

Electronic structure of prototype AFe_2As_2 and 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 AFe_2As_2 ($A=\text{Ba}, \text{Sr}$) and compared it with previously calculated electronic spectra of ReOFeAs ($\text{Re}=\text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}$). 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 AFe_2As_2 are also very similar to that of ReOFeAs . This makes the more simple AFe_2As_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 26 K [1–4] and even more high values of $T_c = 41\text{--}55$ 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 38 K in potassium doped ternary iron arsenides BaFe_2As_2 [9] and SrFe_2As_2 [10], with further synthesis of superconducting AFe_2As_2 ($A=\text{K}, \text{Cs}, \text{K}/\text{Sr}, \text{Cs}/\text{Sr}$) [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 LaOFeAs were calculated in a number of papers (see e.g. [13, 14], [15]) producing results qualitatively similar to that first obtained for LaOFeP [16]. We have performed LDA calculations for the whole series of ReOFeAs ($\text{R}=\text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}$) [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 (~ 2 eV) around the Fermi level.

First LDA results for the density of states (DOS) of BaFe_2As_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 AFe_2As_2 ($A=\text{Ba}, \text{Sr}$) with the aim to compare it with the previously discussed ReOFeAs series. We present LDA DOS, energy disper-

sions 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 $A=\text{Ba}$ and $A=\text{Sr}$, as well as for the whole Re series, below we present data mainly for $A=\text{Ba}$ and $\text{Re}=\text{La}$.

Both BaFe_2As_2 and 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 $(\text{LaO})^+$. Fe^{2+} ions are surrounded by four As ions forming a tetrahedron. The crystal structures of BaFe_2As_2 and 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 BaFe_2As_2 undergoes structural phase transition from tetragonal ($I4/mmm$) to orthorhombic ($Fmmm$) space group [20]. The same transition takes place for 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. It can be seen that for BaFe_2As_2 compound the Fe-As distance is smaller than for LaOFeAs . So one would expect more considerable Fe-*d*-As-*p* hybridization for BaFe_2As_2 system in comparison with LaOFeAs and as a result wider Fe-*d* bandwidth. The distance between nearest Fe atoms within FeAs layers is also significantly smaller in BaFe_2As_2 as compared with LaOFeAs system. After the phase transition of BaFe_2As_2 system to the

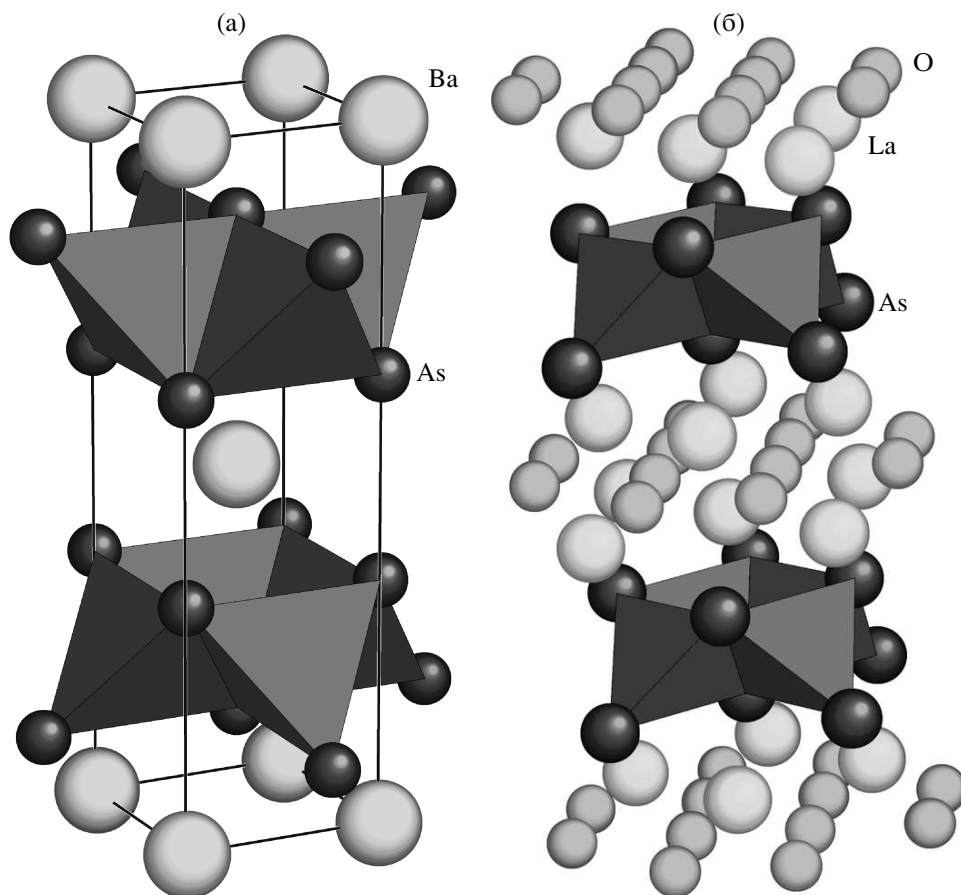


Fig.1. Crystal structure of BaFe_2As_2 and LaOFeAs . FeAs tetrahedra form two-dimensional layers (violet) sandwiched by Ba ion (cyan) or LaO (yellow and green) layers

Crystal structure data for BaFe_2As_2 and LaOFeAs compounds. Atomic positions for BaFe_2As_2 are Ba (0, 0, 0), Fe (0.5, 0, 0.25), As (0, 0, z) and for LaOFeAs are La(0.25, 0.25, z La), Fe (0.75, 0.25, 0.5), As (0.25, 0.25, z As), O (0.75, 0.25, 0)

Parameter	BaFe_2As_2	LaOFeAs
group	$I4/mmm$	$P4/nmm$
a , Å	3.9090(1)	4.03533(4)
c , Å	13.2122(4)	8.74090(9)
z_{La}	–	0.14154(5)
z_{As}	0.3538(1)	0.6512(2)
Source	Ref. [9]	Ref. [1]
Ba-As, Å	$3.372(1) \times 8$	–
La-As, Å	–	3.380×4
Fe-As, Å	$2.388(1) \times 4$	2.412×4
Fe-Fe, Å	$2.764(1) \times 4$	2.853×4
As-Fe-As	$109.9(1)^\circ$	113.6°
	$109.3(1)^\circ$	107.5°

orthorhombic structure the four equal Fe-Fe distances break into two bond pairs of 2.808 and 2.877 Å length. Moreover the two As-Fe-As angles are quite different in

the case of LaOFeAs system (113.6° and 107.5°) and have very close values ($\sim 109^\circ$) for BaFe_2As_2 . Such differences in the nearest surrounding of Fe ions should evoke the distinctions in the electronic structure of these two compounds.

The electronic structure of BaFe_2As_2 and LaOFeAs was calculated within the local density approximation (LDA) by using linearized muffin-tin orbitals basis (LMTO) [22]. For BaFe_2As_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 BaFe_2As_2 and 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.

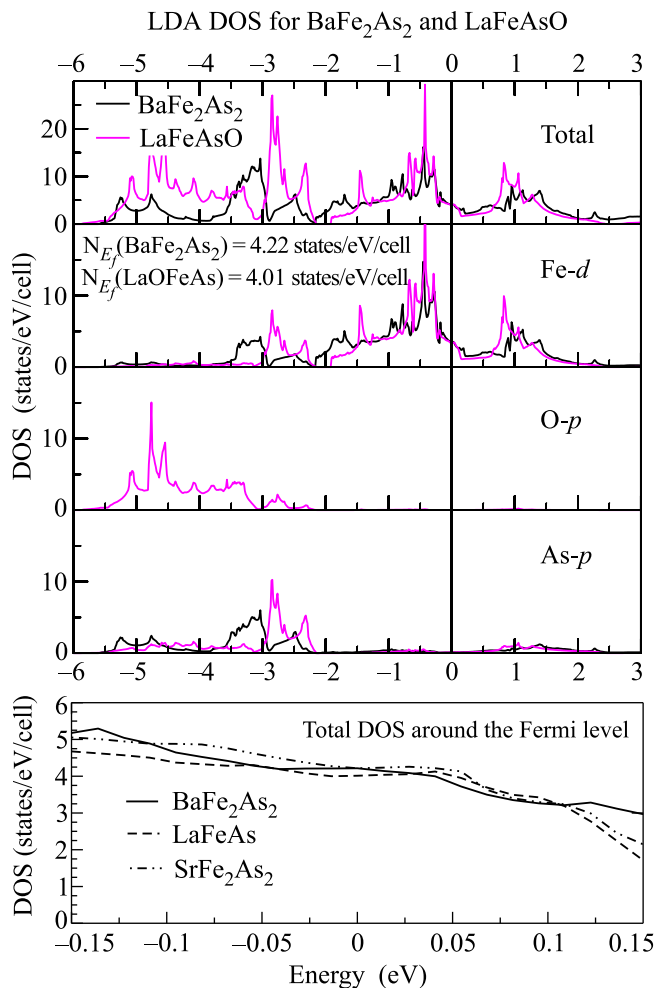


Fig.2. Total and partial LDA DOS for BaFe_2As_2 (black lines) and LaOFeAs (light lines) compounds. Lower panel presents total DOS for different FeAs systems in the vicinity of the Fermi level. The Fermi level corresponds to zero

The orbital projected Fe- d DOS for two compounds is shown in Fig.3. 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 BaFe_2As_2 in comparison with 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. 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 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 LaOFeAs system As- p

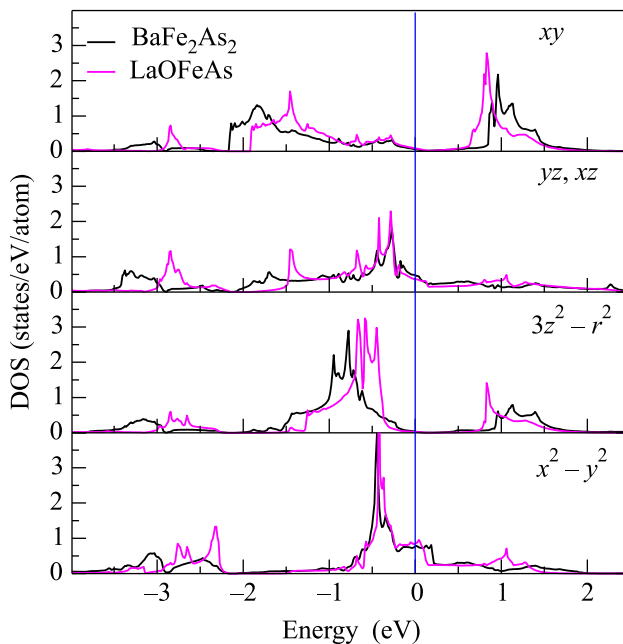


Fig.3. Orbital projected Fe- d DOS for BaFe_2As_2 (black lines) and LaOFeAs (light lines) compounds. The Fermi level corresponds to zero

states are also hybridized with O- p states and the corresponding bands are separated from the Fe- d ones. On the contrary in BaFe_2As_2 Fe- d and As- p bands are entangled. The lower two panels of Fig.4 compare band dispersions for both systems 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 Γ -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 LaOFeAs there are two degenerate bands.

In Fig.5 relative on-site energies of hybridized Fe- $3d$ and As- $4p$ states are presented. A bird’s eye view tells us that this picture for both BaFe_2As_2 (left) and LaOFeAs (right) is rather similar. There are two groups of states – antibonding (mostly Fe- $3d$) and bonding (mostly As- $4p$) states. However, there are some fine differences. First of all for BaFe_2As_2 hybridization between Fe- $3d$ - z^2 and As- $4p_z$ orbitals is about 0.4 eV weaker. It leads to a swap of the energy positions of Fe- $3d$ - z^2 and x^2 orbitals and similarly for corresponding As- $4p$ orbitals. Secondly, Fe- d - t_{2g} orbitals are degenerate for BaFe_2As_2 in contrast to LaOFeAs .

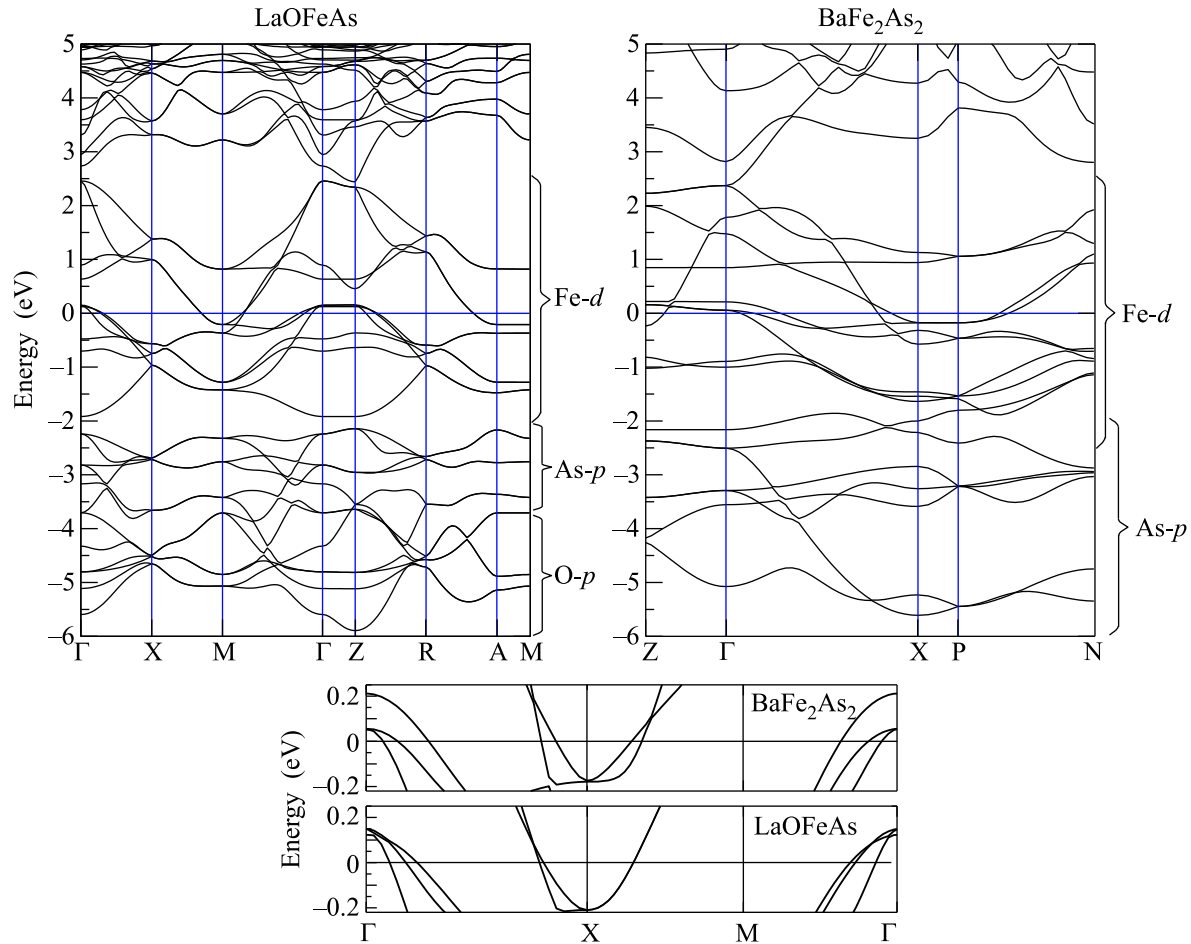


Fig.4. Energy bands for LaOFeAs (left) and BaFe₂As₂ (right) compounds. Lower two panels present (k_x, k_y) dispersions for BaFe₂As₂ and LaOFeAs systems in the vicinity of the Fermi level. The Fermi level corresponds to zero

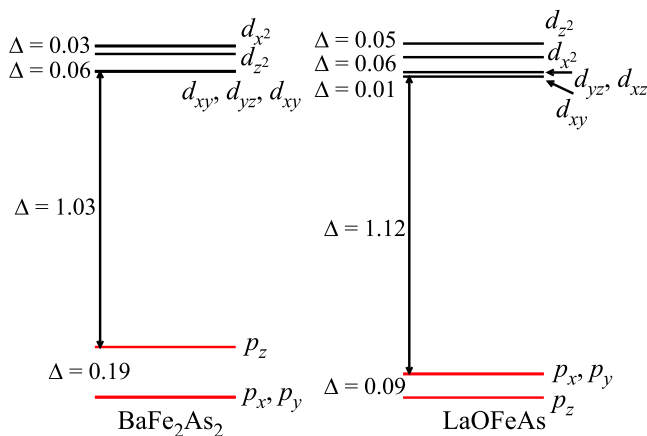


Fig.5. Relative on-site energies of hybridized Fe-3d and As-4p states obtained from LDA dispersions for BaFe₂As₂ (left) and LaOFeAs (right). Δ stands for the corresponding energy distances in eV

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 Fe-*d* states of FeAs layers, making the states of A-ions or rare-earths Re more or less irrelevant for superconductivity. Thus, superconductivity of FeAs layered compounds may be studied within the minimal model, taking into account only essential Fe-*d* bands close to the Fermi level. The variants of such model proposed e.g. in Refs. [23, 24] for LaOFeAs system may also be used for AFe₂As₂ with only slight modification of model parameters, such as transfer integrals.

The role of electronic correlations 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 Fe. However, LDA+DMFT calculations of 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 LDA approach used here is reliable enough.

Fermi surfaces obtained from LDA calculations for $BaFe_2As_2$ and $LaOFeAs$ are shown in Figs. 6 and 7, correspondingly. There are five sheets of Fermi surface

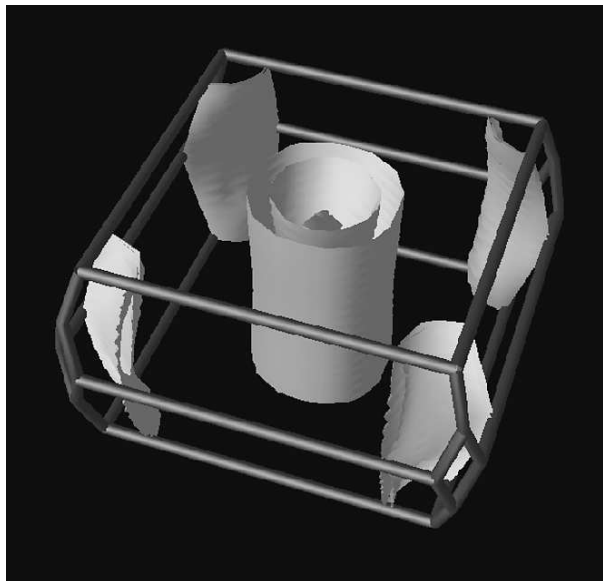


Fig.6. Fermi surface of $BaFe_2As_2$

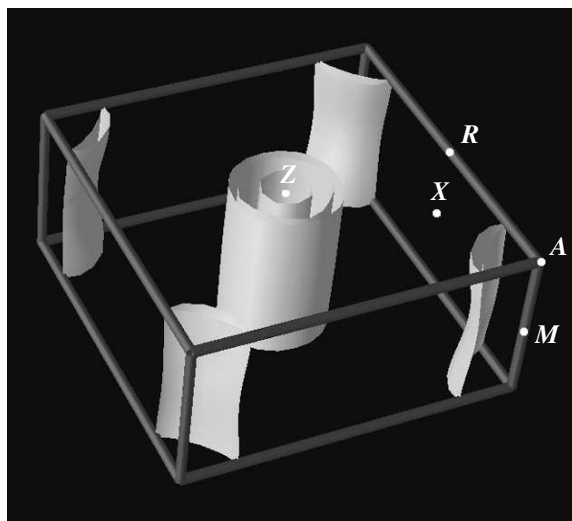


Fig.7. Fermi surface of $LaOFeAs$ shown in the first Brillouin zone centered at Γ point

for both compounds. Qualitatively, Fermi surfaces are similar to that reported for $LaOFeAs$ 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.

Tetragonal to orthorhombic phase transition taking place in undoped compounds is usually attributed to 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 $LaOFeAs$ related with apparently itinerant nature of magnetism were recently discussed in Ref. [28].

In conclusion, we have presented the results of LDA calculations of new prototype high-temperature superconductor AFe_2As_2 ($A=Ba, Sr$) and compared it with previously discussed $ReOFeAs$ series, demonstrating essential similarity of electronic states close to the Fermi level and most important for superconductivity. These states are formed mainly by Fe orbitals in the two-dimensional FeAs layer, which is the basic structural motif where superconducting state is formed. Thus, rather simple AFe_2As_2 system may be considered generic for the studies of high-temperature superconductivity in whole class of FeAs-layered compounds.

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