

FIRST PRINCIPLES STUDY OF $\text{Ba}(\text{Fe}_{1-x}\text{Ni}_x)_2\text{As}_2$ SUPERCONDUCTOR

Kamaliati Hanum Kamaruddin¹, Noriza Ahmad Zabidi², Ahmad Nazrul Rosli³,
Muhd Zu Azhan Yahya¹ and Mohamad F. Mohamad Taib⁴

¹*Defence Science Department, Faculty of Defence Science and Technology,
Universiti Pertahanan Nasional Malaysia, Kuala Lumpur 57000, Malaysia*

²*Centre for Foundation Studies, Universiti Pertahanan Nasional Malaysia,
Kem Sungai Besi, Kuala Lumpur 57000, Malaysia*

³*Faculty of Science and Technology, Universiti Sains Islam Malaysia,
Nilai 71800, Negeri Sembilan*

⁴*Faculty of Applied Sciences, Universiti Teknologi MARA,
Shah Alam 40450, Selangor, Malaysia*

Corresponding author: kamaliatihanum@gmail.com

ABSTRACT

The band structure and density of states of $\text{Ba}(\text{Fe}_{1-x}\text{Ni}_x)_2\text{As}_2$ has been studied using density-functional theory (DFT) calculation within generalized-gradient approximation (GGA) with Perdew-Perke-Ernzerhof (PBE) exchange correlation functional. We report a comparison study for spin and non-spin polarization band structure shows such anisotropic is a fundamental property of the iron based compounds. Our result suggests that ferromagnetic configuration upon introducing Nickel (Ni) to the parent compound of BaFe_2As_2 . The ferromagnetic behavior transforms into the superconducting states as the temperature of cooling increases. The density of states DOS is mostly contributed by the *d* bands of the Fe and Ni ions. The contributions of the electron valence by Ni doping results in shifted of energy bands level and DOS to the lower energy values. The results can serve as a useful approximation in studying general features of the electronic structure.

Keywords: Iron pnictide superconductor; first principles; band structure; energy gap;

INTRODUCTION

Recently, iron oxypnictides have brought a lot of attention to compound containing FeAs layer after the discovery of its high transition temperature, $T_c \sim 55$ K [1]. Similar to the cuprate superconductors, the FeAs-based superconductors have to be doped or, differently from the cuprates, have to be set under pressure to yield superconductivity. A further difference between cuprates and iron pnictides is that the parent compounds in the latter are not

antiferromagnetic Mott-Hubbard insulators but metals with an antiferromagnetic ordering [2-4]. High T_c superconductivity has been found in many related phases that can be generally to their parent compounds: (i) LaFeAsO, (ii) BaFe₂As₂, (iii) LiFeAs and (iv) α -FeSe [5]. The precise mechanism of superconductivity in these compounds is yet to be established, but is strongly thought to be unconventional.

The parent compounds have spin-density wave (SDW) order with a possible exception BaFe₂As₂ phase, exist in a tetragonal structure exhibit static antiferromagnetic (AF) long-range order with a collinear spin structure [6]. Doped BaFe_{1.9}Ni_{0.1}As₂ with $T_c = 20$ K was confirmed by clear dispersion of the mode energy possess iron in a three-dimensional (3D) character as compared to conventional iron arsenide superconductor which is having two-dimensional (2D) nature [7]. Electron-doped superconducting materials suggest static AF long-range order is completely suppressed rather than large anisotropy spin gaps found in the parent compounds. It is generally believed that magnetism plays an important role in the superconductivity of these materials.

Several experimental determination on the superconducting, electronic, magnetic and thermal properties of electron-doped BaFe_{2-x}T_xAs₂ (T = Ni, Co) has been done [8-10]. These compounds have many complex phase diagrams including magnetic, structural, and superconducting phase transitions. The intriguing phenomenon is the existence of superconducting state with Fe and other magnetic ions, where small doping of these elements completely destroys the superconductivity in conventional superconductors and the high- T_c cuprates. Superconducting state of BaFe_{2-x}Ni_xAs₂ emerges about $x = 0.05$ upon Ni doping and reach to a maximum T_c value of 20.5 K at the optimal doping level $x = 0.1$ where the AF state is completely suppressed. The experimental studies also show that for $x = 1.0$ there is no superconducting state to 0.4 K [8] and for $x = 2.0$ the existence of superconducting is found at T_c about 0.7 K [10].

In this paper, we present the details of our calculations of the electronic structure results of Ni doping in the crystal structure of BaFe₂As₂. The band structure and density of state (DOS) of BaFe_{2-x}Ni_xAs₂ as a function of Ni doping x is similar to that reported.

EXPERIMENTAL

Computational details

The first principle geometry optimization of BaFe_{2-x}Ni_xAs₂ structure has been carried out using the CASTEP [11] package. The calculations are done within generalized gradient approximation (GGA) [12] using Perdew Burke Enzerhof (PBE) [13]. Results have been obtained by using ultrasoft pseudopotential [14] and plane wave basis set with kinetic-energy cut off $\hbar^2|\mathbf{k} + \mathbf{G}|^2/2m = 300$ eV. Brillouin zone is sampled in the $\mathbf{k}$

space within Monkhorst-pack scheme [15] in self-consistent potential for all the calculations. The numbers of these k points are the grid size of ($6 \times 6 \times 2$) for $I4/mmm$ space group. Atomic positions in all structures are optimized. Convergence of structure optimization is achieved when the difference of the total energies of last two consecutive steps is less than 10^{-6} eV and the maximum force allowed on each atom is less than 10^{-3} eV/Å. The pressure is less than 0.05 kbar.

Crystal Structures

The unit cell of the tetragonal structure is having the ZrCuSiAs-type structure, isostructural with 122-type iron pnictides BaFe_2As_2 [1] in the space group of tetragonal $I4 / mmm$ obtained from the Rietveld fitting of the x-ray diffraction pattern [8]. The Ba atom is found at position $2a$ site, Fe occupied $4d$ site and As atoms at position $4e$ site. The crystallographic data of parent compound BaFe_2As_2 as a function of Ni doping are listed in the Table 1. A primitive unit cell which only included one formula unit cell is shown in Figure 1(a) for the BaFe_2As_2 tetragonal structure. We induce electron-doped $x = 0.0, 1.0$ and 2.0 to the FeAs layer which adopts a well-defined lattice parameter. The c -axis lattice constant decreases with increasing x which indicates that the distribution of Ni doping is quite uniform except $x = 0.0$ is less than 4% (relative difference) smaller than the reported experimental data [8].

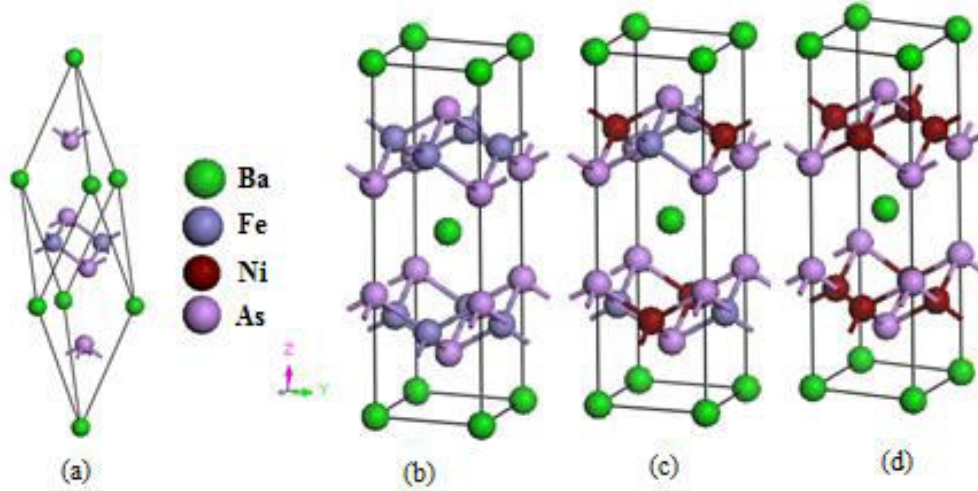


Figure 1: The crystal structure of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.0, 1.0, 2.0$) in tetragonal non-magnetic phase: (a) primitive unit cell of BaFe_2As_2 , and the conventional unit cell for (b) $x = 0.0$ (c) $x = 1.0$ (d) $x = 2.0$

Table 1: Structural parameters of non-spin polarized BaFe_{2-x}Ni_xAs₂ ($x = 0.0, 1.0, 2.0$)

Compound		$x = 0.0$	$x = 1.0$	$x = 2.0$
a=b (Å)	This study	3.949	3.917	4.1525
	Ref 1 ^a	3.945	3.897	4.112
	Ref 2 ^b	3.963	4.002	4.1474
c (Å)	This study	12.626	12.990	11.8086
	Ref 1 ^a	12.429	12.823	11.830
	Ref 2 ^b	13.022	12.676	11.6192
Z_{As} (Å)	This study	0.343	0.3476	0.3461
	Ref 1 ^a	0.355	0.348	0.3464
	Ref 2 ^b	0.35405	0.353	0.3533
Volume (Å³)	This study	196.929	199.318	203.6187
	Ref 1 ^a	193.432	194.737	200.031
	Ref 2 ^b	204.515	202.836	199.861

^a The structural parameters were taken from theoretical work [16]

^b The structural parameters were taken from experimental data [8]

Bands

The non-magnetic band structure calculation in the high symmetry lines of the first Brillouin zone for BaFe_{2-x}Ni_xAs₂ ($x = 0.0, 0.1, 2.0$) is shown in Figure 2. In BaFe₂As₂, three energy bands cross the Fermi level; two small electron-like regions near X point and one hole-like near Z point (Figure 2(a)). At least three small hole-like near the N and G points are obtained when $x = 1.0$. The number of Fermi level crossing bands is increase to five bands in $x = 2.0$; two electron-like near X point, two hole-like regions near N point and one hole-like is found near G point. The hole-like bands near G point tends to enlarge as the function of Ni doping increase and the energy bands founds to shift at lower energy is consistent with other studies [16]. The surface volume results in higher value compared to the other studies [8, 16].

Spin polarized orbital calculations were obtained in Figure 3 shows the existence of superconducting (SC) and antiferromagnetic (AF) states [9] with Fe and Ni ions which playing a role of magnetism in the superconductivity. The electron doping dependence shows the AF long-range order is completely suppressed at $x = 2.0$ (Figure 3(c)) is in agreement with Ref. [9]. The compounds is in a metallic phase by which a number of electron and hole bands crossing the Fermi level and zero semiconducting gap is observed in the whole spectrum range from far infrared to ultraviolet.

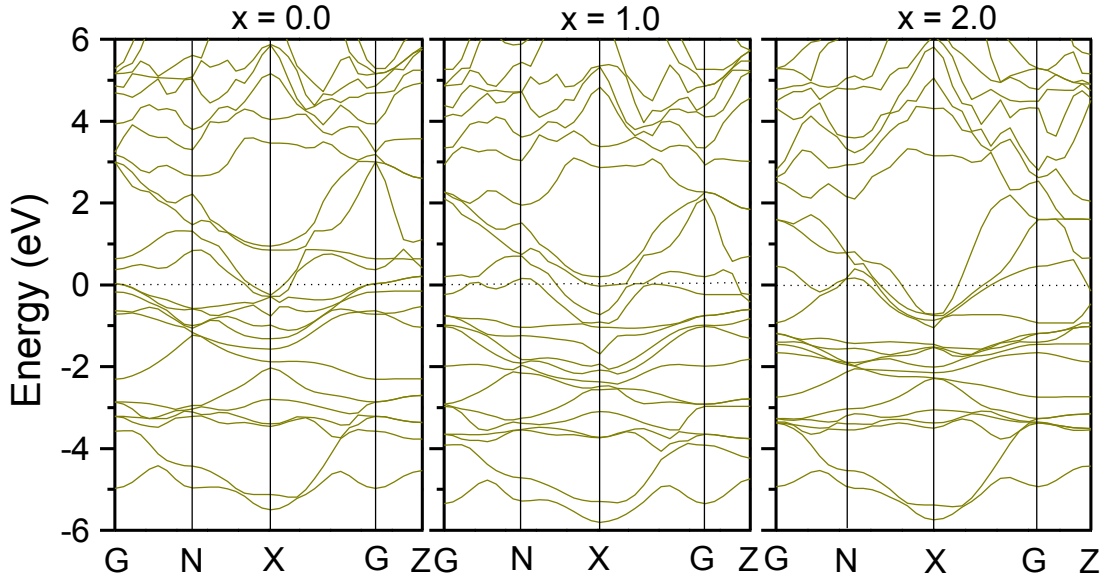


Figure 2: Band structure of non-spin polarized $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.0, 1.0, 2.0$) for high symmetry lines of irreducible Brillouin zone

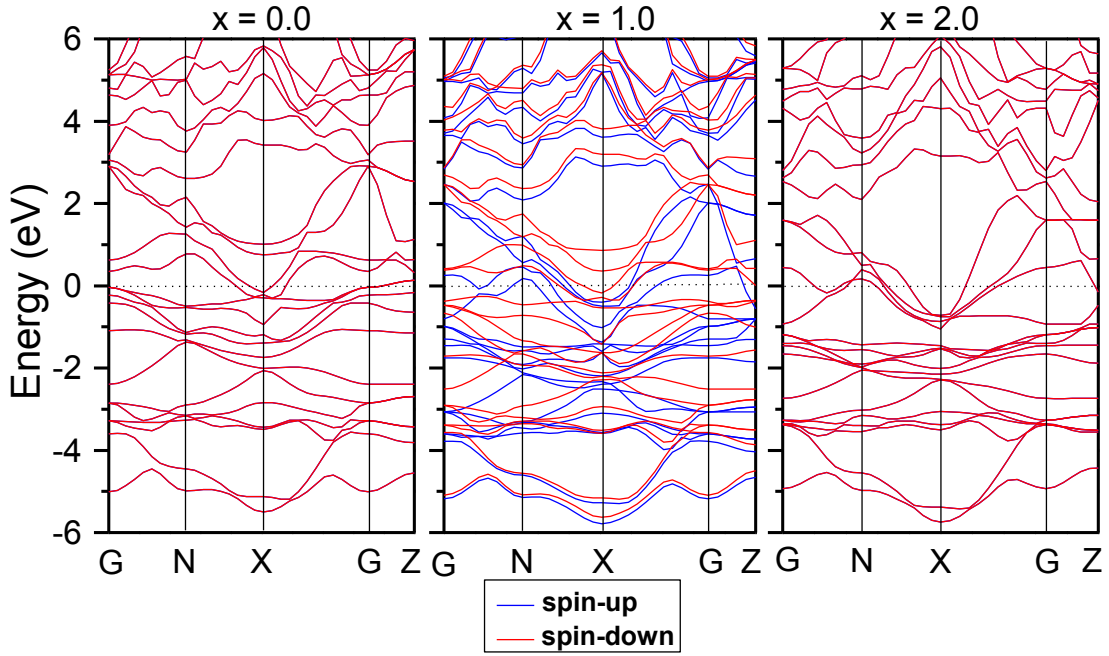


Figure 3: Band structure of spin polarized $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.0, 1.0, 2.0$) for high symmetry lines of irreducible Brillouin zone

Density of state

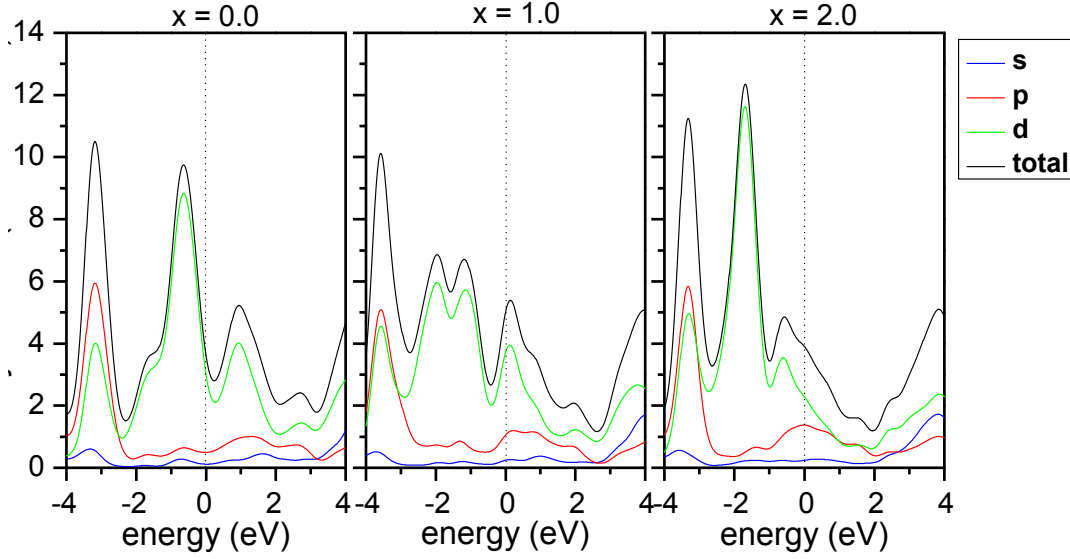


Figure 4: Total and partial density of states of non-spin polarized $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.0, 1.0, 2.0$) in the range -4 to 4 eV relative to the Fermi level

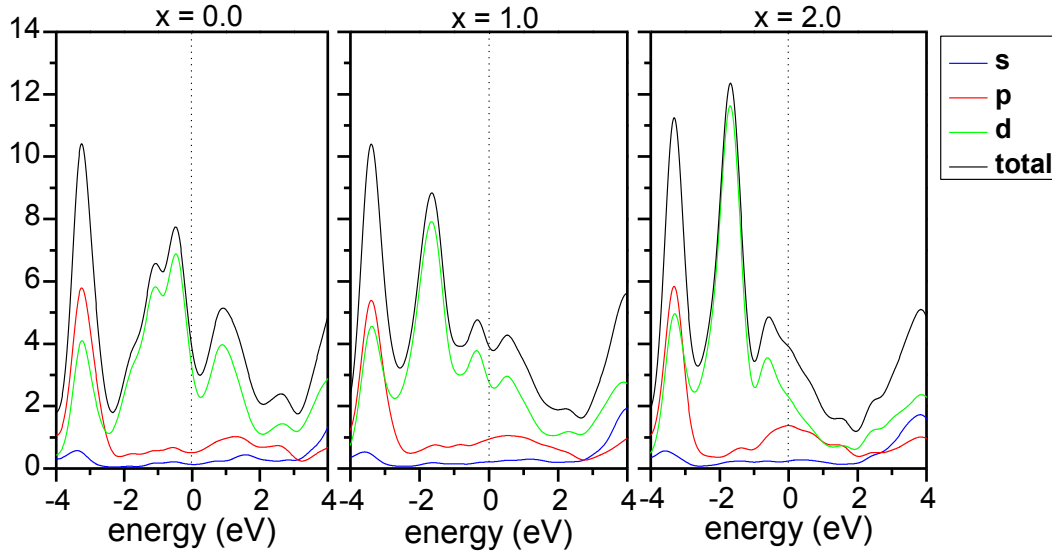


Figure 5: Total and partial density of states of spin-polarized $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ ($x = 0.0, 1.0, 2.0$) in the range -4 to 4 eV relative to the Fermi level

Figure 4 shows the results of density of states (DOS) calculation for $x = 0.0, 0.1, 2.0$ from -4 eV to 4 eV relative to the Fermi level. Two significant peaks and one smaller peak are found near the Fermi level and the d states are shifted to the lower energies for optimal doping. For $x=2.0$ the data are in agreement with experimental result [8].

The results also found similar when spin polarized orbital calculation introduced to the system as shown in Figure 5 except for $x = 1.0$, smaller peaks exist at the Fermi level. This observation indicates the key role of Fe/Ni3d and As4p hybridization and FeAs layer in superconductivity properties of this system. The higher valence electron count in $\text{Ni}^{2+}(3d^8)$ relative to $\text{Fe}^{2+}(3d^6)$ [8] also contribute to the shifted states in the DOS calculation.

ACKNOWLEDGMENT

The author would like to thank Centre for Foundation Studies, Universiti Pertahanan Nasional Malaysia for supporting the project. The author acknowledges the support by the Ministry of Education for the Fundamental Research Grant Scheme (FRGS) (FRGS/1/2013/ST05/UPNM/02/2).

REFERENCES

- [1] Y. Kamihara, T. Watanabe, M. Hirano, H. Hosono, *J. Am. Chem. Soc.* **130** 3296 (2008)
- [2] N. A. Zabidi, H. A. Kassim and K. N. Shrivastava, The Electronic Structure Band Structure of KFe_2As_2 , AIP Conf. Proc. 1169 (2009) 261-264.
- [3] N. A. Zabidi, H. A. Kassim and K. N. Shrivastava, *Sains Malaysiana* **40** (2) 177-180 (2011)
- [4] N. A. Zabidi, M. Z. Azhan, A. N. Rosli and K. N. Shrivastava, The Energy Band Structure of $\text{A}_x\text{Fe}_2\text{Se}_2$ (A = K, Rb) Superconductors, AIP Conf. Proc. 1588 (2014) 281.
- [5] H. Oh, J. Moon, D. Shin, Y. Moon, C. Moon, and H. J. Choi, *Prog. Supercond.* **13** 65 (2011)
- [6] C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. Ratcliff II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, *Nature (London)* **453** 899 (2008)
- [7] S. Chi, A. Schneidewind, J. Zhao, L. W. Harriger, L. Li, Y. Luo, G. Cao, Zhuan Xu, M. Loewenhaupt, J. Hu, and P. Dai, *Phys. Rev. Lett.* **102** 107006 (2009)
- [8] A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. Mandrus, F. Ronning, E. D. Bauer, Y. Mozharivskyj, *Phys. Rev. B* **79** 094508 (2009)
- [9] M. Wang, H. Luo, J. Zhao, C. Zhang, M. Wang, K. marty, S. Chi, J. W. Lynn, A. Schneidewind, S. Li and P. Dai, *Phys. Rev. B* **81** 174524 (2010)
- [10] Z. Xu, Q. Tao, L. Li, J. Shen, X. Lin, G. Cao, *Physica C* **470** 447 (2010)
- [11] M. D. Segall, P. J. D. Lindan, M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark, and M. C. Payne, *J. Phys. Condens. Matter* **14** 2717 (2002)
- [12] M. C. Payne, M. P. Teter, D. C. Allan, T. A. Arias and J. D Joannopoulos, *Rev. Mod. Phys.* **64** 1045 (1992)
- [13] J. P. Perdew, K. Burke, and M. Ernzerhof, *Phys. Rev. Lett.* **77** 3865 (1996); **78** 1396 (1997) (E)
- [14] D. Vanderbilt, *Phys. Rev. B* **41** 7892 (1990)
- [15] H. J. Monkhorst, and J. D. Pack, *Phys. Rev. B* **13** 5188 (1976)

- [16] Z. Inanloo, H. Khosroabadi, M.R. Abolhassani, M. Akhavan, *Physica C* **506** 151-153 (2014)