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The thickness dependence of the magneto-elastic coupling B_1, the intrinsic film stress, and the magnetic in-plane anisotropy K_4 of Fe films on W(100) are measured with an *in situ* combination of a highly sensitive optical deflection technique with magneto-optical Kerr-effect measurements. We find that both B_1 and K_4 depend strongly on the Fe film thickness. The thickness dependence of B_1 can be described by considering a second order magneto-elastic coupling constant $D=1\,\mathrm{GJ/m^3}$ as a strain dependent correction of B_1. We tentatively ascribe the deviation of K_4 from its bulk value to the tetragonal lattice distortion caused by an effective tensile *in-plane* strain of 5.3% in the pseudomorphic region and of 0.2% in thicker films. © *1999 American Institute of Physics.* [S0021-8979(99)40308-1]

In this article we show that the magneto-elastic (ME) coupling B_1 of epitaxial Fe films differs drastically in value and sign from its respective bulk behavior for film thicknesses below 30 nm. This result proves that the assumption of bulk ME constants for the description of the magnetic film properties is wrong. According to a phenomenological model,[1,2] we ascribe this thickness dependence to the epitaxial film strain, which is induced by the lattice mismatch between the Fe film and the W substrate. To study the effects of epitaxial strain in ultrathin Fe films on both the ME coupling and the in-plane anisotropy, we measured the thickness dependence of the intrinsic film stress τ_F, the magneto-elastic coupling B_1, and the magnetic *in-plane* anisotropy K_4 by combining film stress measurements with magneto-optical Kerr-effect (MOKE) measurements.

The magneto-crystalline, elastic, magneto-elastic, and shape energy density contribute to the total energy density $F=f_{\mathrm{MC}}+f_{\mathrm{el}}+f_{\mathrm{ME}}+f_{\mathrm{shape}}$, which is a function of the direction of saturation magnetization M_s described by the direction cosine α_i with respect to the cubic axis, the film strain ϵ_i, and the film thickness t_F. For thin films with cubic structure F can be written as[3]

$$\begin{aligned}F=&K_4(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_1^2\alpha_3^2)+2K_2\alpha_3^2/t_F\\&+1/2c_{11}(\epsilon_1^2+\epsilon_2^2+\epsilon_3^2)+c_{12}(\epsilon_1\epsilon_2+\epsilon_2\epsilon_3+\epsilon_1\epsilon_3)\\&+1/2c_{44}(\epsilon_4^2+\epsilon_5^2+\epsilon_6^2)+B_1(\epsilon_1\alpha_1^2+\epsilon_2\alpha_2^2+\epsilon_3\alpha_3^2)\\&+B_2(\epsilon_4\alpha_1\alpha_2+\epsilon_5\alpha_2\alpha_3+\epsilon_6\alpha_1\alpha_3)+1/2\mu_0M_s^2\alpha_3^2.\end{aligned}\tag{1}$$

Here the c_{ij} denote the elastic constants of the film in the contracted Voigt's notation. Direction 3 is assumed to be parallel to the film normal. A Néel-type uniaxial interface contribution K_2 to the magneto-crystalline anisotropy f_{MC} is included. For the case of simple epitaxy considered here, the shear strains are zero: $\epsilon_4=\epsilon_5=\epsilon_6=0$. The strain perpendicular to the film plane $\epsilon_3=\epsilon_\perp$ can be expressed as a function of the isotropic in-plane strain $\epsilon_1=\epsilon_2=\epsilon_\parallel$ as $\epsilon_\perp=-2\epsilon_\parallel c_{12}/c_{11}$. Minimization of Eq. (1) with respect to ϵ_i gives the magnetostrictive deformation of a magnetized free solid. In contrast to freely deformable samples, the magnetostrictive deformation of epitaxial films is hindered by the bonding to the substrate. The strain derivatives of Eq. (1) give the resulting in-plane stresses τ_1 and τ_2 and the equilibrium deformation perpendicular to the film plane from $\tau_3=0$

$$\begin{aligned}&\partial F/\partial\epsilon_i=c_{11}\epsilon_\parallel+c_{12}(\epsilon_\parallel+\epsilon_\perp)+B_1\alpha_i^2=\tau_i,\quad i=1,2\\&\partial F/\partial\epsilon_3=c_{11}\epsilon_\perp+2c_{12}\epsilon_\parallel+B_1\alpha_3^2=0.\end{aligned}\tag{2}$$

Equation (2) shows that both the epitaxial strain and the magnetization direction dependent terms contribute to the total in-plane stress. To obtain B_1 experimentally, we switch the magnetization direction in-plane between [100] and [010] while measuring the resulting magnetostrictive stress along [100] as described later. The measured difference in stress follows from Eq. (2) to be $\Delta\tau=\tau_1(\alpha_1=1)-\tau_1(\alpha_1=0)=B_1$. Thus, a magnetostrictive stress measurement is an appropriate technique to determine the ME coupling B_1 in thin films *directly*. In contrast to bulk samples, for epitaxial films the correlation between B_1 and the magnetostrictive constant λ_{100} is given by $\lambda_{100}^F=-2B_1/3c_{11}$ and not by $\lambda_{100}^B=-2B_1/3(c_{11}-c_{12})$, as already pointed out in Ref. 4. The magnetic in-plane and out-of-plane anisotropies $f_\parallel$ and $f_\perp$ follow directly from Eq. (1) as

$$f_\parallel=K_4/4$$

and

$$f_\perp=-B_1\epsilon_0(1+2c_{12}/c_{11})+2K_2/t+\mu_0M_s^2/2.\tag{3}$$

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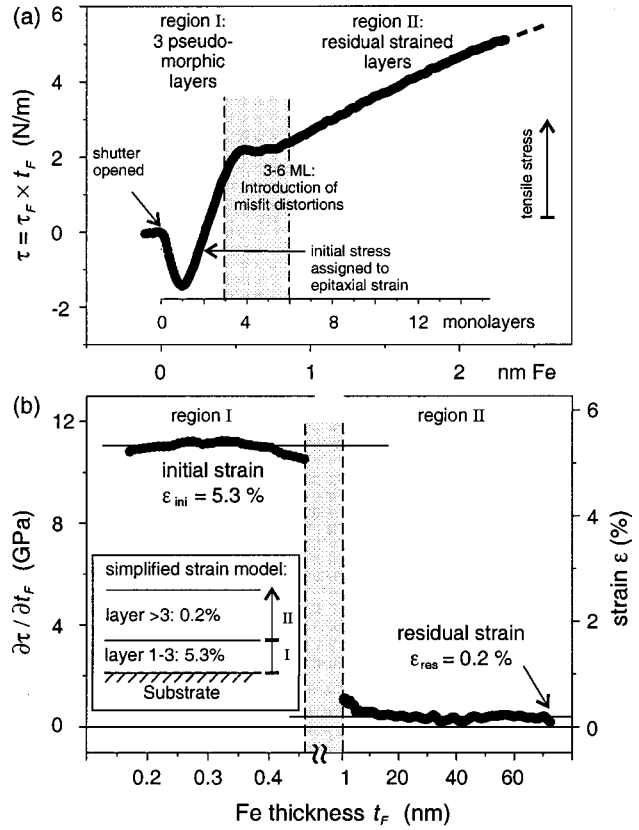

FIG. 1. (a) Stress measurement during Fe growth on W(100) at 300 K. The kink in the curve at 3.5 ML Fe separates two regions of different slope. (b) The thickness derivative of a stress curve for a 73 nm Fe film reveals the stress contribution of each growing layer to the film stress. The stress per layer changes from 11 GPa in region I to 0.4 GPa in region II in a transition region between 0.5 and 4 nm. Inset: for the discussion of the magneto-elastic coupling in nm films a simplified strain model is assumed with constant strains $\epsilon_{\mathrm{I}}=5.3\%$ and $\epsilon_{\mathrm{II}}=0.2\%$.

For $f_{\perp}<0$, a perpendicular magnetization of the film is preferred. Note, that Eq. (3) states that the in-plane anisotropy is not affected by a homogeneous film strain, whereas in cases of strong ME-coupling the in-plane strain $\epsilon_{\parallel}$ can favor an out-of-plane magnetization, as suggested for Fe double layers on W(110).[5]

Film stresses and ME coupling were determined with a highly sensitive optical deflection technique.[6] In short: a stress induced bending of a thin tungsten single crystal is detected by measuring the deflection of a reflected laser beam. The relaxation of an epitaxially induced film strain due to the substrate bending with curvature R is of the order $t_F/R\approx 10^{-4}$, thus a strain relief is practically not expected and Eq. (2) is justified. A typical stress measurement during the growth of 2.2 nm Fe (=15 Fe-bulklike monolayers (ML)[7]) on a W(100) substrate at RT is depicted in Fig. 1(a). Since the substrate curvature is proportional to the product of film stress τ_F and film thickness t_F, the position signal increases linearly with t_F for a constant τ_F. Despite the large misfit between Fe and W of 10.1%, pseudomorphic growth is observed during the deposition of the first 3 ML, as checked by low-energy electron diffraction (LEED) and investigated previously.[8] Thus, strong tensile stress of order $\tau_{\mathrm{init}}=(Y/1-\nu)_{\mathrm{Fe}}\epsilon_{\parallel}=21\,\mathrm{GPa}$ is expected $[(Y/1-\nu)_{\mathrm{Fe}}=207.3\,\mathrm{GPa}]$.

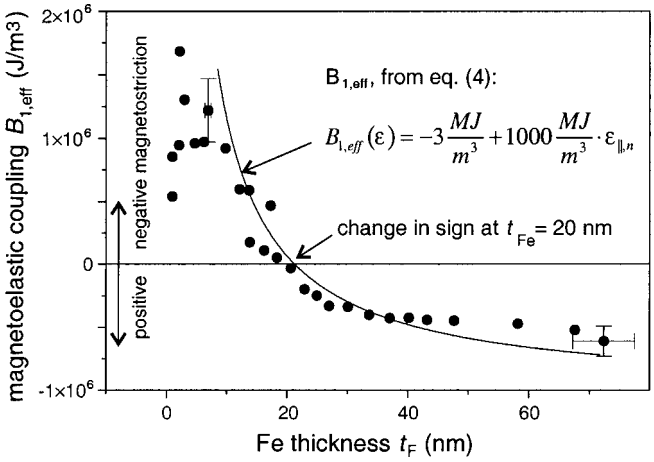

FIG. 2. Experimentally determined effective magneto-elastic coupling $B_{1,\mathrm{eff}}$ as a function of Fe film thickness. The solid line results from a summation over the strain dependent contributions of each layer to $B_{1,\mathrm{eff}}$, as given by Eq. (4).

However, compressive stress is observed as indicated by the negative slope in Fig. 1(a) for submonolayer Fe coverages. A detailed study reveals that surface stress effects during the formation of the Fe–W interface are responsible for this compressive stress.[9,10] We ascribe the constant slope in the stress curve for coverages between 1 and 4 ML to the tensile stress in the pseudomorphic film. The kink in the stress curve indicates the beginning of stress relaxations at a thickness of $t_C=3.5$ ML. We suggest that the increasing elastic energy of the film with increasing film volume favors the formation of misfit distortions.[11] However, an atomistic picture of the processes at t_C remains to be investigated and is not the scope of this work. The kink separates two regions I and II of vastly different film stress. In region I the constant slope indicates a tensile stress of 11 GPa, whereas the stress decreases to a constant residual value of 0.4 GPa in region II. To discuss the film stress, we plot the derivative of a stress curve for a much thicker film of $t_{\mathrm{Fe}}=73$ nm with respect to t_F in Fig. 1(b). The striking point of this plot is that the stress per layer drops from the initial stress of 11 GPa to the residual stress value of 0.4 GPa within a narrow thickness range between 0.5 and 4 nm. This result of two constant values for the film stress in regions I and II is in contrast to the model of a gradual strain relief that we used earlier.[12]

We attribute the in-plane stress to the film strain $\epsilon_{\parallel}$ and calculate $\epsilon_{\parallel}$ from τ_F with $\epsilon_{\parallel}=\tau_F/(Y/1-\nu)_{\mathrm{Fe}}$. The measured film stress of 11 GPa deviates by a factor of 2 from the predictions of elasticity theory. The simple strain analysis assumes layer by layer growth, bulk elastic constants, and no strain relaxation at the island edges. These assumptions are clearly not fulfilled for the film in the monolayer range at RT, and the calculated *effective* strain amounts to 5.3%, only half of the epitaxial misfit strain of 10%. The residual strain is calculated to be 0.2%. For the discussion of the ME coupling we assume the simplified strain model in the inset of Fig. 1(b) with constant strains in regions I and II.

Successive magnetization of our sample along its length and width causes a magnetostrictive stress along the sample length, which equals exactly B_1, as already pointed out. We

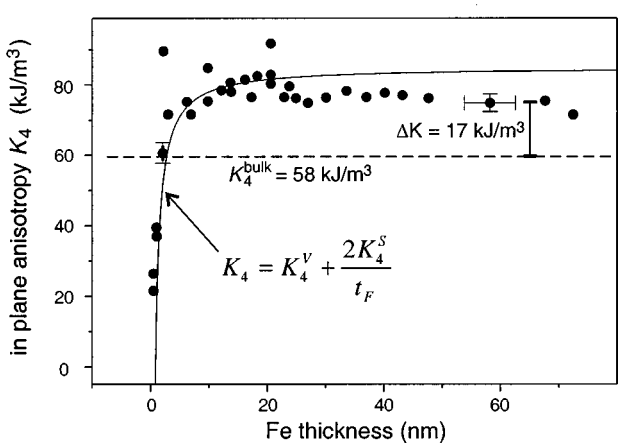

FIG. 3. Magnetic *in-plane* anisotropy K_4 as a function of Fe thickness. The best fit of the experimental data comes from a thickness dependent anisotropy model with $K_4^V = 85\ \mathrm{kJ/m^3}$ and $K_4^S = -0.035\ \mathrm{mJ/m^2}$. Even for high film thicknesses a 30% deviation from the bulk value of $K_4^{bulk} = 58\ \mathrm{kJ/m^3}$ is observed.

measured the B_1 as a function of the Fe thickness in the thickness range between 0.5 and 73 nm of Fe, as plotted in Fig. 2. In contrast to the respective bulk behavior, we find an *effective* ME coupling $B_{1,\mathrm{eff}}$ for epitaxial Fe films on W(100) which depends strongly on the Fe film thickness: nearly constant negative values of $B_{1,\mathrm{eff}} = -0.5\ \mathrm{MJ/m^3}$ ($\lambda_{100}^F = 1.5 \times 10^{-6}$) are measured for $t_F > 40$ nm, whereas $B_{1,\mathrm{eff}}$ changes its sign at $t_F = 20$ nm. The maximum positive value of $B_{1,\mathrm{eff}} = +1.7\ \mathrm{MJ/m^3}$ is reached at 5 nm, and below 5 nm the absolute value of $B_{1,\mathrm{eff}}$ decreases again. Following a model of O'Handley,[1] which was also applied by Koch,[2] we ascribe this thickness dependence of $B_{1,\mathrm{eff}}$ for $t_F > 10$ nm to a strain dependence of $B_{1,\mathrm{eff}}$. A strain dependent contribution to the bulk ME coupling $B_{1,\mathrm{bulk}}$ is considered by the second order ME coupling constant D, as $B_{1,\mathrm{eff}} = B_{1,\mathrm{bulk}} + D\epsilon_{\parallel}$. We now assume that every layer contributes to the effective ME behavior of the whole film $B_{1,\mathrm{eff}}$, which can be estimated for a film of N layers by summation over the $B_{1,\mathrm{eff}}$ of each layer and inserting the simplified strain model from Fig. 1(b):

$$B_{1,\mathrm{eff}}(N) = 1/N \sum_{n=1}^{N} (B_{1,\mathrm{bulk}} + D\epsilon_{\parallel,n}). \tag{4}$$

The solid curve in Fig. 2 shows a reasonable agreement of this model with the experimental data with $B_{1,\mathrm{bulk}} = -3\ \mathrm{MJ/m^3}$ and $D = 1000\ \mathrm{MJ/m^3}$. These values of $B_{1,\mathrm{bulk}}$ and D are within 10% of the bulk ME coupling ($B_1 = -3.44\ \mathrm{MJ/m^3}$) and the previously reported strain correction of $D = 1100\ \mathrm{MJ/m^3}$ (Ref. 2) of Fe. This model proposes a huge B_{eff} for monolayer films, which was, however, not found in experiment. The decrease of B_{eff} for $t_F < 5$ nm indicates that surface contributions might play a dominant role, as shown by Bochi.[13] The same solid curve in Fig. 2 can be obtained by inserting the average strain of the whole film and different values for D and B_{bulk} in the expression of B_1 as published earlier[12] instead of a splitting in single layer contributions, but the respective bulk behavior for an unstrained film can only be reproduced by Eq. (4).

The magnetic in-plane anisotropy K_4 was determined from MOKE loops during magnetization along [100] while a constant bias field was applied along [010]. By this procedure proposed by Allenspach,[14] a bias field induced hard axis loop along an easy film axis is measured. From the slope of this hard axis MOKE loop K_4 is calculated, and plotted as a function of the Fe thickness in Fig. 3. We find a constant value of $K_4^V = 85\ \mathrm{kJ/m^3}$ for films thicker than 10 nm and a strong deviation from K_4^V for $t_F < 10$ nm, that can qualitatively be described with a Néel-type surface contribution to K_4 with $K_4^S = -0.035\ \mathrm{mJ/m^2}$, as indicated by the solid line in Fig. 3. However, K_4^V deviates by 30% from bulk value of K_4^{bulk}, which leads to a 30% too high value for the experimentally determined K_4 even at higher film thicknesses. Higher order anisotropy contributions due to the tetragonal deformation of the Fe unit cell in the film might be the reason for this discrepancy, as was discussed in Ref. 15. Since the information depth of our MOKE signal is of the order of 13–15 nm,[16] a strong deviation from bulk anisotropy due to the contribution of the highly tetragonal distorted first 3 ML below this thickness is expected, as well as a gradual decay of their influence above 15 nm.

In conclusion, we have shown that the thickness dependence of the experimentally determined $B_{1,\mathrm{eff}}$ can qualitatively be understood by considering strain corrections to $B_{1,\mathrm{bulk}}$ in a layer by layer way. The thickness dependent deviation of K_4 from the Fe bulk value is tentatively explained by strain dependent tetragonal lattice distortions that are expected even in thicker films due to the residual epitaxial misfit strain of order 0.2%.

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