

# Electronic Structure of Prototype $\text{AFe}_2\text{As}_2$ and $\text{ReOFeAs}$ High-Temperature Superconductors: a Comparison

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We have performed *ab initio* LDA calculations of electronic structure of newly discovered prototype high-temperature superconductors  $\text{AFe}_2\text{As}_2$  (A=Ba,Sr) and compared it with previously calculated electronic spectra of  $\text{ReOFeAs}$  (Re=La,Ce,Pr,Nd,Sm). In all cases we obtain almost identical densities of states in rather wide energy interval (up to 1 eV) around the Fermi level. Energy dispersions are also very similar and almost two-dimensional in this energy interval, leading to the same basic (minimal) model of electronic spectra, determined mainly by Fe *d*-orbitals of FeAs layers. The other constituents, such as A ions or rare earths Re (or oxygen states) are more or less irrelevant for superconductivity. LDA Fermi surfaces for  $\text{AFe}_2\text{As}_2$  are also very similar to that of  $\text{ReOFeAs}$ . This makes the more simple  $\text{AFe}_2\text{As}_2$  a generic system to study high-temperature superconductivity in FeAs - layered compounds.

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The recent discovery of the new superconductor  $\text{LaO}_{1-x}\text{F}_x\text{FeAs}$  with the transition temperature  $T_c$  up to 26K [1, 2, 3, 4] and even more high values of  $T_c = 41\text{-}55\text{K}$  in  $\text{CeO}_{1-x}\text{F}_x\text{FeAs}$  [5],  $\text{SmO}_{1-x}\text{F}_x\text{FeAs}$  [6],  $\text{NdO}_{1-x}\text{F}_x\text{FeAs}$  and  $\text{PrO}_{1-x}\text{F}_x\text{FeAs}$  [7, 8] was recently followed by the discovery of high-temperature superconductivity with  $T_c$  up to 38K in K doped ternary iron arsenides  $\text{BaFe}_2\text{As}_2$  [9] and  $\text{SrFe}_2\text{As}_2$  [10], with further synthesis of superconducting  $\text{AFe}_2\text{As}_2$  (A=K, Cs, K/Sr, Cs/Sr) [11]. Relatively large single crystals of superconducting  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  were also grown [12], providing a major breakthrough in the studies of anisotropic electronic properties of FeAs - layered superconductors.

The LDA electronic structure of  $\text{LaOFeAs}$  were calculated in a number of papers (see e.g. [13], [14], [15]) producing results qualitatively similar to that first obtained for  $\text{LaOFeP}$  [16]. We have performed LDA calculations for the whole series of  $\text{ReOFeAs}$  (R=La,Ce,Pr,Nd,Sm) [17], demonstrating a very weak (or absent) dependence of electronic spectrum on the type of the rare-earth ion Re in rather wide energy interval ( $\sim 2\text{eV}$ ) around the Fermi level.

First LDA results for the density of states (DOS) of  $\text{BaFe}_2\text{As}_2$  were recently presented in Refs. [18, 19]. Here we present the results of our *ab initio* calculations of electronic structure of the newly discovered prototype high-temperature superconductors  $\text{AFe}_2\text{As}_2$  (A=Ba, Sr) with the aim to compare it with the previously discussed  $\text{ReOFeAs}$  series. We present LDA DOS, energy dispersions and Fermi surfaces of these compounds and briefly discuss possible conclusions with

respect to the minimal model of electronic spectrum and superconductivity. As all results are quite similar for both A=Ba and A=Sr, as well as for the whole Re series, below we present data mainly for A=Ba and Re=La.

Both  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  crystallize in tetragonal structure with the space group  $I4/mmm$  and  $P4/nmm$ , correspondingly. Both compounds are formed of  $(\text{FeAs})^-$  layers alternating with  $\text{Ba}_{0.5}^{2+}$  or  $(\text{LaO})^+$ .  $\text{Fe}^{2+}$  ions are surrounded by four As ions forming a tetrahedron. The crystal structures of  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  are shown in Fig. 1. The quasi two-dimensional character of both compounds makes them similar to the well studied class of superconducting copper oxides. At 140 K  $\text{BaFe}_2\text{As}_2$  undergoes structural phase transition from tetragonal ( $I4/mmm$ ) to orthorhombic ( $Fmmm$ ) space group [20]. The same transition takes place for  $\text{LaOFeAs}$  system at 150 K:  $P4/nmm$  (tetragonal)  $\rightarrow$   $Cmma$  (orthorhombic) [21]. The crystallographic data for tetragonal phase of two compounds is collected in Table 1. It can be seen that for  $\text{BaFe}_2\text{As}_2$  compound the Fe-As distance is smaller than for  $\text{LaOFeAs}$ . So one would expect more considerable Fe-*d*-As-*p* hybridization for  $\text{BaFe}_2\text{As}_2$  system in comparison with  $\text{LaOFeAs}$  and as a result wider Fe-*d* bandwidth. The distance between nearest Fe atoms within FeAs layers is also significantly smaller in  $\text{BaFe}_2\text{As}_2$  as compared with  $\text{LaOFeAs}$  system. After the phase transition of  $\text{BaFe}_2\text{As}_2$  system to the orthorhombic structure the four equal Fe-Fe distances break into two bond pairs of 2.808 Å and 2.877 Å length. Moreover the two As-Fe-As angles are quite different in the case

of LaOFeAs system ( $113.6^\circ$  and  $107.5^\circ$ ) and have very close values ( $\sim 109^\circ$ ) for  $\text{BaFe}_2\text{As}_2$ . Such differences in the nearest surrounding of Fe ions should evoke the distinctions in the electronic structure of these two compounds.

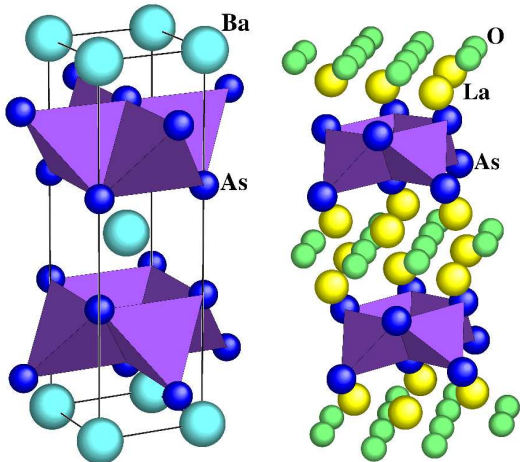


Fig. 1. Crystal structure of  $\text{BaFe}_2\text{As}_2$  (left) and  $\text{LaOFeAs}$  (right). FeAs tetrahedra (violet) form two-dimensional layers sandwiched by Ba ion (cyan) or LaO layers (yellow and green).

Table 1. Crystal structure data for  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  compounds. Atomic positions for  $\text{BaFe}_2\text{As}_2$  are Ba (0, 0, 0), Fe (0.5, 0, 0.25), As (0, 0,  $z$ ) and for  $\text{LaOFeAs}$  are La(0.25, 0.25,  $z$ ), Fe (0.75, 0.25, 0.5), As (0.25, 0.25,  $z$ ), O (0.75, 0.25, 0).

Parameter	$\text{BaFe}_2\text{As}_2$	$\text{LaOFeAs}$
group	I4/mmm	P4/nmm
$a$ , Å	3.9090(1)	4.03533(4)
$c$ , Å	13.2122(4)	8.74090(9)
$z_{\text{La}}$	-	0.14154(5)
$z_{\text{As}}$	0.3538(1)	0.6512(2)
Source	Ref. [9]	Ref. [1]
Ba-As, Å	$3.372(1) \times 8$	-
La-As, Å	-	$3.380 \times 4$
Fe-As, Å	$2.388(1) \times 4$	$2.412 \times 4$
Fe-Fe, Å	$2.764(1) \times 4$	$2.853 \times 4$
As-Fe-As	$109.9(1)^\circ$	$113.6^\circ$
	$109.3(1)^\circ$	$107.5^\circ$

The electronic structure of  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  compounds was calculated within the local density approximation (LDA) by using linearized muffin-tin orbitals basis (LMTO) [22]. For  $\text{BaFe}_2\text{As}_2$  we used the

structure data for K-doped system and temperature  $T=20$  K [9]. The LDA calculated total and partial densities of states for  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  are shown in Fig. 2. In the lower panel of Fig. 2 we show magnified behavior of total DOS around the Fermi level for three different systems under discussion. In all cases DOS is almost flat. It is well known that DOS of two dimensional (nearly free) electrons is a constant defined just by the renormalized electron mass. Thus, our results support the two-dimensional nature of these compounds.

The values of density of states at Fermi level are very similar in both compounds. The 0.3 eV wider Fe- $d$  bandwidth in the case of  $\text{BaFe}_2\text{As}_2$  in comparison with  $\text{LaOFeAs}$  arises from the shorter Fe-As bonds and hence stronger Fe- $d$ -As- $p$  hybridization for this system. The partial As- $p$  DOS is splitted into two parts in the case of Ba system. The orbital projected Fe-3 $d$  DOS for two compounds is shown in Fig. 3. One can see that for both systems three Fe- $d$  orbitals of  $t_{2g}$  symmetry –  $xz$ ,  $yz$ ,  $x^2 - y^2$  mainly contribute to the bands crossing the Fermi level. We call here the  $x^2 - y^2$  (basically rotated  $xy$  orbital) as one of  $t_{2g}$  orbitals following the established earlier terminology for  $\text{ReOFeAs}$  systems.

Energy bands along the high symmetry directions of the Brillouin zone are pictured in Fig. 4. The bands around the Fermi level for both compounds are primarily formed by Fe- $d$  states. In  $\text{LaOFeAs}$  system As- $p$  states are also hybridized with O- $p$  states and the corresponding bands are separated from the Fe- $d$  ones. On the contrary in  $\text{BaFe}_2\text{As}_2$  Fe- $d$  and As- $p$  bands are entangled. The lower two panels of Fig. 4 compare band dispersions for both system close to the Fermi level. Here only  $k_x, k_y$  dispersion is shown. Taking into account different notations of high-symmetry points for these two different crystal structures one can find these dispersions pretty similar to each other. There are three hole-like bands around  $\Gamma$ -point and two electron bands around  $X$ -point. Thus one can define a minimal model of “bare” electronic bands to treat e.g. superconductivity, similar to that discussed in Ref. [23]. Let us mention that along  $X - M$  direction in  $\text{LaOFeAs}$  there are two degenerate bands.

In Fig. 5 relative on-site energies of hybridized Fe-3 $d$  and As-4 $p$  states are presented. A bird’s eye view tells us that this picture for both  $\text{BaFe}_2\text{As}_2$  (left) and  $\text{LaOFeAs}$  (right) is rather similar. There are two groups of states – antibonding (mostly Fe-3 $d$ ) and bonding (mostly As-4 $p$ ) states. However, there are some fine differences. First of all for  $\text{BaFe}_2\text{As}_2$  hybridization between Fe-3 $d$ - $z^2$  and As-4 $p_z$  orbitals is about 0.24 eV weaker. It leads to a swap of the energy positions of Fe-3 $d$ - $z^2$  and  $x^2$  orbitals and similarly for corresponding As-4 $p$  orbitals.

Secondly,  $\text{Fe-}d\text{-}t_{2g}$  orbitals are degenerate for  $\text{BaFe}_2\text{As}_2$  in contrast to  $\text{LaOFeAs}$ .

Neglecting small difference, the overall picture of the energy spectrum in the vicinity of the Fermi level is very similar for both compounds and is determined mainly by  $\text{Fe-}d$  states of  $\text{FeAs}$  layers, making the states of  $A$ -ions or rare-earths  $\text{Re}$  more or less irrelevant for superconductivity. Thus, superconductivity of  $\text{FeAs}$  layered compounds may be studied within the minimal model, taking into account only essential  $\text{Fe-}d$  bands close to the Fermi level. The variants of such model proposed e.g. in Refs. [23, 24] for  $\text{LaOFeAs}$  system may also be used for  $A\text{Fe}_2\text{As}_2$  with only slight modification of model parameters, such as transfer integrals.

The role of electronic correlations in  $A\text{Fe}_2\text{As}_2$  and  $\text{ReOFeAs}$  compounds remains at the moment disputable. On general grounds it can be expected to be rather important due to large values of Hubbard and Hund interactions on  $\text{Fe}$ . However,  $\text{LDA+DMFT}$  calculations for  $\text{LaOFeAs}$  reported in Refs. [25, 26] have produced rather contradictory claims. Obviously, this problem requires further studies. Assuming that correlations in these compounds are most likely in the intermediate range, we may hope that standard  $\text{LDA}$  approach used here is reliable enough.

Fermi surfaces obtained from  $\text{LDA}$  calculations for  $\text{BaFe}_2\text{As}_2$  and  $\text{LaOFeAs}$  are shown in Figs. 6 and 7, correspondingly. There are five sheets of Fermi surface for both compounds. Qualitatively, Fermi surfaces are similar to that reported for  $\text{LaOFeAs}$  in Ref. [13] (see also [15]). There are three hole cylinders in the middle of the Brillouin zones and two electron sheets at the corners of Brillouin zone. Smallest of hole cylinders is usually neglected in the analysis of superconducting pairings [23, 27] and analysis is restricted to minimal two [27] or four bands [23] models, reproducing two hole and two electron cylinders.

$P4/nmm$  (tetragonal)  $\rightarrow$   $Cmma$  (orthorhombic) phase transition taking place in undoped compounds is usually attributed to  $\text{SDW}$  formation due to nesting properties of electron and hole Fermi surfaces [15, 24] or due to excitonic instability in triplet channel [23]. The difficulties of calculating magnetic state of  $\text{LaOFeAs}$  related with apparently itinerant nature of magnetism were recently discussed in Ref. [28].

In conclusion, we have presented the results of  $\text{LDA}$  calculations of new prototype high-temperature superconductor  $A\text{Fe}_2\text{As}_2$  ( $A=\text{Ba}, \text{Sr}$ ) and compared it with previously discussed  $\text{ReOFeAs}$  series, demonstrating essential similarity of electronic states close to the Fermi level and most important for superconductivity. These states are formed mainly by  $\text{Fe}$  orbitals

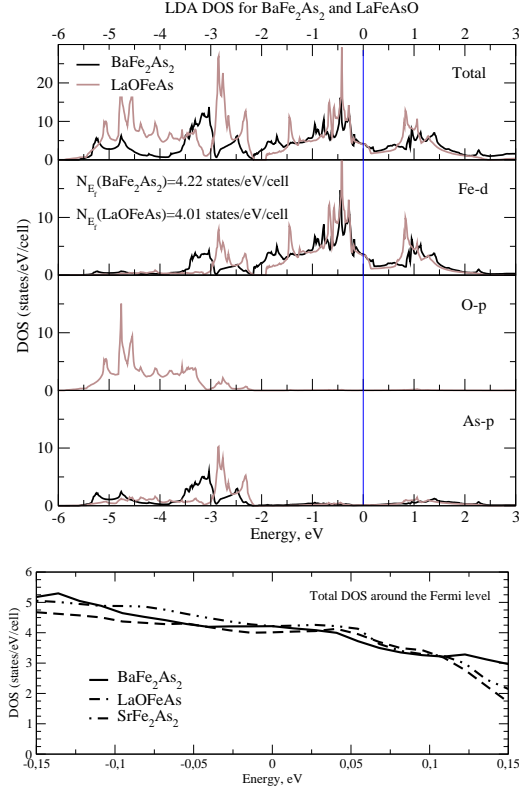


Fig. 2. Total and partial  $\text{LDA}$  DOS for  $\text{BaFe}_2\text{As}_2$  (black lines) and  $\text{LaOFeAs}$  (light lines) compounds. Lower panel presents total DOS for different  $\text{FeAs}$  systems in the vicinity of the Fermi level. The Fermi level corresponds to zero.

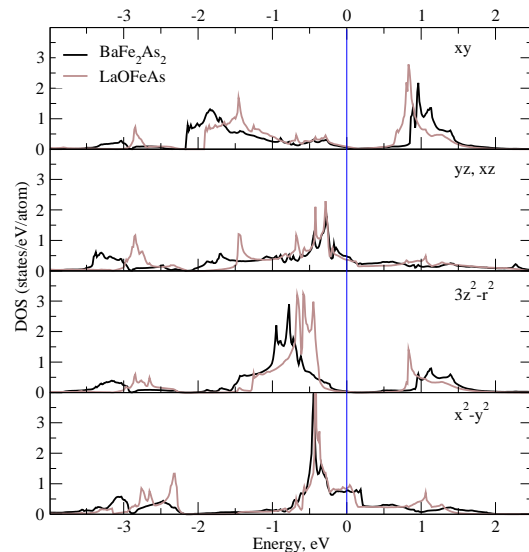


Fig. 3. Orbital projected  $\text{Fe-}d$  DOS for  $\text{BaFe}_2\text{As}_2$  (black lines) and  $\text{LaOFeAs}$  (light lines) compounds. The Fermi level corresponds to zero.

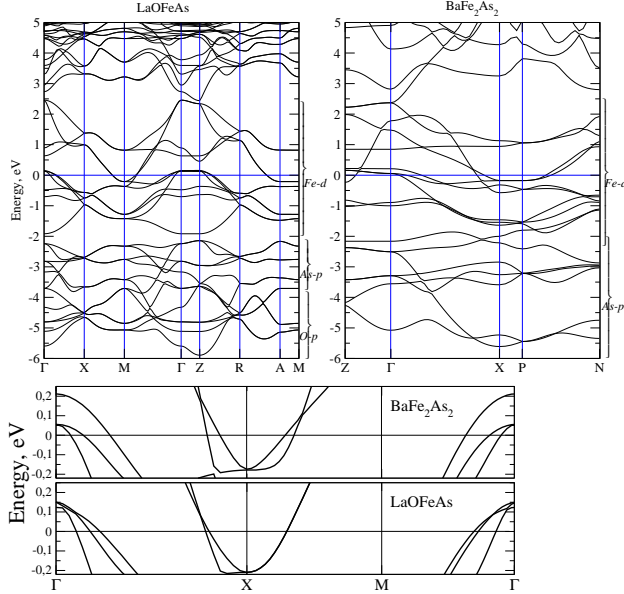


Fig. 4. Energy bands for LaOFeAs (left) and BaFe<sub>2</sub>As<sub>2</sub> (right) compounds. Lower two panels present  $k_x, k_y$  dispersions for BaFe<sub>2</sub>As<sub>2</sub> and LaOFeAs systems in the vicinity of the Fermi level. The Fermi level corresponds to zero.

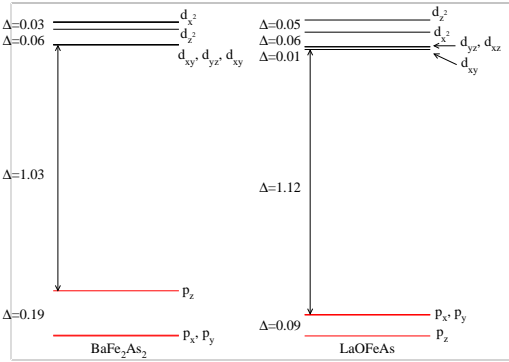


Fig. 5. Relative on-site energies of hybridized Fe-3d and As-4p states obtained from LDA dispersions for BaFe<sub>2</sub>As<sub>2</sub> (left) and LaOFeAs (right).  $\Delta$  stands for the corresponding energy distances in eV.

in the two-dimensional FeAs layer, which is the basic structural motif where superconducting state is formed. Thus, rather simple AFe<sub>2</sub>As<sub>2</sub> system may be considered generic for the studies of high-temperature superconductivity in whole class of FeAs-layered compounds.

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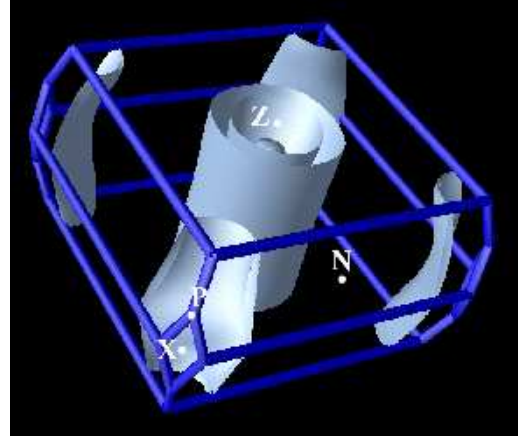


Fig. 6. Fermi surface of BaFe<sub>2</sub>As<sub>2</sub> shown in the first Brillouin zone centered at  $\Gamma$  point.

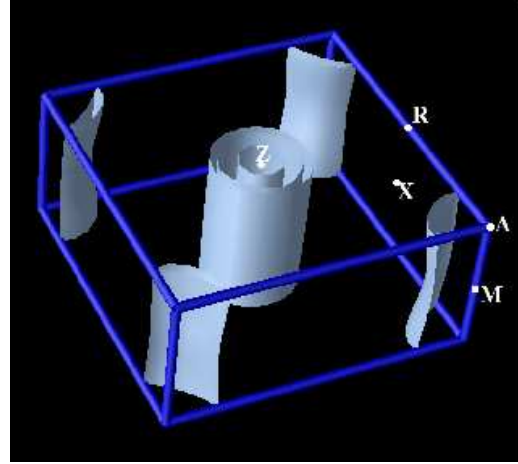


Fig. 7. The same as Fig. 6 but for LaOFeAs.

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