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LDA'+DMFT scheme:
A way to treat the double counting problem

I.A. Nekrasov, N.S. Pavlov, M.V. Sadovskii

KFe2Se2:	Nekrasov, Sadovskii	JETP Letters	(arXiv:1101.0051)
	Nekrasov, Pavlov, Sadovskii	JETP Letters	(arXiv:1211.3499)
	Nekrasov, Pavlov, Sadovskii	JETP	(arXiv:1304.6030)
LDA'+DMFT:	Nekrasov, Pavlov, Sadovskii	JETP Letters	(arXiv:1204.2361)
	Nekrasov, Pavlov, Sadovskii	JETP	(arXiv:1208.4732)

ES&ES, May 2013, Kiev

Content

- The LDA+DMFT double counting problem
- Formulation of consistent LDA'+DMFT approach

$K_{1-x}Fe_{2-y}Se_2$:LDA+DMFT and LDA'+DMFT results

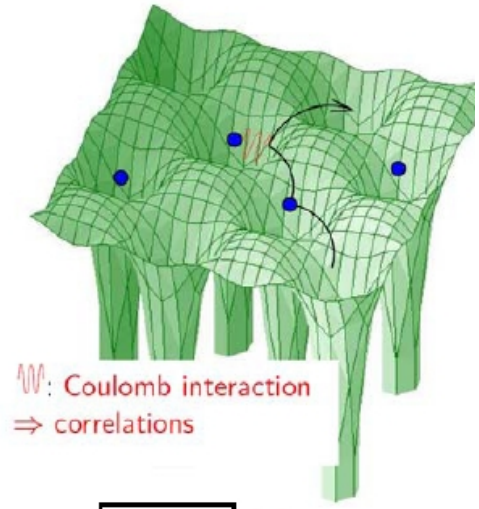
a. Comparison of LDA and LDA' band structures

b. Densities of states and spectral functions

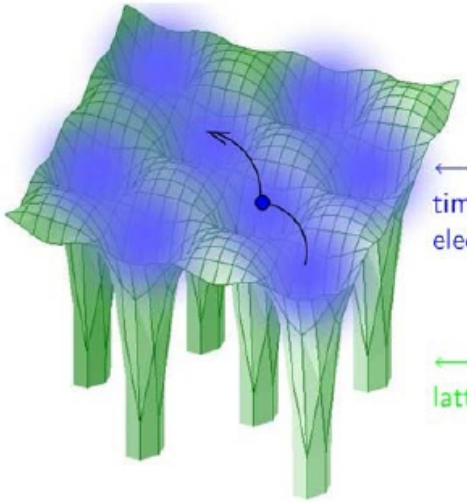
c. Comparison with ARPES data

- Conclusions

Main idea of the LDA approximation



LDA



Hohenberg-Kohn theorem

$$E[\rho] = T_{kin}[\rho] + E_{ext}[\rho] + E_{ion}[\rho] + E_{Hartree}[\rho] + E_{xc}[\rho]$$

$(\delta E[\rho]_{\rho=\rho_0(\vec{r})} = 0$ — At the ground state

Kohn-Sham equations

$$\left[-\nabla^2 + V_{one-electron}(\vec{r}) + V_{xc}(\vec{r})\right]\varphi_i = \varepsilon_i\varphi_i,\ \ \rho(\vec{r}) = \sum_{i=1}^{N}|\varphi_i(\vec{r})|^2$$

Local (Spin) Density Approximation

$$V_{xc}^{uniform}[\rho(\vec{r})] := V_{xc}^{uniform}[\rho^{real}(\vec{r})];\ \ \rho^{real}(\vec{r}) = \sum_{i=1}^{N}|\varphi_i^{real}(\vec{r})|^2$$

Main disadvantage – LDA has simplified electron-electron interaction.

Main advantage – LDA allows to describe kinetic part of the Hubbard Hamiltonian and to calculate interaction parameters.

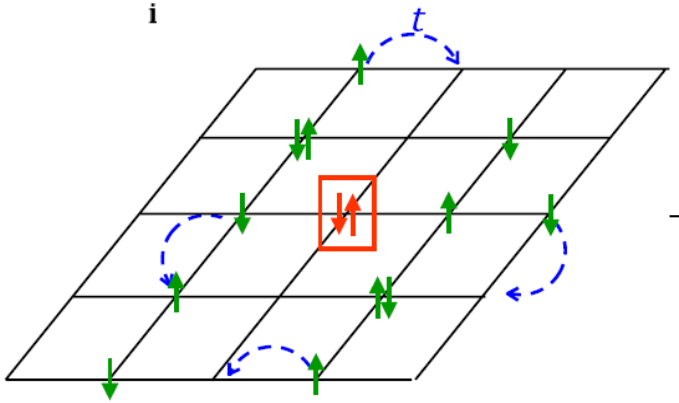
Main idea of DMFT

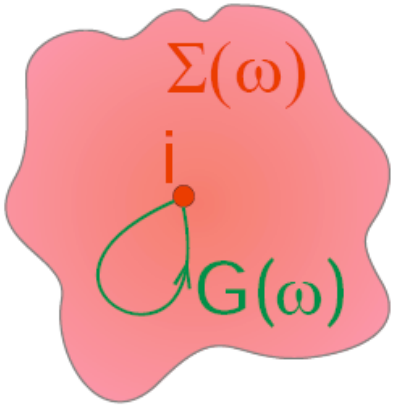
Hubbard model $\xrightarrow{Z \to \infty}$ Effective Anderson Single Impurity Model

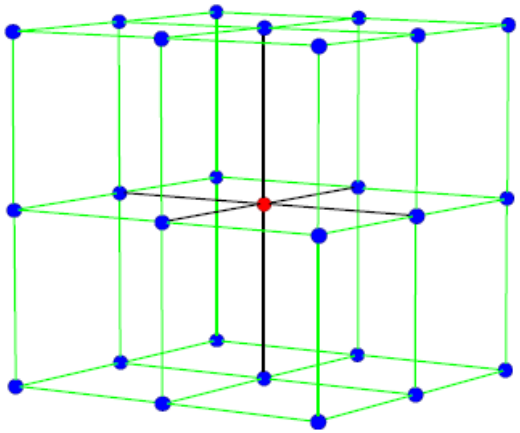
$$H = \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} n_{\mathbf{k}\sigma} + U \sum_{\mathbf{i}} n_{\mathbf{i}\uparrow} n_{\mathbf{i}\downarrow}$$

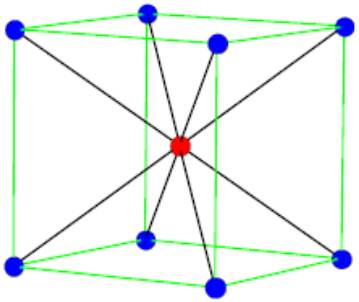
Metzner&Vollhardt'89

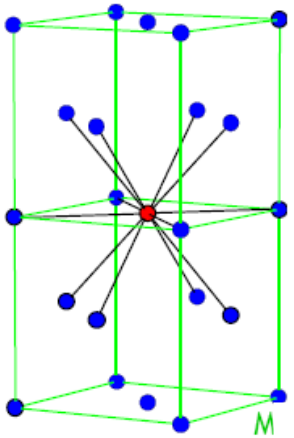
W/U~1









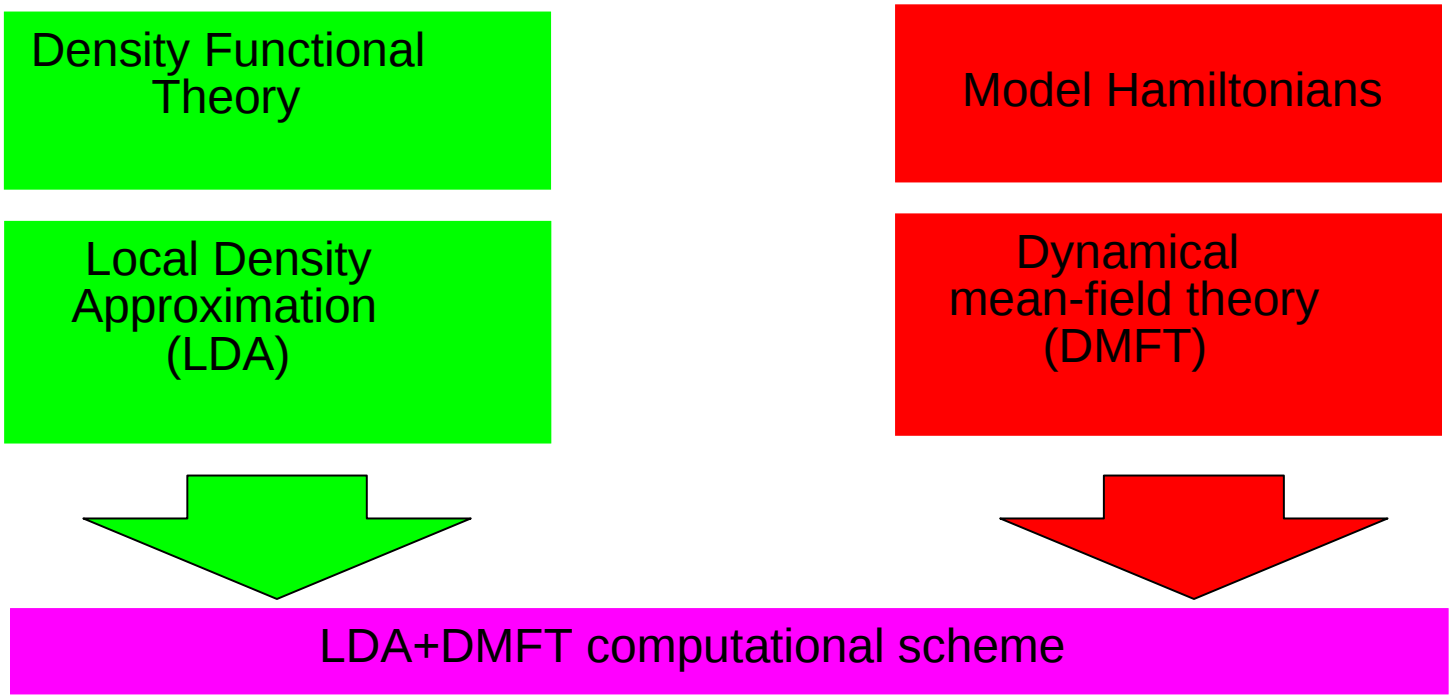


H_{dc} is internal parameter of LDA+DMFT

LDA contains two contributions to local electron-electron interaction:

1. Hartree term;
2. Local exchange-correlation energy of the uniform electron gas

DMFT contains full Hubbard (local) interaction.



Anisimov, Poteryaev, Korotin, Anokhin, Kotliar'97

$$\hat{H}_{LDA+correl} = \hat{H}_{LDA}\{t_{ilm,jl'm'}, \varepsilon_{ilm}\} + \hat{H}_{correl}\{U^{\sigma\sigma'}_{mm'}, J_{mm'}\} - \hat{H}_{DC}$$

The LDA+DMFT double counting problem

The LDA+DMFT double counting problem arises since there is no direct microscopic or diagrammatic relation between LDA and the Hubbard model.

- No unique definition of the H_{dc} term.
- Several *ad hoc* definitions of double counting term were proposed.
- Some of them works only for particular compounds.
- Additional free parameter in LDA+DMFT in a sense of choosing either H_{dc} term form or simply a number or even it is not mentioned at all.

Different types of double counting treatment

If we take Hubbard interaction in the form:

$$\hat{H}^{Hub} = U \sum_m \sum_i \hat{n}_{im\uparrow}\hat{n}_{im\downarrow} + \sum_i \sum_{m \neq m'} \sum_{\sigma\sigma'} \left(U' - \delta_{\sigma\sigma'} J\right) \hat{n}_{im\sigma}\hat{n}_{im'\sigma'}$$

Within LDA+U method there was postulated “around mean-field approach” which assumes that LDA is some kind of Hubbard model mean-field solution

$$\hat{H}^{DC}_{AMF} = \frac{1}{2} U \sum_\sigma n_{d\sigma}(n_d - n^0) - \frac{1}{2} J \sum_\sigma n_{d\sigma}(n_{d\sigma} - n^0_\sigma)$$

with the average occupancies $n^0 = \frac{1}{2(2l+1)} \sum_{m,\sigma} n_{m\sigma}$, $n^0_\sigma = \frac{1}{(2l+1)} \sum_m n_{m\sigma}$ and total number of electrons on interacting orbitals (per spin projection) $n_{d\sigma} = \sum_m n_{il_dm\sigma} = \sum_m \langle \hat{n}_{il_dm\sigma} \rangle$ and $n_d = \sum_\sigma n_{d\sigma}$.

V.I. Anisimov, J. Zaanen and O.K. Andersen,Phys. Rev. B **44**, 943 (1991); V. I. Anisimov, F. Aryasetiawan, and A. I. Lichtenstein, J. Phys. Cond. Matter **9**, 767 (1997).

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One can do the Hartree decoupling for this Hamiltonian then we get so called fully localized limit expression

$$\hat{H}^{DC}_{FLL} = \frac{1}{2} U n_d (n_d - 1) - \frac{1}{2} J \sum_{\sigma} n_{d\sigma} (n_{d\sigma} - 1)$$

M.T.Czyżyk and G.A.Sawatzky, Phys. Rev. B, **49**, 14211 (1994).

Different types of double counting treatment

Alternative way to derive or guess the $\hat{H}^{DC}$ term is to express it through the characteristics of intrinsic single DMFT impurity problem, such as impurity self-energy $\Sigma^{imp}_{mm'}$ or impurity Green's function $G^{imp}_{mm'}$. A popular way is to define double counting energy as a static part of the impurity self-energy [21]:

$$E_{dc} = \frac{1}{2}\mathrm{Tr}_\sigma(\Sigma^{imp}_\sigma(0)). \qquad (9)$$

Some of LDA+DMFT papers used this definition in calculations of metallic magnetic and non-magnetic systems.

[21] A.I. Lichtenstein, M.I. Katsnelson, G. Kotliar, Phys. Rev. Lett. **87**, 067205 (2001); M.I. Katsnelson, A.I. Lichtenstein, Eur. Phys. J. B **30**, 9 (2002).

Different types of double counting treatment

Hartree energy can be determined from LDA+DMFT self-energy as its real part in the high frequency limit value. In Ref. [25] it was proposed to use thus defined Hartree energy as a double counting correction, using the constraint

$$\mathrm{ReTr}(\Sigma^{imp}_{mm'}(i\omega_N)) = 0, \qquad (10)$$

where ω_N is the highest Matsubara frequency (used in calculations). Physically similar definition of double counting term $E_{dc} = \Sigma(\omega \to \infty)$ was successfully applied to metallic ferromagnetic $SrCoO_3$[23].

[25] M. Karolak, G. Ulm, T. Wehling, V. Mazurenko, A. Poteryaev, A. Lichtenstein, Journal of Electron Spectroscopy and Related Phenomena, Volume **181**, 11 (2010).

[23] J. Kunes, V. Krapek, A.V. Kozhevnikov, arXiv:1202.0110.

Different types of double counting treatment

For metallic systems it was suggested to fix the double counting correction by equating the number of particles of non-interacting problem and impurity problem as expressed via corresponding Green's function [24]:

$$\mathrm{Tr}\ G^{imp}_{mm'}(\beta) = \mathrm{Tr}\ G^{0,loc}_{mm'}(\beta), \qquad (11)$$

where $G^{0,loc}_{mm'}$ is local non interacting Green function.

Some of LDA+DMFT works treated double counting energy E_{dc} as a free parameter. The authors of Ref. [25] found that most of described $\hat{H}^{DC}$ terms proposed in the literature are not completely satisfactory in the case of charge transfer insulator NiO and proposed a *numerical* way to define the necessary double counting correction.

[25] M. Karolak, G. Ulm, T. Wehling, V. Mazurenko, A. Poteryaev, A. Lichtenstein, Journal of Electron Spectroscopy and Related Phenomena, Volume **181**, 11 (2010).

Main Idea of LDA' approach

The LDA Hamiltonian is expressed via local charge density:

$$\hat{H}_{\mathrm{LDA}} = -\frac{\hbar^2}{2m_e}\Delta + V_{\mathrm{ion}}(\mathbf{r}) + \int d^3r' \rho(\mathbf{r}')\, V_{ee}(\mathbf{r}-\mathbf{r}') + V_{\mathrm{xc}}^{\mathrm{LDA}}(\rho(\mathbf{r}))$$

$$\rho(\mathbf{r}) = \sum_{i=1}^{N} |\varphi_i(\mathbf{r})|^2$$

It seems somehow inconsistent to use LDA to describe correlation effects in narrow (strongly cor- related) bands from the very beginning, as these should be treated via more elaborate schemes like DMFT. To overcome this difficulty for these states, we propose to redefine charge density as follows:

$$\rho'(\mathbf{r}) = \sum_{i \neq i_d} |\varphi_i(\mathbf{r})|^2$$

$$\hat{H}_{\mathrm{LDA}'} = -\frac{\hbar^2}{2m_e}\Delta + V_{\mathrm{ion}}(\mathbf{r}) + \int d^3r' \rho(\mathbf{r}')\, V_{ee}(\mathbf{r}-\mathbf{r}') + V_{\mathrm{xc}}^{\mathrm{LDA}}(\rho'(\mathbf{r}))$$

The DC term should be consistently taken in the Hartree form

$$H^{DC} = \frac{1}{2}Un_d(n_d-1) - \frac{1}{2}J\sum_{\sigma} n_{d\sigma}(n_{d\sigma}-1).$$

Consistent LDA'+DMFT calculations

In our work we used:

1) LMTO(ASA) to get LDA band structure, with Von Barth-Hedin exchange-correlation potential

2) LDA Hamiltonian contains all valence states (no “projections”)

3) HF-QMC to solve five orbital DMFT equations

KFe2Se2 vs. BaFe2Se2

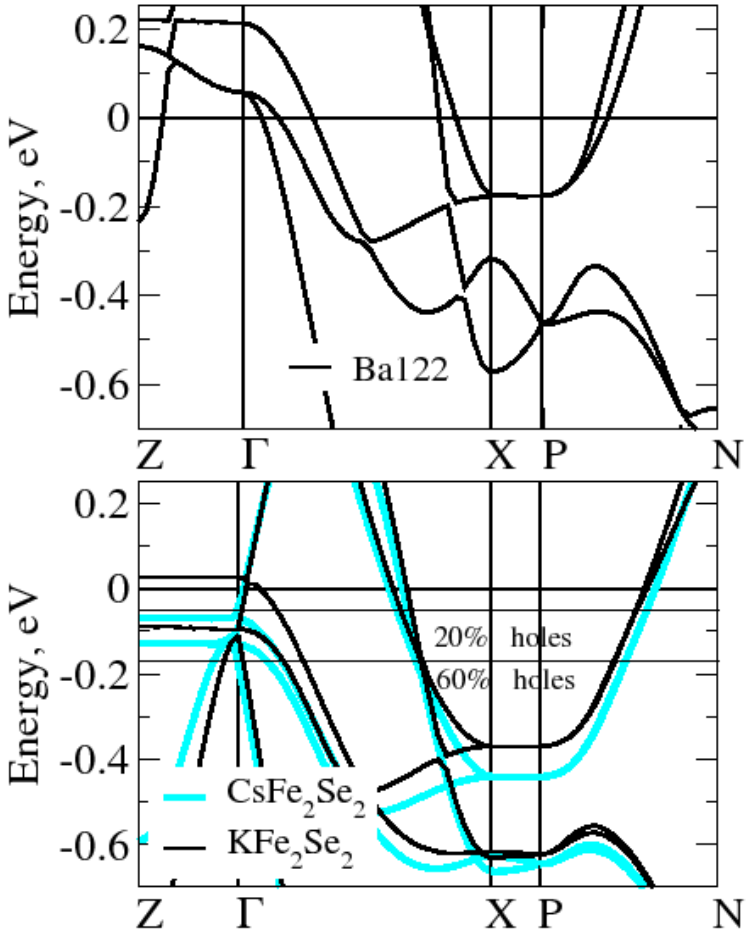


Fig. 2. Top panel – LDA calculated band dispersions in the vicinity of the Fermi level for Ba122; Bottom panel – $K_xFe_2Se_2$ (black lines) and $Cs_xFe-2Se_2$ (gray lines). The Fermi level is at zero energy. Additional horizontal lines correspond to Fermi level position for the case of 20% and 60% hole doping.

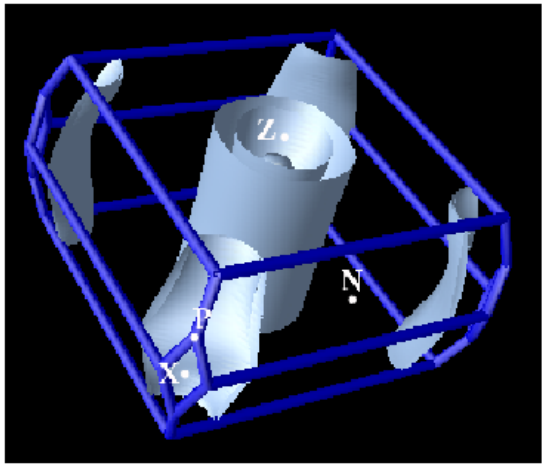
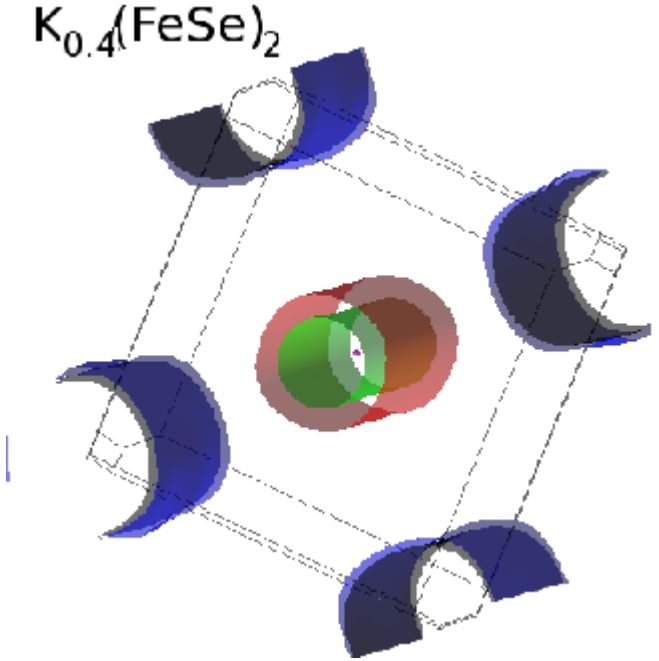
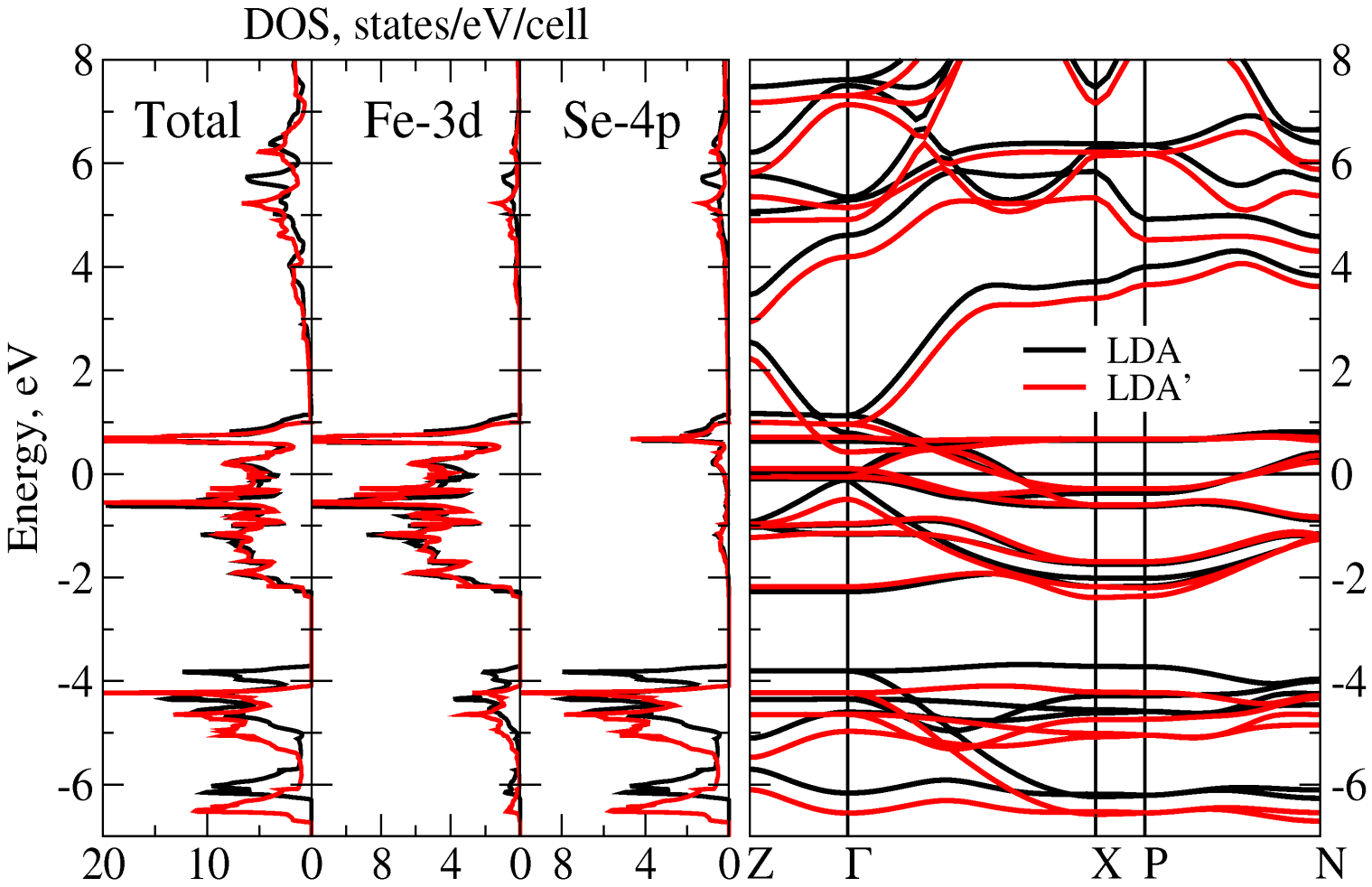


Fig. 6. Fermi surface of $BaFe_2As_2$ shown in the first Brillouin zone centered at Γ point.

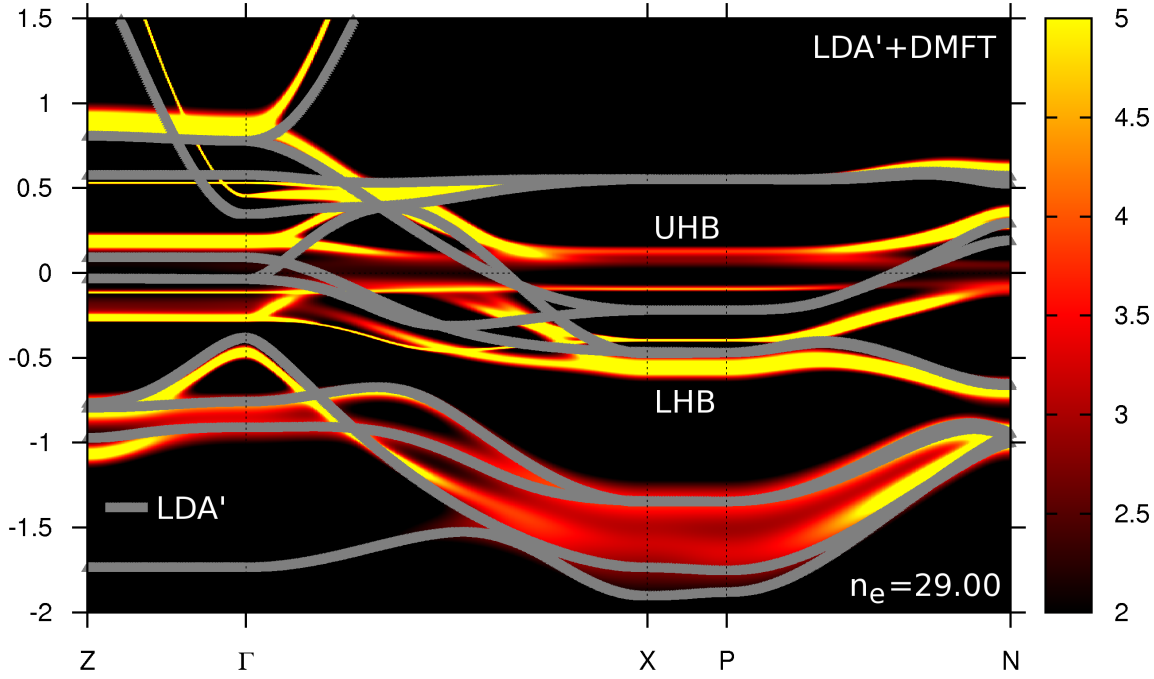


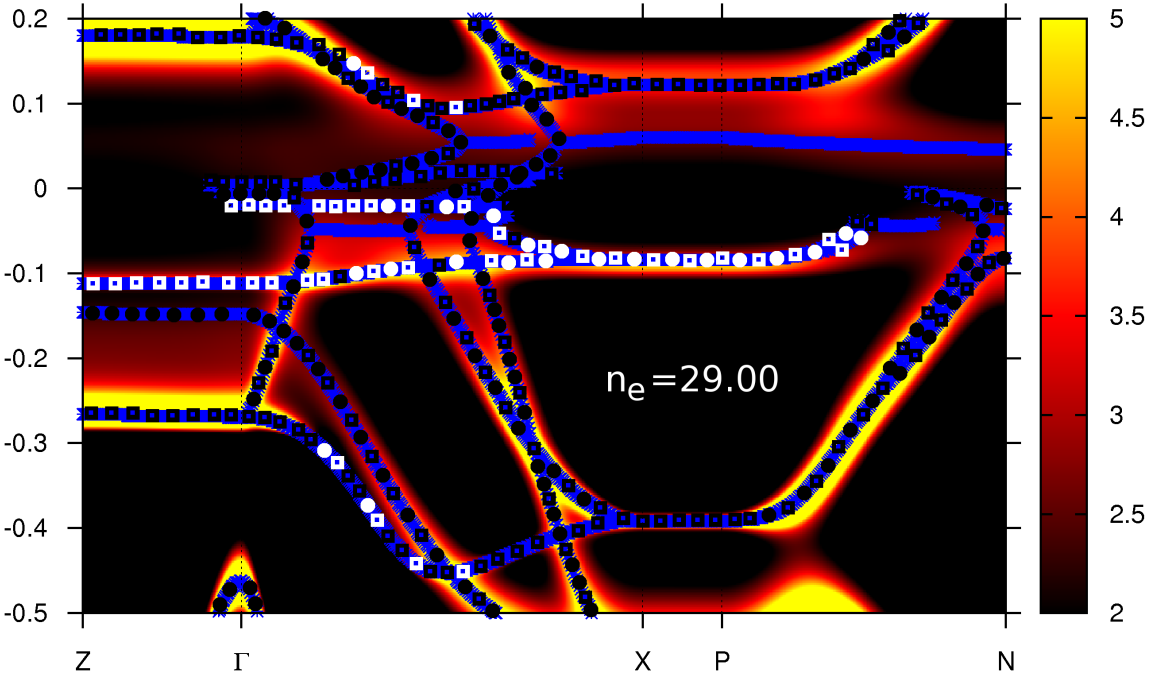
Comparison of LDA and LDA' results for stoichiometric KFe2Se2



Because of more repulsive potential in LDA' splitting between O-2p and Me-3d increases, however band shape stays almost the same. Since Me-3d states are pinned to the Fermi level O-2p states go down.

Comparison of LDA+DMFT and LDA'+DMFT results

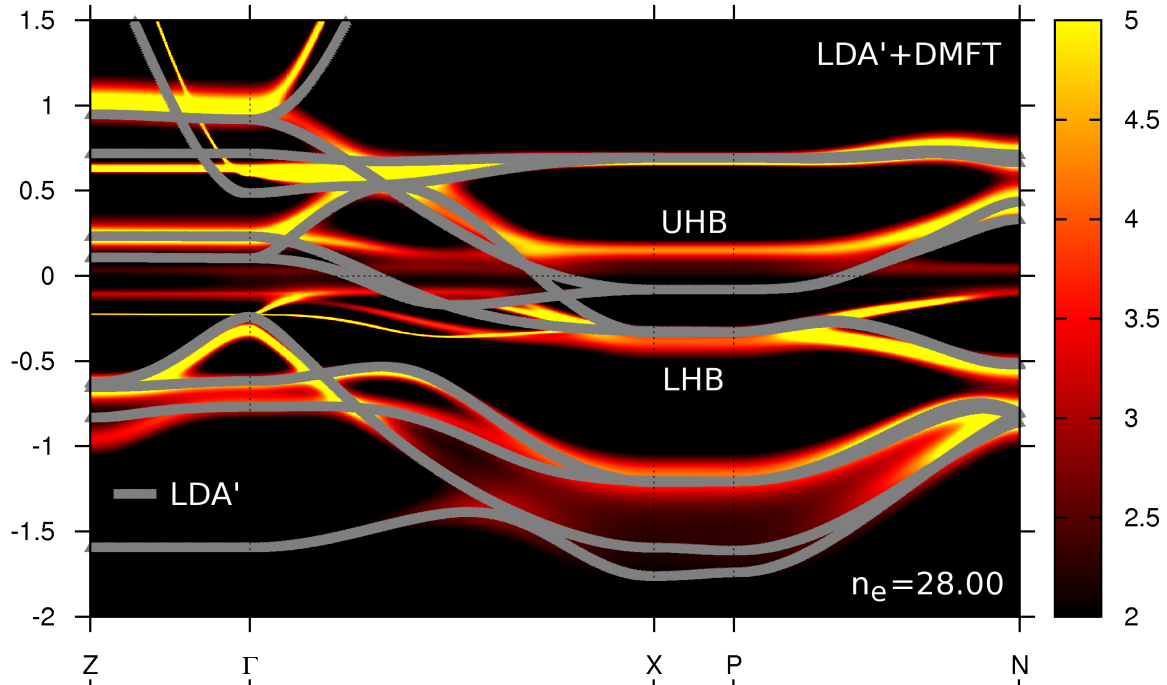


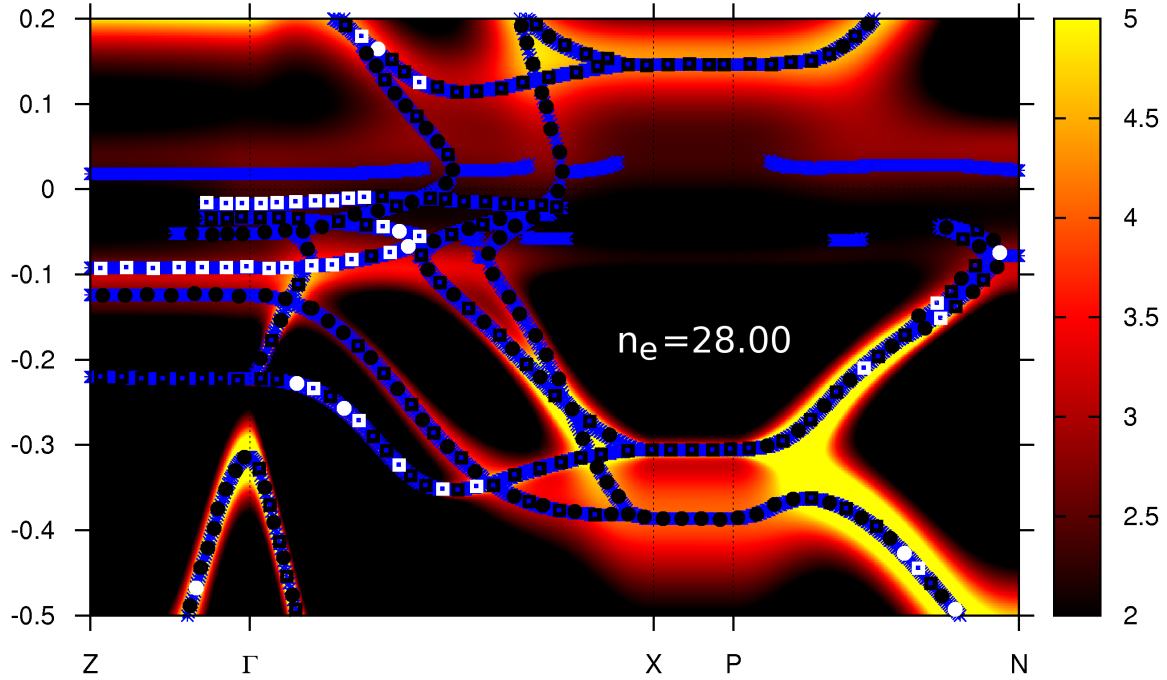


● – xy , □ – xz,yz

● – $3z^2$-r^2 □ –x^2-y^2

Comparison of LDA+DMFT and LDA'+DMFT results





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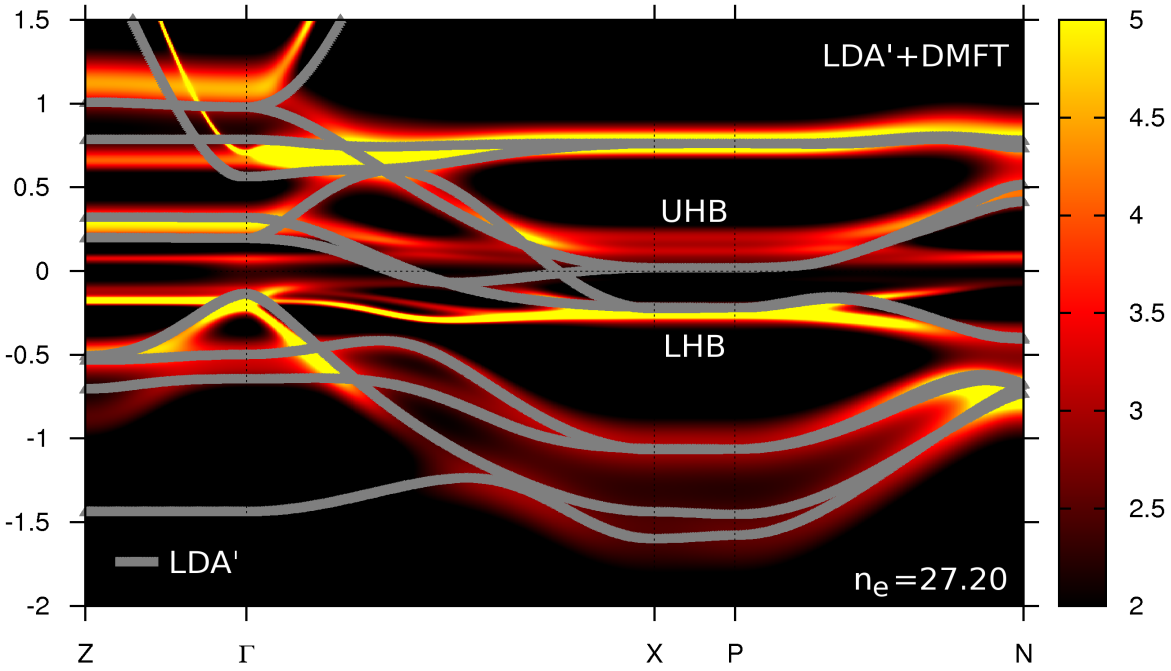

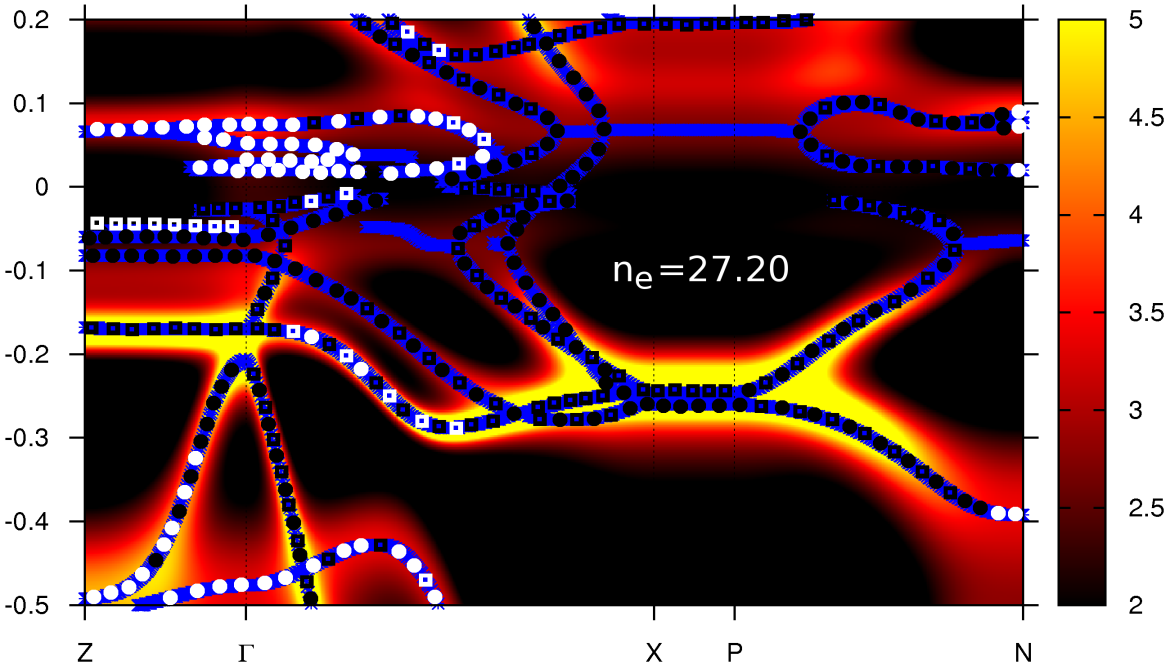

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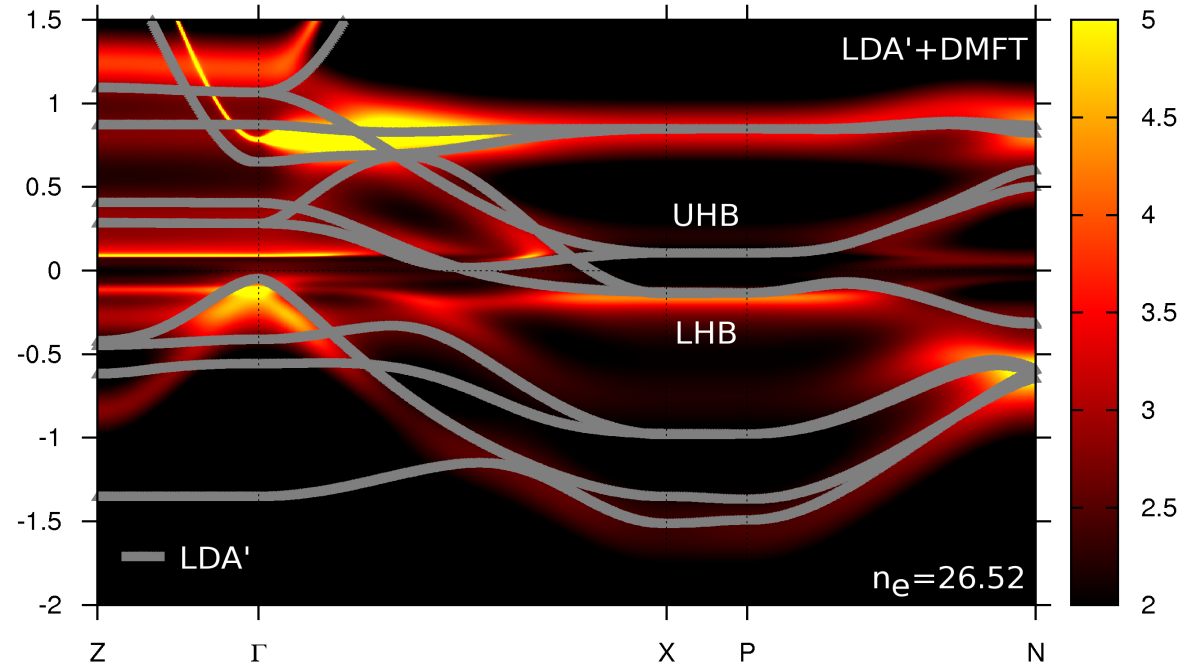
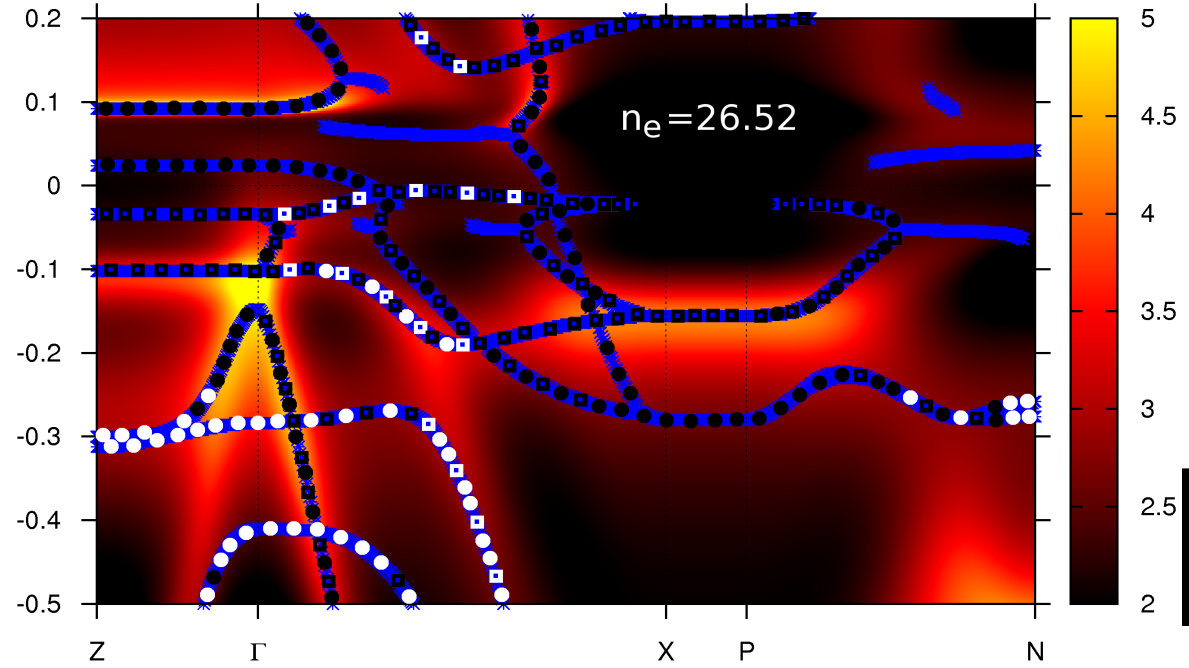




● – xy , □ – xz,yz

● – $3z^2$-r^2 □ –x^2-y^2

LDA'+DMFT self-energy effects

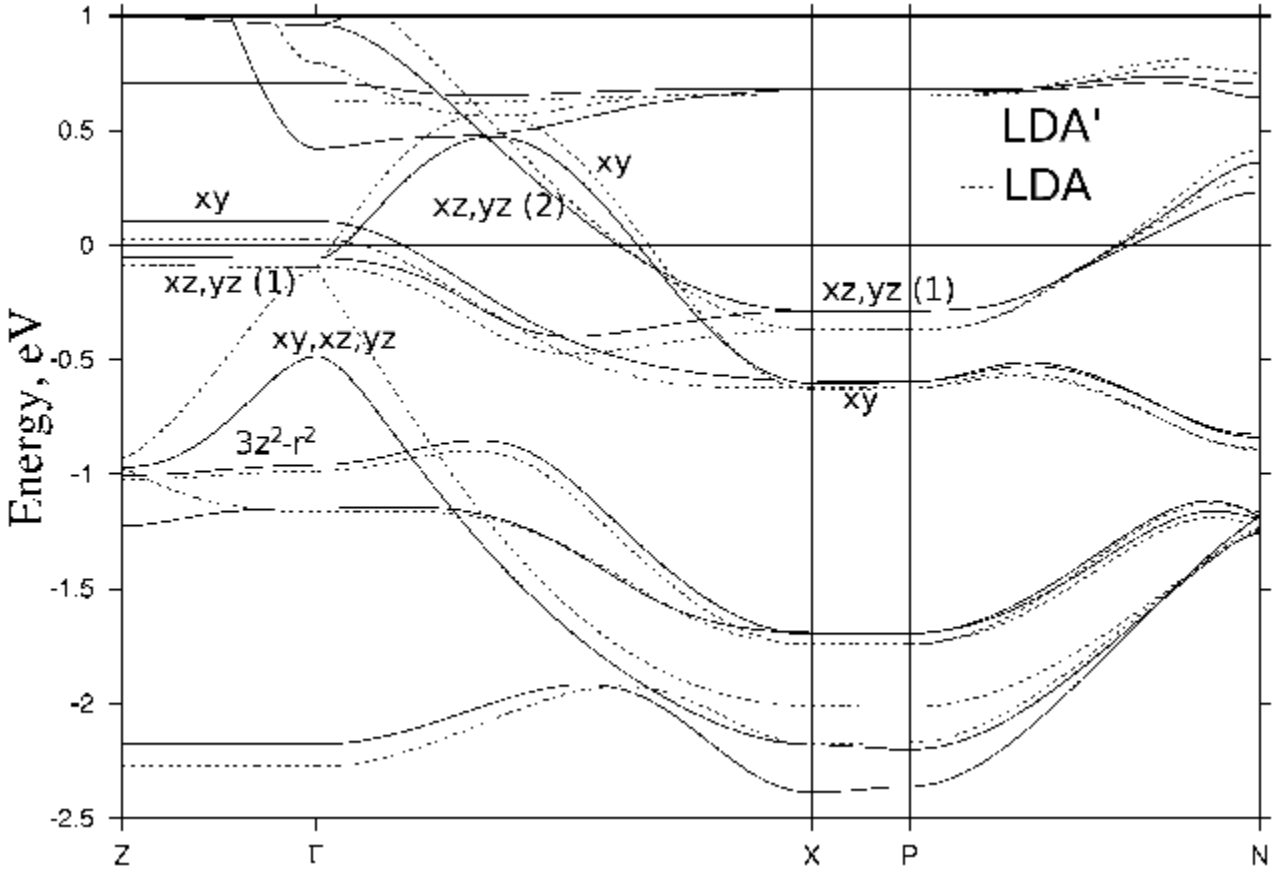
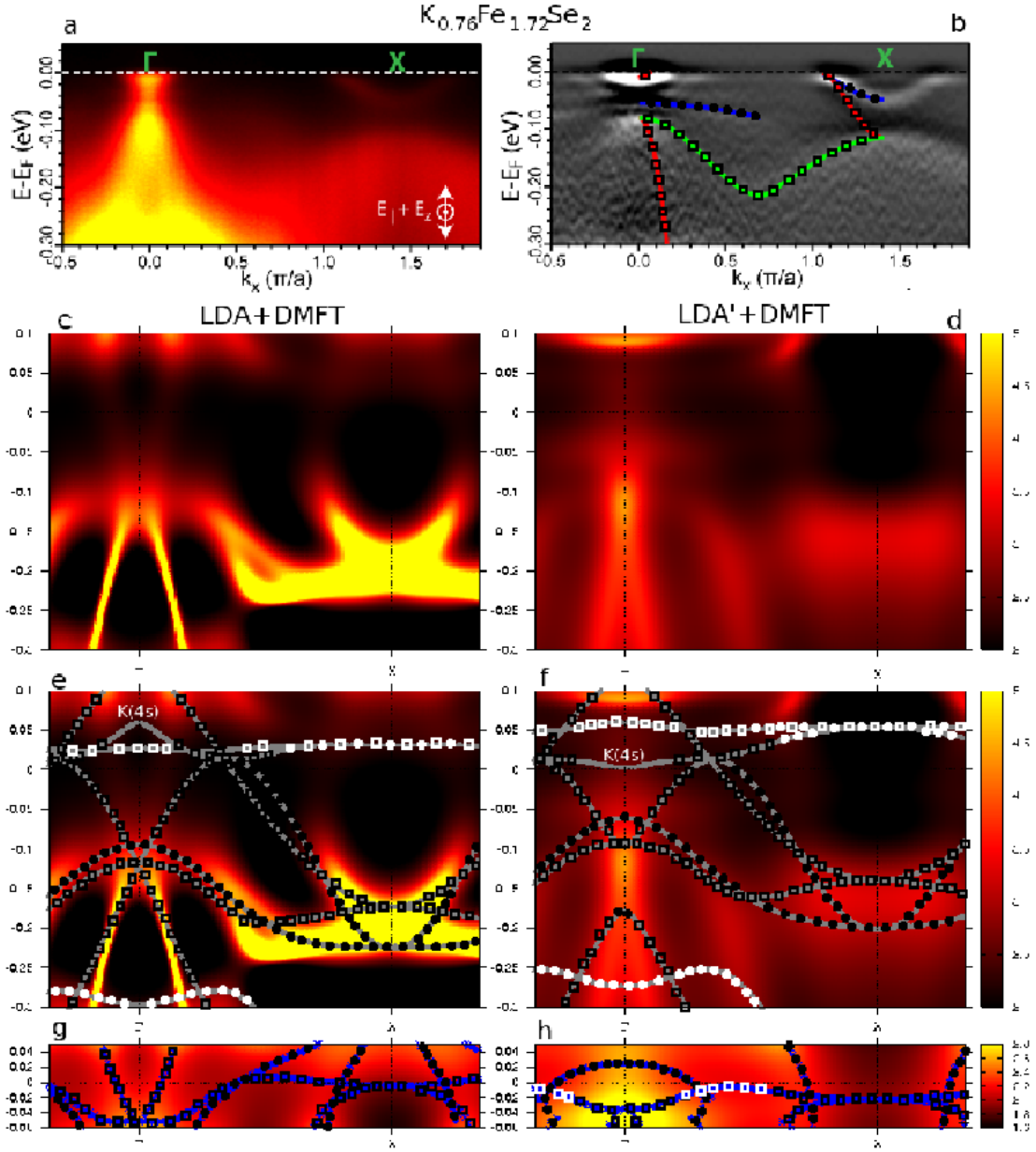


TABLE I: Quasiparticle energy scale renormalization factors and corresponding energy shifts (in eV, in round brackets) for different bare Fe-3d LDA′ orbitals for all hole doping levels n_e in the LDA′ scale energy interval from -1.0 eV to 0.4 eV.

Orbital chracter	n_e=26.52	n_e=27.20	n_e=28.00	n_e=29.00
xy	1.5 (-0.23)	3.9 (-0.73)	2.65 (-0.61)	1.7 (-0.35)
xz, yz (1)	4.2 (-0.78)	3.0 (-0.75)	2.6 (-0.69)	1.7 (-0.38)
xz, yz (2)	2.3 (-0.48)	2.5 (-0.60)	2.6 (-0.69)	1.7 (-0.38)
xy, xz, yz	1.2 (-0.10)	1.3 (-0.10)	1.3 (-0.10)	1.4 (-0.17)
$3z^2 - r^2$	4.7 (-0.85)	2.0 (-0.30)	1.3 (-0.03)	1.25 (0.0)

Comparison of LDA+DMFT and LDA'+DMFT results with ARPES data



ARPES data from

ArXiv: 1208.5192

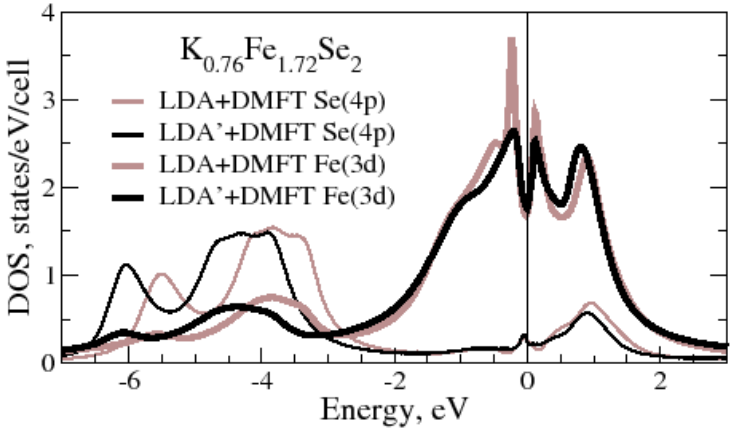


Fig. 4. Comparison of LDA+DMFT (grey lines), LDA′+DMFT (black lines) Se-4p (thin lines) and Fe-3d states (thick lines) densities of states for $K_{0.76}Fe_{1.72}Se_2$. The Fermi level E_F is at zero energy.

Conclusions:

- Proposed consistent LDA'+DMFT method to treat the LDA+DMFT double counting problem.
- LDA+DMFT and LDA'+DMFT calculations are done for charge transfer insulators NiO, MnO, CoO and metallic systems SrVO3 and Sr2RuO4. LDA'+DMFT results give better O-2p states position in comparison with X-ray data. For NiO and CoO LDA'+DMFT improves LDA+DMFT where metallic solutions are obtained.
- Correlation effects in KFe2Se2 grows with hole doping in according to LDA'+DMFT results.
- As can be seen from LDA'+DMFT results Fe-3d bands of different symmetry have different renormalization also dependent on the part of the Brillouin zone.
- LDA'+DMFT spectral functions describes recent ARPES experiments on hole doped iron chalcogenide superconductor KFeSe in the normal state on a semiquantitative level.