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Magnetic Properties of Strained $L1_0$-ordered FePt and CoPt: An *ab initio* Study

Heechae Choi*

Center for Computational Science, Korea Institute of Science and Technology, Seoul 136-791

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Using *ab initio* calculations, the effects of uniaxial, biaxial, and hydrostatic strains on the magnetocrystalline anisotropy of $L1_0$-orderd FePt and CoPt alloys were systematically investigated. Interestingly, the rates and the signs of magnetocrystalline anisotropy changes of FePt and CoPt were determined by the directions and dimensions of strains. The calculation results are consistent with the previous experimental observations and are expected to provide directions to tailor magnetic properties of various types of $L1_0$-ordered FePt and CoPt systems.

Keywords : ab initio, DFT, Strain, FePt, CoPt, Magnetic anisotropy energy, MAE

I. Introduction

$L1_0$-ordered ferromagnetic fct alloys have attracted substantial attention during the last decade for applications to high density recording media up to 1Tbit/in^2 [1]. In particular, fct $L1_0$ FePt and CoPt are among the best candidate materials for nonvolatile recording systems due to their huge magnetocrystalline anisotropies (MCA) K_u ($> 5\times10^7$ erg/cm^3), high Curie temperature ($>$750 K), and good chemical stability [2-5]. The MCA values of $L1_0$ fct alloy are observed even in ultrathin films, and this enables their application to ultra-high density nonvolatile memories such as spin-transfer torque magnetic random access memory [6,7]. Control of the MCA of FePt and CoPt systems is one of the main areas of research on these systems because it determines the energetic stability of storage and switching current density [8,9]. The magnetic properties of fabricated nanoparticles or thin films are the result of a complex interplay of many factors (strains, interface effect, compositions, etc.). It is therefore important to ascertain the contribution of each factor to the magnetic properties of the fabricated FePt alloy systems. The effects of strains on MCA when the fabrication conditions are changed have been discussed in previous studies [10-12]. However, it is extremely difficult to study the effects of strain on the magnetic properties of systems in isolation since such factors vary simultaneously in the fabrication processes [5]. Density functional theory (DFT) [13,14] based *ab initio* studies have successfully provided theoretical analyses on the magnetic alloy systems because this method is capable of clarifying the effect of each factor on the changes of magnetic properties of FePt CoPt alloy systems. For example, a theoretical work by Zhu et al. [5] reported that expansion of the $L1_0$ FePt alloy system in lateral directions reduces the MCA and the switching current. However, another previous theoretical work by Lee et al. [15] reports that volume expansion occurs three-dimensionally near the surface region of a $L1_0$-orderd

* [E-mail] heechae@kist.re.kr
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FePt thin film when the c-axis of the film is along the in-plane direction and this type of volume change has a negligible effect on the MCA change of the $L1_0$ FePt surface alloys. Since the unit cell volume changes of $L1_0$ FePt thin films or nanoparticles occur in various ways depending on the choice of substrates for growth, composition, and the film crystallographic direction, it is necessary to perform a systematic study on the effects of volume changes on the magnetic properties of the system.

Here, to comprehensively address the issue of strain effects on the MCA of $L1_0$ FePt and CoPt, we present the changes of the MCA of systems with uniaxial, lateral, and hydrostatic strains using DFT based *ab initio* calculations. We demonstrate that the linearly changing MCA and magnetic moments of $L1_0$ FePt and CoPt with strains are dependent on the direction and dimension of strains. This systematic study is expected to provide clear direction for the fabrication of ferromagnetic $L1_0$ FePt and CoPt where strain effects on MCA should be discussed.

II. Calculation Methods

The DFT calculations in this study were performed using generalized gradient approximation (GGA) [16] with the projector augmented wave (PAW) method [17]. The PAW method allows the use of moderate cut-off energies in the construction of the plane wave basis. All calculations were performed with the Vienna *ab initio* simulation package (VASP) [18] using a plane wave cut-off energy of 400.0 eV (29.4 Ry). The Brillouin-zone integrations were performed using the tetrahedron method and 14×14×14 Monkhorst-Pack scheme [19]. To study the effects of strains on the magnetic properties and the electronic structures of the alloy systems, the experimental values of the lattice constants (a=3.86 Å/c=3.72 Å for FePt and a3.81 Å/c=3.68 Å for CoPt) were used in our calculations [20,21]. The strains are applied by changing the lattices constants of fct FePt and CoPt unit cells to provide 6% volume changes by both compression and expansion. To study the effects of the volume changes on the magnetic properties of $L1_0$ FePt and CoPt systems, four types of strains were considered (Fig. 1). According to the experimental observations of the c/a ratio evolution by c-axis elongation and compression (Type I) [22], two-dimensional (2D) strains by the lattice mismatches with substrates (Type II and Type III) [5,23], and the three-dimensional (3D) hydrostatic volume changes (Type IV) [15,24], we applied consistent strains by changing the lattice constants. For the cases of $L1_0$ FePt structures with perpendicularly precipitated Fe and Pt planes, we also consider the 2D strains on a- and c-axes (Type III).

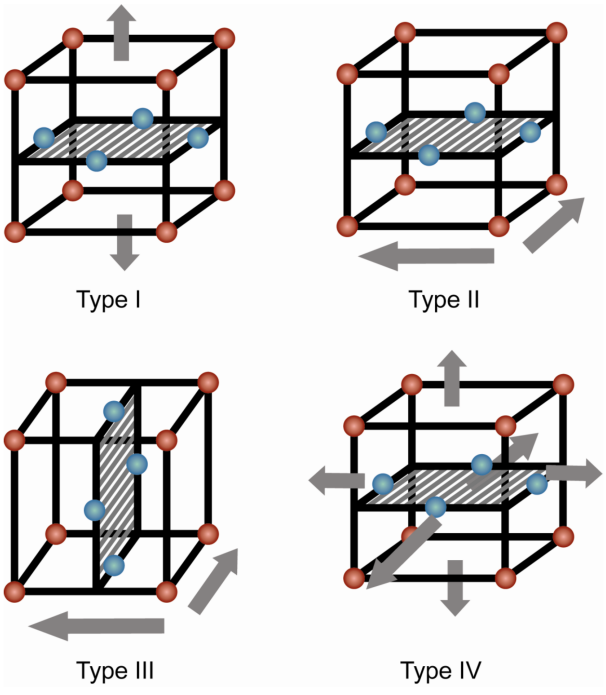

Figure 1. The types of strains on $L1_0$ alloy system. Type I strain is c-axis elongation, Type II (III) strain produces lateral strain in the a- and b-axes (a- and c-axes), and Type IV strain gives a hydrostatic volume change. The brown (blue) spheres represent Pt (Fe or Co) atoms, or vice versa.

III. Results and Discussion

The calculated magnetic moments of FePt and CoPt alloys are depicted for the fraction of volume change (Fig. 2). As shown in the graphs for FePt (Fig. 2(a)) and CoPt (Fig. 2(b)), the magnetic moments of Fe and Co in FePt and CoPt are linearly increased with increment of the cell volumes, under all strain types. The magnetic moments of Fe, Co, and Pt in $L1_0$ FePt and CoPt alloys in this study are consistent with reports based on previous experimental and theoretical works [5,6,15,23]. Type II strain increases both the Fe and Co magnetic moments most sharply. The Pt magnetic moment in FePt increased most rapidly with biaxial strains on the a- and b- axes (Type II) and decreased with uniaxial strain along the c-axis (Types I). It is quite interesting that a biaxial strain in the a- and c-axes on $L1_0$ FePt decreases the Pt magnetic moment. In other words, the Pt magnetic moments in $L1_0$ FePt show a different tendency of changes depending on the strain direction. The increasing magnetic moments of $L1_0$ FePt by 2D strains in this study are consistent with a previous experimental work by Lee et al. [23], who reported varying saturate magnetization of $L1_0$ FePt depending on the lattice mismatch with the underlying substrate. Hence, we reason that the varying magnetic moments of the grown $L1_0$ FePt thin films are due to the dominant intrinsic property of the $L1_0$ FePt alloy system, which has been predicted from experimental works [6,10] on the effects of interfaces. In a theoretical work Lee et al. [15] reported that the c/a ratio is increased and the cell volume of $L1_0$ FePt is expanded near the surface. It can therefore be inferred that the ultra-thin films have large portions of a high c/a ratio and expanded volume and increased Fe magnetic moments. The Co magnetic moment also linearly in-

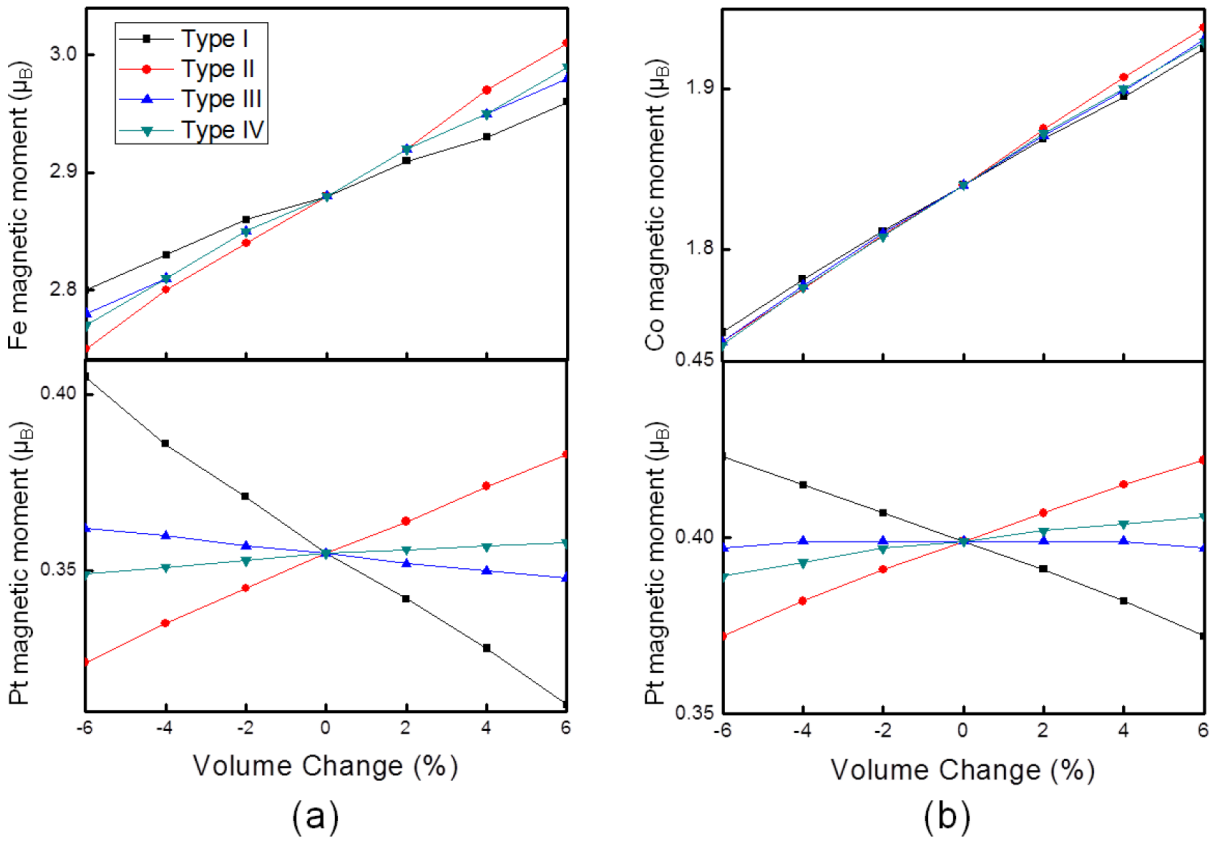

Figure 2. The magnetic moments of each atom in $L1_0$ (a) FePt and (b) CoPt.

creased with the unit cell volume for all types of strain (Fig. 2(b)). The changes of the Pt magnetic moment in CoPt also show clear linearity with a similar tendency with that of the FePt system.

The decomposed density of states (DOS) for 3*d*-orbitals of Fe and Co show how the magnetic moments are changed by the cell volume expansion and compression (Fig. 3). The increased (decreased) magnetic moment of Fe in $L1_0$ FePt by Types I and II strains originated from the enhanced (reduced) spin-majority states of Fe $3d_{yz}$ and $3d_{zx}$ orbitals (Fig. 3(a)). The magnetic moments of Co in the $L1_0$ CoPt system are increased by a different mechanisim: the changed electron occupancies in $3d_{x2-y2}$ orbital and overall 3*d*-orbital under Types I and II strains, respectively (Fig. 3(b)).

The changes of MCA of $L1_0$ FePt and CoPt under volume changes are depicted in Fig. 4. The MCAs of the $L1_0$ FePt and CoPt were obtained using the equation,

$$\mathrm{MCA} = \mathrm{E}(\vec{M}_{\mathrm{a-xis}}) - \mathrm{E}(\vec{M}_{\mathrm{c-xis}}) \quad (1)$$

where $\mathrm{E}(\vec{M}_{\mathrm{a-xis}})$ and $\mathrm{E}(\vec{M}_{\mathrm{c-xis}})$ are the total energies of the alloy systems with magnetization in the a- and c-axes, respectively.

The calculated MCAs of the unstrained $L1_0$ FePt and CoPt (5.2 and 1.3 meV/cell) were consistent with previous experimental and theoretical studies [4,15]. The MCA of FePt increased only with volume expansion by elongation in the c-axis (Type I). With the Types II and IV strains, the MCAs of $L1_0$ FePt decreased steadily. However, the effect of compression with Type III strain on the MCA of $L1_0$ FePt is negligible (Fig. 4(a)). The decreased MCA of the $L1_0$ FePt by Type II strain is consistent with previous theoretical [5] and

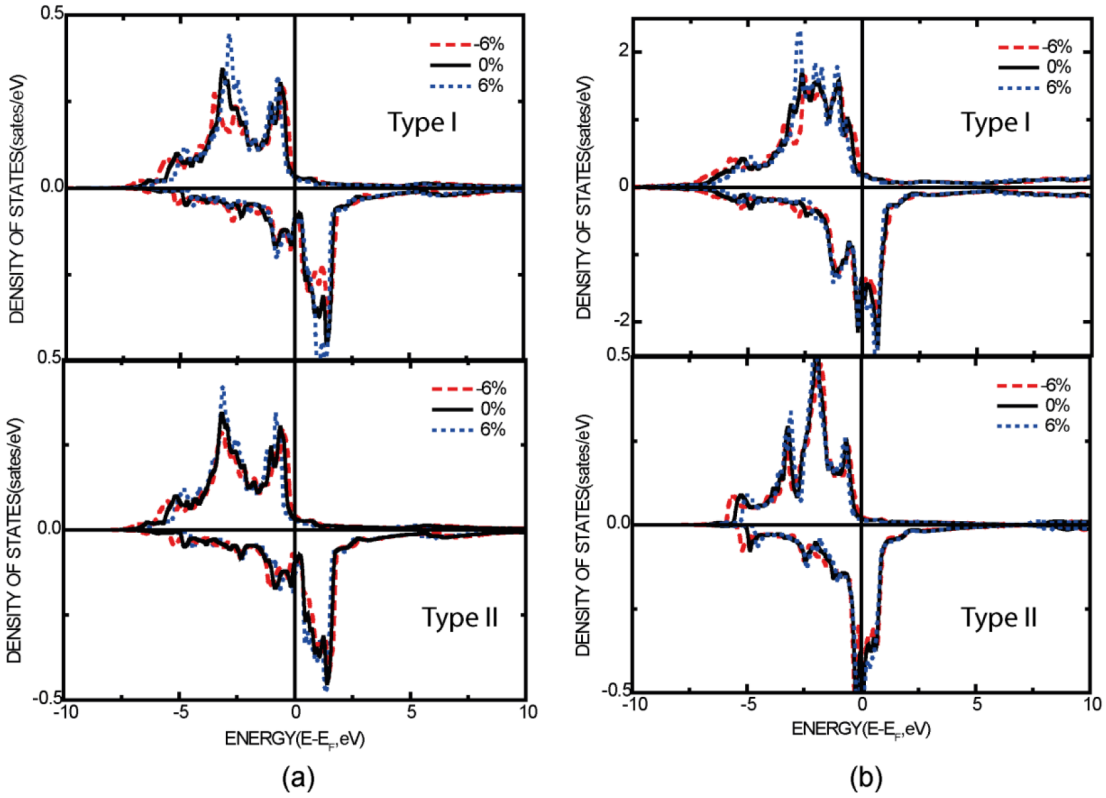

Figure 3. The decomposed density of states of (a) the $3d_{yz}$- and $3d_{zx}$- orbitals of Fe in $L1_0$ FePt under strain Types I and II, and (b) Co in $L1_0$ CoPt under Type I strain (total 3d-orbital) and Type II strain ($3d_{x2-y2}$ orbital).

experimental [6] works. Recent theoretical works using a Monte Carlo simulation [9] and DFT calculations [15] reported that the in-plane alignment of the c-axis of $L1_0$ FePt thin films is also a stable phase. Reorientations of the c-axis of $L1_0$ FePt in in-plane directions are observed in experiments according to the choice of substrate materials [23]. From the calculation results of this study, reorientation of the c-axis of $L1_0$ FePt alloy to the in-plane direction weakens the effect of the lattice mismatch on the MCA of the system relative to the effect of the lattice mismatch on the MCA of perpendicularly oriented $L1_0$ FePt.

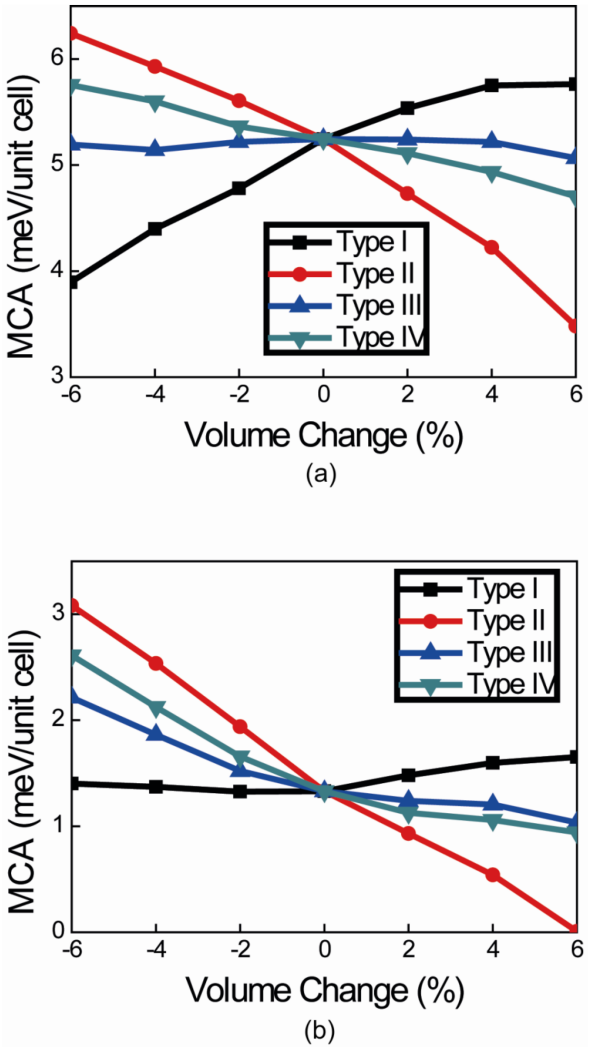

Figure 4. Magnetocrystalline anisotropy (MCA) of $L1_0$ (a) FePt and (b) CoPt under the four strain types.

The MCA of $L1_0$ CoPt also increased only with volume expansion by Type I strain, while c-axis elongation and other types of strains decrease the $L1_0$ CoPt MCA (Fig. 4(b)). For $L1_0$ CoPt, compressive strain (Type IV) was found to have a negligible effect on the CoPt MCA. Type II strain decreases the CoPt MCA most sharply and turns the magnetic easy axis from the c-axis to the a-axis at a 6% volume change. Elongation of the $L1_0$ FePt in c-axis by a boron additive was recently reported [25]. From the calculation results of this study, it was clearly demonstrated that the c-axis elongation increases the MCA of $L1_0$ CoPt and the effect of additives is potentially another worthwhile topic to study. Spin-reorientation in a $L1_0$ CoPt film was observed in a previous experimental work [4], where a $L1_0$ CoPt film deposited on a MgO(111) substrate displayed a switched spin direction, from the c- to a-axis, at high temperature. Hence, it can be reasoned that the magnetic easy axis of $L1_0$ CoPt can be altered by strains in lateral directions (Type II) regardless of the crystallographic direction of the film. Spin-reorientation of the $L1_0$ CoPt film to the in-plane direction occurs by change of the crystallographic direction, too [26]. In such a case, the lateral lattice mismatch of deposited $L1_0$ CoPt with the substrate corresponds to Type III strains. Similarly to the $L1_0$ FePt alloy system, the effect of lattice mismatch is weaker than the film with the c-axis in the surface-normal direction, as shown in Fig. 4(b).

IV. Conclusion

In summary, we systematically investigated the effects of strains on the magnetic properties of $L1_0$ FePt and CoPt alloy systems using DFT calculations. The magnetic moments of Fe and Co in FePt and CoPt linearly increased with volume changes with all types of strain. The calculated MCAs of both $L1_0$ FePt and CoPt increased with volume changes only by uniaxial strain in the c-axis. The effects of 2D strains on the MCAs of

FePt and CoPt were found to vary significantly depending on the crystallographic direction. The hydrostatic volume expansions decreased the MCAs of both the FePt and CoPt system, and the MCA of the latter was not affected by compression from hydrostatic pressure. The results of this study are expected to provide a clear direction to design magnetic memory device materials with tailored magnetic properties.

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