Magnetoelastic coupling in epitaxial magnetic films: An ab-initio study

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Abstract

A method is developed which allows to determine the first-order and the second-order magnetoelastic coefficients of a magnetic bulk material from the ab-initio calculation of the magnetocrystalline anisotropy energy as function of a prestrain $\epsilon_0$. Explicit results are given for bcc Fe, and they agree well with experimental data obtained from the magnetostrictive stress measurements for epitaxial Fe films.

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In recent years magnetic devices based on magnetic films technologies have attracted a considerable interest, e.g., magnetooptical recording media or magnetoresistive devices based on the giant magnetoresistive and the tunnel magnetoresistive effect designed for sensors or magnetostrictive random access memories. Thereby the magnetic anisotropy plays an important role, for instance, the issue of perpendicular anisotropy for the magnetooptical recording or the demand for soft magnetic layers with weak anisotropy as part of the magnetoresistive devices. It has been shown by numerous investigations that the magnetic anisotropies of magnetic films grown epitaxially on a substrate may strongly deviate from those of the respective bulk materials. The reason for this deviation is in general ascribed to several different effects. First, there are contributions to the anisotropy originating from the free surface of the magnetic layer and from the interface between layer and substrate, as well as from the morphology of the film due to a heterogeneous film growth. The influence of all these effects must decrease with increasing thickness of the film. What remains for a film of thickness larger than typically 10 nm is the effect of the magnetoelastic coupling to the film strain induced by the lattice mismatch between film and substrate. Because the epitaxial film strain may be of the order of several % which is much larger than typical magnetostrictive strains of $10^{-6}$ to $10^{-4}$ and because of the dependence of the magnetoelastic coupling energy on the direction of the magnetization (see below), this may result in new magnetic anisotropies different from that of the unstrained bulk. The numerous experiments on the effect of epitaxial strain on the magnetic properties of magnetic films are reviewed in Ref. [1]. An ab-initio study of this effect within the framework of density functional theory is the purpose of the present letter.

To be more specific we consider a material which is cubic in the unstrained state (Fe, for instance). Then the density of the magnetoelastic coupling energy may be written (up to the second order in the strain $\epsilon_{ij}$, omitting the terms including the shear strain $\epsilon_{ij}, i \neq j$, which are not required for the situation discussed below) as [2]

$$f_{me} = B_1 (\epsilon_{11} \alpha_1^2 + \epsilon_{22} \alpha_2^2 + \epsilon_{33} \alpha_3^2) +$$

(1)
\[ \frac{1}{2}D_{11} (\epsilon_{11}^2 \alpha_1^4 + \epsilon_{22}^2 \alpha_2^4 + \epsilon_{33}^2 \alpha_3^4) + D_{12} (\epsilon_{11} \epsilon_{22} \alpha_1^2 \alpha_2^2 + \epsilon_{22} \epsilon_{33} \alpha_2^2 \alpha_3^2 + \epsilon_{33} \epsilon_{11} \alpha_3^2 \alpha_1^2). \]

Here \( B_1 \) and \( D_{11}, \) \( D_{12} \) represent magnetoelastic coupling coefficients of the first and the second order, and the \( \alpha_i \) denote the direction cosines of the magnetization referred to the cubic axes. For Fe grown epitaxially on a cubic (100) surface which is a prototype system experimentally investigated intensively [1,3,4] there are epitaxial strains \( \epsilon_{11} = \epsilon_{22} \) which may be different for various atomic layers of the film [3,4]. In the following we adopt a simple model where we consider only the average strain of the film which depends on the thickness of the film, i.e., we assume \( \epsilon_{11} = \epsilon_{22} = \epsilon_0 \). The strain \( \epsilon_{33} \) then may be obtained by minimizing the total energy density \( f = f_{\text{me}} + f_{\text{el}} \), with the elastic energy density (again omitting the terms containing the shear strains):

\[ f_{\text{el}} = \frac{1}{2} C_{11} (\epsilon_{11}^2 + \epsilon_{22}^2 + \epsilon_{33}^2) + C_{12} (\epsilon_{11} \epsilon_{22} + \epsilon_{22} \epsilon_{33} + \epsilon_{33} \epsilon_{11}) \]  

(2)

and with the cubic elastic stiffness constants \( C_{ij} \). This yields

\[ \epsilon_{33} = -\frac{2C_{12}\epsilon_0 + B_1\alpha_3^2}{C_{11} + D_{11}\alpha_3^4}. \]  

(3)

Because the magnetostrictive contribution to the strain originating from \( f_{\text{me}} \) is much smaller than \( \epsilon_0 \) we find \( \epsilon_{33} \approx -2C_{12}/C_{11} \).

From (1), (3) we obtain the strain dependent part, \( f_{\text{mca}}(\epsilon_0) \), of the magnetocrystalline anisotropy energy density:

\[ f_{\text{mca}}(\epsilon_0) = f_{\text{me}}(\epsilon_0, \alpha_1 = 1) - f_{\text{me}}(\epsilon_0, \alpha_3 = 1) = k_0 + k_1 \epsilon_0 + k_2 \epsilon_0^2, \]  

(4)

with

\[ k_0 = \frac{1}{2} B_1^2 \frac{1}{C_{11} + D_{11}}, \]  

(5)

\[ k_1 = B_1 \left(1 + 2C_{12} \frac{1}{C_{11} + D_{11}}\right), \]  

(6)

\[ k_2 = \frac{1}{2} D_{11} \left(1 - 4 \frac{C_{12}^2}{C_{11} C_{11} + D_{11}}\right). \]  

(7)

The term \( k_0 \) is negligible and arises from the fact that we fix the strains \( \epsilon_{11} = \epsilon_{22} = \epsilon_0 \) independently on the direction of the magnetization. Furthermore, when changing the
direction of the magnetization there is a change in the magnetostrictive stress \( \tau_1 = \frac{\partial f_{\text{mea}}}{\partial \epsilon_{11}} \)
according to

\[
\Delta \tau_1 = \tau_1(\alpha_1 = 1) - \tau_1(\alpha_2 = 1) = B_1 + D_{11} \epsilon_0. \tag{8}
\]

An experimental determination of \( \Delta \tau_1 \) (exploiting the change of the bending moment that is created by the film onto the substrate \([1,3,4]\)) as function of the layer thickness and hence as function of \( \epsilon_0 \) then yields the two magnetoelastic coupling coefficients \( B_1 \) and \( D_{11} \). For Fe on MgO (100) Koch et al. \([3]\) obtained \( B_1 = -3.2 \text{ MJ/m}^3 \), \( D_{11} = 1.1 \text{ GJ/m}^3 \) (±10\%), and for Fe on W(100) Enders et al. \([4]\) found \( B_1 = -3 \text{ MJ/m}^3 \), \( D_{11} = 1 \text{ GJ/m}^3 \). The values for \( B_1 \) extracted from the film experiments agree rather well with the bulk value of \( B_1 = -3.44 \text{ MJ/m}^3 \). There are no values of \( D_{11} \) obtained from bulk measurements for comparison and therefore the two experiments were considered as the first determination of the second-order magnetoelastic coupling coefficient of bulk Fe by a film experiment.

It was pointed out \([1,3,4]\) that due to the large strains accessible by epitaxial film growth the effective first-order coefficient \( B_{\text{eff}} \) defined as \( B_{\text{eff}} = B_1 + D_{11} \epsilon_0 \) changes sign from negative to positive for Fe with decreasing film thickness, i.e., increasing \( \epsilon_0 \), and this clearly demonstrates that the magnetic anisotropy energy depends dramatically on the film thickness, a result which is most relevant for the design of the magnetic film devices (see introduction). The linear strain dependence of \( B_{\text{eff}} \) failed to describe the experimental data for film thicknesses below 10 nm, most probably because then the effects of the surface, interface and film morphology become relevant.

In the present paper we determine the magnetoelastic coupling coefficients \( B_1 \) and \( D_{11} \) for cubic Fe by the ab-initio density functional theory. To do this, we calculate the magnetocrystalline anisotropy energy density \( f_{\text{mea}}(\epsilon_0) \) as function of the strain \( \epsilon_{11} = \epsilon_{22} = \epsilon_0 \) imposed to the bulk material, represent the data by a quadratic polynomial in \( \epsilon_0 \) according to eq. (4) and determine \( B_1 \) and \( D_{11} \) from eqs. (6,7), inserting the elastic stiffness constants \( C_{12} \) and \( C_{11} \) which we have also obtained ab initio.

We have performed the calculations using the WIEN97 \([4]\) code which adopts the
full-potential linearized augmented plane-wave (FLAPW) method \cite{6}. For the exchange-correlation potential the local-spin-density (LSDA) functional by Perdew and Wang \cite{7} and the generalized-gradient-approximation (GGA) functional by Perdew et al. \cite{8} were used. The total energy minimizations on the non-strained bcc Fe gave us the equilibrium lattice parameters $a = 5.2 \ a_0$ for LSDA and $a = 5.34 \ a_0$ for GGA where $a_0$ denotes Bohr’s radius. The calculated ratio $2C_{12}/C_{11}$ is 1.08 for LSDA and 1.13 for GGA. The experimental values are $a = 5.42 \ a_0$ and $2C_{12}/C_{11} = 1.17$.

Numerically the most difficult step is the calculation of $f_{\text{mca}}$ which is due to the spin-orbit coupling (SOC). First, we calculate the self-consistent electronic structure in the scalar-relativistic approximation \cite{9} using $N^3_k \ k$ vectors with $N_k = 21$ in the total Brillouin zone (BZ) which correspond to the 762 $k$ vectors in the irreducible part of the Brillouin zone (IBZ). The criterion for the self-consistency is the difference in the charge densities after the last two iterations being less than $2 \times 10^{-6} \ e/(\text{a.u.})^3$. The contribution of the SOC is determined perturbatively using the second variational method \cite{10,11}. The quantity $f_{\text{mca}}$ is calculated by applying the force theorem \cite{12,13} as the difference between the sums of the perturbed eigenvalues for the different magnetization directions. Fig. 1 represents the convergency test for the calculation of $f_{\text{mca}}$ with respect to $N_k$. The data are for the case with $a = 5.4 \ a_0$ and $c = 5.2 \ a_0$, using GGA. The modified tetrahedron \cite{14} and the Gaussian smearing \cite{15,16} integration schemes were used. The proper convergency with the Gaussian smearing was achieved by setting the smearing parameter as $\Gamma/N_k$. The suitable values of $\Gamma$ for the particular case are roughly from the interval between 6.8eV and 10.1eV which follows from the curves in Fig. 1. All the final calculations of $f_{\text{mca}}$ were performed with $N_k = 51$ (17576 $k$ vectors in the IBZ) using both the tetrahedron and the Gaussian smearing method in order to minimize the numerical uncertainties. The estimated accuracy is $\pm 1 \ \mu eV$/unit cell marked by the horizontal lines in Fig. 1.

Fig. 2 shows the calculated magnetocrystalline anisotropy energy density $f_{\text{mca}}$ with respect to the lateral strain $\epsilon_0$. The numerical data are well fitted by quadratic polynomials as predicted by eq. (4). From the calculated parameters $k_1$ and $k_2$ the magnetoelastic
coefficients $B_1$ and $D_{11}$ are determined according to eqs. (6,7). Table I summarizes the theoretical results in comparison with the experimental data from [14]. There is a big discrepancy between the LSDA result for $B_1$ and the experimental result, whereas the GGA result is much closer to the experiment. This is in line with the calculation of the magnetoelastic coefficient $\lambda_{100}$ of unstrained bulk Fe by Wu et al. [17] who also obtained a strong deviation from the experiment when using LSDA but a satisfactory agreement when using GGA. The calculated second-order magnetoelastic coupling coefficient $D_{11}$ for bulk bcc Fe matches the experimental value obtained from the measurements on epitaxial thin films very well, especially the value from the GGA calculation. The agreement represents the direct proof that the experimental results [13,4] can be really ascribed to the pure strain effect on the magnetoelastic properties and that the measurements of the magnetostrictive film stress as function of the film thickness can provide the second-order coupling constant $D_{11}$ of the bulk which is hard to obtain by bulk measurements.

We close with an important warning. The experiments [13,4] and the present theory demonstrate that the magnetoelastic properties of thin epitaxial films may deviate significantly from that described by the first-order magnetoelastic coupling coefficients of the bulk and that one has to take into account the second-order terms of the bulk. The change of the magnetostrictive stress $\Delta \tau_1$ obtained when switching the magnetization from [010] to [100] as function of the epitaxial strain $\epsilon_0$ then may be expressed by an effective first-order coefficient $B_{\text{eff}} = B_1 + D_{11}\epsilon_0$. However, the correct result for the coefficients $k_0$, $k_1$ and $k_2$ of the polynomial expansion (4) of $f_{\text{mca}}$ may not be obtained by neglecting the second-order terms in eq. (1) and instead replacing $B_1$ in the first-order term by $B_{\text{eff}} = B_1 + D_{11}\epsilon_0$. This would yield $f_{\text{mca}} = k_0 + k_1\epsilon_0 + k_2\epsilon_0^2$ with $k_1 = B_1(1 + 2C_{12}/C_{11})$ and $k_2 = D(1 + 2C_{12}/C_{11})$ instead of eqs. (6,7). Inserting the value of $2C_{12}/C_{11}$ for Fe it becomes obvious that $D$ and $D_{11}$ even have a different sign; the sign of $D$ being opposite to the one of the experimentally determined $D_{11}$ according to eq. (8). Guo et al. [18] have performed an ab-initio calculation of the magnetoelastic properties of epitaxial Co and Ni films, and they indeed proceeded on this line, i.e. they neglected the second-order term in eq. (1) and instead replaced the
constant $B_1$ by a strain-dependent term $B_1(\epsilon_0)$. It should be cautioned that the coefficient $D$ obtained from the linearization of $B_1(\epsilon_0)$ is not identical to the second order coefficient $D_{11}$ of the bulk material but it may deviate strongly.

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FIGURES

FIG. 1. The convergency test for $f_{\text{mca}}$ with respect to $N_k$. The horizontal lines represent the estimated accuracy $\pm 1 \mu\text{eV/unit cell}$; $+$, modified tetrahedron method; $\times$, $\circ$ and $\square$, Gaussian smearing with $\Gamma = 10.2 \text{ eV}$, $6.8 \text{ eV}$ and $1.02 \text{ eV}$. The proper convergency using the Gaussian smearing is obtained for the smearing parameter $\frac{\Gamma}{N_k}$ where $\Gamma$ is from the interval between $6.8 \text{ eV}$ and $10.1 \text{ eV}$.

FIG. 2. The calculated magnetocrystalline anisotropy energy density $f_{\text{mca}}$ as function of the prestrain $\epsilon_0$. The solid lines represent fits to quadratic polynomials according to eq. (4). Note that $k_0$ is indeed negligible; $\square$, LSDA; $\times$, GGA.
TABLES

TABLE I. The calculated results in comparison with the experimental data from the thin film experiments.

|       | \( a [a_0] \) | \( 2 \frac{C_{12}}{C_{11}} \) | \( B_1 [\text{MJ/m}^3] \) | \( D_{11} [\text{GJ/m}^3] \) |
|-------|---------------|-----------------|-----------------|-----------------|
| LSDA  | 5.20          | 1.08            | -9.26           | 1.5             |
| GGA   | 5.34          | 1.13            | -2.39           | 1.1             |
| exp. [1,4] | 5.42      | 1.17            | -3              | 1.0             |
| exp. [3]   |              | -3.2            |                 | 1.1             |
Figure 1:
Figure 2: