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LDA′+DMFT investigation of electronic structure of $K_{1-x}Fe_{2-y}Se_2$ superconductor

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We investigate electronic structure of the new iron chalcogenide high temperature superconductor $K_{1-x}Fe_{2-y}Se_2$ (hole doped case with $x = 0.24$, $y = 0.28$) in the normal phase using the novel LDA′+DMFT computational approach. We show that this iron chalcogenide is more correlated in a sense of bandwidth renormalization (energy scale compression by factor about 5 in the interval ±1.5 eV), than typical iron pnictides (compression factor about 2), though the Coulomb interaction strength is almost the same in both families. Our results for spectral densities are in general agreement with recent ARPES data on this system. It is found that all Fe-3$d(t_{2g})$ bands crossing the Fermi level have equal renormalization, in contrast to some previous interpretations. Electronic states at the Fermi level are of predominantly xy symmetry. Also we show that LDA′+DMFT results are in better agreement with experimental spectral function maps, than the results of conventional LDA+DMFT. Finally we make predictions for photoemission spectra lineshape for $K_{0.76}Fe_{1.72}Se_2$.

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The iron based FeAs(Se) high-temperature superconductors [1] are one of the hottest topics of the present day condensed matter research [2–4]. Recent discovery of iron chalcogenides $K_xFe_2Se_2$ [5], $Cs_xFe_2Se_2$ [6] and (Tl,K)Fe_xSe_2 [7] with superconducting critical temperatures T_c around 30 K, which is typical for the most studied 122 iron pnictides [8–11] initiated intensive studies of these new systems, which was further stimulated by the discovery of nontrivial antiferromagnetic ordering with very high Neel temperature about 550 K and Fe vacancies ordering at approximately the same temperatures in $K_{0.8}Fe_{1.6}Se_2$ (the so called 245 phase) [12]. Because of the complicated picture of microscopic phase separation in this system, there is still no consensus on the composition of the phase, responsible for superconductivity, though the majority point of view indicate to KFe_2Se_2 (122 phase) as a parent compound for superconductivity (while 245 phase is insulating) [13–15]. There is also experimental evidence for some other phases being present in this system [16].

From crystal structure point of view AFe_2As_2, Fe(Se,Te), and AFe_2Se_2 systems are formed by identical layers of $Fe(As,Se)_4$ tetrahedra. AFe_2As_2 and AFe_2Se_2 compounds are isostructural. LDA (local density approximation) calculated electronic band structures of Fe(Se,Te) [17] and AFe_2As_2 [18–20] are quite similar to each other, especially if we are dealing only with bands in the vicinity of the Fermi level. LDA electronic structure of AFe_2Se_2 is significantly different [21, 22]. Direct comparison of LDA band structures of Fe pnictides and chalcogenides can be found in Refs. [22, 13].

From the early days of iron based superconductors, it was noted that electronic correlations are important for understanding the physics of pnictide materials [23–26]. Electronic correlations for these materials were taken into account within LDA+DMFT hybrid computational scheme [27]. It is rather common opinion now, that the main effect of correlations onto band structure of Fe pnictides reduces to the simple LDA bandwidth renormalization (narrowing) by the factor of 2 or 3. There are only few papers on LDA+DMFT in Fe chalcogenides up to now [28, 29].

The AFe_2Se_2 systems were rather extensively studied by angular resolved photoemission spectroscopy (ARPES) [30]. In contrast to AFe_2As_2 compounds, with several, more or less well defined, hole cylinders of the Fermi surface around Γ-point, ARPES data for AFe_2Se_2 show rather weak indications for Fermi surface around Γ-point. Around (π,π) point in both classes of superconductors electron Fermi surface sheets are well observed. These ARPES results are in rough agreement with LDA predictions [21, 22, 13].

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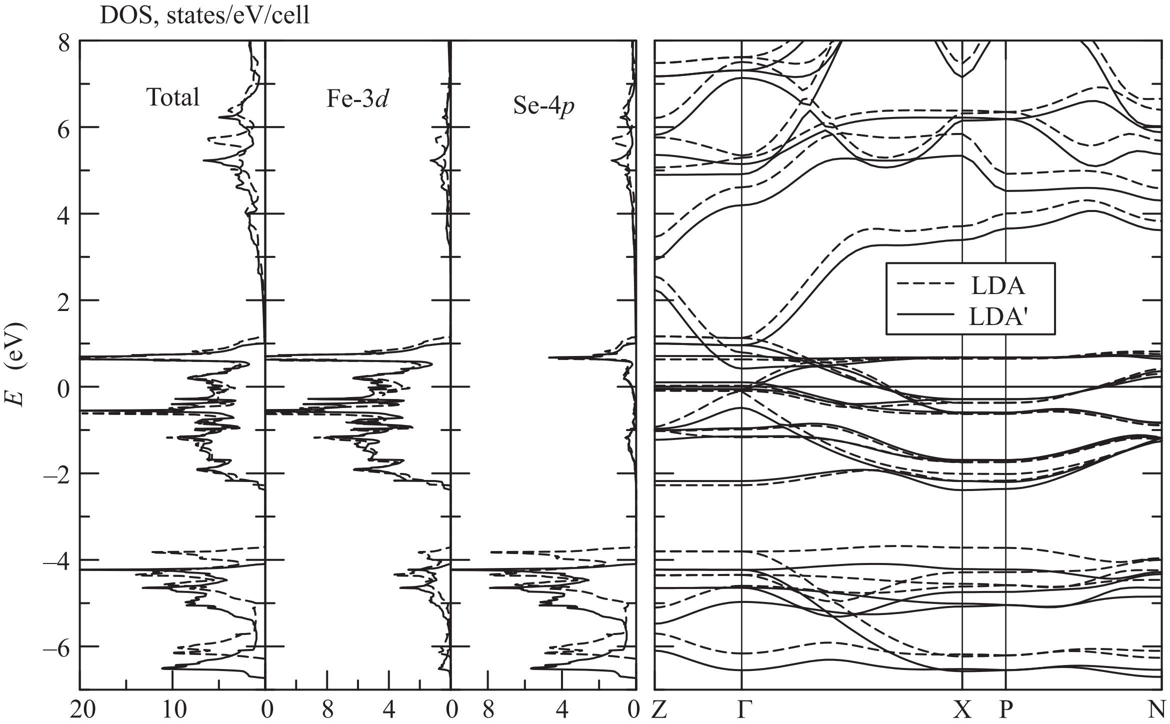

Fig. 1. LDA (dashed lines) and LDA′ (solid lines) calculated band dispersions (right panel) and total, Fe-3d and Se-4p densities of states (left panel) for KFe_2Se_2. The Fermi level E_F is at zero energy

This paper was inspired by the recent ARPES work [31] on $A_xFe_{2-y}Se_2$ ($A = K,Rb$). Here we present our LDA+DMFT and LDA′+DMFT [32] results for hole doped $K_{0.76}Fe_{1.72}Se_2$. LDA and LDA′ calculations were performed using Linearized Muffin-Tin Orbitals method [33], with settings described in Ref. [22]. To solve DMFT effective five orbital impurity problem we used the Hirsh-Fye Quantum Monte-Carlo algorithm [34], with temperature about 280 K. LDA+DMFT and LDA′+DMFT densities of states and spectral functions were obtained as proposed in Ref. [32]. Coulomb parameter U was taken to be 3.75 eV and Hund parameter – $J = 0.7$ eV [31]. These parameters agree well with those calculated in Ref. [35]. To define DMFT lattice problem we used the full (i.e. without any downfolding or projecting) LDA Hamiltonian, which included Fe-3d, Se-4p and K-4s states.

In Fig. 1 we plot LDA (dashed lines) and LDA′ (solid lines) calculated bands dispersions (right panel), as well as total, Fe-3d and Se-4p densities of states (left panel) for stoichiometric KFe_2Se_2. The Se-4p states are located between -7 eV and -3.5 eV. The Se-4p states are well separated in energy from Fe-3d states which cross the Fermi level. The Fe-3d states expand from -2.2 up to $+1$ eV. This is the same or similar to previous LDA results of Refs. [21, 22]. For LDA′ results we observe band shapes, that are almost identical to those of LDA, with approximately rigid shift of Se-4p states down in energy for LDA′ [32]. Note the effect of LDA′ – splitting of xy bands around 0.55 eV.

In Fig. 2 we show orbitally resolved densities of states (DOS) for Fe-3d orbitals. Thin gray lines represent LDA results and thin black lines – LDA′. We see that the main contribution at the Fermi level comes from Fe(3d)-t_{2g} bands – xy and degenerate xz, yz (similar to the case of Ba122 pnictide [18]). In general LDA′ DOS'es are similar to those of LDA, except that LDA′ DOS'es are few tenths of eV narrower. In contrast to Ba122 pnictide [18] both LDA and LDA′ DOS'es here have rather well developed "pseudogap" at the Fermi level. Also at the Fermi level LDA′ DOS is slightly higher than that of LDA.

By thick gray and black lines we show LDA+DMFT and LDA′+DMFT DOS'es for corresponding Fe-3d orbitals of $K_{0.76}Fe_{1.72}Se_2$. This hole doping level of parent compound KFe_2Se_2 corresponds to 6.08 electrons per Fe site within LDA+DMFT calculations. Despite correlations are moderate as compared to Fe-3d bandwidth of $K_{0.76}Fe_{1.72}Se_2$, we observe rather remarkable renormalization of the spectral weight. Interestingly, the contributions of Fe(3d)-e_g bands (x^2-y^2 and $3z^2-r^2$) in our LDA+DMFT and LDA′+DMFT results become larger at the Fermi level, as compared to LDA and LDA′, while t_{2g} bands contribution at the Fermi level remains

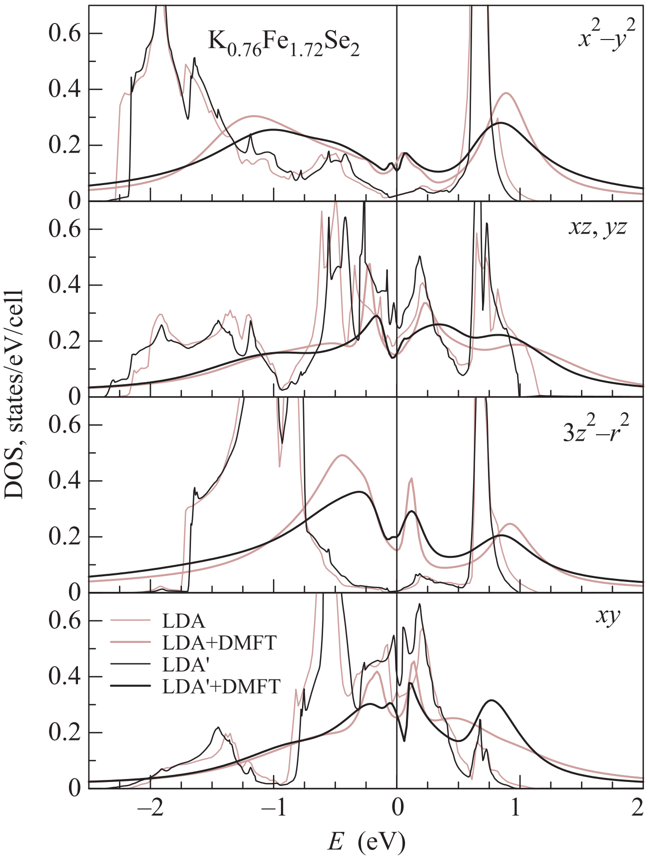

Fig. 2. Comparison of LDA (thin gray lines), LDA′ (thin black lines) and LDA+DMFT (thick gray lines), LDA′+DMFT (thick black lines) densities of states for $K_{0.76}Fe_{1.72}Se_2$ for different Fe-3d orbitals. The Fermi level E_F is at zero energy

nearly the same. From LDA and LDA′ DOS'es one can note, that LDA+DMFT and LDA′+DMFT results can roughly be obtained just by bands compression around the Fermi level by a factor of 2 to 2.5, which is comparable to Ba122 pnictide [26]. However, we shall see below, that situation is not so simple.

More detailed picture is revealed in Fig. 3. Here we show LDA+DMFT and LDA′+DMFT spectral function maps for $K_{0.76}Fe_{1.72}Se_2$ along Γ-X(π,π) high symmetry directions, compared with experimental data of Ref. [31]. Both theoretical and experimental data are shown in a narrow energy interval from −300 to +100 meV. A common feature of theory and experiment is rather low intensity of the spectral function close to the Fermi level, where there are (almost) no well defined quasiparticle bands. It is the main difference of $K_{0.76}Fe_{1.72}Se_2$ from similar Ba122 pnictide, where quasiparticle bands are clearly seen close to the Fermi level [26]. This fact can be explained by "pseudogap" behavior of LDA+DMFT and LDA′+DMFT DOS'es in Fig. 2 at ± 100 meV around the Fermi level. This pseudogap structure is related to rather short lifetime (imaginary part of self-energy), together with positive inclination of the real part of the self-energy (Cf. Ref. [36]). This corresponds to a kind of non Fermi-liquid behavior close to the Fermi level.

At the same time, both experiment and our theory show the pronounced quasiparticle bands at energies about −200 meV. These bands are easily described by LDA or LDA′ bands with the energy scale, which is compressed by a factor of ∼ 5 for the energy interval ± 1.5 eV. Corresponding results are shown on panels (e) and (f) of Fig. 3. Thus, $K_{0.76}Fe_{1.72}Se_2$ has much stronger quasiparticle mass renormalization than similar 122 pnictides [26]. We conclude, that $K_{0.76}Fe_{1.72}Se_2$ is more correlated, than Fe pnictides and is rather close to Mott insulator (see also Refs. [29, 31]). To clarify LDA+DMFT and LDA′+DMFT spectral function maps orbital character, on panels (e) and (f) of Fig. 3 we show with different symbols the predominant contributions of different Fe-3d orbitals (black circles – xz, yz, black squares – xy, white circles – $3z^2-r^2$, white squares – x^2-y^2). It is also important to note, that right above the Fermi level (at +50 meV) there is rather flat Fe(3d)-e_g band which consists of x^2-y^2 and $3z^2-r^2$ contributions. Thus, small changes of doping level may lead to rather dramatic changes of electronic properties, as discussed previously in Refs. [22, 13].

Now we take a closer look at the "dark region" of spectral density, close to the Fermi level. If we plot the region between ± 50 meV of the Fermi level on a smaller intensity scale (panels (g) and (h) of Fig. 3), we can distinguish some "band" structure, in a sense of dispersive maxima of spectral density, which are shown in both panels. We can see here a rather narrow "band" of xy symmetry. However it is not just the LDA xy band, renormalized by a factor of 10, as proposed in Ref. [31]. This band can be seen from second derivative of rough ARPES data from panel (a) of Fig. 3. Its line shape differs quite considerably from those of LDA. One has to remember, that around the Fermi level all quasiparticle bands are rather ill defined. We also note, that it is pretty strange that in Ref. [31] xz and yz bands are found to be different along (0,0)–(π,π) direction, because it is just prohibited by the symmetry (see also panel (a) of Fig. 3).

In Fig. 4 we present 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 LDA+DMFT and LDA′+DMFT results for the DOS are quite similar. At the Fermi level, there is a kind of narrow "pseudogap"

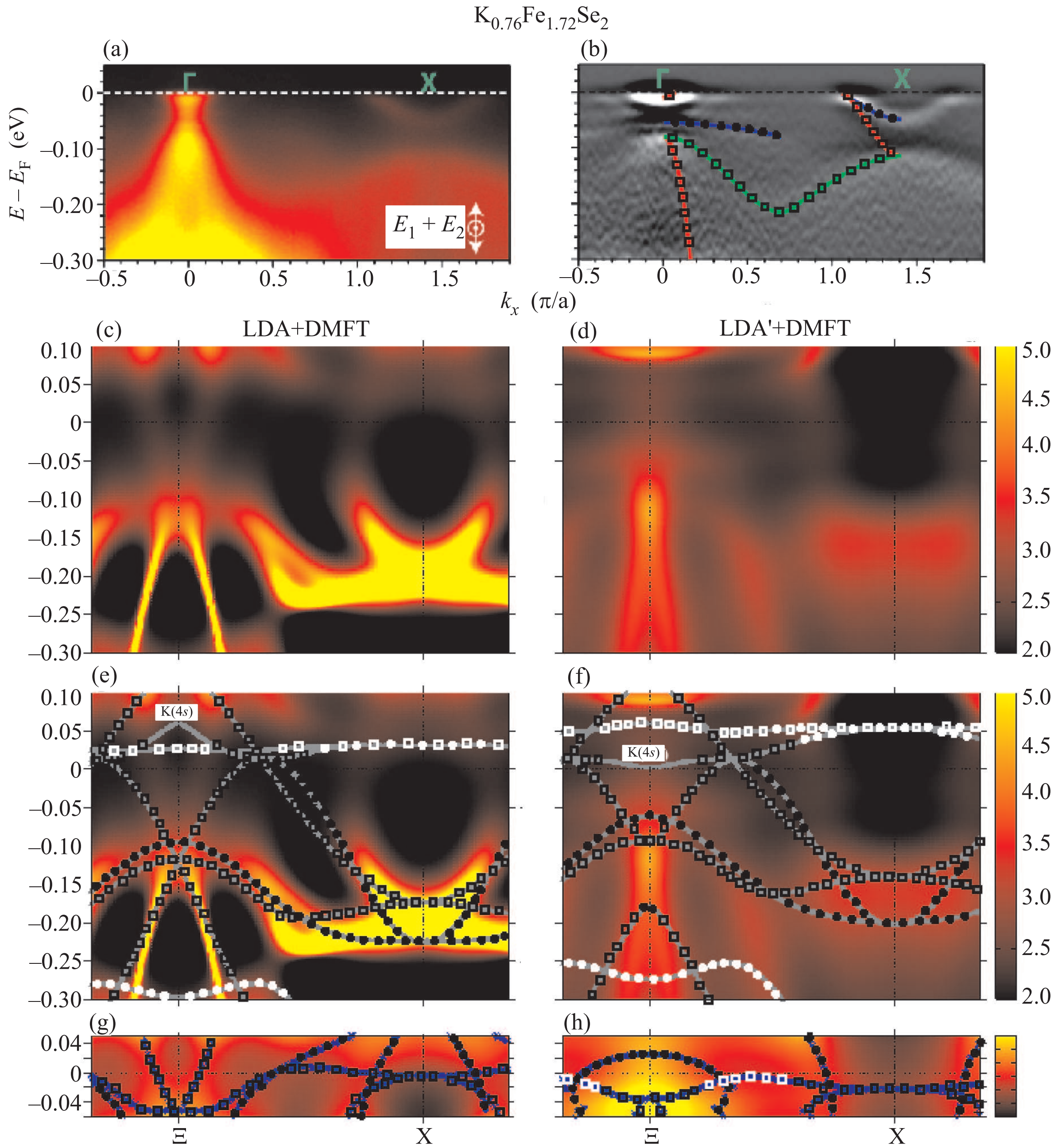

Fig. 3. $K_{0.76}Fe_{1.72}Se_2$: comparison LDA+DMFT (c) and LDA′+DMFT (d) spectral functions with rough experimental ARPES data (a) and corresponding second derivative (b) from Ref. [31]. Panels (e) and (f) show LDA and LDA′ bands (compressed by factor of 5) on top of LDA+DMFT and LDA′+DMFT results. Maxima of LDA+DMFT and LDA′+DMFT spectral functions near the Fermi level are presented on panels (g, h). Symbols show predominant contributions of different Fe-3d orbitals into the spectral function (black circles – xz, yz, black squares – xy, white circles – $3z^2 - r^2$, white squares – $x^2 - y^2$). The Fermi level E_F is at zero energy

structure for Fe-3d states. However, LDA′+DMFT DOS for Se-4p states are shifted about 0.5 eV lower in energy, as compared to those obtained in LDA+DMFT. This suggests the importance of the measurements of the lineshape of X-Ray photoemission spectra, as we are unaware of any experiments of this kind for the $K_{0.76}Fe_{1.72}Se_2$ system.

Conclusion. In this paper we investigated the electronic structure of hole doped high temperature iron chalcogenide superconductor $K_{0.76}Fe_{1.72}Se_2$ in normal phase by means of LDA+DMFT and LDA′+DMFT. It

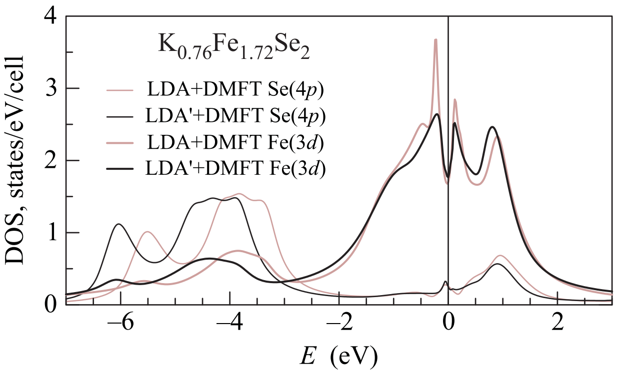

Fig. 4. Comparison of LDA+DMFT (grey lines), LDA$'$+DMFT (black lines) Se-4p (thin lines) and Fe-3d states (thick lines) densities of states for $K_{0.76}Fe_{1.72}Se_2$. The Fermi level E_F is at zero energy

was found, that $K_{0.76}Fe_{1.72}Se_2$ is more correlated, than similar isostructural 122 Fe pnictides, in the sense of bandwidth renormalization (energy scale compression by a factor about 5 in the interval ± 1.5 eV). Contrary to the conclusions of Ref. [31], we observe that all Fe-3d bands have the same bandwidth renormalization.

Also in contrast to Fe 122 pnictides, $K_{0.76}Fe_{1.72}Se_2$ compound does not have well defined quasiparticle bands around the Fermi level and is apparently quite close to Mott insulating phase (see also Ref. [31]). Both within LDA+DMFT and LDA$'$+DMFT we observe a kind of "pseudogap" at the Fermi level, which is inherited from LDA band structure. The presence of this "pseudogap" leads to relatively low intensity spectral function near the Fermi level, which is clearly seen both in theory and in ARPES data [31]. Similarly to Ref. [31], within this "pseudogap" region, we can distinguish some very low intensive "band" structure (both in LDA+DMFT and LDA$'$+DMFT) — there is rather ill defined quasiparticle band of xy symmetry, which is not simply renormalized (by a factor of 10) LDA xy band, as proposed in Ref. [31]. Finally we stress the importance of measurements of photoemission spectra lineshapes.

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