Effects of composition and chemical disorder on the magnetocrystalline anisotropy of Fe$_x$Pt$_{1-x}$ alloys

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Abstract - We perform first-principles calculations of the magnetocrystalline anisotropy energy (MAE) of the L1$_0$-like Fe$_y$Pt$_{1-y}$ samples studied experimentally by Barmak and co-workers (see J. Appl. Phys., 98 (2005) 033904). The variation of composition and long-range chemical order in the samples was studied in terms of the coherent potential approximation. In accordance with experimental observations, we find that, in the presence of long-range chemical disorder, Fe-rich samples exhibit a larger MAE than stoichiometric FePt. By considering the site- and species-resolved contributions to the MAE, we infer that the MAE is primarily a function of the degree of completeness of the nominal Fe layers in the L1$_0$ FePt structure.

Due to its extraordinarily high magnetocrystalline anisotropy energy (MAE), L1$_0$ FePt is of considerable interest to the development of ultrahigh-density magnetic-recording applications, in particular, for heat-assisted magnetic recording (HAMR). The L1$_0$ phase of Fe$_{50}$Pt$_{50}$ is a layered face-centered tetragonal structure, exhibiting alternating Fe and Pt layers along the (001) direction. FePt also exhibits stable FePt$_3$ and Fe$_3$Pt phases as well as a chemically disordered, cubic phase [1,2]. Accordingly, FePt exhibits phase transitions with respect to composition as well as to chemical order and understanding the related effects on the magnetic properties is an important issue. The large effect of chemical disorder on the MAE of Fe$_{50}$Pt$_{50}$ has already been outlined both experimentally [3–5] and theoretically [6–8]. Chemical disorder also has significant impact on the MAE of other L1$_0$ alloys (see, e.g., [9–11].)

The degree of long-range chemical order is quantified in terms of a chemical order parameter [12,13]. The L1$_0$ Fe$_y$Pt$_{1-y}$ alloy is modelled by a repeating sequence of two atomic layers, characterised by compositions Fe$_{r_F}$Pt$_{1-r_F}$ and Fe$_{1-r_F}$Pt$_{r_F}$, respectively. The fractions, $r_F$ and $r_P$, are related to each other through the condition, $1 + r_F - r_P = 2x$. Furthermore, set by the requirement, $r_F \geq 1 - r_P$ (the case of $r_F < 1-r_P$ can simply be obtained by interchanging the two types of layers), the range of $r_F$ is confined to $r_F \geq x$ ($r_P \geq 1-x$), whereby obviously $r_F \leq \min(1,2x)$ ($r_P \leq \min(1,2-2x)$). The chemical order parameter $s$ is then defined by

$$s = 2(r_F - x) = 2(r_P - 1 + x),$$

and ranges from 0 to max$(2 - 2x, 2x)$. Denoting the compositions of the two repeating layers as $(A,B)$, the case of complete disorder refers to the compositions (Fe$_x$Pt$_{1-x}$, Fe$_2$Pt$_{1-x}$) and the maximum order to (Fe, Fe$_{2x-1}$Pt$_{2-2x}$) for $x \geq 0.5$ and to (Fe$_{2x}$Pt$_{1-2x}$, Pt) for $x \leq 0.5$. Note that only in case of $x = 0.5$ can the order parameter reach the value $s = 1$. In the following, we refer to the two layers as the nominal Fe layer and the nominal Pt layer, respectively.

Our present study was motivated by the work of Barmak and co-workers [5], who investigated the MAE of four Fe$_x$Pt$_{1-x}$ samples differing in composition and degree of chemical order. Table 1 summarizes the experimental geometrical and compositional data, as well as the measured MAE values for the FePt samples studied in [5]. Note that for sample No. 4 the MAE could not be determined. One of the main conclusions of ref. [5] is that slightly Fe-rich samples may be preferable to Fe$_{50}$Pt$_{50}$ for obtaining a large MAE. In terms of first-principles calculations we
Table 1: Summary of the experimental data obtained for four samples of Fe$_x$Pt$_{1-x}$ in [5].

| Sample | $x$ (%) | $a$ (Å) | $c$ (Å) | $c/a$ | $s$ | $K$ (meV/atom) |
|--------|---------|---------|---------|-------|----|----------------|
| 1      | 46.2    | 3.870   | 3.721   | 0.961 | 0.89| 0.453          |
| 2      | 51.1    | 3.863   | 3.710   | 0.960 | 0.93| 0.709          |
| 3      | 52.0    | 3.857   | 3.706   | 0.961 | 0.89| 0.775          |
| 4      | 55.4    | 3.839   | 3.704   | 0.965 | 0.72| N/A            |

Fig. 1: (Colour on-line) A comparison of the experimental MAE values of the Fe$_x$Pt$_{1-x}$ samples studied by Barmak et al. in [5] (open circles) and theoretical MAE values calculated using SKKR-CPA as follows: using the experimental lattice parameters for each sample, but assuming $x = 0.5$ and $s = 1$ (+); using the lattice parameters and the compositions $x$ as given in the experiment, but keeping $s$ constant at 0.89 (●); and using the lattice parameters as well as the values of $x$ and $s$ as given in the experiment (×). Solid lines serve as a guide for the eyes.

In order to verify our method against the experiments, first we attempt a direct comparison of our SKKR-CPA calculations to the experimental data of [5], see table 1. As shown in fig. 1, we performed three sets of calculations. The first set only takes into account changes in the lattice geometry (i.e., the variation in the lattice parameters), while assuming stoichiometric composition, $x = 0.5$, and maximum long-range chemical order, $s = 1$. Even by taking into account the “temperature factor” of 0.6, these calculations yield quite high MAE values. Such magnitude differences with respect to the experiment are, however, in agreement with previous first-principles calculations of the MAE of FePt, see, e.g., refs. [22,23]. Furthermore, in this set of calculations only a very moderate change (<3%) of the MAE is obtained across the samples.

In the second set of calculations, we introduce the composition $x$ as given in the experiment, while keeping the degree of chemical order constant at $s = 0.89$. This greatly improves the trend of the MAE; however, the relative change of the MAE from sample No. 1 to sample No. 2 is still underestimated (<15%) as compared to the experiment (∼50%). The overall magnitude of the MAE is significantly decreased, but it is still by a factor of 2–2.5 larger than the measured one. Finally, including also the variation of chemical order as given in the experiment clearly improves the above-mentioned relative change between sample No. 2 and sample No. 3 (∼40%), but, opposite to the experiment, it predicts a slightly decreasing trend from sample No. 2 to 3. Note, however, that these latter changes are within the range of both theoretical and experimental errors.

Having confirmed that the SKKR-CPA calculations satisfactorily reproduce the experimental trends, we next consider the general effects of the chemical composition...
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Fig. 2: (Colour on-line) The variation of the MAE of Fe$_x$Pt$_{1-x}$ alloys as a function of the chemical order parameter $s$ and the composition $x$. The lattice parameters were fixed to $a = 3.857$ Å and $c = 3.706$ Å. Solid lines serve as a guide for the eyes.

$x$ and order parameter $s$ on the MAE of FePt. To this end, we used the lattice parameters measured for sample No. 3 in [5], $a = 3.857$ Å and $c = 3.706$ Å, while we independently varied the chemical order parameter $s$ as well as the composition $x$. Note that for this theoretical study we did not scale down the MAE to mimic temperature-induced effects. The results are shown in fig. 2 for the range of compositions $0.4 \leq x \leq 0.6$. (Beyond this range, the L1$_0$ structure becomes unstable with respect to other phases [1,2].) Our results are in good agreement with the conclusion of Barmak et al. [5], inasmuch as for any given degree of chemical order $s$, the MAE increases monotonically with the Fe-content. However, even maximally ordered Fe$_x$Pt$_{1-x}$ alloys with $x > 50$ cannot achieve the MAE of fully ordered Fe$_{50}$Pt$_{50}$ (3.31 meV per formula unit).

It should be noted that, at $s = 0$, the MAE becomes negative. This is in contrast to [24], which reports a vanishing MAE for completely disordered FePt under the assumption of a cubic unit cell. The “residual” negative MAE we obtain in the case of complete chemical disorder is, therefore, due to the tetragonality of the lattice ($a \neq c$). For real samples, where the lattice parameters cannot be frozen while varying the chemical order and composition, in the case of complete chemical disorder the unit cell is expected to become cubic, removing thus this “residual” MAE.

In order to elucidate the origin of the variation in the MAE with the composition and the chemical disorder, we consider next the species-resolved contributions to the MAE. The MAE per unit cell can be decomposed as

$$K = r_{Fe}D_{Fe}^{Fe} + (1 - r_{Fe})D_{Pt}^{Fe} + (1 - r_{Pt})D_{Fe}^{Pt} + r_{Pt}D_{Pt}^{Pt},$$

where $D_{\gamma}^\beta$ ($\beta, \gamma = Fe$ or Pt) denotes the MAE contribution from an atom of species $\gamma$ when it is positioned in a

Fig. 3: (Colour on-line) Variation in the species-resolved MAE contributions against the chemical order parameter $s$ for compositions $x = 0.40$ (upper panel), $x = 0.50$ (middle panel) and $x = 0.60$ (lower panel). +: $D_{Fe}^{Fe}$, contribution of an Fe atom in nominal Fe layers; *: $D_{Pt}^{Fe}$, contribution of a Pt atom in nominal Fe layers; x: $D_{Fe}^{Pt}$, contribution of an Fe atom in nominal Pt layers; and •: $D_{Pt}^{Pt}$, contribution of a Pt atom in nominal Pt layers. The lattice parameters were fixed to $a = 3.857$ Å and $c = 3.706$ Å. Solid lines serve as a guide for the eyes.

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nominal Fe layers rapidly increases up to about 1 when varying the composition $x$, while keeping the chemical order parameter $s$ constant. However, $\text{Fe}_x\text{Pt}_{1-x}$ with $x \neq 0.5$ cannot attain perfect chemical order ($s = 1$) and perfectly ordered $\text{Fe}_{50}\text{Pt}_{50}$ yields a larger MAE than the Fe-rich alloys with maximum degree of long-range chemical order.

In conclusion, our calculations strongly support the conclusion of Barmak and co-workers in [5], showing that, for a given degree of chemical order, the MAE increases with the Fe concentration of $\text{Fe}_x\text{Pt}_{1-x}$, at least within the range $0.4 \leq x \leq 0.6$. This is due to the strongly positive effect on the MAE of the degree of Fe-filling of the nominal Fe layers, $r_{\text{Fe}}$, which dominates the variation in the MAE when varying the composition $x$, while keeping the chemical order parameter $s$ constant.

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