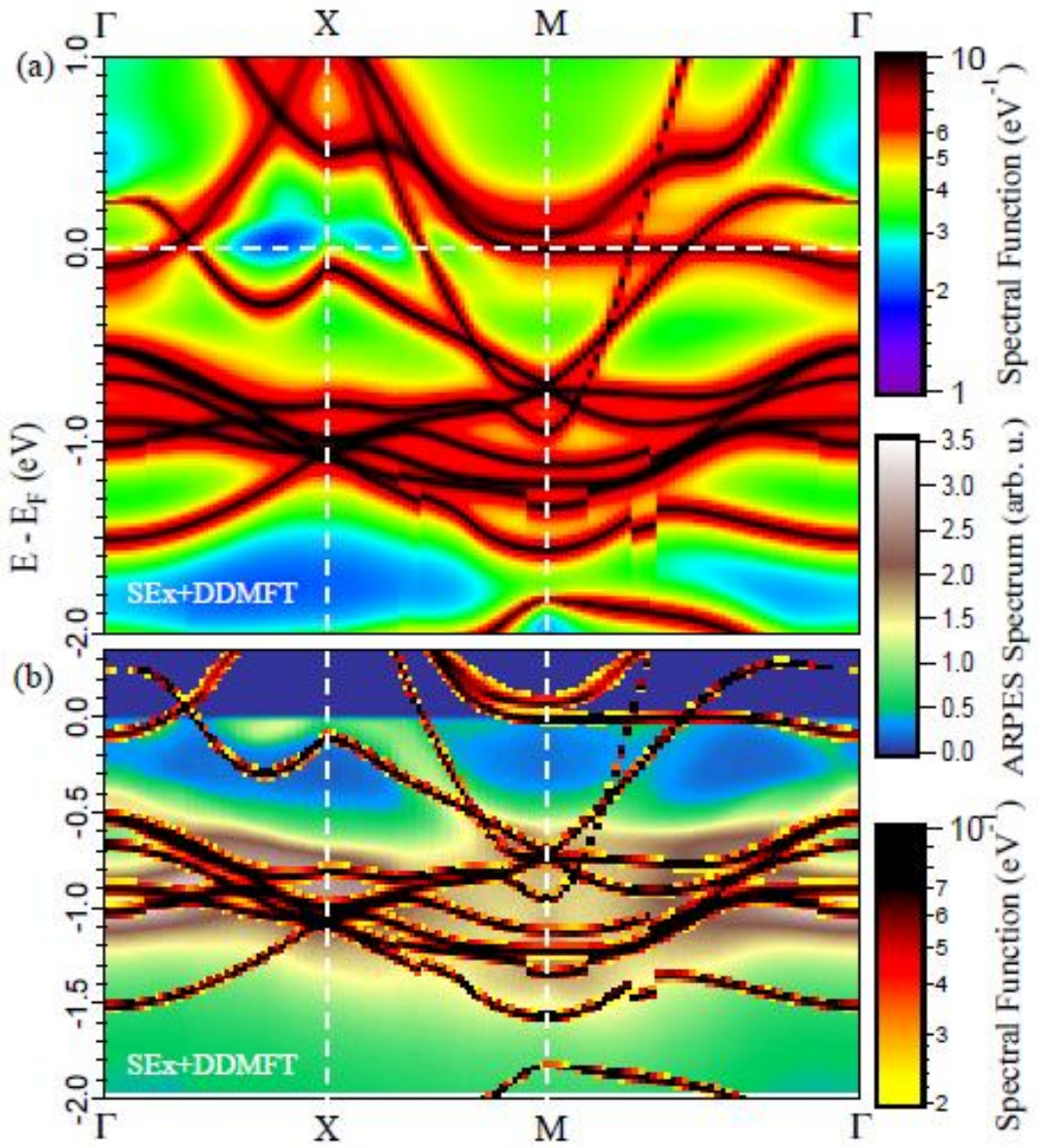
=> Use “screened exchange” as starting point !!

Nonlocal corrections: Screened exchange vs LDA



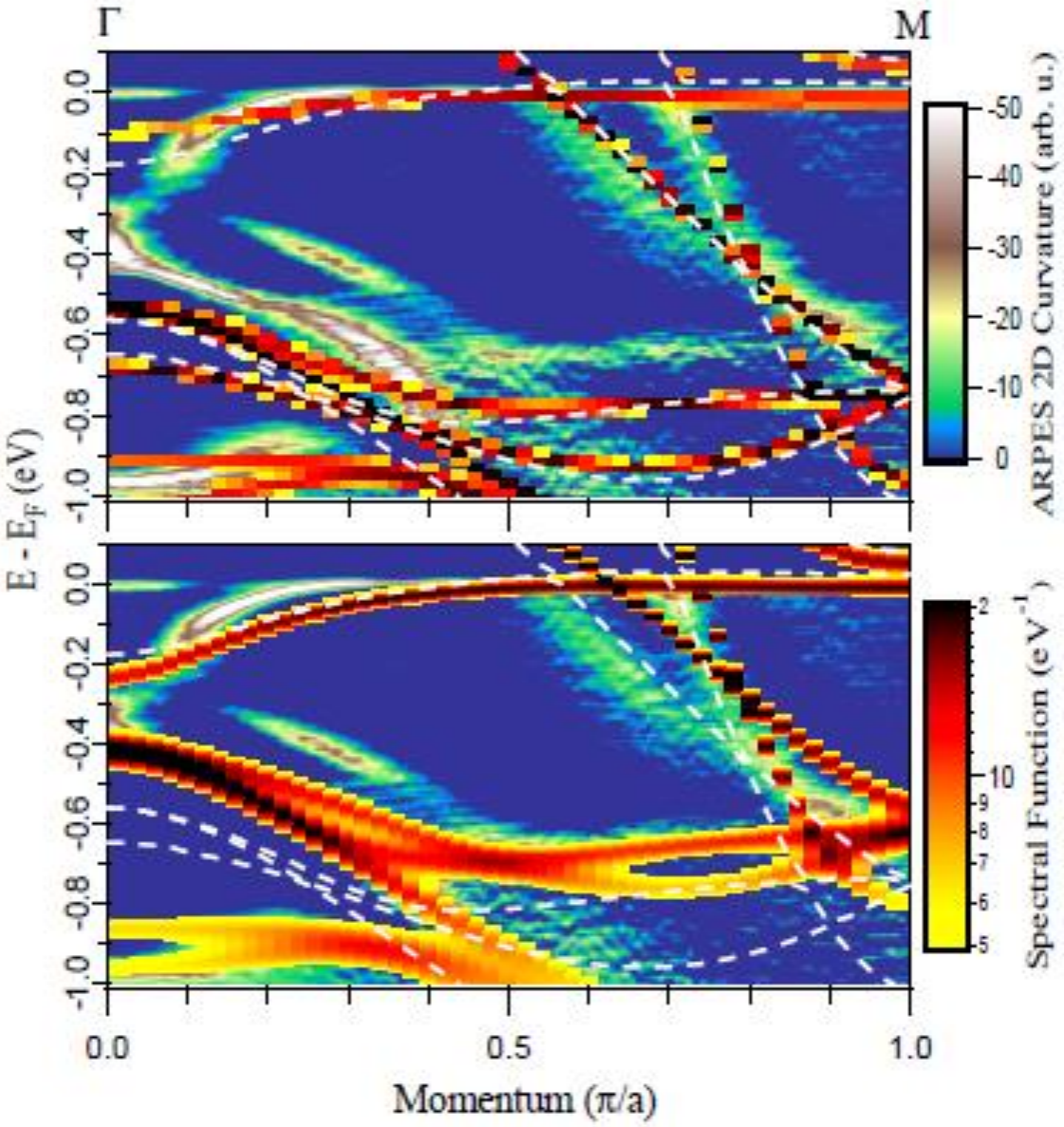
A. van Roekeghem et al., arxiv 14083136, Phys. Rev. Lett. in press

Screened Exchange + DMFT



A. van Roekeghem et al., arxiv 14083136

"Screened exchange+DMFT"
vs.
DFT+DMFT



A. van Roekeghem et al., arxiv 14083136

Side remark: why do DFT bands give a reasonable approximation to single-particle excitations of weakly correlated metals?

- Error cancellation between exchange and correlation ! (well-known for total energies. Here: excitations!)
- Example: n-doped SrTiO3:

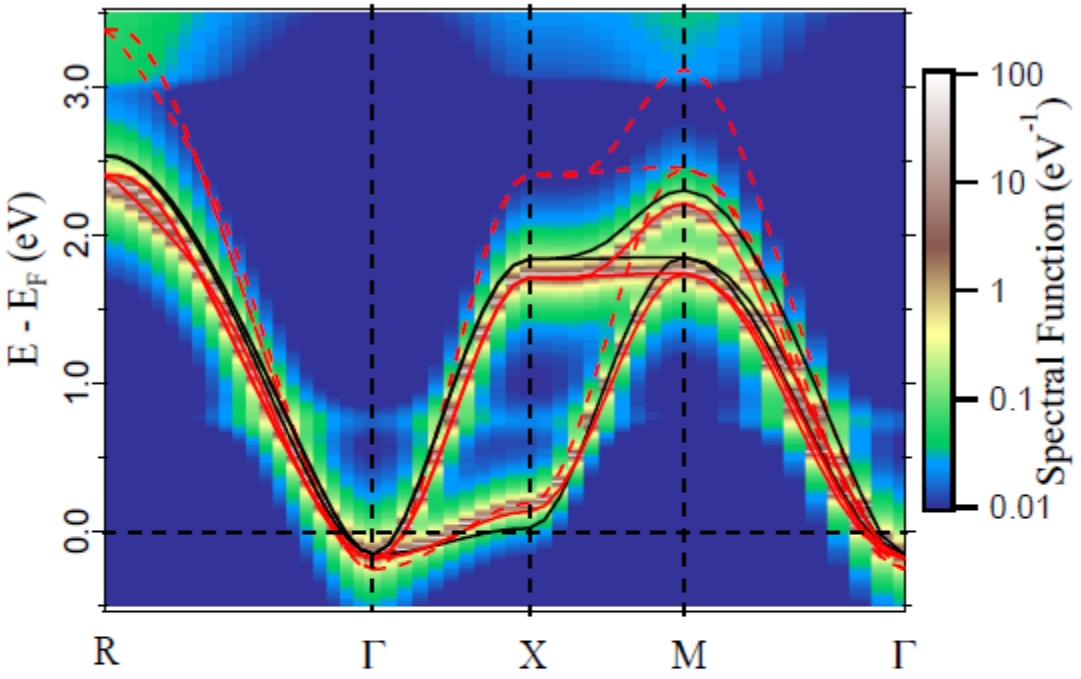


Fig. 4: Band structure of the t_{2g} states of $SrTiO_3$ within LDA (black lines), Screened Exchange (red dashes) and SEx renormalized by a plasmonic factor $Z_B = 0.7$ (red lines) superimposed on the SEx+DDMFT t_{2g} spectral function. The chemical potential corresponds to $n = 0.05$ electron doping per Ti atom, which gives a self-consistent Thomas-Fermi screening-length of $\lambda = 0.6\ a_0^{-1}$ according to the SEx density of states.

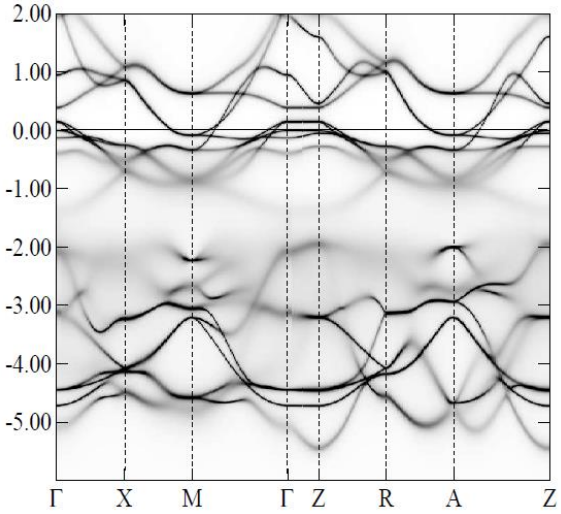
Conclusions

"First principles" calculations of spectral properties of

- **Ceria** — *Tomczak et al., PNAS 2013*
- **Iron pnictides:**
  - **Intermediate correlations in LaFeAsO** — *Aichhorn, et al., PRB 2009*
  - **Hubbard band in FeSe** — *Aichhorn et al., PRB 2010*
  - **Doping-dependent coherence** — *Werner et al., Nature Physics 2012*
  - **Weak correlations in BaCo2As2** — *Xu et al., PRX 2012*
  - **Dynamical screening effects and "Screened Exchange + DMFT"** — *Van Roekeghem et al., arxiv 2014, PRL in press*

Conclusions …

LaFeAsO



Aichhorn et al., PRB (2009)

FeSe: Hubbard band!

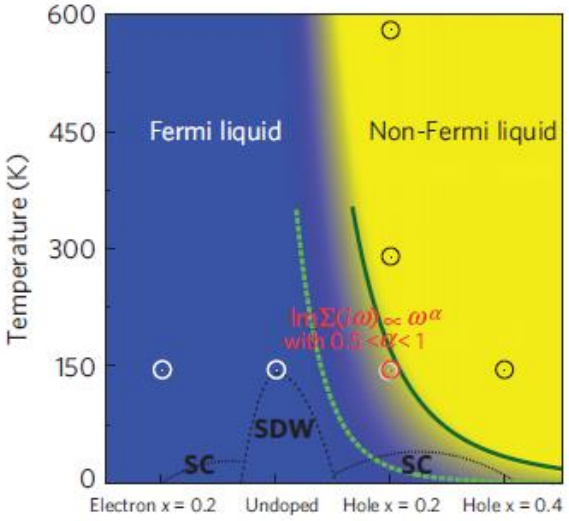
Aichhorn et al., PRB (2010)

f-electron pigments: ceria

(cf. Rhodia's Neolor series)

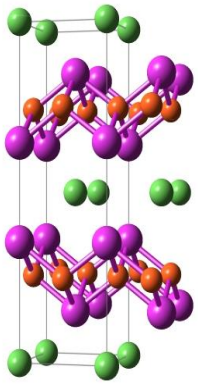
Calculated colour of CeSF:

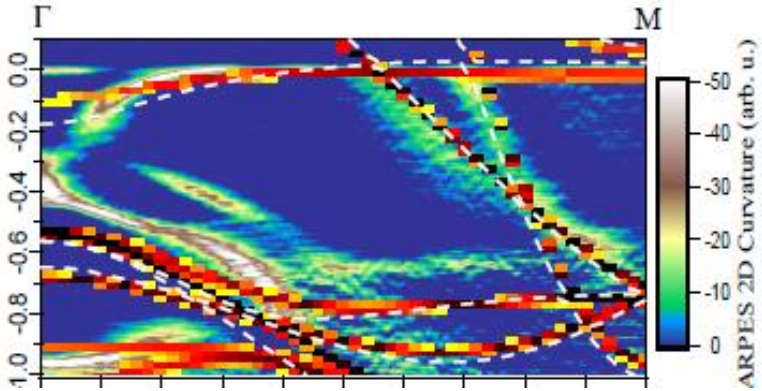
Tomczak et al., PNAS (2013)



Werner et al. (2012)

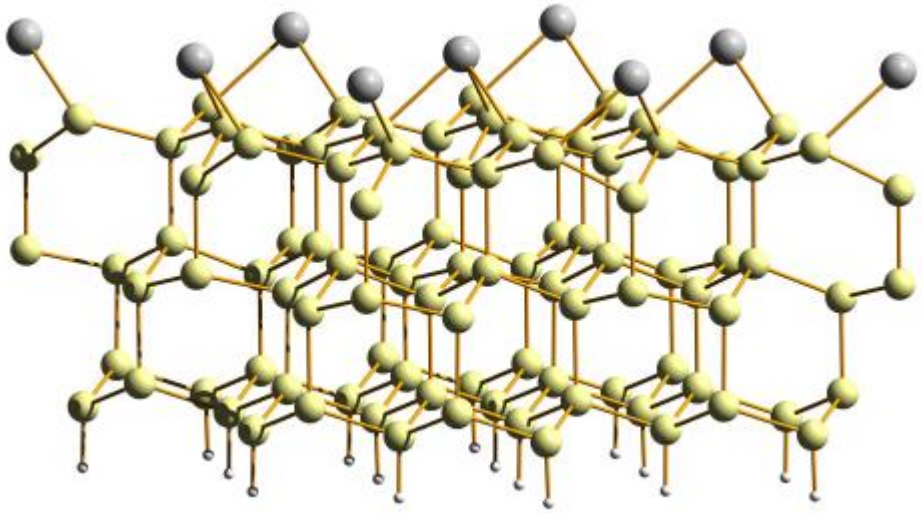
BaCo2As2





Van Roekeghem et al., arxiv2014

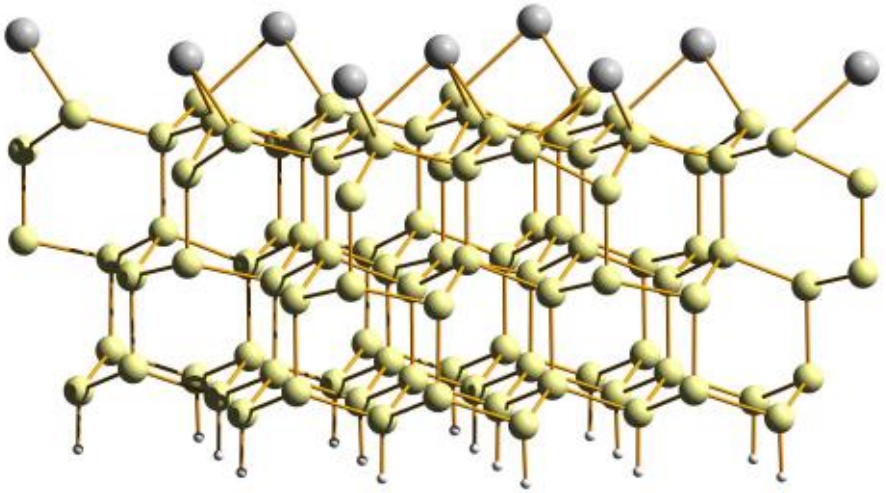
Adatoms on surfaces: Sn/Si(111)
or
How to convert long-range interactions into frequency-dependent ones?

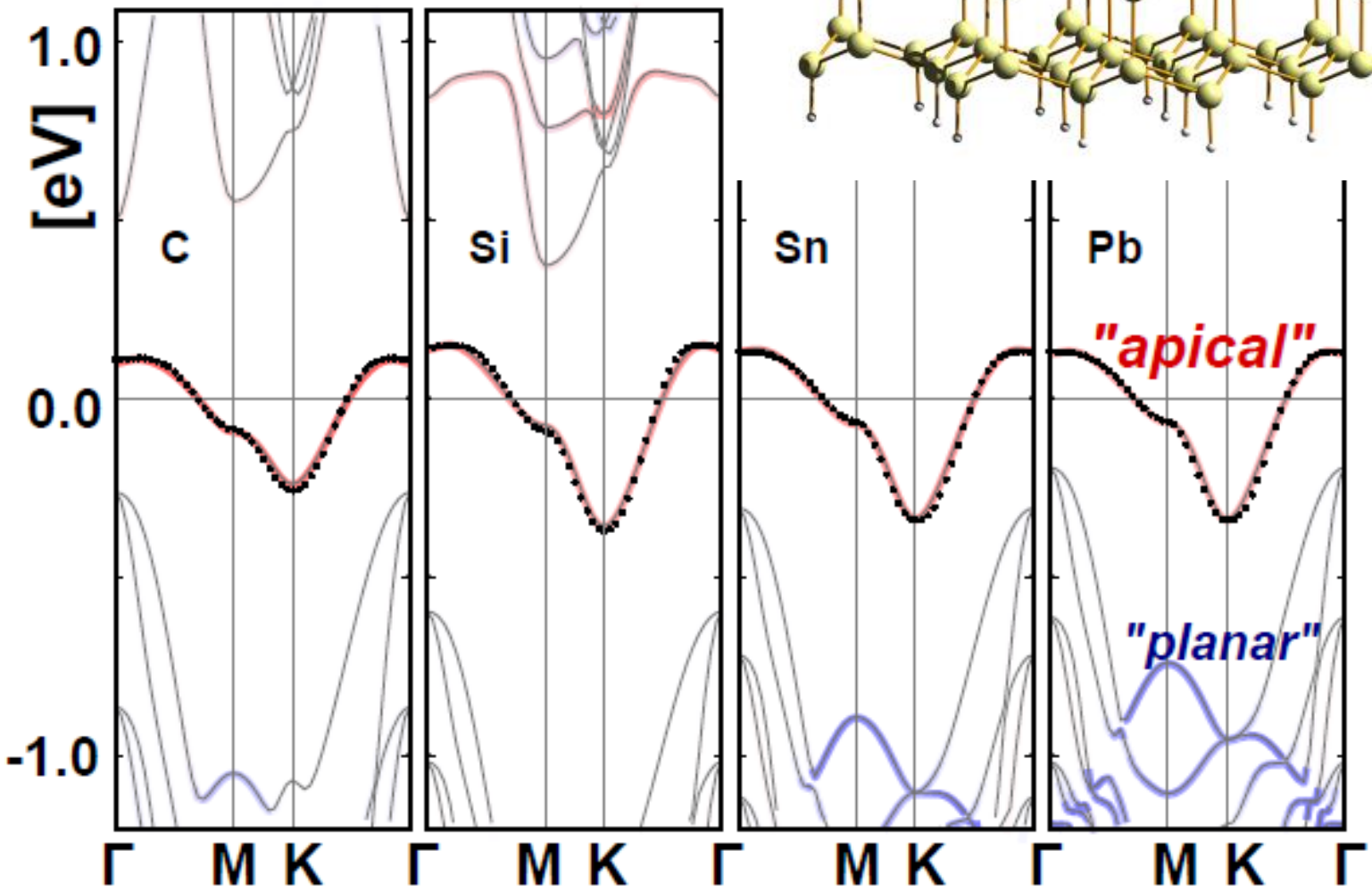


Hansmann et al., Phys. Rev. Lett. 2013 and to be published

See also: 2d extended Hubbard model within Extended DMFT and GW+DMFT:
Ayral, et al., PRL 2012, PRB 2013, Huang et al., PRB 2014

X/Si(111) with X=C, Si, Sn, Pb





P. Hansmann, T. Ayral, L. Vaugier, P. Werner, SB, JPCM 2013 and PRL 2013

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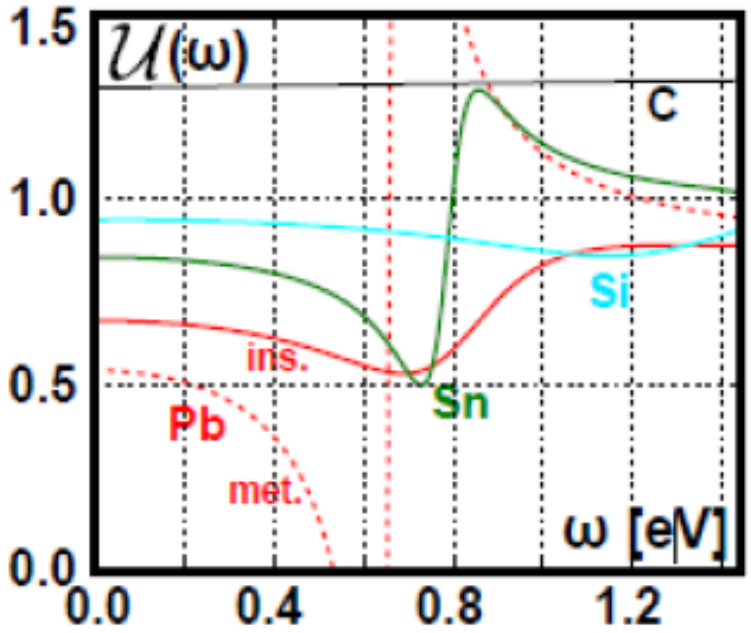

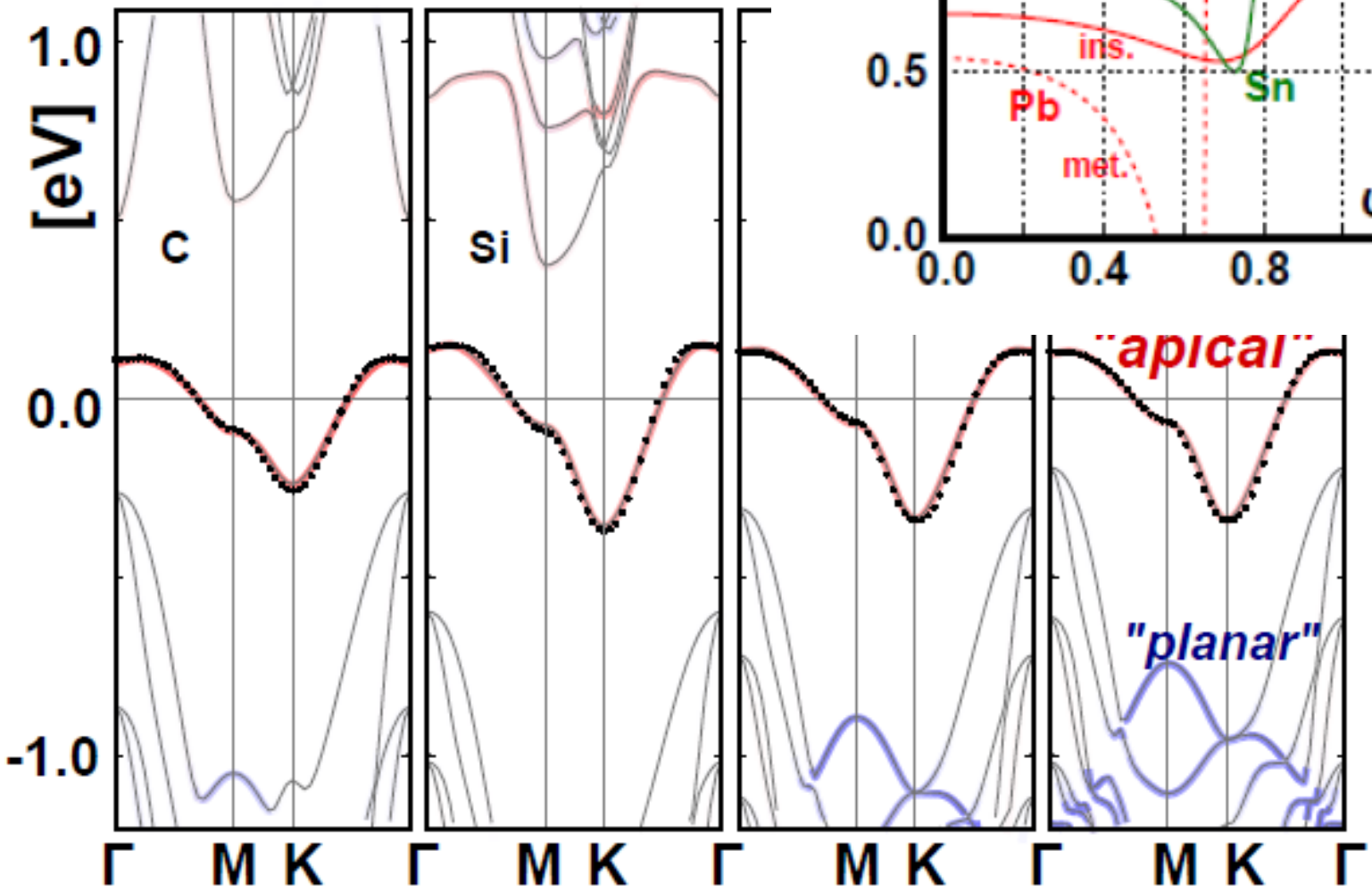

P. Hansmann, T. Ayral, L. Vaugier, P. Werner, SB, JPCM 2013 and PRL 2013

Collaborators and Funding

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LaFeAsO	**M. Aichhorn**, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, A. Georges
FeSe	**M. Aichhorn**, A. Georges, M. Imada
BaFe2As2	**P. Werner**, M. Casula, T. Miyake, F. Aryasetiawan, A. Millis ARPES: Veronique Brouet
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UNIVERSITÉ PARIS-SACLAY

Thank you for your attention !

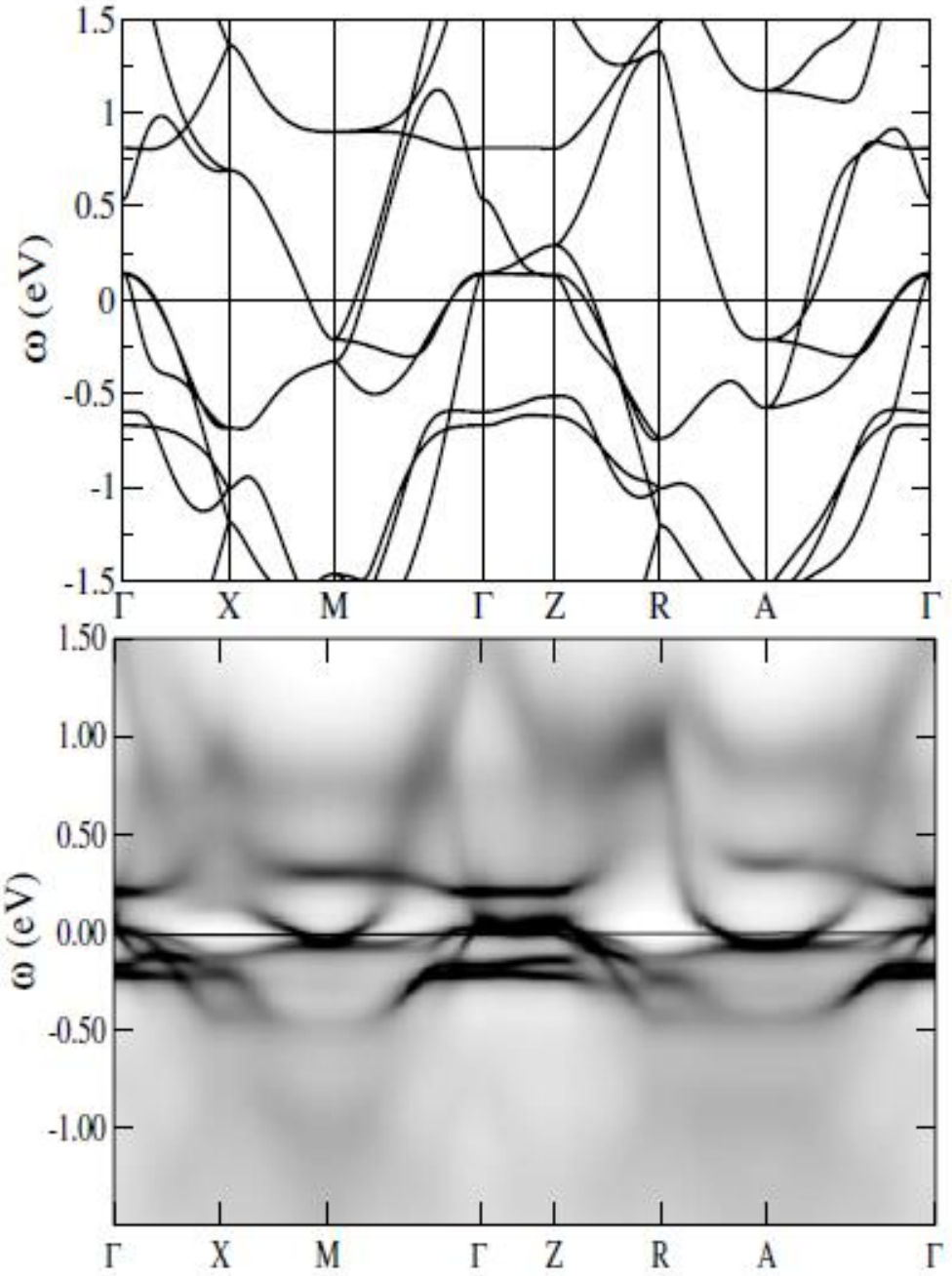
erc

FeSe

LDA

vs.

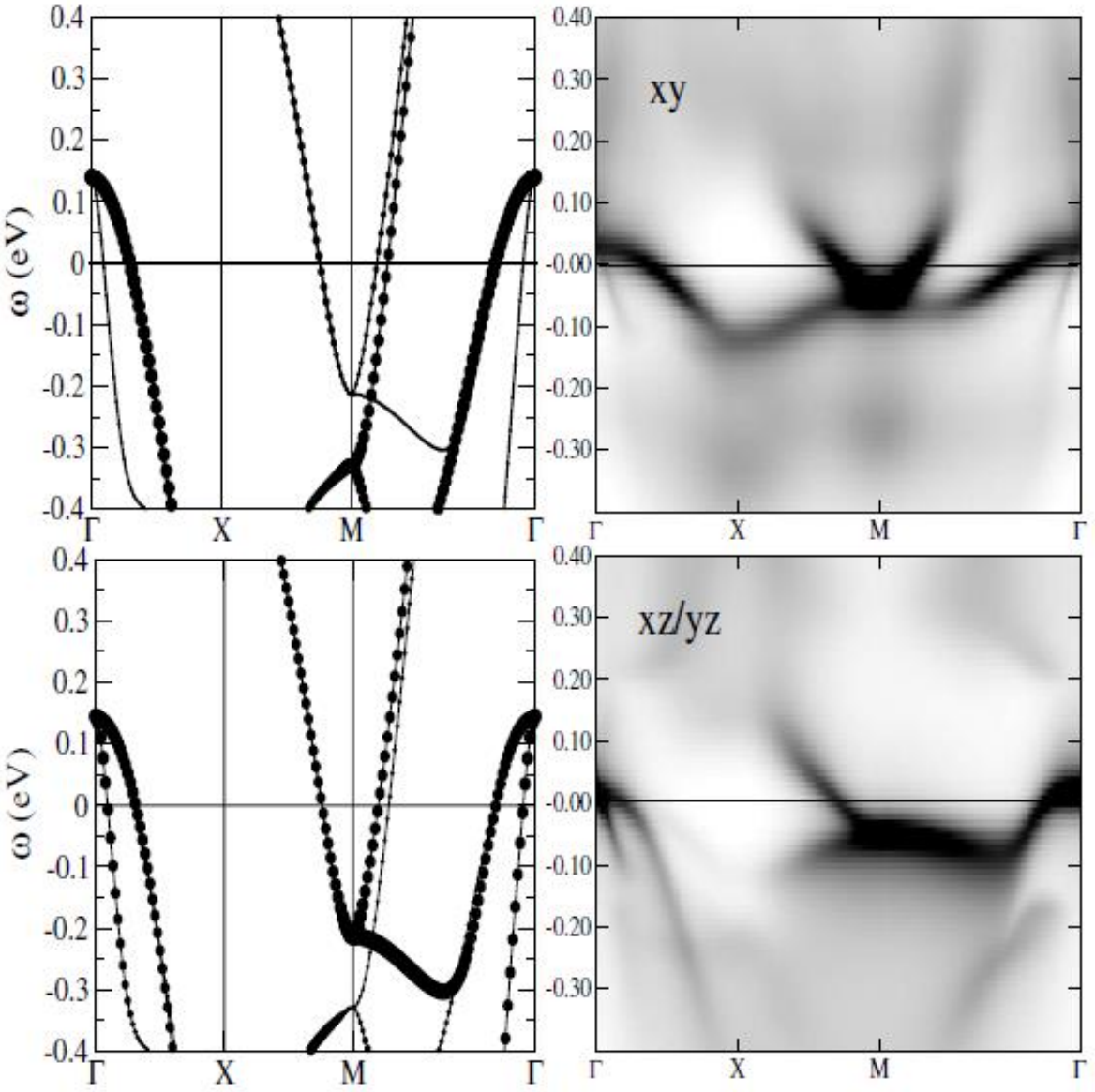
LDA+DMFT



Aichhorn et al., PRB 2010

effective masses: ~ 2 for $x^2 - y^2$, ~ 5 for xy

FeSe



Aichhorn et al., PRB 2010