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Electronic structure, topological phase transitions and superconductivity in $(K,Cs)_xFe_2Se_2$

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We present LDA band structure of novel hole doped high temperature superconductors ($T_c \sim 30$ K) $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ and compare it with previously studied electronic structure of isostructural FeAs superconductor $BaFe_2As_2$ (Ba122). We show that stoichiometric KFe_2Se_2 and $CsFe_2Se_2$ have rather different Fermi surfaces as compared with Ba122. However at about 60% of hole doping Fermi surfaces of novel materials closely resemble those of Ba122. In between these dopings we observe a number of topological Fermi surface transitions near the Γ point in the Brillouin zone. Superconducting transition temperature T_c of new systems is apparently governed by the value of the total density of states (DOS) at the Fermi level.

The FeAs based high-temperature superconductors [1] attracted a lot of attention and huge number of experimental and theoretical investigations have been done (for review see [2, 3]) and many are still going on. Pretty high values of superconducting transition temperature were discovered also in Fe chalcogenides $FeSe_x$ and $FeSe_{1-x}Te_x$ [4].

Structurally FeSe systems are similar to FeAs compounds and consist of layers of $FeSe_4$ tetrahedra. Recent discovery of intercalated $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ compounds produced much higher values of T_c=31 K [5] and 27 K [6] similar to those in FeAs 122 systems [2, 3]. This was followed by $T_c = 31$ K in $(Tl,K)Fe_xSe_2$ [7].

Electronic structure of Fe(S,Se,Te) materials was described in details in Ref. [8]. However $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ have different crystal structure and are actually isostructural to $BaFe_2As_2$ (Ba122). Its electronic structure was reported in Refs. [9 – 11]. First calculations of electronic spectrum of $K_xFe_2Se_2$ were described in a recent preprint [12].

In this work we present comparative study of electronic structure, densities of states for Ba122 and $K_xFe_2Se_2$, $Cs_xFe_2Se_2$ systems, demonstrating changes of Fermi surface topology upon doping and making some simple estimates of superconducting T_c.

The $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ systems are isostructural to Ba122 (for the last one see Ref. [9]) with ideal body centered tetragonal space group I4/mmm. The $K_xFe_2Se_2$ has $a = 3.9136$ Å and $c = 14.0367$ Å with K ions occupying $2a$, Fe – $4d$ and Se – $4e$ positions with z_{Se}=0.3539 [5]. In case of $Cs_xFe_2Se_2$ lattice parameters are a=3.9601 Å and c=15.2846 Å and z_{Se} is 0.3439 [6]. For given crystal structures we performed band structure calculations within the linearized muffin-tin orbitals method (LMTO) [13] using default settings.

In Fig.1 we compare Ba122 band structure and different densities of states of Ref. [9] (left) and those for $K_xFe_2Se_2$ (black lines) and $Cs_xFe_2Se_2$ (gray lines) (right) for stoichiometric case of $x = 0$. In a bird eye view $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ have nearly the same band dispersions which to some extent are similar to those in Ba122. However, there are some quantitative differences. First of all Fe-3d and Se-4p states in new systems are separated in energy in contrast to Fe-3d and As-4p Ba122. Also Se-4p states are of about 0.7 eV lower than As-4p states.

Similar to Ba122 the Fermi level E_F in K and Cs chalcogenides is crossed by Fe-3d states. Detailed band structure near the Fermi level, which is decisive for the formation of superconducting state, for the new systems is compared with that of Ba122 in Fig.2. To some extent Ba122 bands near E_F (upper part of Fig.2) would match those for $(K,Cs)Fe_2Se_2$ if we shift them down in energy by about 0.2 eV. Main difference between old and new systems is seen around Γ point. For $(K,Cs)Fe_2Se_2$ systems antibonding part Se-$4p_z$ band in the Z-Γ direction forms electron-like pocket. In Ba122 corresponding band lies about 0.4 eV higher and goes much steeper, thus it is quite far away from Γ point. However, if we dope $(K,Cs)Fe_2Se_2$ systems (in a rigid band manner) with holes (as shown by horizontal lines in Fig.2 on lower panel) we obtain bands around Γ point (close to the Fermi level) very similar to those in case of Ba122. Namely at 60% hole doping we obtain three hole-like cylinders while stoichiometric KFe_2Se_2 has one small electron pocket and larger hole like one and $CsFe_2Se_2$ has just one electron pocket near Γ point. Thus, in fact under hole doping we observe several topological transi-

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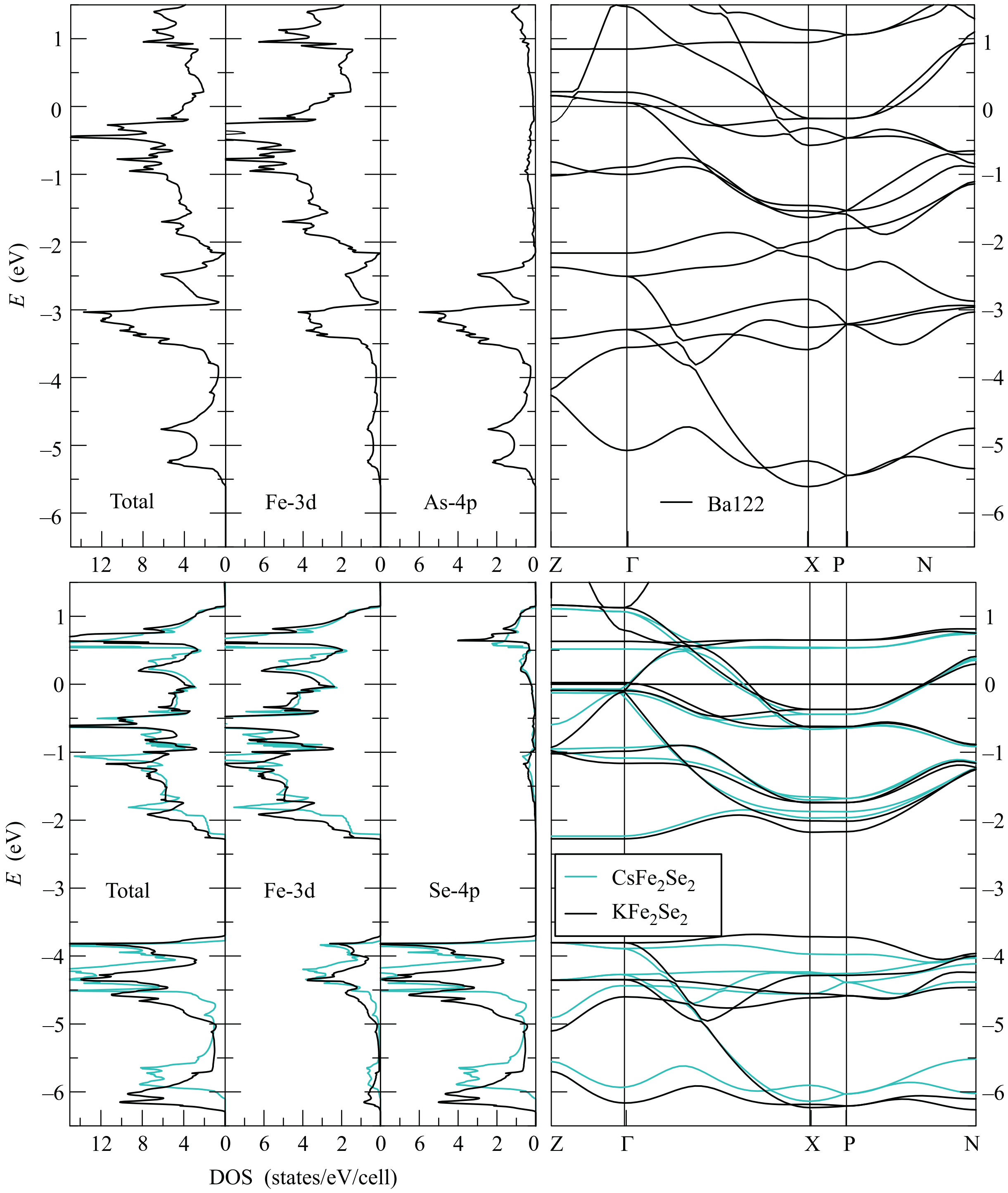

Fig.1. LDA calculated band dispersions and densities of states of Ba122 (upper panel) and KFe$_2$Se$_2$ (black lines) and CsFe$_2$Se$_2$ (gray lines)(lower panel). The Fermi level E_F is at zero energy

tions of the Fermi surfaces which we shall briefly discuss below.

To trace orbital composition of bands if Fig.3 we show orbital resolved densities of states for KFe$_2$Se$_2$. Again as for Ba122 [9] and other iron pnictides mainly t_{2g} states (xy, xz and yz) contribute to the density of states at the Fermi level. The e_g states ($3z^2 - r^2$ and $x^2 - y^2$) are almost absent in the density of states at E_F.

In Fig.4 we present LDA calculated Fermi surfaces (FS) for both K (upper row) and Cs (lower row)

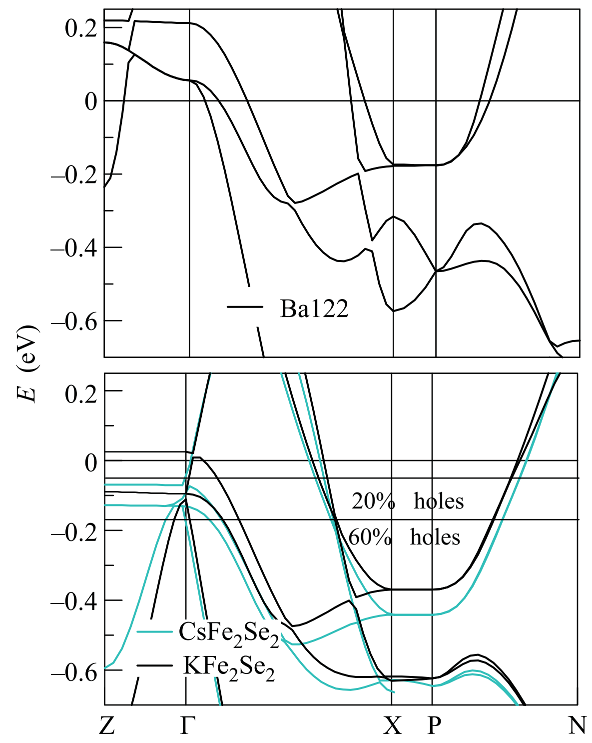

Fig.2. Top panel – LDA calculated band dispersions in the vicinity of the Fermi level for Ba122; Bottom panel – $K_xFe_2Se_2$ (black lines) and $Cs_xFe_2Se_2$ (gray lines). The Fermi level is at zero energy. Additional horizontal lines correspond to Fermi level position for the case of 20% and 60% hole doping

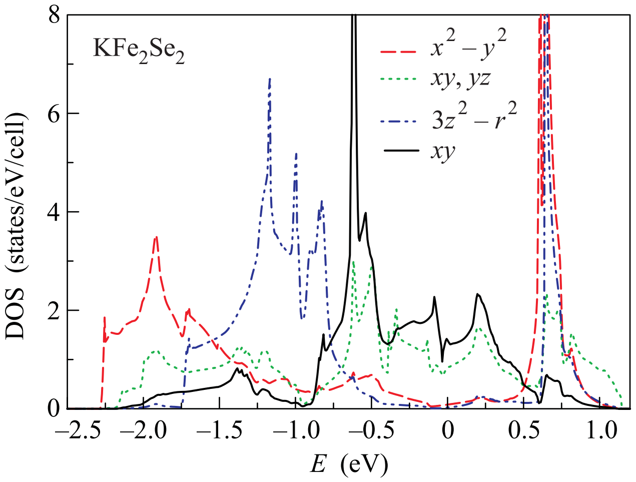

Fig.3. LDA calculated orbital resolved densities of states of KFe_2Se_2. The Fermi level is at zero energy

(K,Cs)Fe_2Se_2 compounds for different hole doping levels of $x = 0$ (left), $x = 0.2$ (middle) and $x = 0.6$ (right). All Fermi surfaces have two almost two dimensional electron-like sheets in the corners of the Brillouin zone with topology independent of doping level. Compared to Ba122 FeAs system the sharp difference in the Fermi surface topology around the center of the Brillouin zone (Γ-point) is observed at $x = 0$ and $x = 0.2$. In fact, KFe_2Se_2 compound has one electron and one hole torus-like FS sheets while $CsFe_2Se_2$ has just one electron-like hourglass FS sheet. With hole doping KFe_2Se_2 torus transforms to electron-like hourglass and hole cylinder. For 20% hole doped Cs compound we get similar picture with smaller volume FS sheets of the same topology. For $x = 0.6$ both K and Cs new FeSe materials have Fermi surfaces quite similar to those in Ba122 iron pnictide (see Ref. [9]), with rather typical hole-lke FS cylinders in the center of the Brillouin zone.

In the Ref. [6] it was shown that K and Cs compounds follow the tendency of T_c dependence on anion height in FeSe plane observed in Ref. [14], which was plausibly explained in Ref. [15] in terms of total density of states change at the Fermi level. Similar observation was made for related compounds $SrPt_2As_2$, $BaNi_2As_2$ and $SrNi_2As_2$ in Ref. [16].

Now we also can make some simple BCS-like estimates of T_c. Taking the LDA calculated value of total DOS at the Fermi level $N(E_F)$ 3.94 states/eV/cell for $K_{x=0}Fe_2Se_2$ and 3.6 states/eV/cell for $Cs_{x=0}Fe_2Se_2$, $\omega_D = 350\,K$ and coupling constant $g = 0.21\,eV$ estimated for Ba122 (as described in Ref. [15]), then using the BCS expression for $T_c = 1.14\omega_D e^{-2/gN(E_F)}$ we immediately obtain $T_c = 34\,K$ and 28.6 K for K and Cs systems respectively (T_c ratio 1.18). That is very close to experimental T_c values 31 K[5] and 27 K (T_c ratio 1.15) [6]. If we take into account the fact that upon hole doping $N(E_F)$ grows for both compounds up to 4.9 states/eV/cell in K and 4.7 states/eV/cell in Cs at 60% doping level superconducting transition temperatures can be estimated in a similar way to give $T_c = 57\,K$ for K system and $T_c = 52\,K$ for Cs system, showing the potential role of doping. Thus, in accordance with our previous work on pnictides [15], the values of T_c apparently well correlate with the total DOS value at the Fermi level $N(E_F)$. It should be stressed that these estimates do not necessarily imply electron-phonon pairing, as ω_D may just denote the average frequency of any other possible Boson responsible for pairing interaction (e.g. spin fluctuations). At the same time the lower values of T_c in Cs compound in comparison to K system can be probably attributed to the usual isotope effect.

To conclude, we investigated the band structure and Fermi surface topology of recently discovered chalgogenide iron superconductors $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ isostructural to Ba122 iron pnictide system at different hole doping levels. We show that at about 60% hole doping level both $K_xFe_2Se_2$ and $Cs_xFe_2Se_2$ energy bands

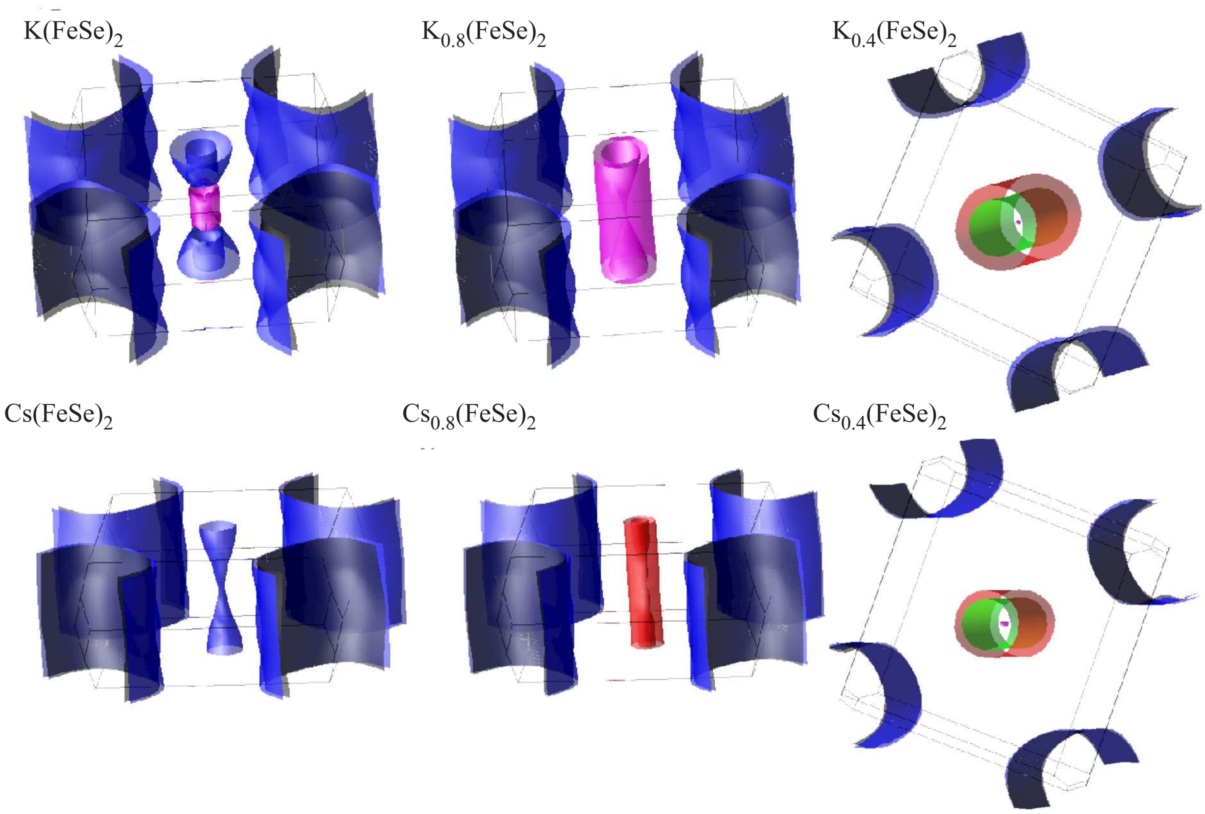

Fig.4. LDA calculated Fermi surfaces of $K_xFe_2Se_2$ (upper row) and $Cs_xFe_2Se_2$ (lower row) for different doping levels: $x = 0$ – left column, $x = 0.2$ – middle and $x = 0.6$ – right

and Fermi surface topologies resemble very much those of Ba122 FeAs system. However, at intermediate dopings there are several topological transitions of the Fermi surfaces with changing of number of (electron-like and hole-like) sheets. Also we demonstrated that T_c values in new superconductors are well correlated with total DOS value at the Fermi level $N(E_F)$, which is related to anion height relative to Fe square lattice, similar to that in other FeAs and Fe(Se,Te) systems.

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