Magnetic anisotropy of FePt: Effect of lattice distortion and chemical disorder

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We perform first principles calculations of the magnetocrystalline anisotropy energy (MAE) in the five samples of L10 FePt that were studied experimentally by Ding and co-workers [J. Appl. Phys. 97, 10H303 (2005)]. The effect of temperature-induced spin fluctuations is estimated by scaling the MAE down according to previous Langevin dynamics simulations. Including chemical disorder as given in experiment, the experimental correlation between the MAE and the lattice mismatch is qualitatively well reproduced. Moreover, we determine the chemical order parameters that reproduce exactly the experimental MAE of each of the samples. Our observations lead to the conclusion that the MAE of the FePt samples is determined by the chemical disorder rather than by lattice distortion. © 2011 American Institute of Physics. [doi:10.1063/1.3644478]

Due to its extraordinarily high magnetocrystalline anisotropy energy (MAE), L10 FePt is of considerable interest to the development of ultrahigh density magnetic recording applications and spintronics devices. From a theoretical point of view, there is an obvious need for a complete first principles model of FePt to be used in generating effective spin Hamiltonians for the purpose of atomistic and multiscale modelling. Amongst many other issues, this requires an understanding of the role of interfacial effects and chemical disorder. The large effect of chemical disorder on the MAE of FePt has already been outlined both experimentally1 and theoretically.2

Recently, the experiments were extended to thin films of FePt deposited on different substrates.3 A strong correlation was revealed between the MAE of the FePt sample and the lattice mismatch of the FePt films with respect to the substrate.3 The experimental data are summarised in Table I. The chemical order parameter, s, is defined as the probability of finding a Fe atom on a nominal Fe site or, equivalently, as the probability of finding a Pt atom on a nominal Pt site. In the experiment, the chemical order parameters were derived from the x-ray diffraction intensities I(001) and I(002) ((xyz denoting the plane of diffraction), through the relationship

\[ s \sim \sqrt{I(001)/I(002)} \]

normalizing s to unity for sample 3. We refer to the experimentally obtained chemical order parameters as \( s_e \) for distinction from the chemical order parameters \( s \) obtained later by fitting calculated MAE-values to experiment.

The aim of the present work is to investigate in detail the effect of lattice distortion and chemical order on the MAE of FePt by means of the relativistic Korringa-Kohn-Rostoker2–7 method as combined with the coherent potential approximation8,9 (KKR-CPA). In order to differentiate between the two main properties characterizing the samples, namely, the lattice distortion and the chemical disorder, we perform calculations with and without the inclusion of chemical disorder. We then fit the calculated MAE to the experimental values using the chemical order parameter, \( s \), as a fitting parameter and draw conclusions from the results of our calculations. We find that the chemical disorder of each sample is the more important factor in determining the experimental MAE.

As the relativistic KKR method is well documented in the literature (see e.g., Ref. 7), here, we merely describe some details of our calculations. We used density functional theory within the local spin-density approximation (LSDA) as parametrised by Vosko et al.10 The effective potentials and fields were treated within the atomic sphere approximation (ASA). As the thin-film samples in the experiment had a thickness of approximately 20 nm (60 f.u.),3 surface contributions to the MAE should be negligible small. We therefore modelled the FePt samples as face-centered-tetragonal (fct) bulk lattices with lattice constants as displayed in Table I. The self-consistent calculations were performed by using the scalar-relativistic approximation, i.e., by neglecting spin-orbit coupling11 and solving the Kohn-Sham-Dirac equation using a spherical wave expansion up to an angular momentum quantum number of \( \ell = 3 \). As in earlier theoretical

<table>
<thead>
<tr>
<th>Sample</th>
<th>a (Å)</th>
<th>c (Å)</th>
<th>( s_e )</th>
<th>( K ) (meV)</th>
<th>( I(001)/I(002) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3.88673</td>
<td>3.69977</td>
<td>0.709</td>
<td>0.493</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>3.88279</td>
<td>3.69387</td>
<td>0.978</td>
<td>0.696</td>
<td>1.9</td>
</tr>
<tr>
<td>3</td>
<td>3.89752</td>
<td>3.68964</td>
<td>1.000</td>
<td>0.841</td>
<td>1.985</td>
</tr>
<tr>
<td>4</td>
<td>3.89646</td>
<td>3.69175</td>
<td>0.965</td>
<td>0.788</td>
<td>1.85</td>
</tr>
<tr>
<td>5</td>
<td>3.86954</td>
<td>3.71378</td>
<td>0.615</td>
<td>0.271</td>
<td>0.7536</td>
</tr>
</tbody>
</table>

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TABLE I. Summary of experimental results by Ding et al. (Ref. 3). Lattice parameters, a and c; chemical order parameter, \( s \); magnetocrystalline anisotropy energy per formula unit, \( K \); and diffraction intensity ratio, \( I(001)/I(002) \).
work,\textsuperscript{2} we used the CPA to elucidate long-range chemical disorder effects in FePt. In combination with KKR, the CPA has proved particularly useful in calculating the physical properties of chemically disordered alloys.\textsuperscript{9} The partially disordered FePt alloy is modelled as a stack of alternating layers with the chemical compositions of Fe\textsubscript{1−s}Pt\textsubscript{s} and Pt\textsubscript{s}Fe\textsubscript{1−s}.

The MAE is then evaluated using the magnetic force theorem,\textsuperscript{12} which states that the difference in a system’s total energy for two different directions of magnetization can be approximated by the corresponding difference of the band energies, neglecting further self-consistency, i.e., keeping the effective potentials and fields fixed. From previous experience, we know that for transition metal systems these potentials and fields can safely be taken from self-consistent scalar-relativistic calculations.\textsuperscript{7} In order to achieve a relative accuracy within 5% for the MAE, the associated energy integration was performed by sampling 20 energy points along a semi-circular contour in the upper complex half-plane. At the energy point closest to the real axis, the \( k \)-integration was shown in Fig. 2. In accordance with earlier work,\textsuperscript{2} long-range chemical disorder drastically reduced the MAE; for sample 1 (\( s \_f = 0.709 \)), we obtained a value of 0.4 meV/f.u., while for sample 5 (\( s \_f = 0.615 \)), the MAE almost vanished. In fact, reducing \( s \) to 0.5 can even cause a change of sign of the MAE. In contrast, for samples 2 and 4 with a high degree of chemical order, the MAE was reduced by less than 10%, and for sample 3 (\( s \_f = 1 \)), the MAE remained unchanged with respect to our previous calculations. Taking into account again a reduction by a factor of 0.6 due to temperature effects, it is obvious that the inclusion of chemical disorder has significantly improved the agreement between experiment and theory: the trend of the MAE across the different samples is now correct and the magnitudes of the MAE are closer to the range reported by the experiment.

As mentioned above, the chemical order parameters in Table I were derived from measured diffraction intensity ratios.\textsuperscript{1,4} However, due to an incomplete rocking curve,\textsuperscript{3} the measured diffraction intensities, and thereby the experimentally obtained chemical disorder parameters, can only be considered

\[ \text{FIG. 1. (Color online) Crosses (solid line): calculated MAE per formula unit for each of the FePt samples in Table I modelled as perfectly ordered alloys. Circles (dashed line): the same values scaled down by a factor of 0.6 in order to account for temperature induced effects. Stars (dotted line): the experimental values.} \]

\[ \text{FIG. 2. (Color online) Crosses (solid line): calculated MAE per formula unit for each of the FePt samples in Table I modelled as partially disordered alloys with the degree of disorder given by the experiment. Circles (dashed line): the same values scaled down by a factor of 0.6 in order to account for temperature induced effects. Stars (dotted line): the experimental values.} \]
Subtle interplay of the dependence of the MAE on the chemical order parameters as indicated by the dashed line in Fig. 3. Obviously, this remarkable linear behavior is the result of a contrast, for samples 1 and 5, an increased degree of chemical disorder, we, furthermore, determined theoretical chemical order parameters that reproduced the experimental MAE values. Interestingly, a linear correlation between the MAE and the best-fit chemical order parameters is found. It should be mentioned that work is underway to perform constrained Monte-Carlo simulations of $K(T)$ for chemically disordered FePt, since this is clearly an important factor in relation to experimental data.

In summary, our first principles calculations imply that lattice distortion in the FePt samples has only a minor effect on the MAE, even opposite to the experimental trend. Calculating the MAE using the highly approximate experimental chemical order parameters significantly improves the agreement between theory and experiment, in particular with regards to the relative differences in the MAE between the samples. This indicates that the substrate-sample lattice mismatch effect on the MAE reported by Ding et al. is mainly due to the variation in chemical disorder. To circumvent the uncertainty of the experimental determination of chemical disorder, we, furthermore, determined theoretical chemical order parameters that reproduced the experimental MAE values. Financial support was provided by the Hungarian Research Foundation (Contract Nos. OTKA K77771 and K84078) and by the New Széchenyi Plan of Hungary (Project ID: TAMOP-4.2.1/B-09/1/KMR-2010-0002). C.J.A. is grateful to EPSRC for the provision of a research studentship.