

Determination of Magneto-crystalline Anisotropy Energy (MAE) Of ordered L1₀ CoPt and FePt nanoparticles

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Abstract: The structural and magnetic properties of both L1₀ ordered FePt and CoPt nanoparticles make them potential candidates for optical-electronic and magneto-optical devices. First, we carried out *an ab initio* total energy minimization study to find the geometrical optimization of both L1₀ phases of FePt and CoPt nanoparticles. Then, we investigated the magnetocrystalline anisotropy energy (MAE) of both systems along special line joining the points of high symmetry (A,B and C points) using super-cell slab approach with alternating layers Fe/Co and Pt along the (001) direction. We found that the point (A) has the highest MAE value for both systems, where the value of MAE in FePt is 8.89×10^7 erg/cm³ and in CoPt is 6.40×10^7 erg/cm³. Our spin density based calculations indicate that large spin-orbit interaction and the hybridization between Pt 5d states and Fe/Co 3d states are the dominant factors in determining the MAE in both systems.

Keywords: Magneto-crystalline Anisotropy Energy (MAE); L1₀ CoPt and FePt nanoparticles; Spin density functional theory; Slap approach; Spin-orbit interaction

1. Introduction

A lot of efforts have been devoted to explore the physics of tiny structure for magnetic materials that can help to utilize its desirable properties [1-3]. In recent years, applications of magnetic nanoparticles have arisen, particularly those that have a high anisotropic energy [1, 4-7]. Because of their potential applications such as data storage, optical filters, and biomedical applications, some of magnetic nanoparticles gain crucial importance [8-10]. The value of magneto-crystalline anisotropy energy (MAE) of a magnetic material indicates the magnetic stability in response to the orientation of the magnetic spin moment. The storage devices based on magnetic materials are of great interest from fundamental and technological points of view due to their low cost [11-26]. When the thermal energy is large enough and able to overcome the anisotropy energy, the phenomenon of super paramagnetic occurs [29]. As a result, the magnetization is not stable which causes the damage and loss of the storage data [30]. To overcome this issue, we must look for magnetic materials that have high anisotropic energy [10, 31-32]. The FePt and CoPt nanoparticles adopting the face center tetragonal phase are considered among promising candidates for the field of data storage due to their high magnetic anisotropy energies [2, 16, 26]. Among the variant magnetic nanoparticles and their alloys, ordered ferromagnetic L1₀ FePt and CoPt alloys have desirable structural and magnetic properties. For instance, they exhibit good response to an external magnetic field; also, they show high magnetocrystalline anisotropy in bulk phase [2, 12]. In addition, The L1₀ FePt and L1₀ CoPt structures have huge values of MAE compared to the other magnetic nanoparticles. The MAE'S values lie in the range ($7-11 \times 10^7$ erg/cm³) for L1₀ FePt structure [11, 13-14] and



approximately 5×10^7 erg/cm³ for L10 CoPt [7, 14-15] structure. The CoPt binary exhibits a ferromagnetic behavior in its disordered A1 phase, while an ordered L12 CoPt phase possesses a cubic anisotropy and is said to be magnetically soft. On the other hand, the L10 FePt and CoPt phases are well known to have very high uniaxial magnetocrystalline anisotropy. The origin of this large anisotropy mainly comes from the significant spin-orbit coupling and the large hybridization between Co (Fe) 3d bands and Pt 5d states [36]. In this work, we attempt to present very accurate calculations of MAE in L10 CoPt and FePt nanoparticles using slab approach and spin density functional theory. The MAE of bulk FePt and CoPt is defined to be the energy difference, $E_{100} - E_{001}$ between the two magnetic configurations in which the magnetic moments are parallel or perpendicular to the Fe (Co) or Pt (0 0 1) planes.

2. Methodology and Calculations

The methodology of the work includes calculations within the spin density functional theory (SDFT) using a plane wave basis set and the Projector augmented wave (PAW) pseudopotentials that are implemented in Vienna ab initio simulation package (VASP) [37-40]. The PAW pseudo potentials include the following valence electrons: Fe: $3p^6 3d^7 4s^1$ and Pt: $5d^9 5s^1$. The generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof (PBE) form was used for the exchange–correlation functional. The used parameters in calculating MAE of both FePt and CoPt SYSTEMS are as following: the plane wave cut-off energy is 400 eV, a $12 \times 12 \times 1$ and $24 \times 24 \times 1$ Monkhorst–Pack k-point meshes are used for both FePt and CoPt, respectively. The relaxed atomic positions were followed by minimizing the total energy (i.e. 0.003eV), and the Hellmann–Feynmann forces. These forces were as small as (0.003eV/Å) at convergence in the unit cell of all different electronic structure relaxations that have been performed. Finally, in order to evaluate the MAE results for both systems we performed self-consistence calculations for total energy for two different magnetic configurations. One configuration is demonstrated by allowing the magnetic moments to be oriented parallel, $E(\parallel)$, and the other by forcing the magnetic moments to be aligned perpendicular, $E(\perp)$, to the easy axis (c-axis). And then MAE can be considered as the energy difference between the two configurations,

$$MAE = E(\parallel) - E(\perp)$$

3. Results

Crystallography Aspects Of The L10 Structure

The prototype structure for CoPt and FePt structures is the CuAu (I) type structure. The CoPt and FePt exhibit an A1 phase (The structure with A1 phase possess a disordered face centered cubic (fcc) structure) at high temperatures and they exhibit an ordered L10 or L12 phase at low temperatures. Figure 1 represents the unit cell of ordered L10 phase of both FePt and CoPt nanoparticles. In L10 FePt and CoPt structures, there are two types of atoms, one type of atoms occupy the corners and corresponding to $[0\ 0\ 0]$ and $[1/2\ 1/2\ 0]$ lattice sites, whereas the second type of atoms are located at the other two face centers with $[1/2\ 0\ 1/2]$ and $[0\ 1/2\ 1/2]$ site positions. In L10 structure, the arrangement of alternately aligned Co or Fe and Pt planes along the c-axis $[001]$ results in the distortion of the cubic symmetry, this symmetry distortion leads to a slight tetragonal distortion in the unit cell. Hence this structure is referred to as an ordered face centered tetragonal (fct) L10 phase. The space group of the L10 structure is $P4/mmm$. We performed an *ab initio* total energy minimization study to find the geometrical optimization of both L10 phases of FePt and CoPt nanoparticles. The calculated values of the equilibrium lattice parameters are $a_0 = 3.880\ \text{\AA}$, $c_0 = 3.780\ \text{\AA}$ and $a_0 = 3.814\ \text{\AA}$, $c_0 = 3.710\ \text{\AA}$ for FePt and CoPt, respectively. These results are in agreement with data reported in the literature (2, 15-17, 31, 33).

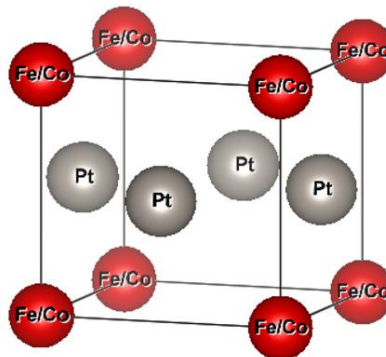


Fig.1. Unit cell of ordered L10 phase of both FePt and CoPt nanoparticles.

Determination of Magneto-crystalline Anisotropy Energy (MAE)

In magnetic anisotropic materials, the magnetic moment tends to be oriented in the energetically favorable direction, this direction is called "easy axis". By definition the magneto-crystalline Anisotropy Energy (MAE) is defined as the total energy E_{tot} associated with the change in the direction of the magnetization

with respect to the easy axis [34-35]. When the magnetic material has one easy axis, the material is called uniaxial. The L10 ordered FePt and CoPt structures are familiar examples of uniaxial magnetic materials. The MAE for L10 (face centered tetragonal) phase can be defined and calculated as the energy difference between two magnetic orientations as shown in equation [2, 16, 24],

$$\Delta E = E_{100} - E_{001},$$

we used super-cell slab approach to compute the MAE of the L10 FePt and CoPt systems. The super cell approach is constructed from a relatively atomic layers (slap) separated by vacuum layers. The super cell is repeated throughout all space using periodic boundary conditions as shown in the Figure 2. Here, each slap in the super cell is constructed from ten atoms of Fe and Co and the eight Pt atoms with vacuum layers of 30 Å in z direction.

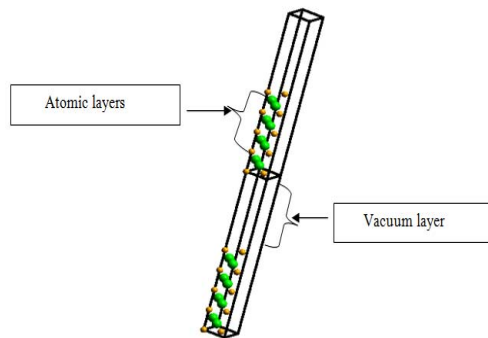


Figure 2: Super-cell slab approach which explain the atomic and vacuum layers. The small spheres denote the Fe/Co atoms while large spheres refer to Pt atoms.

We use this approach in order to compute the MAE of the FePt and CoPt along the high symmetry points. These points are symbolized by A, B, and C as shown in Figure3. The point A represents the regular L10 structure whereas point (B) is obtained by shifting the Fe or Co atoms by a displacement of (0.25, 0.25, 0) while the Pt atoms are shifted alternately between x and y coordinates by (0.75, 0.25,0) and (0.25, 0.75, 0). At point C with the shift of (0.5, 0, 0) the crystallites arranged atoms of Pt directly on top of each other across the interface in such a way that it corresponds to ideal FePt and CoPt lattices where one Fe or Co layer is removed. The structure of point C corresponds to a crack with one nearest neighbor across the interface.

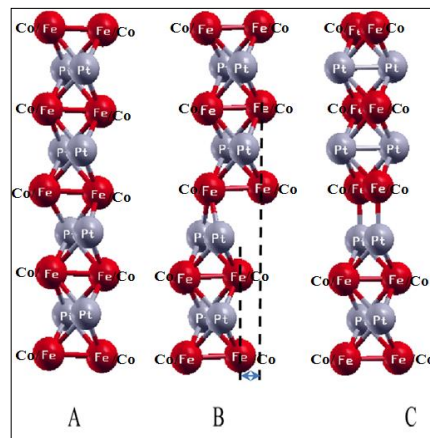


Figure 3: The super-cell slab approach of the ordered L1₀CoPt and FePt systems, the high symmetry points A, B and C from left to right in regular super-cell structure, red spheres are used for Fe or Co atoms.

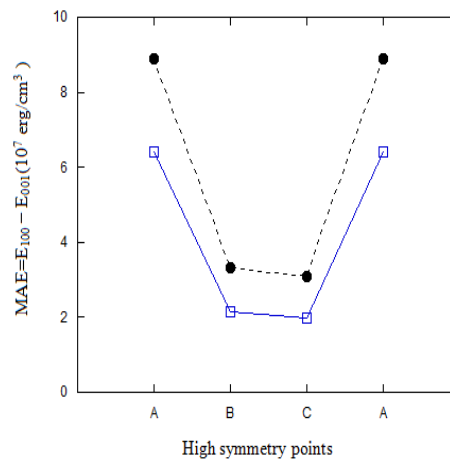


Fig. 4: Magnetocrystalline anisotropy energy (MAE) of L1₀ FePt (Black-dotted) and CoPt (Blue-solid) nanoparticles Plotted versus slip step along high symmetry points A-B-C-A of the regular lattice. Note that 1 J/m^3 is equivalent to $1 \times 10^{-5} \text{ Merg/cm}^3$.

Figure 4, represents the MEA per unit volume of L1₀FePt and L1₀CoPt at the high symmetry points. In FePt, point A has the highest MEA ($8.89 \times 10^7 \text{ erg/cm}^3$), followed by point B ($3.314 \times 10^7 \text{ erg/cm}^3$), and point C has the lowest MEA ($3.087 \times 10^7 \text{ erg/cm}^3$). In CoPt system, the corresponding MAE values are at point A ($6.40 \times 10^7 \text{ erg/cm}^3$), at point B ($2.15 \times 10^7 \text{ erg/cm}^3$), and at point C ($1.98 \times 10^7 \text{ erg/cm}^3$). It is obvious from Figure 3 the MAE decreases in along the line A→

$B \rightarrow C \rightarrow A$ in both $L1_0$ FePt and $L1_0$ CoPt nano systems. The physical interpretation of our striking result comes from the spin-polarized band calculations including spin-orbit interaction for $L1_0$ -FePt and CoPt ordered alloys performed within the framework of local spin density functional approximation. We have found that strong magnetocrystalline anisotropy of both ordered systems along the $A \rightarrow B \rightarrow C \rightarrow A$ line is due to a large spin-orbit coupling of Pt atom with Fe and Co atoms and a strong hybridization of Pt d bands with highly polarized Fe (Co) d bands. These two effects decrease along the $A \rightarrow B \rightarrow C \rightarrow A$ line for both ordered systems. This can be regarded as being associated with the behavior that the MAE of both systems decrease with decreasing of the strength of the spin-orbit coupling of Pt d-band with Fe (Co) bands, as well as, a significant decrease of Pt d-band hybridization with Fe (Co) d-bands.

4. Conclusion

We report our results on the structural and magnetic properties of $L1_0$ -FePt and CoPt ordered systems using spin density functional theory. The spin-polarized band calculations including spin-orbit interaction for $L1_0$ -FePt and CoPt ordered systems have been performed within local spin density functional approximation. Magneto-crystalline Anisotropy Energy (MAE) of $L1_0$ ordered FePt and CoPt nanoparticles were computed using super-cell slab approach within the framework of local spin density approximation. The (MAE) was computed at high symmetry points that are carefully selected in $L1_0$ ordered FePt and $L1_0$ CoPt systems. The points were chosen because of their importance from fundamental and experimental points of view. We found that MAE's of $L1_0$ FePt ordered structures decreases from $8.89 \times 10^7 \text{ erg/cm}^3$ to $3.087 \times 10^7 \text{ erg/cm}^3$ and that of $L1_0$ CoPt decreases from $6.40 \times 10^7 \text{ erg/cm}^3$ to $1.98 \times 10^7 \text{ erg/cm}^3$ along the $A \rightarrow B \rightarrow C \rightarrow A$ line joining points of high symmetry. This significant decrease in MAE's values of both ordered systems can be explained as being related with the observed decrease of the strength of the spin-orbit coupling of Pt d-band with Fe (Co) bands, as well as, a drastic decrease of Pt d-band hybridization with Fe (Co) d-bands. Our results are quite important for magnetic devices whose functionalities depend extremely on the magnetic reversal. This finding is very important and could be a guide for experimental research groups who are interested in growing both $L1_0$ FePt and $L1_0$ CoPt nano systems along the prescribed line joining the symmetry points. Our results could be used as guidelines for studying cracks that are formed during the growth of $L1_0$ FePt and $L1_0$ CoPt nanoparticles.

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