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Research Paper

Lifshitz Transition on $SrFe_2As_2$ and $SrFe_2Bi_2$ Compounds Under Pressure

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In this work, we analyzed the structural, electronic, magnetic and bonding properties of $ThCr_2Si_2$-type compounds, namely $SrFe_2As_2$ and $SrFe_2Bi_2$ with space group I4/mmm (139) under pressure by means of Full Potential-Linearized Augmented Plane Wave (FP-LAPW) method as implemented in WIEN2K code. The optimized lattice parameters, the positional z-parameter, Fermi energy, Density of States (DOSs) at Fermi Level at equilibrium lattice configuration and under compressions are reported. Electron density plots, band structure plots, Density of States histograms (DOS) are illustrated and compared. Different bonding characteristics are noted and it shows anisotropic bonding nature of these compounds. High pressure band structure calculations reveal the possibility of phase transition in these compounds. On application of pressure, the parent $SrFe_2As_2$ compound undergoes structural transition at 10.65 GPa and it agrees well with the experimental value. The novel/ hypothetical $SrFe_2Bi_2$ compound shows a structural transition at compression of 15% of volume (11.84 GPa) from ambient condition and band structure plots provide an evidence of improving conductivity under pressure.

Keywords: Compounds; Lifshitz Transition; High Pressure Study; DFT; Anisotropic Bonding

Introduction

$ThCr_2Si_2$-type compounds crystallize in the well known tetragonal structure with space group I4/mmm (139). These compounds show a variety of interesting properties such as phase transition, different order of magnetism such as diamagnetism, Pauli paramgnetism, valence paramagnetism, Van Vleck paramagnetism, ferromagnetism and antiferro-magnetism etc., (Jeitschko *et al*., 1987), low temperature and high temperature superconductivity and coexistence of superconductivity with anti ferromagnetism. Some $ThCr_2Si_2$-type compounds exhibit superconductivity and conductivity has been improved under chemical doping and under the application of external pressure. Some of the recent research of 122 compounds under pressure are as follows: $BaMn_2As_2$ attains metallization at 5.8 GPa with sharp fall in resistivity indicating a possibility of high conductivity (Satya *et al*., 2011); $CaFe_2As_2$ attains collapsed tetragonal phase under hydrostatic pressure condition (Tuson *et al*., 2008). The collapsed tetragonal phase transition is the sudden change of the *c* lattice parameter and a subsequent increase of the in-plane *a* lattice constant without variation of space group either by doping with 'Th' site or under pressure (Sanna *et al*., 2012). $BaFe_2As_2$ compound revealed a bulk superconductivity under a critical pressure of 2.8 GPa (Awadesh Mani *et al*., 2008); $EuCo_2As_2$ compound changed from tetragonal phase to collapsed tetragonal phase at 4.7 GPa (Matthew Bishop *et al*., 2009); $SrFe_2As_2$ compound loses its resistivity and attains high conductivity at 3 GPa (Kumar *et al*., 2008); $SrFe_2As_2$ compound attains phase transition at 10-18 GPa (Walter O Uhoya *et al*., 2011); possibility of Lifshitz transition in $CaNi_2As_2$ compound at 20 GPa (Jayalakshmi *et al*., 2013), $EuFe_2P_2$ exhibit a second order phase transition under pressure whereas first order structural phase transition is noticed in the compound $EuCo_2P_2$ (Huhnt *et al*., 1997 and 1998). Moreover, the heavier pnictides of antimonides and bismuthides may have a high conductivity compared to the smaller arsenide and phosphide (Saparov *et al*., 2013). Hence this study motivates us to compare the change in conductivity between arsenide and

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bismuthide compounds under pressure.

The main aim of this work is to study the structural, electronic, magnetic and bonding properties of compounds namely $SrFe_2As_2$ and $SrFe_2Bi_2$ with space group I4/mmm (139) under pressure by means of Full Potential-Linearized Augmented Plane Wave (FP-LAPW) method as implemented in WIEN2K code. The motive of the study is to investigate the structural transition and analyze the conductivity nature of the compounds under pressure.

Materials and Methodology

The calculations are carried out by means of Full Potential Linearized Augmented Plane Wave (FP-LAPW) method as implemented in the WIEN2K code (Schwarz *et al*., 2010). The generalized Gradient Approximation (GGA) parameterized by Perdew *et al*. (1966) has been applied to calculate the exchange correlation potential. Muffin tin spheres of radius RMT are chosen in such a way that there is minimal charge leakage from the core. The calculations are performed with 1000 k-points in the Brillouin zone. The plane wave expansion ($R_{MT} * K_{MAX}$) is taken as 7. The calculations are performed with full lattice optimization including Z_{Pn} coordinates. The atomic positions for Sr is (0, 0, 0), Fe is (0, 0.5, 0.25) and for arsenic and bismuth is (0, 0, $Z_{As,Bi}$). Here $Z_{As,Bi}$ is the so-called internal coordinate distance. The self-consistent calculations are carried out to an accuracy of 0.0001 Ry, 0.1 mRy and 0.001 |e| for energy, force and charge respectively. The computations are performed to find the total energy of the given system and the obtained total energies are then fitted into the Birch-Murnaghan equation of state (F. Birch 1947) to get the pressure – volume relation. The pressure is obtained by taking the volume derivative of the total energy. The theoretically calculated equilibrium lattice parameters are then used to optimize the position of the Pn atoms. The computation is repeated with optimization for each compression and expansion of volume from V/V_0 = 1.20 to V/V_0 = 0.85 for both the compounds namely $SrFe_2As_2$ and $SrFe_2Bi_2$.

Results and Discussion

High Pressure Band Structure Study in $SrFe_2As_2$ Compound

The corresponding pressure experienced by the $SrFe_2As_2$ compound at antiferro magnetic state for V/V_0 ratio (1.25-0.85 in steps of 10 % of compression) is obtained from the volume derivative of the Birch-Murnaghan equation of state. This compound is experimentally reported, to verify the same results, the compression is taken in steps of 10%. For each volume, a and c lattice parameter were calculated from c/a ratio of the ambient condition and optimization of position z_{As} was carried out because of the distortion produced inside the change in the volume. All the observed structural and electronic parameters are given in Table 1 for $SrFe_2As_2$ compound at various pressures. We observe from Table 1 that $N(E_F)$ is fluctuating under pressure in $SrFe_2As_2$ compound; value of $N(E_F)$ at V/V_0 = 0.85 reaches the maximum high value 90.28 (states/Ryd) while at V/V_0= 0.95 the $N(E_F)$ value decreases, then till V/V_0 = 1.25 the value gradually increases. This kind of fluctuation in $N(E_F)$ leads to structural

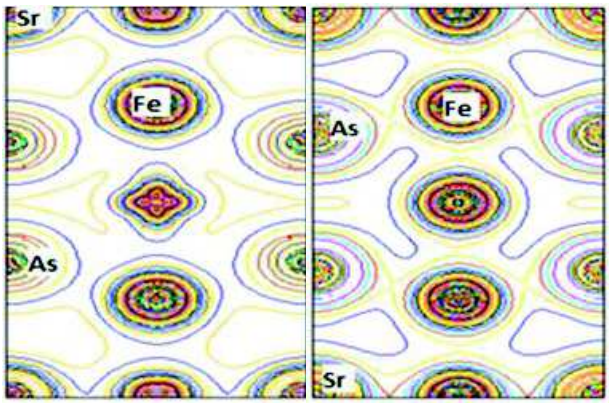
Table 1: Structural and positional Parameters a, b, c and $Z_{As,}$ distance between Fe-As atoms in intralayer d_{Fe-As} ,distance between As-As atoms in interlayer d_{As-As}, Fermi Energy E_F, Density of States $N(E_F)$ and total energy of $SrFe_2As_2$ compound

V/Vo (Pressure in GPa)/ Parameter	0.8 5(10.65)	0.95 (5.002)	1.05 ((-)1.986)	1.15 ((-)13.017)	1.25 ((-)20.94)
a = b (a.u.)	8.9147	9.2514	9.4109	9.8598	9.9447
c (a.u.)	18.5123	19.2114	19.5431	20.4749	20.6513
Z_{As}	0.3505	0.3466	0.3505	0.3470	0.3230
d_{Fe-As} (Bohr)	4.8300	5.0125	5.0989	5.3149	5.1959
d_{As-As} (Bohr)	5.5352	5.5964	5.8443	6.2653	7.3106
E_F (Ryd.)	0.7153	0.61924	0.5778	0.4719	0.4515
N (E_F) (States/Ryd.)	90.28	82.43	82.80	86.31	86.75
Total Energy (Ryd.)	-25586.0830	-25586. 0865	-25586.4426	-25585.9959	-25585.8236

transition (I. M. Lifshitz *et al*., 1960). Even though there is no change in space group of the compound at respective volume. Hence it is expected that there is an existence of transition within the tetragonal structure. The obtained results and literature studies (Zhao *et al*., 2018) evidenced that the tetragonal structure changed to the collapsed tetragonal structure under the pressure 10.65 GPa ($V/V_0 = 0.85$) and the same is explained in the following section. This result shows good agreement with the existing literature (Walter O Uhoya *et al*., 2011).

Bonding Properties of $SrFe_2As_2$ Compound Under Pressure

The interaction between pnictogens of the compound under study is explored in terms of strength and bonding between them. This is explained using the electron density plots drawn at ambient condition (Sundareswari *et al*., 2016) and at pressures 10.65 GPa ($V/V_0 = 0.85$-transition state) and (-) 13.02 GPa ($V/V_0 = 1.15$-stabled state with minimum energy) and is shown in Fig. 1. In Fig. 1(A), there is no charge density contours around Fe-As atoms, it indicates the existence of weak bonding between them and less stability of the compound. The same point is validated by density of states in the following section. In Fig. 1(B), the contours around Fe-As layer show the covalent bonding. The contours around Fe-As layers indicate metallic bonding in between the layers. Fe-As layer and Sr involves ionic bonding. Thus the charge density plots exhibits a mixed covalent, ionic and metallic behavior under pressure. This anisotropic bonding nature of the compound shows good agreement with the existing literature (I.R. Shein *et al*., 2009).



(A) **(B)**

Fig. 1: Electron Density Plot of $SrFe_2As_2$ in 100 plane (a) V/V_0=0.85 (b) V/V_0=1.15

Electron Transport Properties of $SrFe_2As_2$ Compound Under Pressure

To know the electron transport behavior, the densities of states (DOS) of the compound at various compressions are drawn for $SrFe_2As_2$ compounds and shown in Fig. 2. By comparing these figures, one could observe that the Fermi energy lies in the peak (Fig. 2A) at 10.65 GPa ($V/V_0 = 0.85$) which shows the unstable nature of the compound. At expansion of volume 1.15 (V/V_0) Fermi energy gaps lie in the deep valley (pseudo gap) and it indicates the large stability (Moon *et al*., 2014) in structure (Fig. 2B).

Band structure plot (Fig. 3) are drawn to further study about the transport behavior of electron and conducting nature variation in compound at ambient and under pressure conditions. At 10.65 GPa ($V/V_0 = 0.85$) the band at gamma and H point is located below the Fermi level and its corresponding $N(E_F)$ is 90.28 states/Ryd (Fig. 3A) and hence it shows predominant $N(E_F)$ value shown in Table 1. At pressure 13.017 GPa ($V/V_0 = 1.15$) it started to move away from the Fermi level (Fig. 3B), hence $N(E_F)$ value is decreased. Hence it is proved that metallization and conductivity of these compounds are enhanced under 10.65 GPa pressure.

To validate our observations, electrical conductivity (σ) is calculated by using Boltzmann code interfaced to the Wien2k program at ambient temperature with constant relaxation time (Madsen *et al*. (2006) and the values are listed is Table 2. These values explicitly show that, $SrFe_2As_2$ has higher σ value (1.19064 M $\Omega^{-1}m^{-1}$) at 15 % of compression (10.65 GPa) than that at ambient condition (0.84932 M $\Omega^{-1}m^{-1}$).

Table 2: Conductivity values (σ) of $SrFe_2As_2$ and $SrFe_2Bi_2$ compounds at ambient condition and under pressure are listed in Table 2

V/V_0 Compounds	0.85 ($10^6\,\Omega^{-1}m^{-1}$)	1.0 ($10^6\,\Omega^{-1}m^{-1}$)	1.15 ($10^6\,\Omega^{-1}m^{-1}$)
$SrFe_2As_2$	1.19064	0.84832	0.44776
$SrFe_2Bi_2$	1.22143	0.90586	0.82371

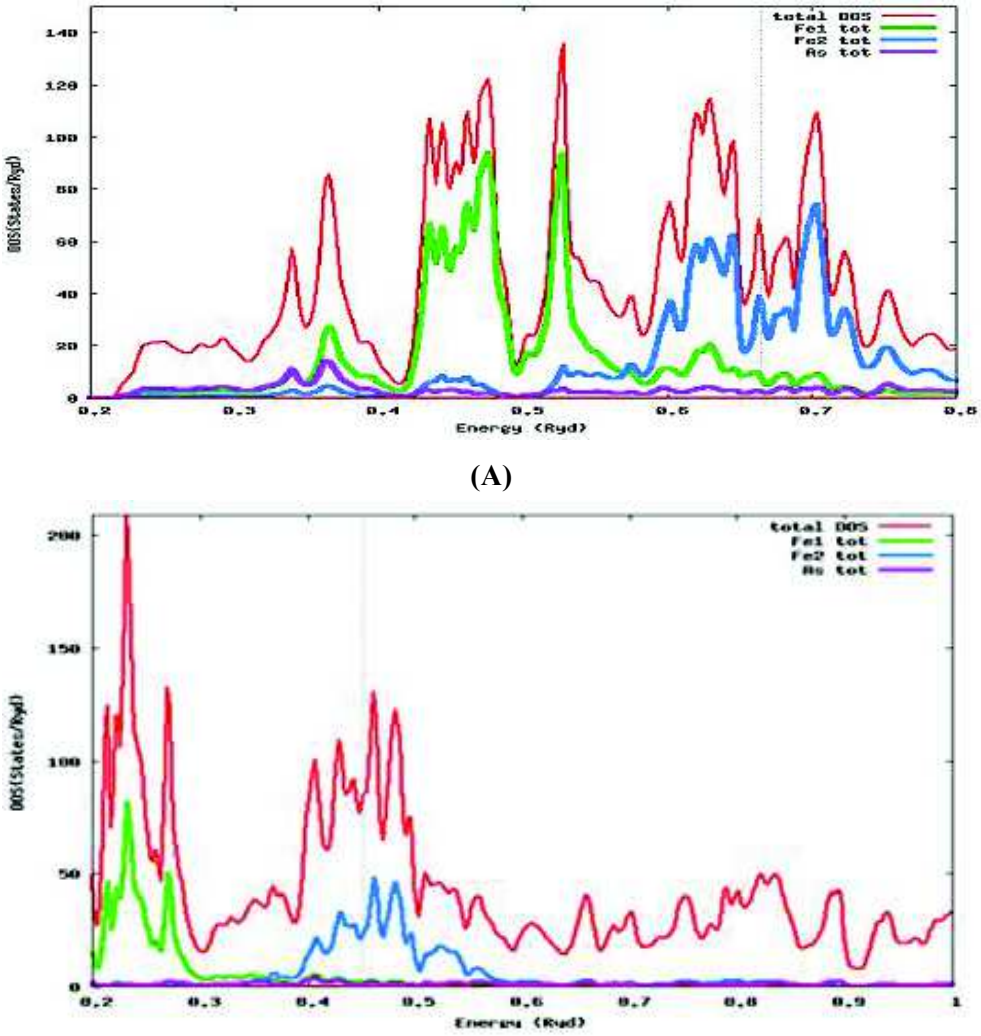


Fig. 2: Density of States histograms of $SrFe_2As_2$ (A) V/V_0=0.85 (B) V/V_0=1.15

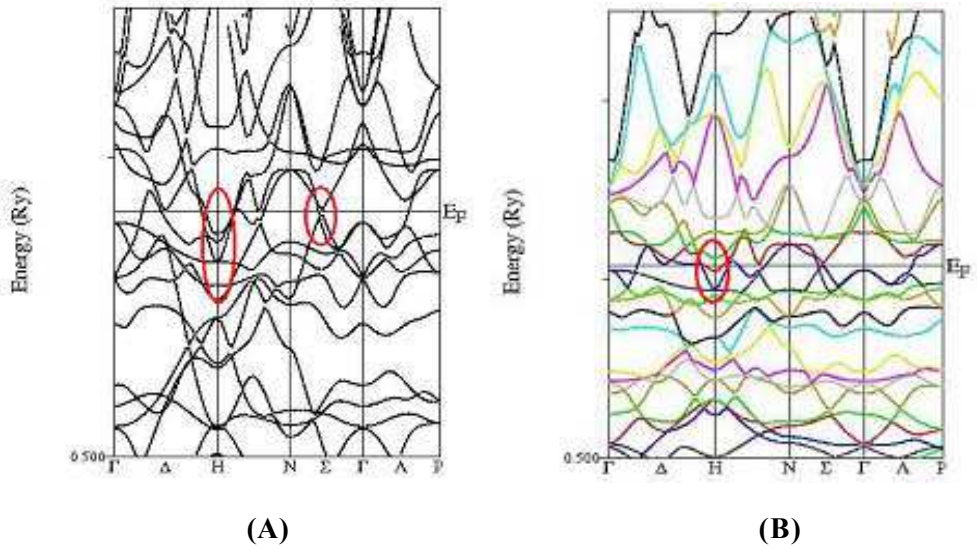


Fig. 3: Band structure plot of $SrFe_2As_2$ (A) V/V_0=0.85 (B) V/V_0=1.15

Replacing of As with Bi in $SrFe_2As_2$ compound

The heavier pnictides of bismuthides may possibly have more conductivity and higher T_c compared to the smaller arsenide (Saparov.B *et al.*, 2013). Hence, in this study arsenide is replaced by bismuthides and it is known as $SrFe_2Bi_2$ compound. The similar structural optimization of parent compound namely $SrFe_2As_2$ has been done in $SrFe_2Bi_2$ compound. The crystal structure of $SrFe_2Bi_2$ compound has tetragonal structure. It is isostructure with its parent compound and its space group remains same.

All the observed structural and electronic parameters are given in Table 3 for $SrFe_2Bi_2$ compound at ambient condition and under pressure in steps of 5% volume compression from 1.20 to 0.85. Since this compound is novel, to observe the complete changes, the compression has been taken in steps of 5 %. We observe from Table 3 that $N(E_F)$ is fluctuating under pressure in $SrFe_2Bi_2$ compound as like its parent compound.

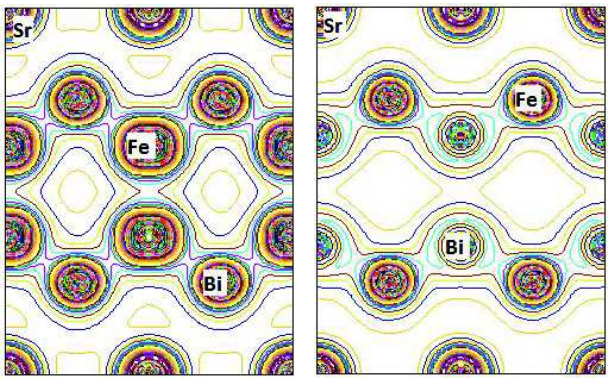
Table 3: Structural and positional Parameters a, b, c and $Z_{Bi,}$ distance between Fe-Bi atoms in intralayer $d_{Fe\text{-}Bi,}$ distance between Bi-Bi atoms in interlayer $d_{A\text{-}As,}$ Fermi Energy $E_{F,}$ Density of States at fermi level $N(E_F)$ and total energy of $SrFe_2Bi_2$ compound are listed in Table 3

V/V_0 (Pressure in GPa)/ Parameter	0.85 (11.84)	0.90 (7.23)	0.95 (3.87)	1.0[a] (0.41)	1.05 ((-) 3.24)	1.10 ((-)12.76)	1.15 ((-)21.32)	1.20 ((-)25.48)
a (a.u.)	7.9906	8.1443	8.2924	8.4354	8.5737	8.7077	8.8377	8.9648
c (a.u.)	22.1244	22.550	22.9599	23.356	23.7390	24.1099	204.469	24.8219
Z_{Bi}	0.3751	0.3595	0.3595	0.3595	0.35285	4.9813	0.3482	0.3469
$d_{Bi\text{-}Fe}$ (Bohr)	4.8603	4.7623	4.8489	4.9325	4.9334	4.9813	5.0299	5.0870
$d_{Bi\text{-}Bi}$ (Bohr)	5.5267	6.3366	6.4517	6.5631	6.9864	7.2146	7.4288	7.6005
E_F (Ryd.)	0.6833	0.63783	0.59215	0.5465	0.51659	0.48362	0.45358	0.42513
$N(E_f)$ (states/Ryd.)	77.27	36.57	39.37	43.50	43.70	48.12	49.59	53.70
Total Energy (Ryd.)	–97776. 34902	–97776. 58532	–97777. 00062	–97777. 05247	–97777. 47903	–97777. 63180	–97777. 85087	–97777. 82873

[a]Sundareswari *et al* (2016)

Bonding Properties of $SrFe_2Bi_2$ Compound Under Pressure

To probe into the bonding properties of these compound, charge density plots along (100) plane are plotted and analyzed as shown in Fig. 4. From the electron density plots one can find the uniformly distributed directional charge density contours that enclose Fe and Bi atoms in $SrFe_2Bi_2$ compound at (-) 21.32 GPa (V/V_0=1.15 - stable state with minimum energy) and the same indicates the large stability regime. The same is substantiated in DOS and band structure plots, in the following section. The existence of such contours is an indication of the presence of covalent bonding. No such directional charge density contours, hence no strong covalent bonding between the pnictogens can be found at 11.84 GPa (V/V_0=0.85 – transition state). The spherical contours around Bi atom show the metallic nature of the compound. The electron clouds around Fe_2Bi_2 blocks are separated from Sr and it indicates the ionic interaction. Thus the charge density plots exhibits a mixed covalent ionic and metallic behavior under pressure alike its group of compounds. (Jayalakshmi *et al*., 2013a).



(A) **(B)**

Fig. 4: Electron Density plot of $SrFe_2Bi_2$ (A) V/V_0=0.85 (B) V/V_0=1.15

Electron Transport Properties of $Srfe_2bi_2$ Compound Under Pressure

To know the electron transport behavior of $SrFe_2Bi_2$ compound, the densities of states (DOS) histograms and band structures are plotted and shown in Fig. 5 and Fig. 6. In $SrFe_2Bi_2$ compound, at V/V_0=0.85 (Fig. 5A) the Fermi level lies at peak and it represents that $N(E_F)$ is high (77.27 states/Ryd.). At V/V_0=1.15 (Fig. 5B) the Fermi level lies in the deep valley and it represents that $N(E_F)$ is minimum (49.59 states/Ryd), hence it attains larger stability. At ambient condition (V/V_0=1.0), the DOS at Fermi level is 43.50 states/Ryd. Hence it is observed that the trend of $N(E_F)$ is decreasing from V/V_0=0.85 to V/V_0=1.0 and above the ambient condition the trend tends to increasing. This kind of fluctuation in $N(E_F)$ leads to structural transition (I.M. Lifshitz *et al*., 1960). In this compound, the transition occurs over the lattice parameters and space group remains same. Hence it is marked as collapsed tetragonal phase transition.

From the Band structure plots it is observed that

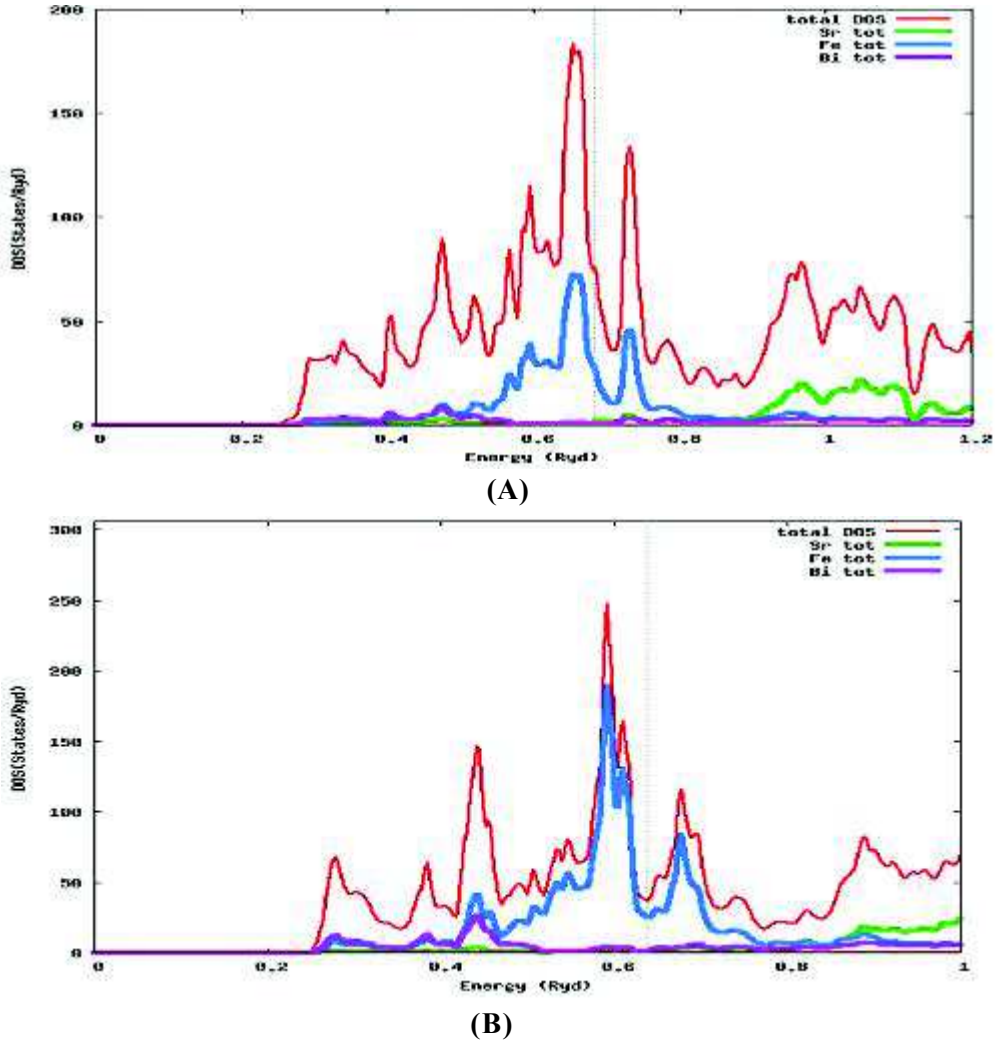


Fig. 5: Density of States of $SrFe_2Bi_2$ (A) V/V_0=0.85 (B) V/V_0=1.15

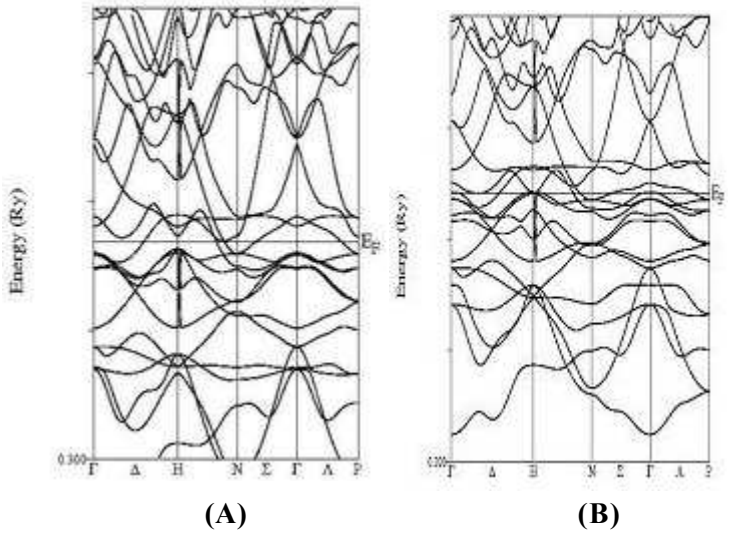


Fig. 6: Band structure plot of $SrFe_2Bi_2$ (A) V/V_0=0.85 (B) V/V_0=1.15

at V/V_0=1.15 (Fig. 6B), the Fe-d and Bi-p bands lie very near to Fermi level at H and gamma point. At successive volume compression the bands move nearer to Fermi level. At 15% compression (V/V_0=0.85) the bands are crossing the Fermi level at H and gamma points and its $N(E_F)$ value is high (Fig. 6A). Hence, it is observed that if the pressure increases the number of band crossing into the Fermi level increases, it shows that the conductivity increases under compression. The same has been validated by the calculated electrical conductivity (σ) by using Boltzmann code (Table 2) that $SrFe_2Bi_2$ has high σ value (1.22143 M $\Omega^{-1}m^{-1}$) at 15 % of compression than that at ambient condition (0.90586 M $\Omega^{-1}m^{-1}$) and successive expansion of volume.

Conclusion

At 10.65 GPa, DOS fluctuations indicates that $SrFe_2As_2$ compound completely transforms to a high pressure collapsed tetragonal structure within the same space group and this agreed well with the existing literature. Number of bands those crossing across the Fermi level increases in $SrFe_2As_2$ compound under pressure. Hence, the conductivity increases when pressure increases and the compound behaves more metallic. From electron density plots of $SrFe_2As_2$ compound it is understood that the compound has anisotropic nature and this agreed well with the existing literature.

At ambient condition, the Fermi level of $SrFe_2Bi_2$ compound lies on the pseudo region which shows the higher stability of the compound. At pressure, we could encounter the DOS fluctuations in $SrFe_2Bi_2$ compound that may lead to phase transition in the compound under study at 11.84 GPa. This indicates about the enhancement of electrical conductivity and metallic behavior of $SrFe_2Bi_2$ under compression and exhibits anisotropic bonding character like its parent compound. The present study concludes that the structural transition in $SrFe_2As_2$ compound is possible under compression of 15 % volume at 10.65 GPa and $SrFe_2Bi_2$ compound under 15 % of compression of volume ($V/V_0 = 0.85$) at 11.84 GPa.

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