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# Study of the magnetic and electronic properties of the $Fe_4N$ with pressure

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ABSTRACT

In the present work, we report ab-initio studies of the magnetic property variations with pressure of both iron sites in the structure of $Fe_4N$, using full-potential linearized augmented plane wave method and the Perdew–Burke–Ernzerhof functional and generalized gradient approximation to describe the exchange-correlation potential are reported. The results show that the magnetic moment of FeI is almost constant while the magnetic moment of FeII presents a discontinuity when the lattice parameter is varied. This is reflected in the compression of the spin up and down energy bands to different concentration points. The variation in the FeII magnetization arises mainly from changes in the $d_{xy}$, $d_{xz}$, $d_{yz}$ and $d_{x2-y2}$ orbitals.



## 1. Introduction

The $Fe_4N$ displays a remarkably rich behaviour that has attracted the attention of applied and basic researchers in the last 80 years. The earliest interest on the synthesis and study of properties was motivated by its possible application as a catalyst for the production of ammonia and for its role in the surface treatment of steels with ammonia [1]. Later, it was considered as a material for high-density magnetic recording component in thin film devices. The interest of the researches has also focus in the variation of the magnetic properties of $Fe_4N$ because it serves as a model for expanded $\gamma$-Fe (FCC). In effect, when applying pressure to $Fe_4N$, the volume per atom can be driven towards the value for $\gamma$-Fe, allowing interesting comparisons and investigations on the stability of the Fe magnetic moment in the FCC structure. These considerations have set off several experimental [2–4] and theoretical studies of compressed $Fe_4N$ [5–7]. The theoretical investigations of the electronic structure of bulk $Fe_4N$ were mainly performed using linearized augmented plane wave (LAPW) [5,8,9], linear muffin-tin orbital (LMTO) [10,11] and augmented spherical wave (ASW) methods [6]. These methods generally reported similar structural and magnetic moment properties, consistent with the experimental data [2,3].

In this work, the behaviour under pressure of the magnetic properties of $Fe_4N$ is studied by analysing the evolution of the density of the electronic states (DOS) of every atomic species. The full-potential linearized augmented plane wave method (FP-LAPW) have been used [12,13].

## 2. Calculations

The $Fe_4N$ has an anti-perovskita structure (space group Pm-3m(2 2 1)) where the Fe occupy the corners (FeI) and the centre of the faces (FeII) while the N atoms are placed at the centre of the cube [14] (see Fig. 1). The FeI atoms are surrounded by 12 FeII atoms nearest neighbours placed at 0.71*a*, as in pure FCC-Fe [15], and 4 N atoms located at 0.87*a* as second neighbours. Otherwise, the FeII atoms have 2 N atoms at 0.5*a* as first neighbours and 12 FeI atoms at 0.71a as second neighbours.

The calculations were performed using FP-LAPW method within the framework of density functional theory (DFT) [12,13] as implemented in the Wien2K code [16]. In this framework, the exchange-correlation potential for structural and electronic properties was calculated using both a local spin density approximation (LSDA) and the generalized gradient approximation (GGA) based on the Perdew–Burke–Ernzerhof expression [13,17]. The parameter $R_{MT}K_{max}$ was keep equal to 8 ($R_{MT}$ is the muffin-tin radius and $K_{max}$, the largest $K$ vector in the plane wave expansion). Muffin-tin radius of 1.79 and 1.59 a.u. for Fe and N were used, respectively. A mesh of 286 $k$-points was taken in the irreducible wedge of the Brillouin zone. The number of $k$-points was varied to ensure a total energy convergence better that $10^{-6}$ Ry. In all calculations relativistic effects and spin polarization were considered.

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## 3. Results and discussion

The total and partial DOS at the equilibrium lattice constant value ($a$) are displayed in Fig. 2. The main part of the DOS(up) of N atoms is in the range from −8.5 to −5.3 eV. Similarly, the main part of FeI DOS(up) lies between −5 eV and the Fermi level ($E_f$), while that of FeII is placed from −8.5 eV to $E_f$. These results indicate a strong superposition of electronic states of the N and FeII atoms in the range from −8.5 to −5.0 eV, approximately and explain the difference between FeI and FeII magnetic moments in terms of the Fe–N 3d–sp hybridization [18].

The equilibrium lattice constant ($a$), obtained from the calculated total energy curves by fitting with the Murnaghan equation [19], and the magnetic moments ($m_{FeI}$ and $m_{FeII}$) of both iron atoms, FeI and FeII, are reported in Table 1. The resulting values are in good agreement with the experimental data [2,3,20], as observed in Table 1. When the lattice parameter is varied from 6.8 to 7.4 a.u., it is observed in Fig. 3 that $m_{FeI}$ remains almost constant. On the contrary, $m_{FeII}$ shows a parabolic behaviour with $a$ up to 7.1 a.u. and then a linear relationship is established between $m_{FeII}$ and $a$, with a small and positive slope. It is worth to

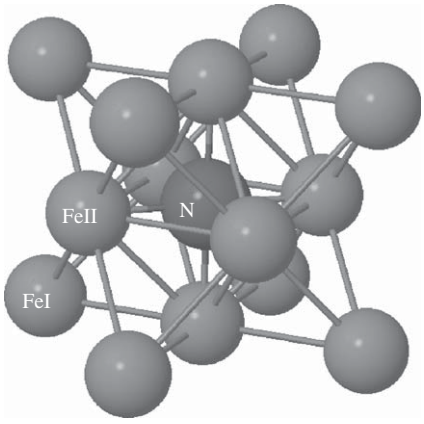


**Fig. 1.** The unit cell of $Fe_4N$ structure.

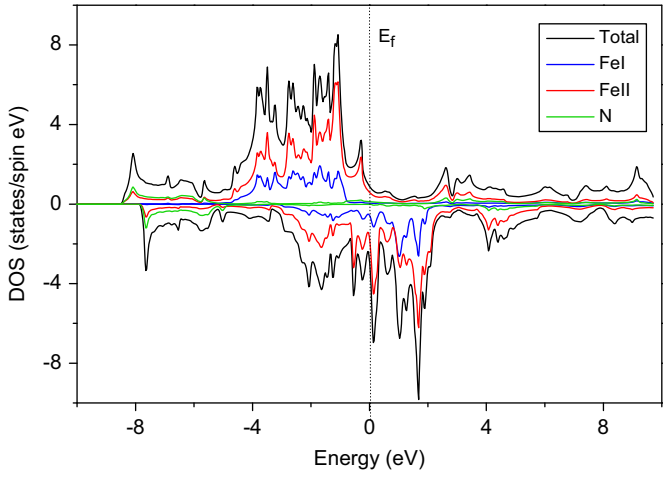


**Fig. 2.** Density of states total and partial of the $Fe_4N$.

**Table 1**
Calculated and experimental lattice parameter ($a$) and magnetic moment ($m$).

| | Present results | Experimental |
|---|---|---|
| $a$ (a.u.) | 7.17 | 7.17 [20] |
| $m_{FeI}$ ($\mu_B$) | 2.84 | 3.0 [3] |
| $m_{FeII}$ ($\mu_B$) | 2.27 | 2.0 [3] |

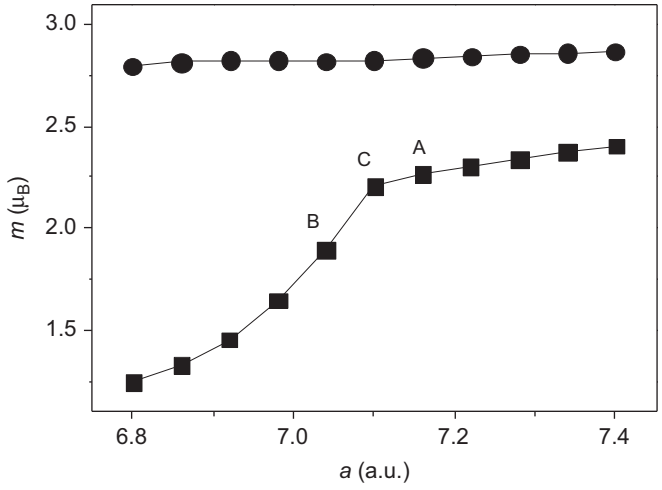


**Fig. 3.** Variation of the magnetic moments vs. the lattice constant: (a) $m_{FeI}$: ● and (b) $m_{FeII}$: ■.

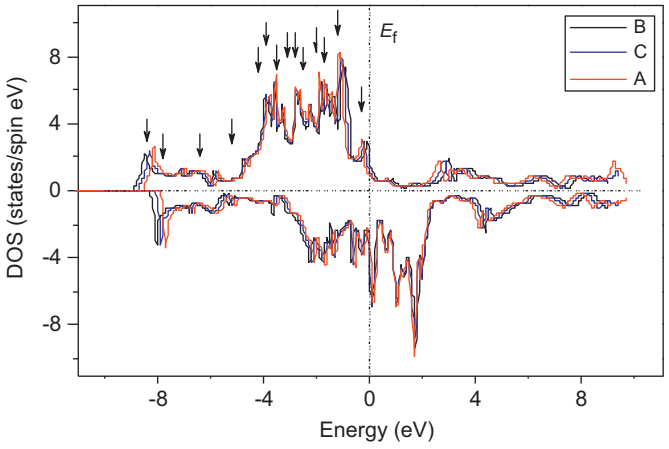


**Fig. 4.** Calculated DOS using different parameters (B, C, and A). The arrows indicate representative peaks.

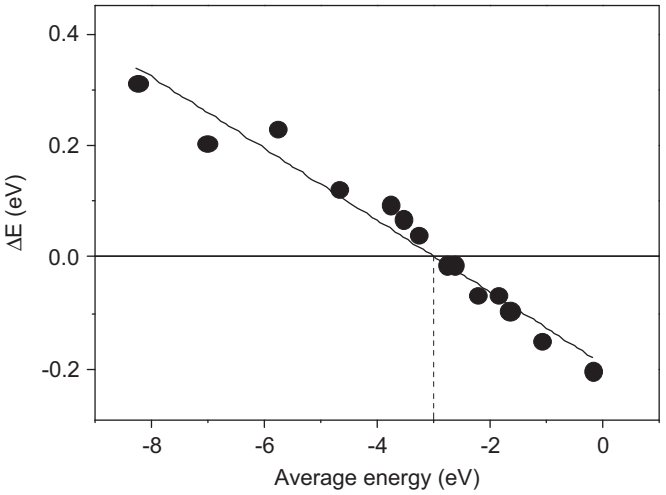


**Fig. 5.** Energy difference ($\Delta E$) of the peaks quoted with arrows in Fig. 4, calculated at the B, C and A points of Fig. 3.