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A DFT-Based Study of the Magnetic, Electronic and Elastic Properties of YFe_2

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Abstract

We present a DFT-based study of the equilibrium volume, bulk modulus, electronic structure and magnetic properties of YFe_2 at ambient and higher hydrostatic pressures. The LSDA and GGA approximations, as implemented in FPLO-09 are used throughout the scalar relativistic calculation in this work. Charge density maps using Wien2k are also reported for selected lattice constants. We demonstrate that the application of pressure on YFe_2 has a prominent effect on its magnetic and electronic properties, e.g. the reduction of the magnetic moment and finally the disappearance of ferromagnetism. The exchange –correlation energy is shown to have an effect on both the cell volume and its magnetic moment.

Introduction

YT_2 system with T= Mn, Fe, Co, and Ni has attracted numerous theoretical [1-8] and experimental [9-11] investigations to elucidate the role of 3d electrons responsible for the diverse magnetic behavior of these alloys.

In particular, several studies were devoted to Y-Fe system e.g. [5-7] where LDA /LSDA approximations have been used to calculate the magnetic moment, electronic structure, DOS and magneto-volume effects. One point of interest in YFe_2 is the magnetic moment carried by the Y atom [9, 10, 12]. The second point is the pressure dependence of the electronic and magnetic properties of this system [13, 14]. In particular, whether it can cause a collapse of the ferromagnetic state, or to a phase transformation from C15 (cubic) to C14 (hexagonal) at elevated pressures [15].

In the present study we present a DFT- based ab initio calculation on YFe_2 using both the Local Spin Density Approximation (LSDA) and Generalized Gradient

Approximation (GGA) as implemented in the electronic code FPLO9[16]. Our purpose is to investigate the effect of these two approximations on the equilibrium volume, magnetic moment, bulk modulus and density-of-states DOS and to study to what extent the application of a hydrostatic pressure will change the elastic, electronic and magnetic properties. Another aim of the present work is to find out the dependence of the exchange-correlation interaction on the volume and magnetic moment $E_{x\text{-}c}$ (V,M) using these two different approximation schemes.

Theory and Computation

Our calculation is based on DFT. The absence of electrons in the f-shell of Y does not demand taking the spin-orbit coupling into consideration. Our calculation is scalar relativistic within the LSDA and GGA approximation schemes of the FPLO-09 code where Perdew and Zunger potential was used. We have used the same set of parameters in our computation to ensure unbiased comparison between the results obtained from LSDA and GGA approximations. The parameters are: the k-mesh subdivision: 24×24×24, the accuracies of the density and total energy are 10^{-6} and 10^{-8} Hartree respectively. The space group is 227/Fd3m and the atomic positions are 1/8,1/8,1/8 for Y and 1/2, 1/2,1/2 for Fe. Two formula units of YFe_2 are present in the conventional unit cell. At first, we make geometrical optimization i.e. determine the equilibrium volume V_0 of the crystal structure at hand from the DFT results of the volume dependence of energy E(V). Next we fit the E(V) data to the Murnaghan equation-of-state[17]. The fitting parameters B_0 (the bulk modulus) , B' (the pressure dependence of B_0) and V_0 are consequently used to determine the volume dependence on the hydrostatic pressure P. The DOS and magnetic moment are calculated at either ambient pressure (P=0) and volume (V_0) or at P>0. The stoner parameter is calculated from the second derivative of the $E_{x\text{-}c}$ (V,M) with respect to the magnetic moment M at M=0 [18]. The charge density maps in the (110) plane are calculated using the Wien2k package [19,20] for two different cell volumes: the equilibrium volume V_0 and $V \approx 0.70\ V_0$

Results and Discussion

The total energy vs. the primitive cell volume is shown in Fig.1 for both magnetic and nonmagnetic phases of YFe_2. The calculation is done in the GGA approximation. Clearly the magnetic phase is more stable than the nonmagnetic one. The equilibrium volumes are 97.25 (lattice constant a = 7.300 Å) and 91.38 $Å^3$ (lattice constant a = 7.15 Å) for these two phases respectively with a spontaneous volume magnetostriction w_s of 6.4 %. The calculated lattice parameter of the magnetic phase 7.300 Å is in good agreement with the experimental 7.363 Å[10]

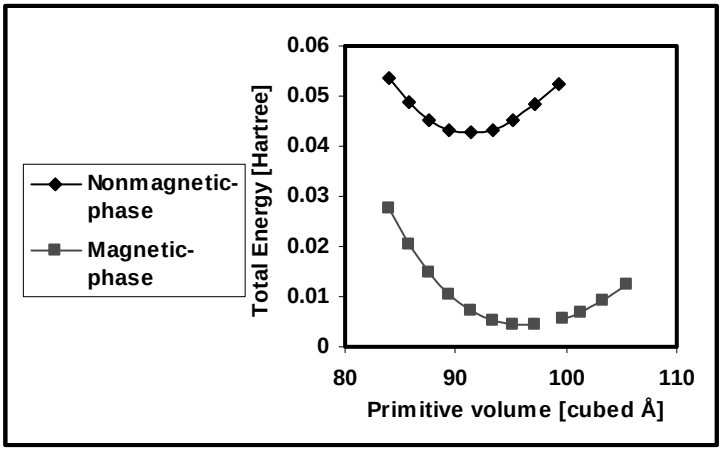

Fig.1: Dependence of the total energy vs. the primitive cell volume for magnetic and nonmagnetic YFe_2 using GGA approximation.

The dependence of the total moment on volume is shown in Fig.2, together with individual contributions of the Y and Fe sublattices. The overall decreases of the magnetic moment with decreasing the unit cell volume is a feature of this GGA calculations and LSDA one as well (Fig.3). The unit cell loses its magnetic moment with volume almost linearly in the volume range ~ 100 – 140 $Å^3$ and then drops faster to smaller values for $V \leq 100$ $Å^3$. This overall decrease of the total magnetic moment with decreasing the volume supports the itinerant nature of magnetism in this system and is in agreement with the experimental work of Armitage et al [21].

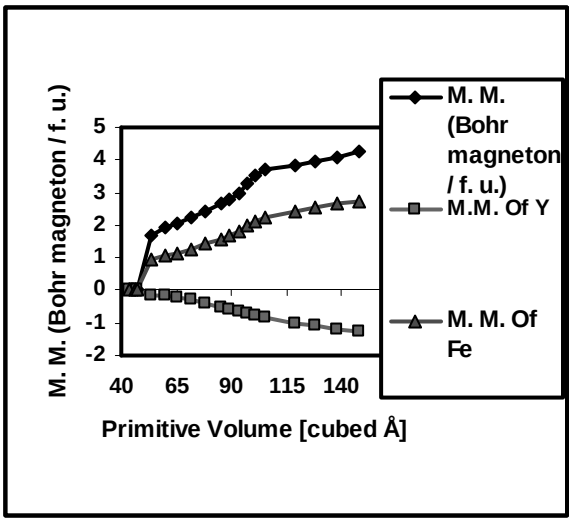

Fig.2: Dependence of the total moment on the primitive volume for magnetic YFe_2 using GGA approximation, in together with individual contributions of the Y and Fe sublattices.

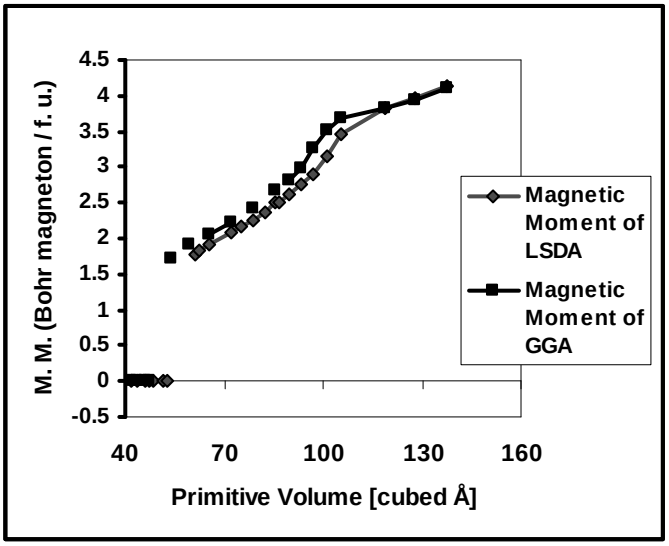

Fig.3: Dependence of the total moment on the primitive volume for magnetic YFe_2 using GGA and LSDA approximation.

The Fe and Y sublattices have a ferrimagnetic coupling but Fe atoms are ferromagnetically coupled with a net moment of 3.27 μ_b /formula unit. The partial moments are Y= -0.699 μ_b and Fe = 1.98μ_b. The negative magnetic moment of Y has been confirmed by neutron scattering [10].

Our magnetic moment value is close to the experimental value and to the values of the calculation by Yamada et al [12] and Armitage et al [9] of 2.91 μ_b. It is also in fair agreement with 2.93 μ_b calculated by Buschow et al [22] and to 2.94 μ_b calculated by Erikson et al [23].

We have repeated the same calculation in the LSDA approximation and found the magnetic phase to be more stable as well, however the GGA calculated magnetic moment is closer to the experimental values. Table 1 shows a summary of calculated structural and / or magnetic properties for magnetic and nonmagnetic phases of YFe_2. We present in table 2, for comparison, the corresponding data for pure bcc iron.

In order to obtain the equation of state i.e. P (V) where P is the hydrostatic pressure on YFe_2, we have fitted the energy vs. primitive volume in the LSDA data to the Muranghan equation EOS [17]. The resulting fit is shown in Fig.4 for the magnetic phase. The P-V relationship shows a continuous decrease from the volume at ambient pressure (P=0) to ~ 70 $Å^3$ at P in excess of 50 GPa (Fig.5). The bulk modulus B_0 and its first pressure derivative B' are 148.4 GPa and 4.93 respectively. The GGA data turned out, however, to give B_0 = 94.90 GPa and B' = 1.5, compared to the experimental B_0 = 129 GPa. The relative error, defined as:

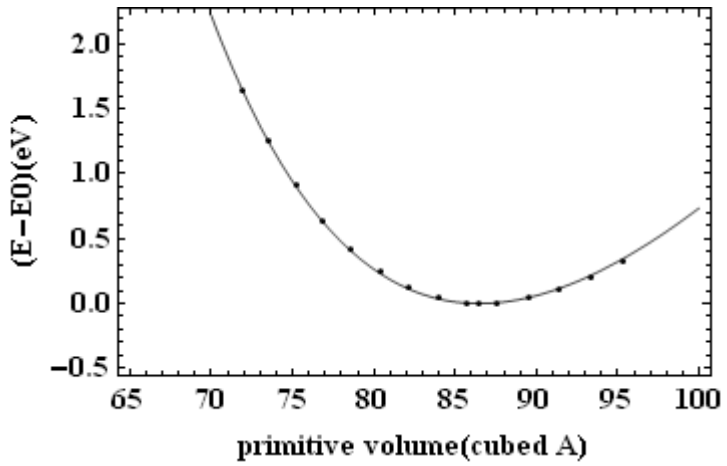

Fig.4: The energy vs primitive volume in the LSDA approximation fitted to the Muranghan equation for magnetic YFe_2.

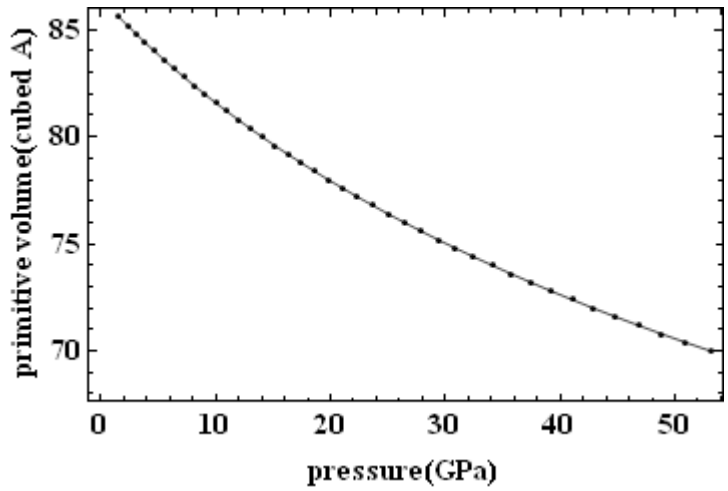

Fig.5: The P-V relationship in the LSDA approximation for magnetic YFe_2.

$(B_0 - B_{exp})/B_{exp}$, is approximately 27% and 15 % in GGA and LSDA approximations respectively. This indicates that LSDA approximation is more adequate in this case. Our LSDA value is close to 143 GPa calculated using LMTO method [3] and LMTO-ASA [5].

The dependence of the total magnetic moment on pressure is shown in Fig.6. The material loses its magnetic moment rather rapidly for pressures in excess of ≈ 100 GPa becoming nonmagnetic for P ≥ 250 GPa. This behavior is similar to that calculated for YFe_2 and $LuFe_2$ by W. Zhang [13]. We have performed fixed spin moment calculation in both LSDA and GGA approximation. The results are shown in Figs. 7a, b respectively. As we have seen that GGA predicts higher moment at equilibrium. We've used the exchange- correlation dependence on the magnetic moment in the 0-5 μ_b range in order to check the Stoner criterion for ferromagnetism

(see for example the book by P. Mohn [18]). Our results of the dependence of $E_{x\text{-}c}$ on volume and magnetic moment in both LSDA and GGA approximation are shown in Figs.8a, b respectively. It is apparent that a decrease in the unit cell volume leads to an increase in both of the magnitude of the negative $E_{x\text{-}c}$ and the magnetic moment as shown in Fig.9. We have calculated the product $D(E_f)I$ at V_0 where the density of states is evaluated for the nonmagnetic phase and I is the Stoner parameter evaluated by taking the second derivative of $E_{x\text{-}c}$ (V,M) with respect to M at M = 0. Our value of 2.04 is in good agreement with 2.12 reported by Taga [3].

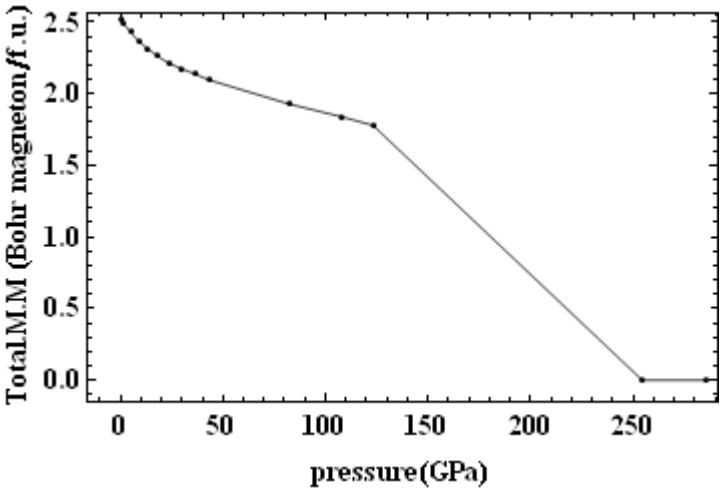

Fig.6: The dependence of the total magnetic moment on pressure in the LSDA approximation for magnetic YFe_2.

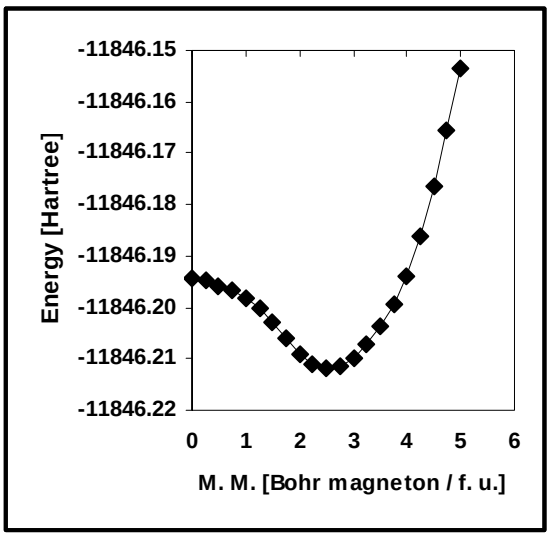

Fig.7a: The fixed spin moment calculation in the LSDA approximation for magnetic YFe_2.

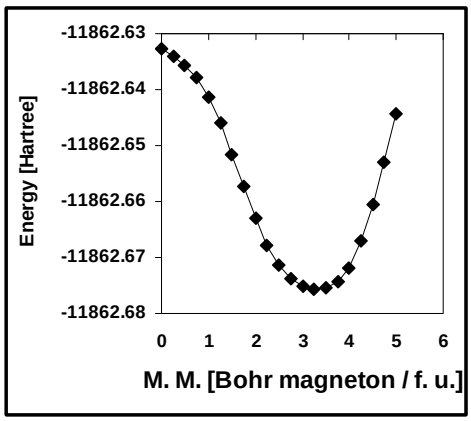

Fig.7b: The fixed spin moment calculation in GGA approximation for magnetic YFe_2.

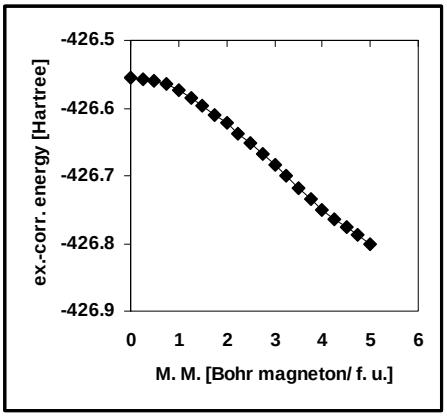

Fig.8a: The dependence of $E_{x\text{-}c}$ on magnetic moment in LSDA approximation for magnetic YFe_2.

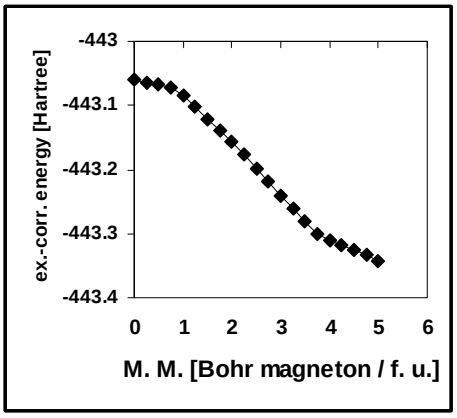

Fig.8b: The dependence of $E_{x\text{-}c}$ on magnetic moment in GGA approximation for magnetic YFe_2.

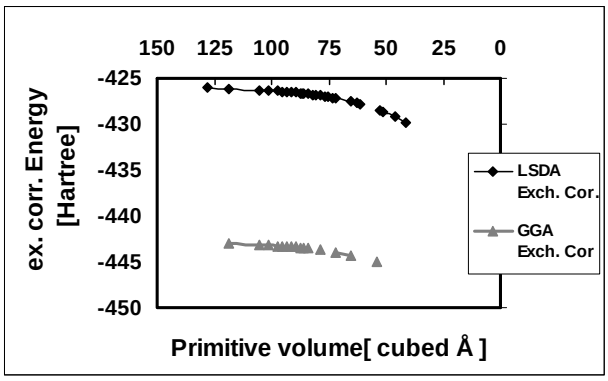

Fig.9: The dependence of $E_{x\text{-}c}$ on volume in both LSDA and GGA approximation for magnetic YFe_2.

The total and partial DOS's of magnetic YFe_2 at the equilibrium volume of 97.25 $Å^{3,}$ as calculated in the GGA approximation is shown in Figs.10(a-c). The Fermi level is located at a peak in spin- up DOS. The total DOS counts at E_F = 7.8 states /eV. The Fe and Y partial DOS's show that the total DOS is dominated by 3d electrons, the contributions of 4d electrons are negligible. The corresponding band structure is shown in Fig.11. The flat bands close to E_F correspond to peaks in the DOS (Fig.10a). The DOS at V=46.3 $Å^3$ is shown in Fig.(12), which clearly shows that YFe_2 loses its magnetic moment completely as it is compressed to this small volume. We have calculated the DOS at other volumes in the range 46.3-97.25 A^3 and found that the compound loses its magnetic moment gradually until becoming non-magnetic rather abruptly in the vicinity of V≈ 46 A^3. In order to study the effect of pressure on the charge density we calculated the charge density maps in the (110) plane using the wien2k package. These maps are shown in Figs.(13 a, b) for a = 7.3Å (V=97.25 $Å^3$) and a = 6.50Å (V=68.66 $Å^3$) respectively. The smaller volume corresponds to a pressure in excess of approximately 50 GPa. It is evident the charge density in Fig. 13b is higher than that at the ambient pressure (Fig.13a), given the fact that the total charge is of course conserved.

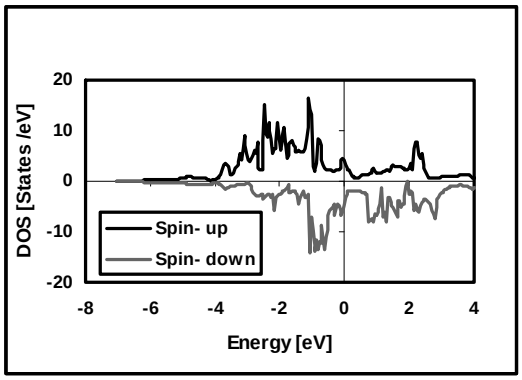

Fig.10a: The total density of states (DOS) for magnetic YFe_2 at the equilibrium volume of 97.25 $Å^3$ as calculated in the GGA approximation.

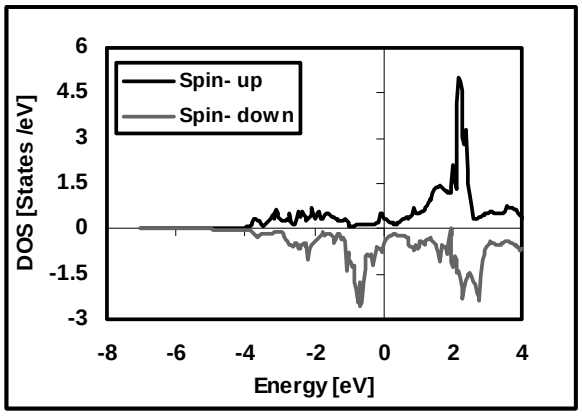

Fig.10b: The partial density of states (DOS) for 4d of Y for magnetic YFe_2 at the equilibrium volume of 97.25 $Å^3$ as calculated in the GGA approximation.

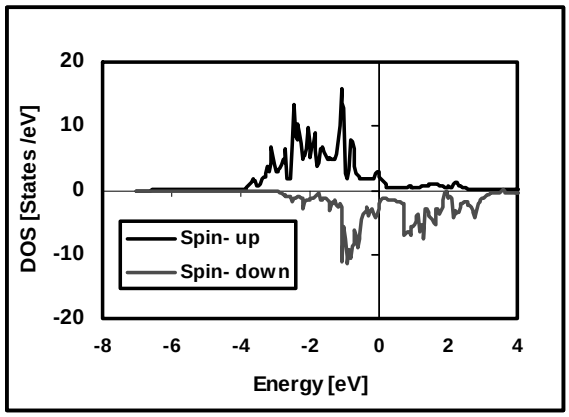

Fig.10c: The partial density of states (DOS) for 3d of Fe for magnetic YFe_2 at the equilibrium volume of 97.25 $Å^{3,}$ as calculated in the GGA approximation.

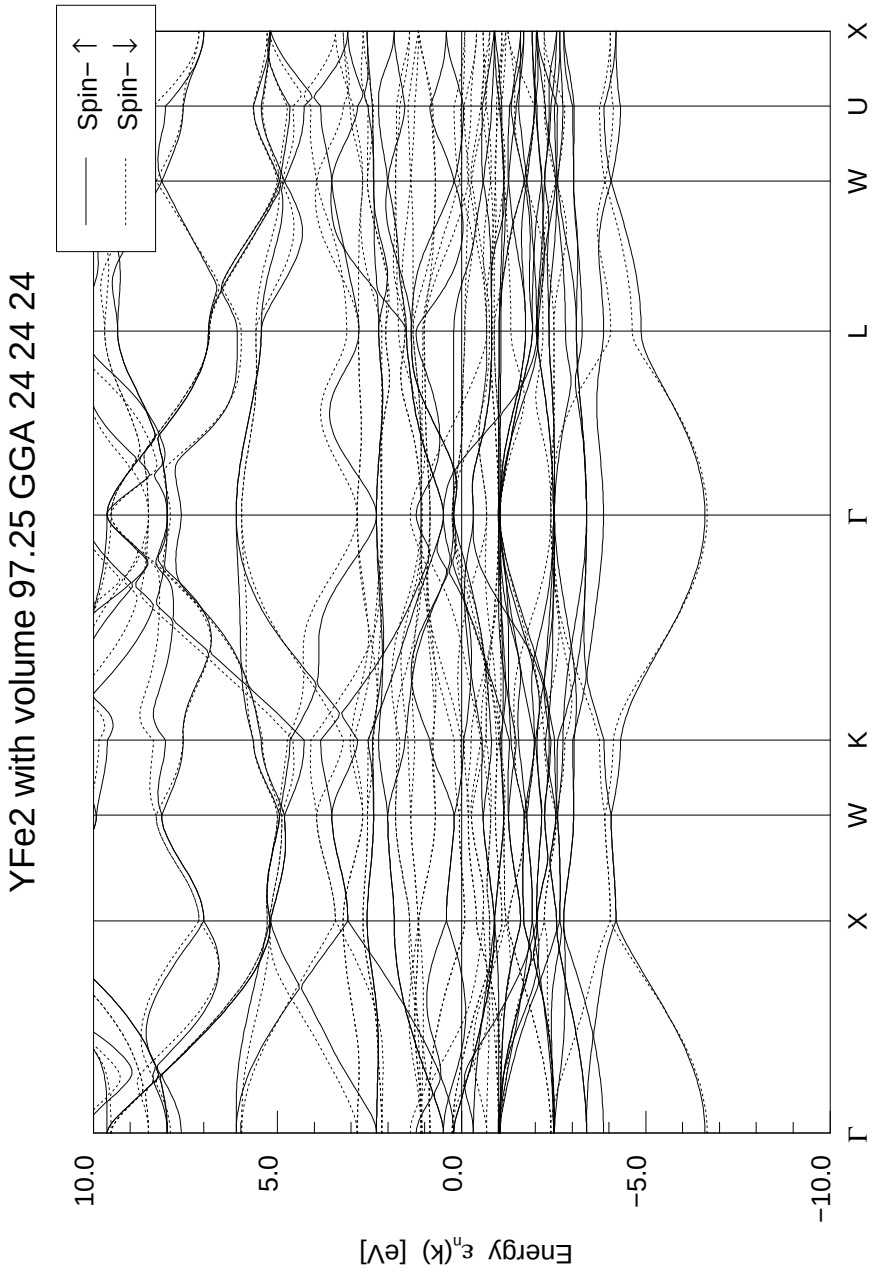

Figure 11: The band structure for magnetic YFe2 at the equilibrium volume of 97.25 Å3, as calculated in the GGA approximation.

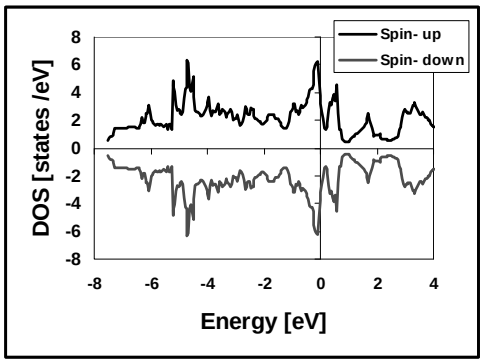

Fig.12: The total density of states (DOS) for magnetic YFe_2 at volume of 46.3 $Å^{3,}$ as calculated in the GGA approximation.

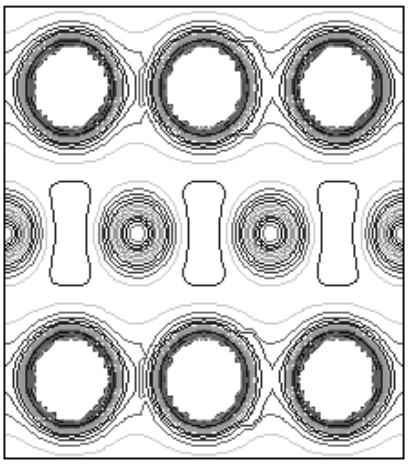

Fig.13a: Charge density map in the (110) plane of YFe_2 at a= 7.3 Å. The six large circles are Y atoms and the smaller ones are Fe atoms.

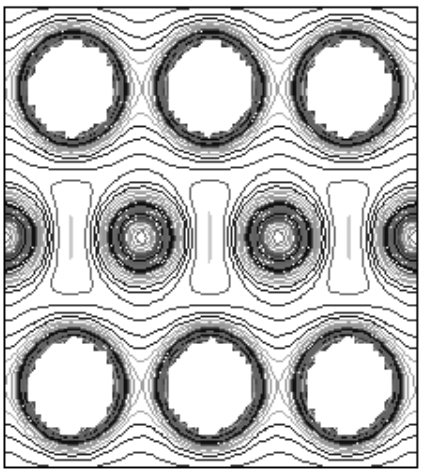

Fig13b: Charge density map in the (110) plane of YFe_2 at a= 6.5 Å. The six large circles are Y atoms and the smaller ones are Fe atoms.

Conclusions

We have done ab initio calculations of the magnetic moment, bulk modulus and electronic structure of YFe_2 , in both LSDA and GGA approximations of the DFT theory.

Our work indicates the collapse of the ferromagnetic state at high pressures. The exchange-correlation energy increases as the inter-atomic distance (and hence the cell volume) decreases. On the other hand, the magnetic moment shows a continuous increase upon increasing the exchange-correlation energy. The effect of increasing the pressure on increasing the charge density is demonstrated by the results of the charge density maps at two different volumes. The results of FPLO and wien2k package are in good accord with those of previous theoretical and experimental investigations.

Table 1. Our calculed results, experimental data and other calculations of lattice constant, total magnetic moment, Fe magnetic moment, Y magnetic moment, bulk modulus and first pressure- derivative of the bulk modulus of magnetic and nonmagnetic fcc YFe_2 .References are given in brackets.

Method	Phase	Lattice constant [Å]	Total M. M. / f. u.[μ_B]	M. M. of Y [μ_B]	M. M. of Fe [μ_B]	Bulk modulus [GPa]	B'
LSDA	magnetic	7.02	2.52	-0.346	1.43	148.4	4.9
	nonmagnetic	6.95	------	------	-------	167.8	7.0
GGA	magnetic	7.3	3.27	-0.696	1.98	94.9	1.53
	nonmagnetic	7.15	-------	------	------	140.7	2.7
Experimental	magnetic	7.363 [5], 7.35 [10]	2.9[12]	-0.50 [9, 10]	1.45[25]	129[3], 90.66[11], 97[26]	7.363[5], 7.35[10]
	nonmagnetic	-------	------	------	------	------	------
Other	magnetic		3.10[13], 2.87[21]	-0.45 [21]	1.36[5], 1.66[21]	143[3], 133[24], 123[21]	
	nonmagnetic	------	------	------	------	160[5]	------

Table 2. Our calculated, experimental and other calculation of lattice constant, magnetic moment, bulk modulus and first pressure- derivative of the bulk modulus of magnetic and nonmagnetic bcc Fe. References are given in brackets.

Method	Phase	Lattice constant	B (GPa)	B\	Magnetic moment [Bohr magneton]
LSDA	magnetic	a =2.75 Å	255.6	4.33	1.965
	nonmagnetic	a =2.7 Å	321.6	4.00	
GGA	magnetic	2.83 Å	201.53	4.96	2.17
	nonmagnetic	2.76 Å	272	3.91	
Experimental	magnetic	2.87 [27]	168 [27]		2.22 [27]
Others	magnetic		230 [5]		2.16 [5]
	nonmagnetic		300 [5]		

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