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Letter to the Editor

Magnetoelastic coupling in Co thin films on W(0 0 1)

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Abstract

The magnetoelastic coupling in Co films on W(001) in the range from 3 to 200 monolayers has been measured. The film stress during film growth and the magnetoelastic stress are determined in situ with a highly sensitive bending beam method. The magnetoelastic coupling in Co films as thin as three atomic layers has been determined. We find a strong variation of the magnetoelastic coupling between 32 and 15 MJ/m³ for films of three monolayers and 30 nm thickness, respectively. This deviation of the magnetoelastic coupling from its bulk value of 37.5 MJ/m³ is ascribed to the large strain of the film and changes in the crystalline structure. Relations between the magnetoelastic coupling coefficients B_j and the magnetostrictive strains λ_i are derived for hexagonal symmetry, which correct misprinted relations found in the literature. © 2000 Elsevier Science B.V. All rights reserved.

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The magnetoelastic (ME) coupling plays an important role for the magnetic anisotropy of thin epitaxial films. It describes the contribution to the magnetic anisotropy energy resulting from the interaction between film strain and magnetization direction. For thin Ni films on Cu(001), for example, the ME coupling is responsible for a perpendicular magnetic anisotropy [1] for a film thickness between 10 monolayers (ML) [2] and several tens of ML [3]. In many publications, the bulk ME constants have been applied to discuss the anisotropy of thin films. This approach turned out to be not applicable. Recent experiments have

In bulk materials, one consequence of the ME coupling is the well-known effect of magnetostriction, i.e., a minute change in length (e.g. $\Delta l/l = 10^{-4}$ for Co along [0001]) and volume of a material upon a change in magnetization. Due to the bonding of a film to its substrate, however, a change in length of thin films is only possible in the direction perpendicular to the surface plane. Within the film plane, a change in the magnetization leads to an additional stress in the film. We

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shown conclusively that the ME coupling in thin films is in general very different from the bulk value [4–8]. Even a change in sign of the ME constants has been observed [7,9]. In this contribution, we discuss the ME coupling of hexagonal Co on W(001). A clear deviation from the bulk behavior is found. Our experiments indicate the decisive role of film strain for the modified ME behaviour.

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measured this magnetostrictive stress for film thicknesses between 3 ML and 30 nm and deduce the ME coupling coefficients B_i directly.

For bulk materials, the magnetostriction constants λ_i are tabulated in the literature. These magnetostriction constants λ_i are transformed into the ME coupling coefficients B_j to compare the bulk values with the thin film measurements. In the case of hexagonal crystalline symmetry, this transformation is given by a set of equations that have been published by Bruno [10], but unfortunately have been misprinted. The correct transformations are given.

In order to obtain the relations between magnetostrictive strains λ_i and the magnetoelastic coupling B_j , an expression of the energy density is minimized with respect to the strain for various magnetization states, which are characterized by different magnetization directions [11]. For the hexagonal crystal symmetry the i=3 direction corresponds to the c-axis. The elastic energy density can be written as [6]

$$f_{e1} = \frac{1}{2}c_{11}(\varepsilon_1^2 + \varepsilon_2^2) + \frac{1}{2}c_{33}\varepsilon_3^2 + c_{12}\varepsilon_1\varepsilon_2 + c_{13}(\varepsilon_1\varepsilon_3 + \varepsilon_2\varepsilon_3) + \frac{1}{2}c_{44}(\varepsilon_4^2 + \varepsilon_5^2) + \frac{1}{4}(c_{11} - c_{12})\varepsilon_6^2 + \cdots,$$
 (1)

with the elastic constants c_{ij} and the strain variables ε_i in the Voigt notation [6]. The magnetoelastic contribution to the free energy density can be written as [6,10]

$$f_{\text{me}} = B_1(\alpha_1^2 \varepsilon_1 + 2\alpha_1 \alpha_2 \varepsilon_6 + \alpha_2^2 \varepsilon_2) + B_2(1 - \alpha_3^2)\varepsilon_3$$
$$+ B_3(1 - \alpha_3^2)(\varepsilon_1 + \varepsilon_2) + B_4(\alpha_2 \alpha_3 \varepsilon_4$$
$$+ \alpha_1 \alpha_3 \varepsilon_5) + \cdots. \tag{2}$$

Here, the B_i are the ME coefficients and α_i are the direction cosines of the magnetization direction. For moderately strained bulk materials higher order terms that are indicated by the dots in Eqs. (1) and (2) can be neglected. Their importance for thin films, however, is discussed below in Eqs. (7) and (8).

The equilibrium strains are obtained from minimizing the free energy density $f = f_{el} + f_{me}$ as a function of strain. In order to determine the ME

coefficients, four magnetostriction constants have to be known. Bozorth [12] used the constants λ_A to λ_B :

$$\lambda_{A} = \lambda_{100}^{100} - \lambda_{100}^{001},$$

$$\lambda_{B} = \lambda_{010}^{100} - \lambda_{010}^{001},$$

$$\lambda_{C} = \lambda_{001}^{100} - \lambda_{001}^{001},$$

$$\lambda_{D} = \lambda_{011}^{011} - \lambda_{011}^{001}.$$
(3)

The upper index in λ_{ijk}^{lmn} indicates the direction of magnetization and the lower the direction in which the strain is measured, e.g. $\lambda_{100}^{001} = \varepsilon_1$ with $\alpha = (0,0,1)$. All λ_{ijk}^{lmn} are derived from the free energy density and finally the $\lambda_A - \lambda_D$ can be written as a function of the c_{ij} and B_i :

$$\lambda_{A} = \frac{B_{2}c_{13} - B_{3}c_{33} - \frac{1}{2}B_{1}c_{33}}{(c_{11} + c_{12})c_{33} - 2c_{13}^{2}} - \frac{B_{1}}{2(c_{11} - c_{12})},$$

$$\lambda_{B} = \frac{B_{2}c_{13} - B_{3}c_{33} - \frac{1}{2}B_{1}c_{33}}{(c_{11} + c_{12})c_{33} - 2c_{13}^{2}} + \frac{B_{1}}{2(c_{11} - c_{12})},$$

$$\lambda_{C} = \frac{(B_{1} + 2B_{3})c_{13} - B_{2}(c_{11} + c_{12})}{(c_{11} + c_{12})c_{33} - 2c_{13}^{2}},$$

$$\lambda_{D} = \frac{1}{4}\left(\frac{(\frac{1}{2}B_{1} + B_{3})(2c_{13} - c_{33}) - B_{2}(c_{11} + c_{12} - c_{13})}{(c_{11} + c_{12})c_{33} - 2c_{13}^{2}}, \dots - \frac{B_{1}}{2(c_{11} - c_{12})} - \frac{B_{4}}{c_{44}}\right).$$
(4)

These equations are solved for B_i

$$B_{1} = (\lambda_{A} - \lambda_{B})(c_{12} - c_{11}),$$

$$B_{2} = -(\lambda_{A} + \lambda_{B})c_{13} - \lambda_{C}c_{33},$$

$$B_{3} = -\lambda_{A}c_{12} - \lambda_{B}c_{11} - \lambda_{C}c_{13},$$

$$B_{4} = (\lambda_{A} + \lambda_{C} - 4\lambda_{D})c_{44}.$$
(5)

The terms for λ_A , λ_B and λ_C in Eqs. (4) and for B_1 , B_2 and B_3 in Eqs. (5) are consistent with Eqs. (A.7) in Ref. [10] and Eqs. (3.4) in Ref. [6]. The terms for λ_D and B_4 in Eqs. (4) and (5) have been corrected as compared to the terms in Refs. [6,10,13,14].

The ME coefficients are calculated with Eqs. (5) for hexagonal Co from the magnetostriction constants of Hubert et al. [15]. The result

is $B_1 = -8.1 \text{ MJ/m}^3$, $B_2 = -29.0 \text{ MJ/m}^3$, $B_3 = 28.2 \text{ MJ/m}^3$ and $B_4 = 37.5 \text{ MJ/m}^3$. The elastic constants for the calculation have been taken from Ref. [16].

In order to measure the magnetoelastic coupling of thin films, a bending beam method with a laser deflection technique is used. The experimental setup has been described in detail before [6,17]. With this method, the mechanical stress of the deposited film material as well as the magnetoelastic stress is determined. From the latter, the magnetoelastic coefficients are calculated directly.

The sample is a thin W(001) single crystal (length: 15 mm, width: 3 mm, thickness: 0.1 mm). The edges are along [100] directions. All experiments have been performed at room temperature in an ultra-high-vacuum chamber at a base pressure of 5×10^{-11} mbar. The Co film is deposited from an electron beam evaporator at a maximum pressure of 2×10^{-10} mbar. We determine the film thickness by calibrating the evaporator before and after deposition with a quartz balance. The film stress is measured during film deposition. A detailed discussion of the growth mode and the film stress of Co/W(001) will be published [18]. The first two monolayers (ML) of Co grow pseudomorphically on W before a HCP growth mode sets in. The HCP Co is oriented with the $(11\bar{2}0)$ -plane in the W(001)-plane and Co[0001] parallel to W[110]or W[1 $\overline{1}$ 0] [18,19], as indicated in Figs. 1(a) and (b).

After deposition of a Co film of a certain thickness, the magnetoelastic coupling is determined from the magnetostrictive stress by first magnetizing the film along the sample width (W[010]-direction) and then along the sample length (W[100]). The resulting change of curvature along the sample length is measured. A magnetooptical Kerr-effect setup is used to control the state of magnetization. Measuring the state of magnetization on different lateral positions on the sample ensures that the fields are sufficient to saturate the whole film.

A minute bending of the substrate during the change of magnetization is detected. The radius of curvature of the W substrate for a 3 ML Co film is of the order of 40 km. From the bending, the magnetoelastic stress is calculated. The ME measure-

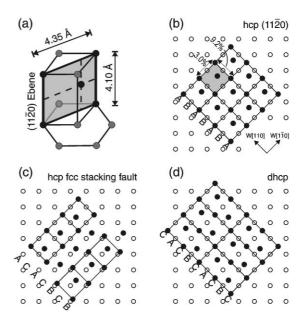


Fig. 1. (a) Section of a HCP cell. The grey marked area indicates an $(1\,1\,\bar{2}\,0)$ plane. It is almost quadratic $(\sqrt{3}a=4.35\,$ and $c=4.10\,$ Å). (b) Crystalline orientation of HCP Co on W(001). The hollow dots represent the positions of W atoms, the filled dots the positions of Co atoms. The same $(1\,1\,\bar{2}\,0)$ area as in (a) is marked again in grey. The HCP(0001) glide planes are oriented perpendicular to the W surface. The stacking sequence of the HCP structure is characterized by the letters ABAB.... (c) At the boundary of two shifted HCP structures the stacking sequence changes from ACAC... to CBCB.... This includes the stacking sequence of an FCC structure which is ACBACB.... (d) A dHCP (double HCP) lattice is formed by the stacking sequence CACBCACBC.... The unit cell is twice as large as in a HCP lattice. The same CACBC sequence is also found in (c) at the transition between the two HCP structures.

ments have been performed for films thicker than two monolayers. For films of two monolayers or less no magnetism was observed upon cooling to 180 K.

Due to the crystalline orientation of the Co-film, the stress change corresponds to the value of the magnetoelastic coefficient B_4 , as will be derived below. The determination of other magnetoelastic coupling constants requires other crystal orientations.

The results of the ME stress measurements are plotted in Fig. 2. The magnetoelastic coupling varies with film thickness and does not show the bulk value $B_4^{\text{bulk, HCP}} = 37.5 \,\text{MJ/m}^3$, as calculated from

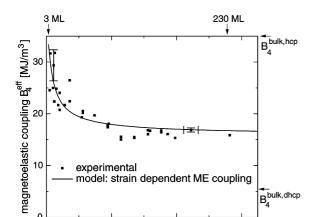


Fig. 2. Experimental data of the ME coupling (squares) as a function of film thickness. The solid line is calculated from the model of a strain-dependent ME coupling. The values of the magnetoelastic coupling of bulk HCP Co, $B_4^{\text{bulk}, \text{HCP}}$ and of a dHCP Co-rich Co-Fe alloy [24,25], $B_4^{\text{bulk}, \text{dHCP}}$ are indicated by arrows.

film thickness [nm]

20

30

10

Eq. (5), even for films of several tens of nm thickness. Such a deviation of the magnetoelastic coupling from its bulk behavior has also been observed for other systems [4,5,7,8,20]. The apparent thickness dependence of the ME coupling can be ascribed to a strain dependent correction of B_j , as will be discussed next.

It has been proposed that the change in magnetoelastic coupling is due to the large strain of the film material [4,21]. In epitaxy, strains of the order of several percent are easily obtained, which is much more than the elastic limit of bulk materials. Thus, the inclusion of higher order strain corrections for the discussion of strain related properties seems appropriate and is performed here for the ME coupling.

This ansatz has been confirmed by experimental results on the ME coupling of Ni/Cu(001) [8,20], Fe/W(001) [7] and Fe/MgO(001) [4]. The more complex structure and morphology of hexagonal cobalt on the quadratic W surface makes the distinction between structural and strain induced changes in the magnetoelastic coupling more cumbersome.

We start with a very simplified approach where the film is treated as a smooth homogenous film with a HCP(1 $\overline{120}$) orientation as mentioned above and indicated in Fig. 1(b). The magnetoelastic stress is measured along the W[100] direction and is given by its thermodynamical definition as the strain derivative of the total energy density $\tau'_1 = /\partial \varepsilon'_1$. Note, that the coordinates of the Co film system have to be transformed into the W substrate system (marked with ') by the appropriate tensor transformation, see e.g. Ref. [6].

When the magnetization is switched from the direction parallel to the sample width to the direction parallel to the sample length, the change in stress is given by

$$\Delta \tau' = \tau'_1 \lceil \alpha' = (1, 0, 0) \rceil - \tau'_1 \lceil \alpha' = (0, 1, 0) \rceil = B_4.$$
 (6)

The HCP[$11\overline{2}0$] cell is not four fold symmetric, since the inner atom is slightly off center. Thus, the crystalline orientation of the cobalt allows for four possible structural domains. Starting from the orientation as indicated in Fig. 1(b), other orientations are obtained by rotating the HCP[$11\overline{2}0$] plane by 90, 180 or 270° in the film plane. However, since the ME stress from Eq. (6) is independent of this orientation, we can treat the film as one structural entity. The influence of the structural domain boundaries on the ME coupling is neglected for simplicity. The influence of structural stacking faults on the ME coupling will be discussed below.

In order to take the large strain into account, we expand the term of the ME energy density [Eq. (2)] to quadratic terms in ε_1' . ε_1' is the film strain along the sample length, which is an average of the strains ε_1 and ε_3 along the Co[1 $\bar{1}$ 00] and [0001] directions, respectively. The magnetoelastic stress, that is measured in the experiment, is the derivative of this energy density. Thus, the second-order terms lead to an effective magnetoelastic coupling that has a linear strain dependence. This can be written by

$$B_4^{\rm eff} = B_4 + D_4 \varepsilon_1'. \tag{7}$$

The film strain ε'_1 is calculated from the measured film stress that is obtained during the deposition. The inset in Fig. 3 shows the epitaxial film stress. Each data point corresponds to a single prepared film. The gradual decrease of the film stress with increasing film thickness is typical for epitaxial systems [22,23]. The solid line serves as

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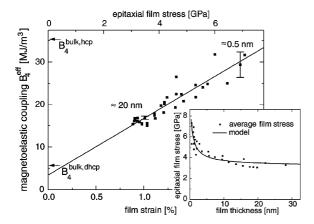


Fig. 3. Measured ME coupling (squares) as a function of measured film stress (upper scale) and linear fit (line). The film strain is calculated with the elastic constants from the film stress. The arrows indicate bulk reference values, see caption of Fig. 2. The inset shows the measured average film stress (squares) as a function of film thickness. The line serves as a guide to the eye.

a guide to the eye. In Fig. 3, the magnetoelastic coupling $B_4^{\rm eff}$ is plotted vs. the strain ε_1 . The experimental data points indicate a linear relation between $B_4^{\rm eff}$ and the strain, as in Eq. (7). The slope of the fitted line is $D_4 = 1346 \pm 120 \,\mathrm{MJ/m^3}$. The thickness dependence of the film strain as obtained from the inset of Fig. 3 is inserted into Eq. (7) and gives rise to an apparent thickness dependence of the ME coupling. The result is shown as a solid line in Fig. 2. The experimental data of Fig. 2 are well described by a strain-dependent ME coupling. Thus, what appears to be a thickness dependence of B_4^{eff} is ascribed to a strain-dependent correction of B_4^{eff} as indicated by Eq. (7). However, the axis intercept in Fig. 3 of $B_4 = 3.4 \pm 1.3 \,\mathrm{MJ/m^3}$ at $\varepsilon = 0$ does not correspond to the bulk value $B_4^{\text{bulk, HCP}} = 37.5 \text{ MJ/m}^3 \text{ of HCP Co, as would be}$ expected from the strain model. Thus, a more detailed look at the structure of the film seems necessary and is discussed next.

The large and anisotropic misfit of Co on W(001) leads to a rather irregular growth. The misfit along $Co[1\bar{1}00]$ is 3.0%. In the perpendicular direction, along Co[0001], it is 9.2%, as indicated in Fig. 1(b). This leads to a locally anisotropic strain that results in islands with additional rows of atoms. Combined investigations with

scanning tunneling microscopy and low-energy electron diffraction [18] suggest, that the additional rows lead to a strain relaxation preferably along the highly strained c-axis direction of the HCP-Co. In the simplest model, one assumes that the strain of 3.0% along Co[1 $\bar{1}$ 00] is conserved. In order to take the anisotropy of the strain into account, Eq. (7) is rewritten as

$$B_4^{\text{eff}} = B_4 + D_4^{(1)} \varepsilon_1 + D_4^{(3)} \varepsilon_3. \tag{8}$$

In a simple picture, it is assumed that ε_1 (along Co[1 I 0 0]) is approximately constant and the film stress relaxation is mainly due to a change in ε_3 (along Co[0 0 0 1]). Thus, $D_4^{(1)}\varepsilon_1$ in Eq. (8) is an additional constant giving rise to a shift of the relation between the ME coupling and the strain. Accordingly, this leads to a deviation of the ME coupling at zero strain ε_3 from the bulk value. However, the neglect of the strain relaxation along ε_1 seems over simplifying, since it is known from other systems, that even much smaller strains relax by the incorporation of misfit dislocations [8,20]. From our measurements, however, we cannot deduce the relaxations along both strain directions independently.

Additionally, the structure in the vicinity of the stacking faults (see Fig. 1(c)) is changed, which can influence the ME coupling significantly. The stacking faults correspond to a few atomic rows in a double-HCP (dHCP) structure [18]. A dHCP structure is illustrated in Fig. 1(d). Every sixth $HCP(11\overline{2}0)$ elementary cell, an additional row is implemented, thus leading to a significant amount of changes in the stacking sequence. For Co/ W(001) grown at 400 K even an extended dHCP structure has been found [19]. The dHCP structure of bulk Co-rich Fe-Co alloys is known to exhibit a drastically changed ME coupling as compared to the HCP structure [24,25]. Experiments on Co- ≈ 2 at% Fe alloys indicate $B_4^{\text{bulk, dHCP}} =$ 5.6 [24] and $7.7 MJ/m^3 [25]$, which is much smaller than the HCP-Co ME coupling of $B_4^{\text{bulk}, \text{HCP}} =$ 37.5 MJ/m³. This strong correlation between film structure and ME coupling is another possible reason for the small value of the ME coupling of the unstrained material as obtained from the extrapolation of the experimental values to $\varepsilon = 0$. This extrapolation yielded a value of $B_4 = 3.4 \,\mathrm{MJ/m^3}$, which is close to the dHCP value of $\approx 6 \text{ MJ/m}^3$, but it deviates sharply from the HCP value of 37.5 MJ/m^3 .

In extension of former investigations [4,7,8], we find that for Co on W(001) not only the strain, but also the structure plays an important role for the ME coupling. In the case of simple epitaxial systems like Fe/W(001) [7,9], Fe/MgO [4] and Ni/Cu(001) [8] the stress relaxation is expected to be isotropic on average and no structural changes occur besides the formation of dislocations. Thus, for these cubic systems, the change in ME coupling could be fully explained by a strain correction term. In the case of Co/W(001), also a linear dependence between ME coupling and strain has been found, as corroborated by Fig. 3. For a complete description of the non-bulk like ME coupling, all aspects like a strain-dependent correction term, the anisotropic strain relaxation, and the structural changes of the Co film have to be taken into account.

The most important result of this study is that — in contrast to common practice in anisotropy discussions — bulk magnetoelastic coupling coefficients do not apply to ultrathin epitaxial films in general. Previously, surface magnetoelastic coupling coefficients that contribute via a term inversely proportional to the thickness and to the overall magnetoelasticity have been proposed to account for the non-bulk-like magnetoelastic behavior in thin films [26,27] or for the modified magnetoelastic coupling in the surface region of amorphous alloys [28]. However, this study and other recent work on the direct measurement of both magnetoelastic stress and film stress [4-6,8] identify film strain as a key factor for the modified magnetoelastic coupling in epitaxial films.

The strain-induced change of the magnetoelastic properties has a profound impact on the discussion of magnetic anisotropy. The modified magnetoelastic behavior of epitaxial layers can be viewed as the decisive factor for the anisotropy of ultrathin films. The application of the experimentally determined magnetoelastic coupling coefficients can explain the peculiar magnetic anisotropy, e.g., of Fe(110) or Ni(100) films. It is proposed that interface and surface contributions to the anisotropy can be considerably smaller than previously assumed [8]. In some cases, interface anisotropy contributions

are not mandatory to explain the anisotropy of strained films, if the appropriate experimental values of the magnetoelastic coupling are employed [29].

In conclusion, we investigated the ME coupling of thin Co/W(001), which is vastly different from the Co bulk value. A linear correlation between film strain and ME coupling has been found, thus suggesting that the change in the ME coupling is due to the large strain. Both, the anisotropic stress relaxation and the complex structure of the HCP growth have to be considered to explain the difference between the bulk value for HCP-Co and the zero strain ME coupling as extrapolated from the experiment.

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