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Magnetoelastic coupling in Ni and Fe monolayers on Cu(001)

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The correlation between mechanical stress and magnetic anisotropy of Ni and Fe films on Cu(001) is investigated. The magnetoelastic coupling *and* the film stress during the growth are measured *in situ* with a highly sensitive optical bending beam technique. For Ni a dramatically reduced magnetoelastic coupling of $B_1 = 3.5\,\mathrm{MJ/m^3}$ is found for films thinner than 10 ML, roughly one third of the bulk value of 9.4 MJ/m^3. This change is explained by a strain correction to the magnetoelastic coupling. The influence of the interfaces does not significantly contribute to the magnetoelastic coupling. A very small magnetoelastic coupling of 0.4 MJ/m^3 for Fe films in the range from 12 ML to 25 nm is attributed mainly to the crystallographic orientation of the bcc–Fe. © *2000 American Institute of Physics.* [S0021-8979(00)48508-7]

In this article the influence of mechanical stress on the magnetoelastic (ME) coupling of ultrathin films of Ni and Fe on Cu(001) is discussed. In bulk materials the ME coupling is responsible for the magnetostriction, i.e., a minute change in length and volume when the sample is magnetized. On the other hand, an imposed strain on the material leads to an additional magnetic anisotropy. It is generally accepted that this ME coupling is responsible for the perpendicular magnetic anisotropy of Ni/Cu(001)[1] in the range from about 10 (Ref. 2) to several tens of ML.[3] Ni grows pseudomorphically and is strained by 2.5% in the film plane. With the bulk ME coupling constant of $B_1 = 9.4\,\mathrm{MJ/m^3}$ (Ref. 4) and the elastic constants of Ni, an anisotropy of 0.53 MJ/m^3 is obtained, which is larger than the shape anisotropy of -0.15 MJ/m^3. Thus, a perpendicular magnetization is preferred. An additional interface anisotropy is responsible for the in-plane magnetization below 10 ML. The ME anisotropy is reduced in thicker films due to strain relaxation and the magnetization is forced back into the film plane by the shape anisotropy.

In Fe/Cu(001) a structural transformation from a pseudomorphic fcc film to a (110)-bcc film takes place at 12 ML. The structural transition is accompanied by a reorientation of the magnetization from out-of-plane to in-plane.[5]

It has been shown previously that the ME coupling in thin films is dramatically different from bulk materials.[4,6,7] Even a change in sign has been observed.[4,8–10] Two models have been proposed to describe this change in the ME coupling. In the first model, an interface ME coupling term is introduced.[11,12] The second model includes higher order terms in the strain in order to take the large misfit strain into account.[8,13] Several attempts have been made to separate different contributions to the magnetic anisotropy of thin films by measuring the anisotropy as a function of film thickness.[12,14,15] However, the magnetic anisotropy of thin films is influenced by a large set of parameters like the bulk ME coupling, the interface magnetic anisotropy, and the interface ME coupling, which hardly can be separated from each other by performing a measurement of the thickness dependence of the magnetic anisotropy only.

In this contribution we report direct measurements of the ME coupling in thin films by means of a bending beam method. The same technique is used to determine *in situ* the film stress during film preparation from which the film strain is derived.

The film preparation and the ME measurements were performed in an ultrahigh vacuum chamber. The base pressure was below 5×10^{-11} mbar. The film thickness was determined by measuring the deposition rate of the evaporator with a quartz balance right before and after deposition. While evaporating, the pressure was below 2×10^{-10} mbar for Ni and 5×10^{-10} mbar for Fe.

The experimental setup for measuring the epitaxial and magnetoelastic stress has been described in detail elsewhere.[4,16] The substrates are thin (≈100 μm) single crystals of Cu(001) with the [100] direction along the sample length of 15 mm. The sample is clamped at one end, the width is 3 mm.

Due to the biaxial epitaxial strain ϵ of the film, a stress $\tau = Y/(1-\nu)\times\epsilon$ along the sample length [100] is expected from elasticity theory. Y is the Young modulus and ν the Poisson ratio of the film material. The stress is continously recorded during film evaporation. The film strain is calculated from the stress curve.

After deposition the ME coupling is determined by magnetizing the sample along its length and width subsequently and measuring the change in the sample curvature. In the case of a (001)-film, the ME stress τ_{ME} measured along [100] is given by the ME coupling constant B_1.[9]

The magnetoelastic coupling has been measured for Ni films of different thicknesses. The result is shown in Fig. 1(a). For films below 10 ML the ME coupling is close to 3.5 MJ/m^3. It seems to be constant for a film thickness between

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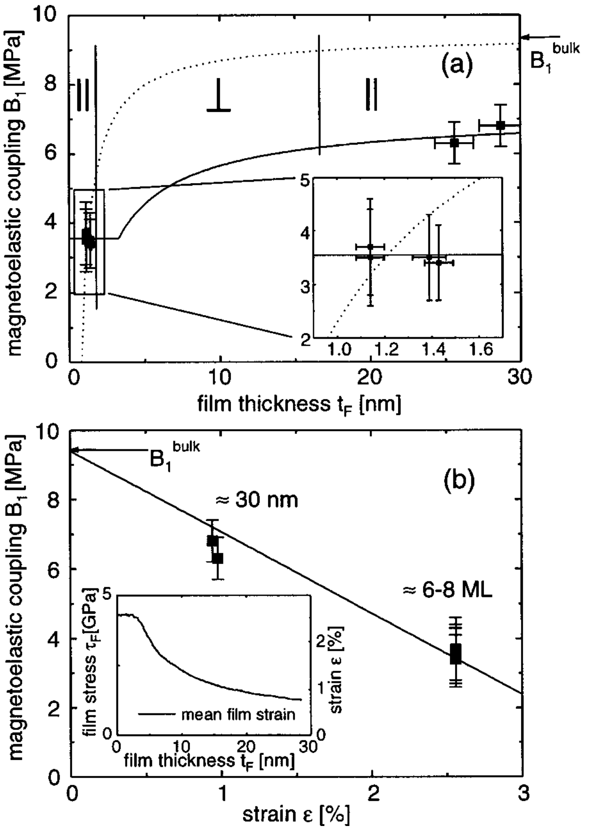

FIG. 1. (a) ME coupling of Ni/Cu(001) as a function of film thickness. The squares are experimental values, the solid line is deduced from a strain dependent correction and the dotted line from a surface term correction to the ME coupling. The regions of perpendicular (⊥) and in-plane (∥) magnetic anisotropy are indicated. (b) Experimental (squares) ME coupling vs film strain deduced from the experimental film stress and model of a strain dependent correction (solid line). The inset shows the experimental film stress vs the film thickness.

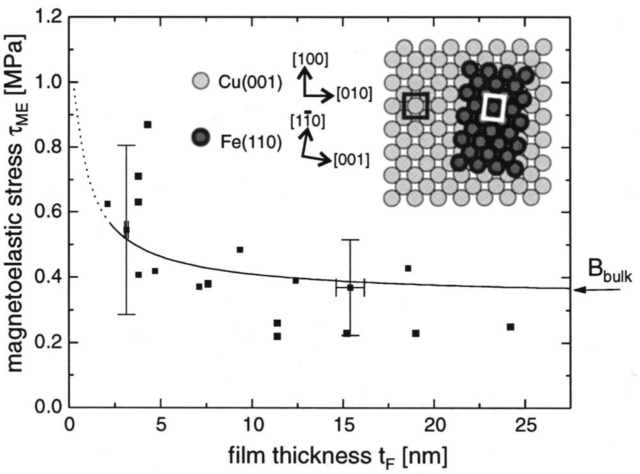

FIG. 2. Measured (squares) ME coupling of Fe/Cu(001) as a function of film thickness. Note the small scale of the ME stress. The solid line results from the strain dependent model. Inset: Orientation of the bcc(110) Fe on Cu(001), after Ref. 18. The Fe[$1\bar{1}0$] and the Cu[100] direction are tilted with respect to each other by an angle of 9.7°. Other structural domains with angles of −9.7°, 80.3° and −80.3° are not shown.

1.1 and 1.4 nm, as can be seen from the inset. For films of more than 20 nm the ME coupling is of order 6.5 MJ/m^3. The solid and the dotted lines are derived from models that will be discussed later. In the region of perpendicular magnetic anisotropy the maximum magnetic field of 0.4 T was not sufficient to magnetize the film in-plane and no values of B_1 could be obtained.

In Fig. 1(b) the experimental magnetoelastic coupling is plotted versus the strain of the film. The strain is derived from the film stress that is measured during evaporation. Due to the large initial strain of 2.5% of the Ni film, third-order terms[17] in the elastic energy density have been considered, thus leading to a reduced biaxial modulus of $Y/(1-\nu) = 168\,\text{GPa}$. With these values a film stress of 4.2 GPa is expected which is very close to the experimental value of 4.3 GPa. Thus, we conclude that the stress measurement is an appropriate measure also for the film strain. The stress and the calculated strain are plotted in the inset of Fig. 1(b) as a function of film thickness. In the initially pseudomorphic region the strain of the film is constant until strain release sets in above a critical thickness of about 17 ML (2.8 nm).

In Fig. 2 we show the experimental values of the ME stress of Fe/Cu(001). Note the small scale of the stress values. The values are all close to 0.4 MPa over the whole thickness range. Only bcc–Fe films thicker than 12 ML have been measured since our fields were not sufficient to magnetize the fcc films below 12 ML in-plane. The small magnetoelastic coupling can be attributed to the crystallographic orientation of the Fe. A schematic drawing[18] is presented in the inset of Fig. 2. From the measurement of the ME stress the average value of four structural domains of possible bcc(110) orientations is obtained. The Fe[$1\bar{1}0$] and Cu[100] direction in the different domains are tilted with respect to each other by an angle of ±9.7° and ±80.3°. The calculation can be simplified by using the approximate tilt angles of ±0° and ±90°. One obtains a magnetoelastic stress of $\tau_{\mathrm{ME}} = (3B_1 + B_2)/4 = -0.62\,\text{MPa}$ with the bulk ME constants $B_1 = -3.43$ and $B_2 = 7.83\,\text{MJ/m}^3$.[4] When using the exact values of ±9.7° and ±80.3° the result is $\tau_{\mathrm{ME}} = 0.35\,\text{MPa}$, which is close to zero as compared to simply B_1 or B_2. Thus, the combination of ME constants leads to a very small ME coupling in this geometry. The experimental values obtained with the Fe film are all close to 0.4 MPa.

We first discuss the results obtained with the system Ni/Cu(001). As already pointed out, the ME coupling of the thin films can deviate significantly from the bulk values. In order to investigate the reason for this change, two models were fit to our data. The first one includes higher order terms in the strain to the ME energy density thus leading to a ME stress that has the form[8,9]

$$\tau_{\mathrm{ME}} = B_1 = B_1^{\mathrm{bulk}} + D_1 \epsilon. \tag{1}$$

From fitting a straight line to the measured values of the ME coupling versus film strain ϵ [see Fig. 1(b)] by varying the slope only, we obtain $D_1 = -234\,\text{MJ/m}^3$. Note, that the intercept at $\epsilon = 0$ is not varied but kept fixed at the bulk value of $B_1^{\mathrm{bulk}} = 9.4\,\text{MPa}$. From the experimental film strain as a function of thickness and Eq. (1), the ME coupling can be calculated as a function of thickness. The result is plotted as a solid line in Fig. 1(a). Before discussing this result in more detail, we apply a second model to our data.

Due to symmetry lowering, one expects interface type contributions to the magnetoelastic coupling that lead to[12,11]

$$\tau_{\mathrm{ME}}=B_1=B_1^{\mathrm{bulk}}+\frac{B^{\mathrm{interface}}}{t_F}. \qquad (2)$$

Again the model was fit to our data by varying the parameter $B^{\mathrm{interface}}$ and keeping the bulk value fixed. The result is shown as dotted line in Fig. 1(a).

Obviously the model of a strain dependent correction to the ME coupling fits our data better than the interface term correction. This is confirmed by two main characteristics. One is that the ME coupling does not reach the bulk value even for thick films of more than 20 nm. This is a consequence of the residual strain that has been found to be present even for films up to 70 nm.[10] The second is the constant value of the ME coupling in the thin film range where also the film strain is constant. An interface term model would predict a significant change in the ME coupling when the film thickness is varied. Thus, interface term contributions seem to play a minor role as compared to the strain dependent corrections. This is in agreement with first principles calculations[19] and results of Ha and O'Handley[13] who included both interface and strain correction terms to the ME coupling of Ni/Cu(001). Their interface correction term to the ME coupling is small and becomes comparable to the bulk value of the ME coupling only for a nominal layer thickness well below 1 ML.

In the following we compare our correction term D_1 with the results of Ref. 13. The (bulk) strain correction to the ME coupling B_1 is $-D_B/(1+2c_{12}/c_{11})\approx-480\,\mathrm{MJ/m^3}$.[13] With $D_1=-234\,\mathrm{MJ/m^3}$ we obtain the same sign and order of magnitude. From a more detailed analysis of second-order ME coupling[20] it follows that D_1 used in Eq. (1) is a combination of constants that is different from the correction term necessary to calculate the magnetic anisotropy as in Ref. 13. Thus, a direct comparison of both values can only give a rough idea about the magnitude of the strain correction. Measurements of the ME stress along different crystallographic directions would be necessary in order to obtain a complete set of second-order ME coupling constants.

The same model of a strain dependent ME coupling can be applied to the bcc–Fe films on Cu(001). From the linear fit to the ME coupling as a function of film strain we obtain a weak strain dependence of the ME coupling of $0.22\,\mathrm{MJ/m^3}\times\epsilon$. Obviously not only the bulk ME coupling but also the strain correction terms almost cancel out in the given crystallographic orientation. The ME coupling as a function of film thickness is calculated from the film stress and the correction term. The result is plotted as a solid line in Fig. 2. It agrees very well with the experimental data.

We have presented experiments in which the magnetoelastic coupling of thin films of Fe and Ni/Cu(001) has been determined. The ME coupling of the Ni film is significantly different from the bulk value. The data are in very good agreement with a model that includes a strain dependent correction term to the ME coupling.

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