

Structural, elastic, electronic and magnetic properties of Fe_3AC ; $\text{A} = \text{Al}$, Ga and In

Y. Medkour^{1,a}, A. Roumili¹, A. Saoudi², L. Louail² and D. Maouche²

¹Laboratoire d'Études des Surfaces et Interfaces des Matériaux Solides (LESIMS), Université de Sétif, Algérie

²Laboratory for Developing New Materials and their Characterizations, University of Setif, Algeria

Abstract. We report first principle calculations on the structural, electronic and magnetic properties of antiperovskite Fe_3AC ; $\text{A} = \text{Al}$, Ga and In . Calculations show that these compounds are more stable in the magnetic states, the estimated equilibrium lattice parameters (a and V) are in agreement with the experimental data. From the single crystal elastic constants, the polycrystalline elastic moduli is estimated. Similar to previous studies on carbides antiperovskite, these compounds are good electrical conductors. The analysis of the total and partial densities of states shows that the conductivity is assured by d electrons of the transition metal atoms. The magnetic character in these compounds is mainly related to the spin polarization of Fe-d electrons. The magnetic moment per unit formula is found to decrease from $3.52 \mu_B$ to $3.06 \mu_B$ corresponding to Fe_3InC and Fe_3AlC respectively.

1. INTRODUCTION

Ternary transition metal carbides (or nitrides) have shown attractive and unusual properties, which made them of potential use in a number of applications, extend from semiconducting to superconducting. Among these carbides and nitrides are the antiperovskite compounds with the general formula M_3AX where X is C or N , while both A and M are metal atoms [1]. The discovery of superconductivity at 8 K for the Ni_3MgC [2], highlights these materials and stimulates more experimental and theoretical works to investigate the origin of this behaviour.

The present work is carried out on Fe_3AC ; $\text{A} = \text{Al}$, Ga and In , both of them have a cubic structure with A elements at the cubic corner, Fe atoms at the face centres and C atom at the body centre [1–4].

2. COMPUTATIONAL METHODS

Our calculations were carried out using the CASTEP (Cambridge Serial Total Energy Package) code [5], in which the density functional theory [6] and the Kohn-Sham approach were used to calculate the fundamental eigenvalue. In order to reduce the basis set of plane wave (PW) functions used to describe the real electronic functions, the pseudopotential (PP) approximation was introduced, where the nucleus and the core electrons were replaced by an effective potential. The exchange correlation energy was estimated using the generalized gradient approximation with the Perdew-Burke-Ernzerhof functional (PBE) [7]. Brillouin zone sampling was done by the Monkhorst-Pack scheme [8] and was sampling on $10 \times 10 \times 10$ irreducible k points. The ultra-soft pseudopotential was employed with a cut off energy $E_{\text{cut,off}} = 400 \text{ eV}$, with an energy tolerance of $5.10^{-6} \text{ eV/atom}$.

^a e-mail: you_med2000@yahoo.fr

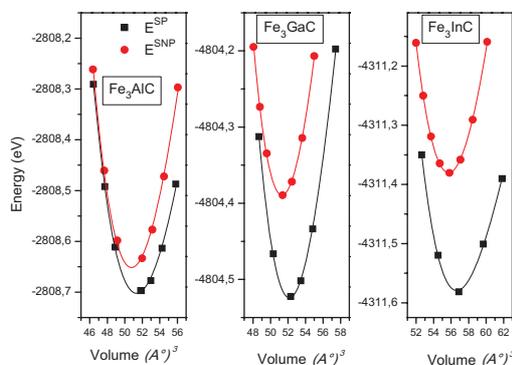


Figure 1. Total energy versus cell volume curves for Fe_3AlC , Fe_3GaC and Fe_3InC for spin polarized and spin nonpolarized calculations.

3. RESULTS AND DISCUSSIONS

3.1. Structural and elastic properties

To achieve the ground state properties, we have performed both spin polarized and non spin polarized calculations as presented in Fig. 1. The energy difference: $\Delta E = E^{SP} - E^{SNP}$ for Fe_3AlC , Fe_3GaC and Fe_3InC is about -0.10 , -0.13 and -0.20 eV respectively.

These quantities ensure a good stability of these compounds in the magnetic state. From Table 1, the calculated lattice parameter a and the equilibrium volume V are in well satisfactory with the experimental results reported in Ref. [1].

According to Hooke's law, single crystal elastic constants of Fe_3AC ; $\text{A} = \text{Al}$, Ga and In , were determined by a linear fit of the stress-strain data. The elastic behaviour can be described by three independent elastic constants [9] C_{11} , C_{12} and C_{44} . C_{11} values are higher about 84%, 84% and 77% than C_{44} for Fe_3AlC , Fe_3GaC and Fe_3InC respectively.

Polycrystalline elastic constants B and G are obtained with the Hill's assumption. The bulk modulus B and G

Table 1. Calculated cell parameters and elastic constants of Fe_3AC ; A = Al, Ga and In.

Comp.		Al	Ga	In
a (Å)	Cal.	3.728	3.739	3.846
	Exp. [1]	3.758	3.74	3.88
V (Å ³) [3]	Cal.	51.834	52.261	56.915
	Exp. [1]	53.072	52.313	58.411
C_{11} (GPa)		490	479	362
C_{44} (GPa)		75	75	83
C_{12} (GPa)		114	115	99
B (GPa)		239	236	187
G (GPa)		110	108	100

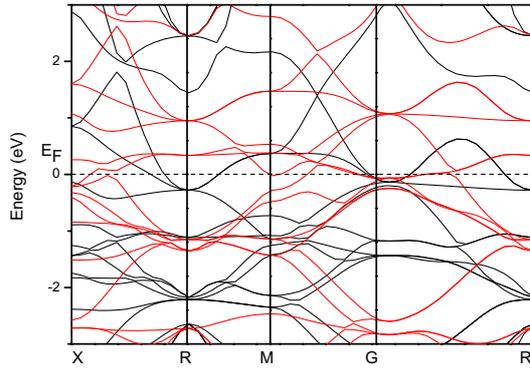


Figure 2. Calculated spin polarized band structure of Fe_3GaC , black and red colors referred respectively to up and down states.

decrease with the atomic number Z of A elements, in the sequence Fe_3AlC , Fe_3GaC and Fe_3InC , our calculated values are very close to the bulk modulus of GaCMn_3 , 224 GPa [3]. The obtained bulk modulus of Fe_3AlC is about 16% higher than the reported one by Ouyang *et al.* [4]. Shear modulus G is defined as the response of a material against hydrostatic pressure within shape change [9], G decreases by 9% from 110 to 100 GPa for Fe_3AlC and Fe_3InC respectively. The results show that the ratio G/B is around 0.5 for all compounds under study suggesting a ductile behaviour of these materials.

3.2. Electronic and magnetic properties

Since the equilibrium ground state was defined to be stable in the magnetic state, we have calculated the band structure (BS), the total (TDOS) and the partial (PDOS) densities of states for both spin up and down states. Only the results for Fe_3GaC are presented. The band structure reported in Fig. 2 shows an important overlapping around the Fermi level, pointing out to the electrical conduction state in these compounds.

The spin polarized results for both TDOS and PDOS are presented in Fig. 3, for Fe_3GaC . The valence band structure of Fe_3AC is formed from two parts. The lower part located under -12 eV originates from C(s), and Fe (s, d) electrons. The upper one, extend from -9 eV to Fermi level, originates from A(s,d), Fe d and C p electrons. The conduction band above the Fermi level is a contribution

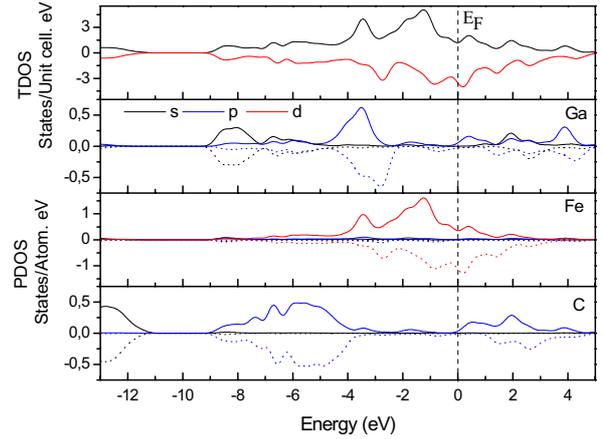


Figure 3. Spin polarized total and partial densities of states for Fe_3GaC .

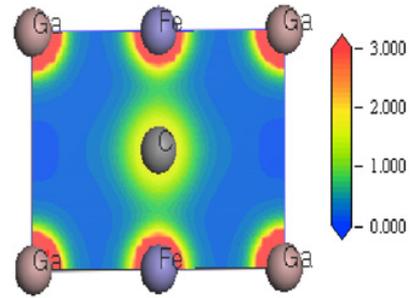


Figure 4. Valence electron density for Fe_3GaC .

from C(p) and Fe(d) states, for which we remark that at the Fermi level only the Fe d electrons are involved in the electrical conduction, whereas A(d) electrons of Ga and In atoms have no contribution. These conclusions are in agreement with the early results reported by Grandjean *et al.* [10].

The estimated magnetic moment of Fe atom is $1.16 \mu_B$ in Fe_3AlC and increases to $1.32 \mu_B$ in Fe_3InC , it is mainly arises from d electrons. The magnetic moment of Fe in Fe_3AlC is in good agreement with that reported in Ref. [4].

From Fig. 4, it was observed that Fe-C bonds in Fe_3AC are very strong and of covalent-ionic nature. Fe-A are less strong and show ionic nature, A-A bonds are the weakest and exhibit metallic character. These results are in agreement with the observed hybridized states in Fig. 3.

4. CONCLUSION

The calculated lattice parameter a and the equilibrium volume V are in agreement with the experimental results. The results show that these compounds are more stable in the magnetic state. The bulk modulus B and G decrease with the atomic number Z of A elements. Based on the BS and the PDOS, we have observed that only the Fe d electrons are involved in the electrical conduction. The plot of the valence charge density shows a mixture of covalent, ionic and metallic bonds which maintain the crystal stability.

References

- [1] V. H. Nowotny, *Prog. Solid State Chem.* **2**, 27 (1970).
- [2] S. Mollah, *J. Phys. Condens. Matter.* **16**, R 1237 (2004).
- [3] S. Q. Wu, Z. F. Hou, and Z. Z. Zhu, *Solid state Sci.* **11** (2009) 251.
- [4] Y. Ouyang, M. Tang, J. Fang, P. Xiang, Y. Du, Y. He and Y. Liu, *Phys. Scr.* **80** (2009) 055603.
- [5] 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** (2002) 2717.
- [6] J. Kohanoff, *Electronic structure calculations for solids and molecules*, Cambridge University Press, 2006.
- [7] J. P. Perdew, K. Burke, M. Ernzerhof, *Phys. Rev. Lett.*, **77** (1996) 3865.
- [8] H. J. Monkhorst and J. D. Pack, *Phys. Rev. B.* **13** (1976) 5188.
- [9] E. Schreiber et al., *Elastic Constants and Their Measurement*. McGraw-Hill, New York, 1973.
- [10] F. Grandjean and A. Gerard, *J. Phys. F: Met. Phys.* **6** (1976) 451.