Magnetization, Magnetostriction and Film Stress of Fe Monolayers on W(100)

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***Abstract*—Film stress, magnetization and magnetoelastic coupling are measured *in situ* by a simple optical deflection technique. This technique reveals that 2 nm thin Fe films grown on W(100) show magnetoelastic coupling that differs in both magnitude and sign from Fe bulk data. Stress contributions to the magnetoelastic coupling are discussed to describe this peculiar magnetoelastic behavior that is observed for Fe films thinner than 20 nm.**

***Index Terms*— Film stress, magnetoelastic coupling, magnetostriction, Fe, W**

I. Introduction

One of the fundamental aspects of heteroepitaxial growth is how the lattice mismatch between film and substrate induces characteristic changes in the structural and magnetic properties of ferromagnetic films. Stress driven structural changes in the growing film such as the formation of misfit dislocations and the change of the growth mode from layer by layer to Stranski-Krastanov have been found to induce characteristic changes in the coercivity or the easy axis of magnetization, as discussed for the growth of Fe monolayers on W(110) [1]. However, the most direct example for the intimate relation between strain and magnetism comes from experiments on the magnetoelastic coupling. We measure the film stress induced by magnetostrictive strain in order to derive the magnetoelastic coupling coefficient directly. We find that the magnitude of the magnetoelastic coupling becomes thickness dependent and changes its sign for Fe films thinner than 20 nm, deposited on W(100). The cantilever bending technique is employed to measure film stress, film magnetization and magneto-elastic coupling *in situ* under ultrahigh vacuum (UHV) conditions.

II. Optical Deflection Measurement

The application of the cantilever technique to measure all three properties: film stress, magnetization and magnetostriction *in situ* with the same apparatus was pioneered by Weber, Koch and Rieder with a capacitive detection of the substrate deflection [2]. Klokholm used the cantilevered substrate as a capacitor in the tuning circuit of a 10 MHz oscillator and he detected magnetostrictive bending of the substrate as a frequency shift of the oscillator [3]. Tam and Schroeder described a high-precision optical deflection technique to measure magnetostrictive strain as small as 10^{-7} in 30 nm films that were deposited on 20 μm thin glass substrates [4]. Optical deflection techniques that measure magnetostrictive strain with a resolution of 10^{-8} in 5 nm films are commercially available [5]. We use a simple optical deflection technique described earlier [6] to measure film stress, magnetization and magnetostriction of monolayers deposited on a 70 μm thick W(100) single crystal. In short, a laser beam is reflected from the bottom end of the W(100) substrate crystal onto a position sensitive detector. Any change of the radius of curvature R will induce a respective displacement Δ of the reflected beam on the detector, inducing a position signal proportional to Δ. From a measurement of R under different experimental conditions, film stress, magnetization and magnetostriction can be calculated.

III. Data Evaluation

Before we discuss our experimental results we briefly compile the necessary equations to derive film stress, film magnetization and magnetostriction from a measurement of the bending of a cantilevered substrate. Young's modulus Y and Poisson's ratio ν have to be calculated for the (100) surface orientation. As discussed by Brantley [7], Y and ν can be derived from the elastic compliances s_{ij}: $Y_{(100)} = 1/s_{11}$, $\nu_{(100)} = -s_{12}/s_{11}$. In Table I these values are given for Fe and W. In the following all elastic properties refer to the (100) plane and the subscript (100) is replaced by S or F, depending on the application to substrate or film properties, respectively.

An isotropic film stress τ_F induced by epitaxial misfit between a film and substrate leads to a bending of the film-substrate composite with a radius of curvature R [8]:

$$\tau_F = \frac{Y_S t_S^2}{6(1-\nu_S) R t_F} \tag{1}$$

The sample thickness is given by $t_S = 70$ μm, the film thickness by t_F. Note that even thick films with $t_F = 100$ nm, the film can be regarded as infinitesimally thin with respect to the substrate. Thus eq.(1) can be employed to calculate the film stress from the radius of curvature without further corrections.

For film magnetization with a total magnetic moment $\boldsymbol{m}_{tot}$ along the length of the substrate, an external magnetic field $\boldsymbol{B}$, which is oriented perpendicularly to the substrate, will induce a torque $\boldsymbol{T} = \boldsymbol{m}_{tot} \times \boldsymbol{B}$, which causes the substrate to bend with a radius of curvature R. Elementary theory of bending [9] gives the relation between torque, radius of curvature, and the flexural rigidity of the sample: $T = Y_S I/R$. The flexural rigidity is given by the product of Young's modulus of the sample with the moment of inertia of the cross section of the sample with the sample width b: $I = t_S^3 b/12$. A measurement of the resulting radius of curvature gives the magnetization [2]:

$$m_{tot} = \frac{Y_S t_S^3 b}{12 R B} \tag{2}$$

Finally, a magnetostrictive strain is induced in the Fe film upon magnetization. However, the film is not free to expand but rather is bonded to the substrate, a magnetostrictive stress is

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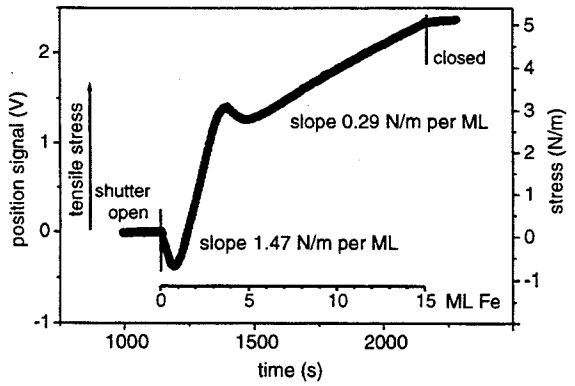

Fig. 1. Film stress during growth of 15 ML Fe on W(100) at 300 K. The kink in the curve around 4 ML is ascribed to the formation of misfit distortion.

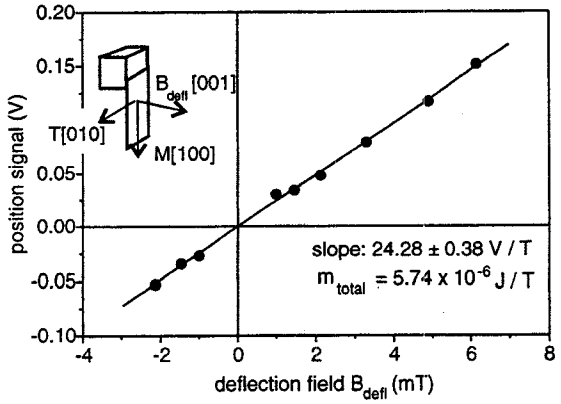

Fig. 2. Bending beam magnetometry of 72 nm Fe on W(100). The substrate bends due to the torque T induced by the deflection field B_{defl} interacting with the film magnetization M.

induced on the front side of the film-substrate composite. To perform a measurement of the magnetoelastic coupling coefficient B_1, we magnetize the film along the sample length and along its width. The resulting change of the radius of curvature $R^L - R^W$ is measured simultaneously. The magnetoelastic coupling coefficient is calculated from:

$$B_1 = -\frac{Y_S(1+\nu_F)t_s^2}{Y_F(1+\nu_S)6t_F}\left(\frac{1}{R^L}-\frac{1}{R^W}\right)(c_{11}^{Fe}-c_{12}^{Fe}) \qquad (3)$$

(c_{11}^{Fe} = 229 GPa, c_{12}^{Fe} = 134 GPa [10]). The results of eq.(3) can be expressed in terms of the magnetostrictive constant λ_{100} [11], which describes the strain due to magnetization along [100] starting from an *ideally demagnetized* sample: $\lambda_{100} = -2B_1/3(c_{11}^{Fe}-c_{12}^{Fe})$.

In the derivation of eq.(1) and eq.(3), a free 2D bending of the cantilevered sample, i.e. bending along the length *and* the width is assumed. A hindered bending along the sample width, as expected for short samples clamped along the width, changes the evaluation drastically: in eq.(3) the factor $(1+\nu_S)$ has to be replaced by $(1-\nu_S^2)$ [12], and the substrate appears stiffer. For cantilevered substrates it can be assumed that an increasing length to width ratio > 2 facilitates 2D bending [8], and we employ eq.(1) and eq.(3) for the data evaluation (substrate length=13 mm, width=3 mm). Note that the magnetostriction results obtained for different sample geometries cannot easily be compared, due to the issue of free 2D bending.

TABLE I
Elastic compliances [10] and calculated Young's modulus Y and Poisson's ratio ν for the (100) surface.

	s_{11} 10^{-11} m^{2}N^{-1}	s_{12} 10^{-11} m^{2}N^{-1}	$Y(100)$ 10^{11} Nm^{-2}	$\nu(100)$
Fe	0.764	–0.281	1.309	0.368
W	0.249	–0.07	4.016	0.281

IV. Experimental Results and Discussion

To measure the stress in an Fe film grown on W(100) we monitor the position signal of the split photodiode during growth. As shown in Fig.1, for depositions of up to 0.6 monolayers (ML, 1 ML = a_{Fe} / 2 = 0.144 nm), a negative position signal indicates compressive stress. For 0.7 ML to 3 ML film thicknesses, a tensile stress of order 1.47 N/m per deposited ML is observed. The compressive stress observed for submonolayer coverages is in contrast to the tensile stress which one might expect from lattice mismatch arguments, where the growth of the first monolayers has been reported to lead to a pseudomorphically strained Fe film [13]. From the lattice mismatch

$f=(a_W-a_{Fe})/a_{Fe}=10.5\%$, $a_W=0.317$ nm, $a_{Fe}=0.287nm$, a tensile stress of order $\tau_F=fY_{Fe}/(1-\nu_{Fe})=21.7$ GPa is expected. We ascribe this compressive stress to a surface stress effect in the Fe-W composite, as discussed in more detail elsewhere [14]. The measured tensile slope 1.47 N/m per ML corresponds to a film stress in the pseudomorphic regime of 10.3 GPa, which is only half of the value expected from lattice mismatch arguments. For films thicker than 5 ML we measure a constant film stress of ~ 2 GPa, as indicated in Fig. 1 by the slope of 0.29 N/m per ML. We find that this slope decreases gradually for increasing film thicknesses. We conclude that the investigated 15 ML (2.2 nm) Fe film is under a tensile stress of order 2 GPa, which could be ascribed to a residual misfit strain of order 1 %.

Before we discuss our results on magnetoelastic coupling we briefly demonstrate in Fig. 2, that the cantilever technique can be used to measure the total magnetic moment m_{tot} of a 72 nm Fe film. As already described above (eq.(2)), and schematically indicated in Fig. 2, a proper orientation of film magnetization M, and deflection field B_{defl}, will induce a bending of the sample, from which m_{tot} can be deduced. A small deflection field is increased in small increments, and for each value of B_{defl}, the resulting position signal is monitored. From the slope of the position signal *vs.* B_{defl} in Fig.2, we calculate $m_{total}=5.74\times10^{-6}$ J/T, giving a magnetic moment per Fe atom of 2.5 μ_{Bohr}. The most significant error in the determination of the total magnetic moment is ascribed to the uncertainty in the determination of the film volume. Based on the calibration of the Fe evaporator with a quartz microbalance, we assume this error to be ± 10 %. A further source of error arises from the low values of the coercivity observed in ultrathin Fe(100) films in combination with the four-fold symmetry of the easy axis of M. Even a slight misalignment between sample and B_{defl} has the

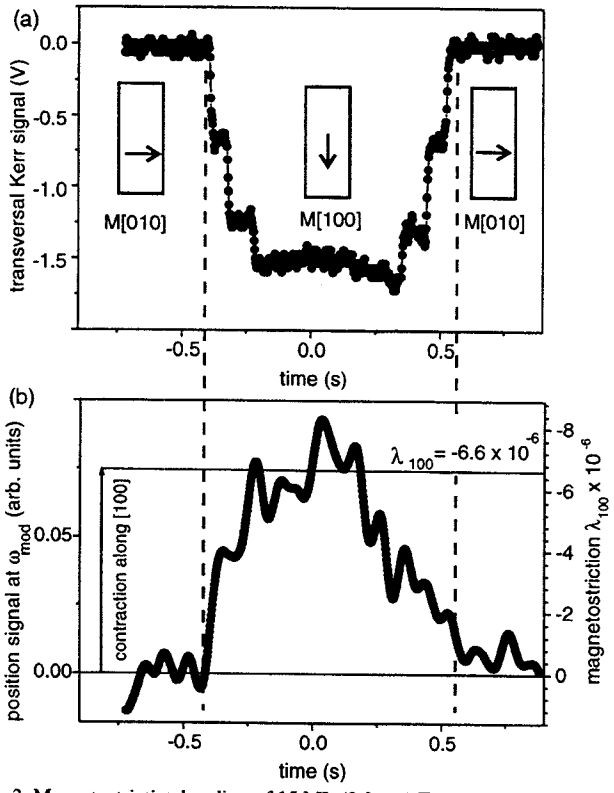

Fig. 3. Magnetostrictive bending of 15 ML (2.2 nm) Fe on W(100). (a) Transversal Kerr effect measurements verify a switching of the magnetization direction. (b) Positive position signals indicate a contraction of the Fe film upon magnetization along its length.

potential to change the sample magnetization. Thus, a simultaneous measurement of M by magneto-optical Kerr effect was required to make sure that the total magnetic moment was pointing along the long axis.

In Fig. 3, we present data on the magnetostrictive stress in 15 ML (2.2 nm) Fe on a 70 μm thick W(100) substrate. As derived above in eq.(3) we can deduce the magnetoelastic coupling constant B_1 or the magnetostricitive constant λ_{100} from a difference in the curvature of the substrate for magnetization along the sample width and along the sample length. Magnetostrictive strains however, are typically a factor of 1000 smaller than misfit strain. Thus the resulting stress is minute and pushes the resolution of this technique to its limits. To enhance the resolution for minute sample bending, we employ a modulation technique of both the magnetizing field and the laser output and use phase sensitive detection [4]. While measuring the magnetostrictive signal, the sample magnetization is monitored by the transversal Kerr effect simultaneously. A constant magnetic field along the width is superimposed with a periodically increasing field along the sample length. As shown in Fig. 3(a), the sample magnetization is modulated at 1 Hz along the width and along the length. Simultaneously, the position signal is monitored, Fig. 3(b). At the beginning of the cycle, the sample is magnetized along the width, and a constant position signal is measured. When the sample magnetization switches to being parallel to the sample length in the middle of the cycle at time = 0, the position signal becomes positive, indicating a tensile magnetostrictive stress. Later in the cycle, the magnetization switches back to the horizontal direction, and the position signal follows correspondingly. The positive position signal for magnetization along the sample length indicates the tendency of the 15 ML thin film to contract upon magnetization, in contrast to thicker Fe films which show a behavior similar to what is observed in the bulk of expansion on magnetization. We measure $\lambda_{100} = -6.6 \times 10^{-6}$, corresponding to $B_1 = 1.13$ MJ/m^3. Both values have to be compared to the data for bulk Fe [11], λ_{100}(bulk)= 20.7×10^{-6}, B_1(bulk)= -3.44 MJ/m^3 to underline the dramatically changed magnitude and sign of the magnetoelastic coupling in 15 ML thin Fe films.

The origin of the deviation of the magnetoelastic properties of ultrathin films from the bulk values can be found in the strain dependence of the magnetoelastic coupling constant B_1 [15]. The misfit stress, as measured in Fig. 1, has been proposed to change B_1 from a constant to be linearly dependent on film stress. According to Koch's *et al.* work, a tensile stress in excess of 0.65 GPa causes the magnetoelastic coupling for epitaxial Fe (100) films to change its sign [15]. This change of sign is supported by our results on the 15 ML Fe film, however the magnitude of our experimental value B_1 differs from their value for 2 GPa stress by roughly a factor of five. Thus we conclude that the results of [15] that were obtained for 100 nm thick Fe films are not necessarily relevant for our 2 nm thin film. A thickness dependence of the magnetoelastic coupling has been proposed [16]. The authors [16] introduce a Néel type surface contribution to the magnetoelastic coupling and they report a strong positive contribution to B_1 with decreasing film thickness, which agrees with our results.

Whereas a qualitative description of the novel magnetoelastic coupling behavior in ML films can be obtained, a first principle investigation of magnetoelastic coupling in epitaxial monolayers is clearly called for [17].

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