The effect of Ir on the magnetic and electronic properties of FePt alloy: A DFT study

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Abstract. $L1_0$-ordered FePt alloy is a promising material for high-density magnetic recording media due to its high magnetic anisotropy energy and coercivity. However, it was reported that this alloy faces challenges including issues with thermal stability and noise. Hence, ternary alloying with Ir was conducted to enhance the stability in response to the orientation of the magnetic spin moment of the binary FePt system. The structural, magnetic and electronic properties of $L1_0$-ordered $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$) were studied using the density functional theory. It was found that the lattice parameters and magnetic moments of the binary $Fe_{50}Pt_{50}$ are well in agreement with previous theoretical and experimental data to within 5 %. In all $Fe_{50}Pt_{50-x}Ir_x$ alloys, the calculated heats of formation were negative, demonstrating their thermodynamic stability. The magnetic moments and density of states were determined to evaluate the magnetic behaviour of $Fe_{50}Pt_{50-x}Ir_x$ alloys. These results contribute to the development of $Fe_{50}Pt_{50-x}Ir_x$ alloys as the next generation of magnets for high-density magnetic recording media.

1 Introduction

The growing demand for higher storage density in magnetic devices drives to the need for materials with high coercivity, strong thermal stability, and optimized magnetic anisotropy [1]. FePt alloy, meet these criteria, particularly in their chemically ordered $L1_0$ phase, exhibiting outstanding magnetic properties such as high magneto crystalline anisotropy energy (MAE), and high coercivity, making them perfect candidates for high-density magnetic recording media [2]. However, it was reported that this alloy faces challenges, including issues with thermal instability and media noise [3]. Among these problems, high ordering temperature problem is well addressed in previous studies, but the problem associated with thermal stability and reduction of media noise remain unsolved [4]. Hence, ternary alloying is proposed to enhance the stability in response to the orientation of the magnetic spin moment of the binary FePt system, which can solve the problem associated with thermal stability and media noise. Previous studies indicated that alloying FePt with elements like Cu, Mn, and Ni can effectively modify structural and magnetic properties [5, 6, 7]. For instance, Singh et al. performed annealing on

$Fe_{50}Pt_{50}$/Cu multilayer and shown that addition of Cu reduces the Fe-Pt ordering temperature. Chiang et al also achieved lower ordering temperature on Fe-Pt by addition of Cu which introduces dynamic stress while Yang et al. showed that doping Ni on Fe-Pt alloy decrease the coercivity of Fe-Pt alloy using experimental method. Furthermore, Gruner et al. indicated that Mn enhances phase stability on Fe-Pt alloy using first principle study. The addition of Ir to $L1_0$ ordered FePt can influence its magnetic and structural properties, including coercivity and MAE. Iridium's high melting point can help to improve phase stability in FePt alloy. In this study, ternary alloying of $L1_0$ ordered FePt with iridium (Ir) has been performed using density functional theory (DFT) approach to investigate the on structural, thermodynamic, magnetic, electronic and mechanical properties.

2 Methodology

All calculations in this work were performed using density functional theory implemented in the Vienna ab initio Simulation Package (VASP) [8, 9]. Electron-ion interactions in the system were described using the Projector Augmented Wave (PAW) pseudopotential [10]. The generalized gradient approximation (GGA) [11] developed by Perdew, Burke, and Ernzerhof (PBE) [12] was applied to account for the exchange correlation function. The total energy's convergence was investigated in terms of plane-waves basis set size and k- spacing. A plane-wave cutoff energy of 400 eV and k- spacing of 0.07 /Å were used in all the calculations. A 2x2x2 supercell with 32 atoms was constructed on $L1_0$ FePt system. The ternary $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$) were constructed using a substitutional search tool within the MedeA software and where platinum (Pt) was substituted with iridium (Ir). Before calculations, all $Fe_{50}Pt_{50-x}Ir_x$ alloys structures were optimized, where internal atom positions were allowed to relax. Then, the density of states and elastic properties were determined from optimized structures. The elastic properties were calculated using strain of 0.005.

3 Results and discussion

3.1 Structural and thermodynamic properties

The equilibrium lattice parameters as shown in Figure 1 were obtained by performing geometry optimization on $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$). Lattice parameters of binary structure ($Fe_{50}Pt_{50}$) were found to be a = b = 3.86 Å and c = 3.76 Å which are in good agreement with the experimental values (a = b = 3.86 Å and c = 3.71 Å) [13]. The equilibrium lattice parameters of the $L1_0$ $Fe_{50}Pt_{50}$ alloy reduces as the Ir composition was increased. This was expected since the atomic radius of Ir (1.36 Å) is less than that of Pt (1.39 Å).

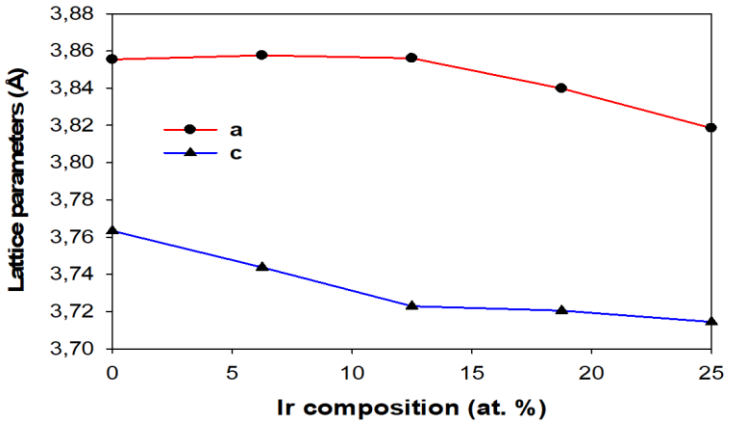


Figure 1: The equilibrium lattice parameters for $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$).

Heats of formation provide valuable information about energy change associated with formation of a crystal/material and can be used to predict the thermodynamic stability of a material. Structures with lower heats of formation values are considered most stable, as they require less energy to form and are more likely to exist in a lower energy state. To investigate the thermodynamic stability the heats of were calculated using equation 1:

$$\Delta H_f = E_C - \sum_i x_i E_i \qquad (1)$$

The calculated heats of formation as shown Figure 2 were negative, showing thermodynamic stability. The binary structure ($Fe_{50}Pt_{50}$) is the most stable structure with heat of formation of -0.47 eV/atom. It observed that the heats of formation increase linearly with the increase in iridium (Ir) composition, suggesting that Ir reduces thermodynamic stability.

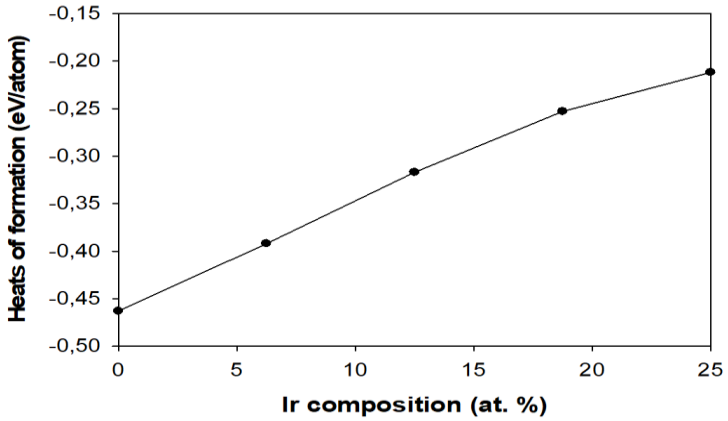


Figure 2: Heats of formation against Ir composition for $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$).

3.2 Magnetic properties

Magnetic moment is one of the magnetic properties that is used to investigate the magnetic strength of a material, which is an important fixture for magnetic recording devices application. Figure 3 shows the calculated magnetic moments for $Fe_{50}Pt_{50-x}Ir_x$ alloys (x= 6.25, 12.50, 18.75 and 25). The calculated magnetic moment of binary structure ($Fe_{50}Pt_{50}$) is 3.31 μB, which in good agreement with the experimental value of 3.24 μB [14]. From Figure 3, it is observed that magnetic moment of $Fe_{50}Pt_{50}$ alloy reduces as the Ir composition was increased, this suggests that Ir does not enhance magnetic strength of FePt alloy. According to Schmidt et al. [15], this behaviour is due to antiferromagnetic coupling between the FePt and Ir moments, resulting in partial reduction of magnetic moment.

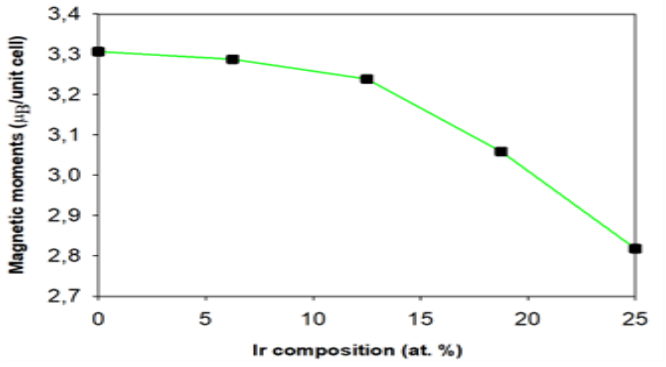


Figure 3: Magnetic moments against Ir composition for $Fe_{50}Pt_{50-x}Ir_x$ alloys.

3.3 Elastic properties

The elastic properties provide important information about mechanical behaviour of materials as they respond to applied stress or strain. Elastic constants are important parameters for predicting the mechanical stability of a material. $Fe_{50}Pt_{50-x}Ir_x$ alloys exhibits tetragonal shape and crystals and have six elastic constants (c_{11}, c_{12}, c_{13}, c_{33}, c_{44}, c_{66}). Tetragonal systems mechanical stability is given the following conditions [16]:

$$c_{44} > 0,\ c_{66} > 0,\ c_{11} > |c_{12}| \text{ and } c_{11}+c_{12} - \frac{2c_{13}^2}{c_{33}} > 0 \qquad (2)$$

Figure 4 represent the calculated elastic constants of the $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$). For the structure/material to be stable, the stability criterion for the elastic constants must be satisfied. All the calculated elastic constants

as shown in Figure 4 are positive, indicating that $Fe_{50}Pt_{50-x}Ir_x$ alloys are stable, does they satisfy the tetragonal stability criterion as indicated by equation 2. From Figure 4, it is observed that addition of iridium enhances mechanical stability since C_{44}, C_{66} and C_{11} increases as the iridium composition was increased. The $Fe_{50}Pt_{25}Ir_{25}$ alloy is observed to be most mechanical stable alloy.

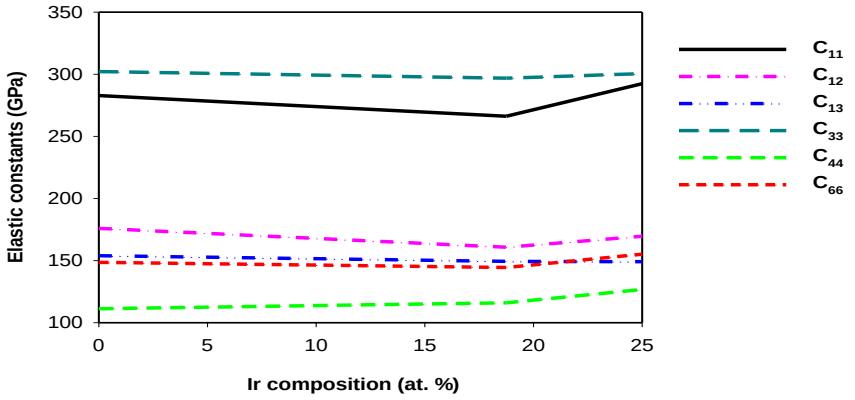
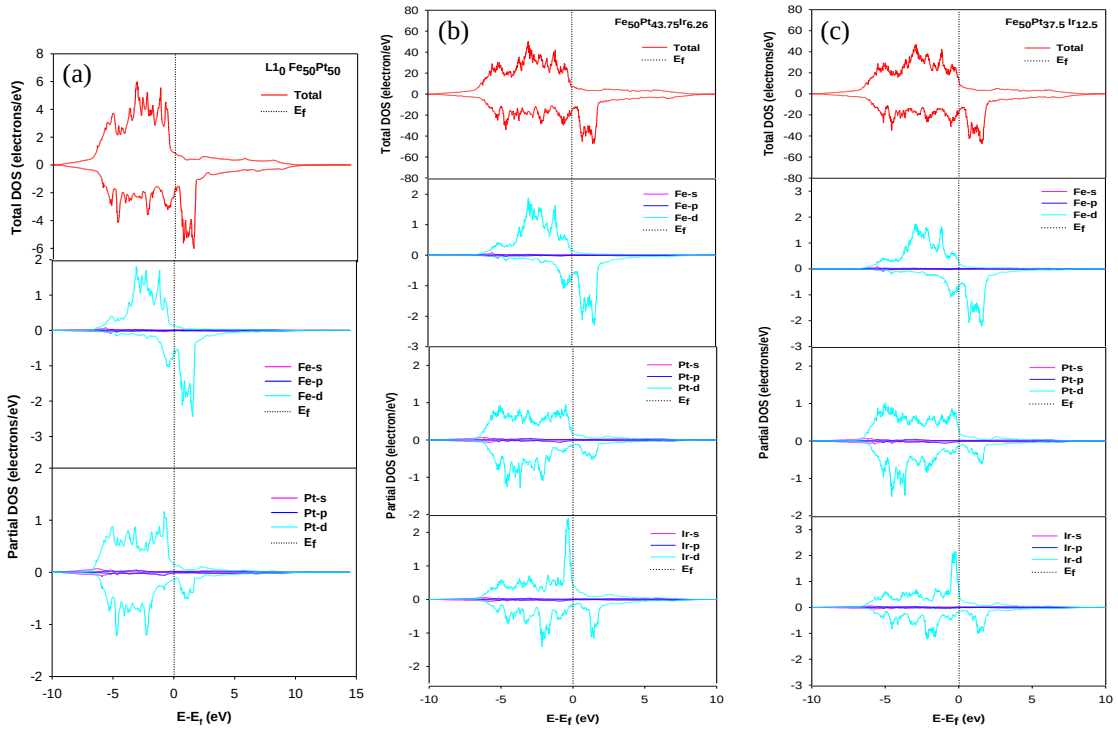


Figure 4: Elastic constants (GPa) against Ir composition for $Fe_{50}Pt_{50-x}Ir_x$ alloys ($0 \leq x \leq 25$).

3.4 Electronic properties

To investigate the electronic structures of $Fe_{50}Pt_{50-x}Ir_x$ alloys, spin polarized density of states (DOS) where computed as shown in figure 5. A significant difference in spin ups and spin downs is observed in the total DOS suggesting ferromagnetic behavior, which was also indicated by the calculated positive magnetic moments. The absence of energy gap around the fermi level indicates that the systems are metallic. From the partial DOS it is observed that iron (Fe) is the one that contribute more to the magnetism in $Fe_{50}Pt_{50-x}Ir_x$ alloys. Furthermore, it observed that Ir increases the DOS at the Fermi level. $Fe_{50}Pt_{50}$ system have less states near fermi level indicating that is more thermodynamically stable, which is in good agreement with the predicted heats of formation. Lastly it observed that the material retains its magnetism, this makes these materials best candidates for application in magnetic recording devices.



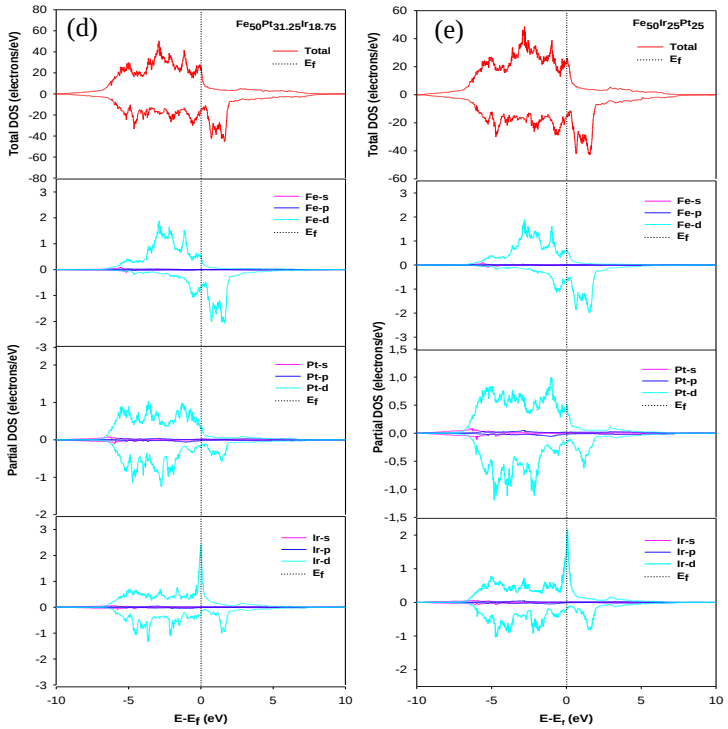


Figure 5 (a-e): The spin-polarized total and partial density of states for $Fe_{50}Pt_{50-x}Ir_x$ alloys.

4 Conclusion

The density functional theory (DFT) was successfully utilised to investigate the effect of iridium (Ir) substitution on the properties of the $L1_0$ ordered $Fe_{50}Pt_{50-x}Ir_x$ ($0 \leq x \leq 25$) alloys. The results of lattice parameters are in good agreement with the experimental values within 5%. In all $Fe_{50}Pt_{50-x}Ir_x$ alloys, the calculated heats of formation were negative, demonstrating thermodynamic stability. It was found that substitution of Ir slightly reduced magnetic strength of FePt alloy. Lastly, iridium (Ir) substitution enhanced mechanical stability of FePt alloy, and the most stable ternary alloy was found to be $Fe_{50}Pt_{25}Ir_{25}$ alloy. The total DOS showed that $Fe_{50}Pt_{50-x}Ir_x$ alloys are ferromagnetic and partial DOS indicated that iron (Fe) contribute more to the magnetism in $Fe_{50}Pt_{50-x}Ir_x$ alloys.

References

[1] G. W. Qin, Y. P. Ren, N. Xiao, B. Yang, L. Zuo, and K. Oikawa, "Development of high density magnetic recording media for hard disk drives: Materials science issues and challenges," *Int. Mater. Rev.*, vol. 54, pp. 157–179, 2009.

[2] N. Lethole, P. Ngoepe, and H. Chauke, "Compositional dependence of magnetocrystalline anisotropy, magnetic moments, and energetic and electronic properties on Fe-Pt alloys," *Materials*, vol. 15, p. 5679, 2022.

[3] K. Shibata, "FePt magnetic recording media: Problems and possibilities for practical use," *Jpn. Inst. Met.*, vol. 44, pp. 1542–1545, 2003.

[4] A. Alsaad, A. A. Ahmad, and T. S. Obeidat, "Structural, electronic and magnetic properties of the ordered binary FePt, MnPt, and $CrPt_3$ alloys," *Heliyon*, vol. 6, p. 3596, 2020.

[5] S. Singh, C. L. Prajapat, M. Gupta, and S. Basu, "Room temperature superparamagnetism in ternary $(Fe_{50}Pt_{50})_{0.42}Cu_{0.58}$ phase at interfaces on annealing of $Fe_{50}Pt_{50}$/Cu multilayer," *J. Magn. Magn. Mater.*, vol. 462, pp. 58–69, 2018.

[6] C.-H. Lai, C. C. Chiang, and C. H. Yang, “Low-temperature ordering of FePt by formation of silicides in underlayers,” J. Appl. Phys., vol. 97, 2005.

[7] M. E. Gruner and P. Entel, “Structural and magnetic properties of ternary $Fe_{1-x}Mn_xPt$ nanoalloys from first principles,” Beilstein J. Nanotechnol., vol. 2, pp. 162–172, 2011.

[8] W. Kohn and L. J. Sham, “Self-consistent equations including exchange and correlation effects,” *Phys. Rev.*, vol. 140, pp. 1133–1138, 1965.

[9] J. Hafner and G. Kresse, “The Vienna ab-initio simulation program VASP: An efficient and versatile tool for studying the structural, dynamic, and electronic properties of materials,” *Appl. Sci.*, vol. 351, pp. 351–358, 1997.

[10] P. E. Blöchl, “Projector augmented-wave method,” *Phys. Rev. B*, vol. 50, pp. 17953–17979, 1994.

[11] J. P. Perdew, K. Burke, and M. Ernzerhof, “Generalized gradient approximation made simple,” *Phys. Rev. Lett.*, vol. 77, pp. 3865–3868, 1996.

[12] J. P. Perdew and A. Zunger, “Self-interaction correction to density-functional approximations for many-electron systems,” *Phys. Rev. B*, vol. 23, pp. 5048–5079, 1981.

[13] A. Alsaad, A. A. Ahmad, and T. S. Obeidat, “Structural, electronic and magnetic properties of the ordered binary FePt, MnPt, and $CrPt_3$ alloys,” *Heliyon*, vol. 6, p. 3596, 2020.

[14] J. Lyubina, I. Opahle, M. Richter, O. Gutfleisch, K. Müller, and L. Schultz, “Influence of composition and order on the magnetism of Fe–Pt alloys: Neutron powder diffraction and theory,” *Appl. Phys. Lett.*, vol. 89, no. 032506, 2006.

[15] N. Y. Schmidt, S. Laureti, F. Radu, H. Ryll, C. Luo, F. d’Acapito, S. Tripathi, E. Goering, D. Weller, and M. Albrecht, “Structural and magnetic properties of FePt-Tb alloy thin films,” *Phys. Rev. B*, vol. 100, pp. 1–8, 2019.

[16] M. J. Phasha, P. E. Ngoepe, H. R. Chauke, D. G. Pettifor, and D. Nguyen-Mann, “Link between structural and mechanical stability of fcc- and bcc-based ordered Mg–Li alloys,” *Intermetallics*, vol. 18, pp. 2083–2089, 2010.