PHYSICAL REVIEW B VOLUME 60, NUMBER 16 15 OCTOBER 1999-II

Stress dependence of the magnetoelastic coupling constants B_1 and B_2 of epitaxial Fe(001)

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(Received 4 June 1999)

Magnetoelastic (ME) coupling, a property of major importance in heteroepitaxy, describes the dependence of the free energy of magnetic materials on strain/stress. Using our versatile UHV cantilever beam magnetometer we have investigated the ME coupling constants B_1 and B_2 of epitaxial Fe(001) films in the thickness range of 2–100 nm, where the films are characterized by the magnetization and the magnetic anisotropy of bulk Fe. Both constants exhibit a strong dependence on the film stress above 0.1 GPa and even change sign at stress values in the GPa range. Whereas B_2 decreases linearly with film stress up to 6 GPa, B_1 saturates after a steep linear increase at 2–3 GPa. Stress-free Fe(001) films exhibit bulk behavior. [S0163-1829(99)50340-8]

Magnetoelastic (ME) energy—though usually negligible in the bulk—may dominate the magnetic contributions to the free energy in thin films, thus offering the challenging opportunity to stabilize new magnetic anisotropies in future magnetic devices (MRAM's, etc.). The perpendicular magnetization of single Co layers on Au(111),[1] for instance, or of Ni(001)/Cu(001) in the thickness range of 7–60 ML,[2–4] most likely are the result of ME effects. The ME energy F_{mel}—contrary to the quadratic elastic energy F_{el}—depends linearly on the strain ϵ_{ij}, and consequently may be positive or negative as well. Therefore when the strain is high enough and furthermore has the proper sign, F_{mel} may compete with the magnetocrystalline energy F_{mcr}.

In the cubic system the two strain dependent contributions of the free energy, F_{el} and F_{mel}, are given by the following equations (tensor notation):[5]

$$F_{el}=\frac{1}{2}C_{11}(\epsilon_{11}^2+\epsilon_{22}^2+\epsilon_{33}^2)+2C_{44}(\epsilon_{12}^2+\epsilon_{23}^2+\epsilon_{31}^2)+C_{12}(\epsilon_{11}\epsilon_{22}+\epsilon_{22}\epsilon_{33}+\epsilon_{33}\epsilon_{11}), \quad (1)$$

$$F_{mel}=B_1\left[\epsilon_{11}\left(\alpha_1^2-\frac{1}{3}\right)+\epsilon_{22}\left(\alpha_2^2-\frac{1}{3}\right)+\epsilon_{33}\left(\alpha_3^2-\frac{1}{3}\right)\right]+2B_2(\epsilon_{12}\alpha_1\alpha_2+\epsilon_{23}\alpha_2\alpha_3+\epsilon_{31}\alpha_3\alpha_1). \quad (2)$$

C_{ij} and B_i are the elastic stiffness and the ME coupling constants, respectively (for bulk Fe: $B_1=-3.44\times10^6$ J/m^3 and $B_2=+7.62\times10^6$ J/m^3); α_i are the direction cosines of the magnetization vector **M** with respect to the cubic axes. In the bulk phase magnetic materials usually change their dimensions with **M** in order to minimize the total free energy ($\partial F_{tot}/\partial\epsilon_{ij}=0$), a phenomenon well known as magnetostriction (λ). Since the lattice distortions $\epsilon=\lambda$ due to magnetostriction of 3*d* metals typically lie between 10^{-4} and 10^{-6}, the involved ME energy, which is proportional to $B_i\lambda$ [compare Eq. (2)], is orders of magnitude smaller than the respective magnetocrystalline anisotropy (Fe:$F_{mcr}\approx K_1=5.48\times10^4$ J/m^3).

A different situation may be found in heteroepitaxial thin films, which — due to the misfit between the lattices of film and substrate — often are strained by a few percent ($\epsilon\approx10^{-2}$). The corresponding magnetoelastic energy ($F_{mel}\approx B_i\epsilon$) then reaches values of 10^4 J/m^3, which is of the same order of magnitude as F_{mcr}. In a previous study of 100 nm thick Fe(001) films,[6] however, we found that at stress values above 0.1 GPa ($\epsilon\approx10^{-3}$) the magnetoelastic coupling constant B_1 differs significantly from the respective bulk value and even changes sign when the stress exceeds 0.7 GPa (see also Fig. 2). Therefore in order to judge and predict the role of ME energy contributions in heteroepitaxial thin films properly the quantitative knowledge of the strain (or stress) dependence of the ME coupling constants is essential.

In this study we report on the ME coupling constants B_1 and B_2 of high quality epitaxial Fe(001) films in the thickness range of 2–100 nm and in a broad range of film stress (0.15-6 GPa). Irrespective of the film thickness we observe a strong dependence of B_1 and B_2 on the film stress, which both change sign at stress values in the GPa range. The key instrument for this study is our sensitive and versatile UHV-CBM (cantilever beam magnetometer),[7] which enables quantitative measurements of the intrinsic stress, the magnetization, magnetic anisotropies, and the ME coupling constants of thin films on one and the same sample. As will be shown the quantitative knowledge of all these properties, which were measured from all samples, actually is necessary to unambiguously verify the stress dependence of the ME coupling constants.

All Fe(001) films were prepared and investigated in UHV (base pressure $<10^{-10}$ mbar). Fe and Cr (see below) were electron beam evaporated from Knudsen-type tungsten sources at a deposition rate of 0.008 ± 0.001 nm/s. The pressure during deposition was better than 2×10^{-9} mbar. The MgO(001) substrates[8] were baked for several hours (4–10) at 1300 K in a stream of oxygen at atmospheric pressure

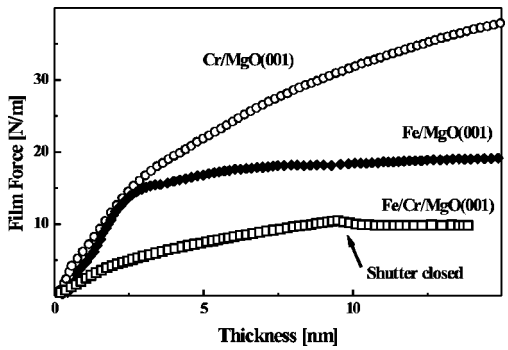

FIG. 1. Film forces (i.e., integral forces in films of unit width) measured during the deposition of Fe and Cr onto MgO(001) at 520 K as well as of Fe onto the Cr buffer layer.

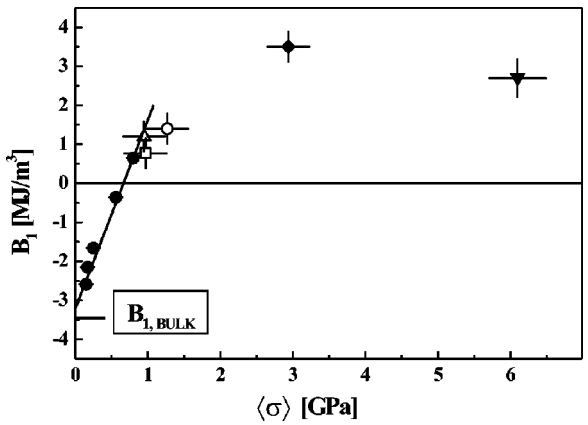

FIG. 2. Effective ME coupling constant B_1 of various Fe(001) films plotted vs the average film stress $\langle\sigma\rangle$, thick Fe/MgO(001) films of Ref. 6 (full circles); Fe/MgO(001), 5.5 nm, 520 K (full diamond); 2 nm, 520 K (full triangle); Fe/Cr/MgO(001), 25 nm Cr and 5.5 nm Fe at 520 K (open triangle), 50 nm Cr and 5.5 nm Fe at 540 K (open circle), 12 nm Cr at 520 K, and 10 nm Fe at 300 K (open square).

prior to mounting into the UHV chamber and shortly outgassed in UHV at 520 K before the film deposition. The quality of the substrate preparation was checked by low energy electron diffraction. For the measurements of B_1 the substrate orientation has to be chosen so that the [100] and [010] directions of the Fe(001) film are parallel to the length and width of the cantilever beam (orientation I); the difference in magnetostrictive stress upon saturation magnetization along [100] and [010] then is equal to B_1 ($\sigma^{[100]}-\sigma^{[010]}=-B_1$).[6,9] For B_2 the [110] direction of Fe(001) runs parallel to the long cantilever beam axis (orientation II); saturation magnetization along [110] and [010] yields $\sigma^{[110]}-\sigma^{[010]}=\frac{1}{2}B_2$. As an example, Fig. 3(d) illustrates the evolution of B_2 as function of the magnetic field strength along [110]. The average film stress was derived from the film forces (i.e., the integral forces in films of unit width), which were measured in real time during the deposition via the bending of the substrate. A typical force curve of Fe at a deposition temperature of 520 K is shown in Fig. 1. At this temperature Fe is known to grow layer by layer on MgO(001).[6,10,11] As discussed in more detail previously[6,12] the film stress at the beginning of deposition is dominated by the misfit of 3.5%, which at 520 K may give rise to tensile stress of 6.8 GPa. Correspondingly the film forces increase nearly linearly with thickness until at a thickness of 2-3 nm relaxation processes set in that suppress the further transmittance of the misfit strain. The experimental thickness exceeds significantly the theoretical critical layer thickness t_c at which strain relief by misfit dislocations becomes energetically favorable. With the formulas of Van der Merwe[13] and Matthews and Blakeslee[14] t_c is estimated to 0.85 nm or 1.25 nm, respectively. It is well known that the incorporation of misfit dislocations is a kinetically controlled process which may even be obstructed for (001) planes by the fact that the glide planes for dislocations in Fe are {110} and {112}.

Figure 2 summarizes the stress dependence of the ME coupling constant B_1 of different Fe(001) films; it includes also the previous results of 100 nm and thicker films (full circles).[6] Up to 1 GPa B_1 depends linearly on the film stress and changes sign at about 0.7 GPa, whereas B_1 of an extrapolated stress-free film (-3.2 M J/m^3) is in good agreement with the respective bulk value (-3.44 M J/m^3). Extending these investigations to low film thicknesses corroborates the results obtained with the thick films. The force curve of Fig. 1, however, illustrates the difficulty of how to prepare low-stress thin films on MgO(001), at which the misfit may induce tensile stress up to 6.5 GPa. To circumvent this dilemma we deposited a Cr buffer layer at 520 K epitaxially onto the MgO(001) substrates before preparing the Fe(001) films. Since Cr and Fe exhibit almost identical lattice parameters, low-stress Fe films may be obtained once the misfit strain has been relaxed by the Cr layer. The force curve of Cr/MgO(001) also shown in Fig. 1 indeed reveals that—analogous to Fe/MgO(001)—the misfit strain is transmitted only to a thickness of about 2–3 nm. At 15 nm the force curve has converged to a slope corresponding to residual stress of about 1.2 GPa, thus substantially reducing the strain imposed on the subsequently grown Fe films (see Fig. 1).[15] B_1 of the two 5.5 nm and the 10 nm thick ''low-stress'' Fe films prepared on the Cr templates indeed fit well into the series of B_1 values of the thick films. The 5.5 nm (full diamond in Fig. 2) and 2 nm (full triangle) thick Fe(001) films deposited directly onto the MgO(001) substrates, on the other hand, exhibit stress of 3 GPa and 6 GPa, respectively. With B_1 values of $+3.5$ M J/m^3 as well as $+2.6$ M J/m^3 these films indicate that at stress values higher than 1 GPa there are significant deviations from the linear behavior, which would imply considerably higher B_1 values of $+12$ M J/m^3 and $+28$ M J/m^3 at 3 GPa and 6 GPa, respectively.

It is worth noting that with respect to the other magnetic properties accessible to our CBM the Fe(001) films behave identically in the entire thickness range down to 2 nm (i.e., 14 ML). The rectangular hysteresis[7] loops measured with our UHV-CBM on substrates with orientation I [e.g., Fig. 3(a)] reveal that all of Fe(001) films presented in this study exhibit a distinct magnetic in-plane anisotropy. The experimental saturation magnetization M_S of 1.7–1.8 MA/m is in good agreement with the Fe bulk value of 1.76 MA/m. Furthermore all films exhibit the magnetic anisotropy of bcc Fe with easy magnetization axes lying along [100] and [010]. Therefore on substrates with orientation I the (measurable) remnant magnetization component parallel to the cantilever beam axis is zero when the magnetizing field is rotated at

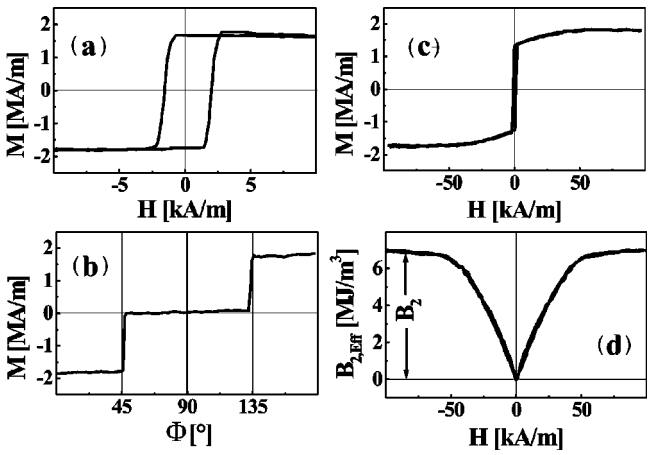

FIG. 3. (a) Magnetic hysteresis loop (magnetization **M** vs magnetic field **H** directed along Fe[100]) measured with our UHV-CBM: 5.5 nm Fe/MgO(001) deposited at 520 K. (b) Dependence of the remnant magnetization along Fe[100] on the in-plane angle of the magnetizing field indicating easy axis along [100] and [010]: 5.5 nm Fe/MgO(001) deposited at 520 K. (c) Magnetic hysteresis loop (magnetization **M** vs magnetic field **H** directed along Fe[110]): 50 nm Fe/MgO(001) deposited at 520 K. (d) Evolution of the ME coupling constant B_2 as function of the magnetic field strength along [110]: 50 nm Fe/MgO(001) deposited at 520 K.

angles of $45° - 135°$ within the film plane [e.g., Fig. 3(b)], because the remnant magnetization switches to the easy [010] direction when the magnetizing field is turned off. Figure 3(c) displays a hysteresis loop measured on substrates with orientation II. In this case a magnetic field strength of about 50 kA/m is necessary to saturate the films in the hard [110] direction, which in this geometry is parallel to the long cantilever beam axis. In remanence the magnetization rotates to the easy [100] or [010] directions with angles of 45° against the cantilever beam axis; therefore only a component $M_x = 1/\sqrt{2} M_S$ (≈ 1.2 MA/m) is detected.

Our results clearly demonstrate a strong dependence of B_1 on the film stress irrespective of the film thickness in the range of 2–100 nm. Sander *et al.* recently investigated B_1 of Fe/W(001) in the thickness range of 0.4–70 nm and obtained good agreement with the Fe/MgO(001) results in the low-stress regime (<1 GPa) (see, e.g., Ref. 16). At higher stress values they also observed a break down of the linear description, though with significantly smaller B_1 values ($\approx +1$ M J/m^3). As the ''high-stress'' films (>1 GPa) of this study are only 0.4-1 nm thin, a direct comparison with the present results is difficult as at film thicknesses below 1 nm a variety of—experimentally and/or physically based—finite size effects may gain in importance.

(i) Below 1 nm (i.e., 7 ML) surface roughness at the scale of a few atomic layers is no longer negligible. As the surface roughness allows for partial relief of the magnetostrictive stress, which is probed by the cantilever beam technique to measure the ME coupling, apparently smaller values of the ME coupling constants are determined.

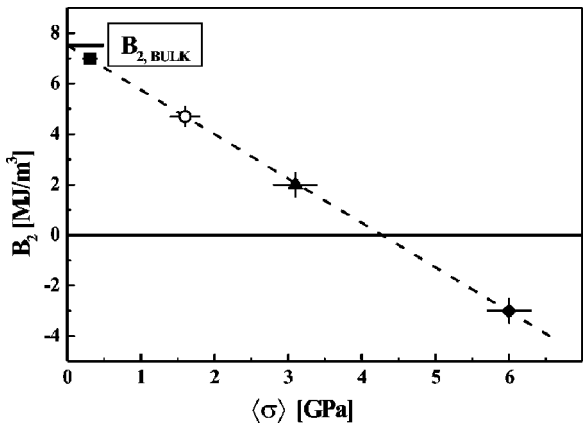

FIG. 4. Effective ME coupling constant B_2 of various Fe(001) films plotted vs the average film stress $\langle \sigma \rangle$, Fe/MgO(001): 50 nm, 520 K (full square), 4.5 nm, 435 K (full triangle), 2 nm, 520 K (full diamond); Fe/Cr/MgO(001), 11 nm Cr at 520 K and 4.5 nm at 300 K (open circle).

(ii) Investigation of 1 nm thick Fe(001) films on MgO(001) reveals that the magnetization is significantly reduced, which also gives rise to smaller B_1 values.

(iii) According to a study of Sun and O'Handley[17] the ME coupling constants of the surface itself may differ markedly from the corresponding bulk values. A more detailed study certainly is necessary in order to separate the contribution of these effects on the ME coupling.

Figure 4 summarizes the stress dependence of the ME coupling constant B_2 of differently stressed Fe(001) films deposited onto MgO(001) or Cr/MgO(001). Also B_2 exhibits a clear dependence on the film stress and changes sign when the stress exceeds 4 GPa. Compared to B_1, however, the stress dependence is less pronounced and remains linear up to stress values as high as 6 GPa. Notice that—as in the case of the ME coupling constant B_1—the extrapolated B_2 value of a stress-free film ($+7.5$ M J/m^3) again is in good agreement with the respective bulk value ($+7.62$ M J/m^3).

In conclusion, we have investigated the ME coupling constants B_1 and B_2 of epitaxial Fe(001) films in the thickness range of 2–100 nm and at stress values up to 6 GPa with our versatile UHV-CBM. For both constants we find a strong dependence on the film stress above 0.1 GPa, while stress-free films still exhibit bulk behavior. Whereas B_2 decreases linearly with the film stress up to 6 GPa, B_1 saturates after a steep increase at 2–3 GPa. Both constants change sign, B_1 at 1 GPa and B_2 at 4 GPa. The magnetization and the magnetic anisotropy of all films stay bulklike in the entire range of thickness and stress. Our study certainly underlines the importance of parallel and quantitative measurements of the ME coupling constants and the film stress as well as other magnetic properties (magnetization, anisotropies) in order to correctly interpret and control the magnetic properties of ultrathin heteroepitaxial films.

The authors wish to thank Professor K. H. Rieder for his support. The work was sponsored by the Deutsche Forschungsgemeinschaft (Sfb 290).

[1] P. Bruno and J.-P. Renard, Appl. Phys. **A49**, 499 (1989).
[2] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13 467 (1994).
[3] M. Farle, B. Mirwald-Schulz, A. N. Anisimov, W. Platow, and K. Baberschke, Phys. Rev. B **55**, 3708 (1997).
[4] G. Bochi, C. A. Ballentine, H. E. Inglefield, C. V. Thompson, and R. C. O'Handley, Phys. Rev. B **53**, R1729 (1996).
[5] We choose here the strain-dependent equations based on the Helmholtz free energy, because they are more familiar in the magnetic community, particulary concerning the ME coupling. In the case of thin films which are clamped at constant in plane strain by the substrate ($\epsilon_{11}, \epsilon_{22}, \epsilon_{12}$) actually the Gibbs free energy with the stress as natural variable is more appropriate ($\sigma_{33}=\sigma_{23}=\sigma_{31}=0$!). However, as both quantities are related Hooke's law, the stress can be easily calculated from the epitaxial strain and vice versa.
[6] R. Koch, M. Weber, K. Thürmer, and K. H. Rieder, J. Magn. Magn. Mater. **159**, L11 (1996).
[7] M. Weber, R. Koch, and K. H. Rieder, Phys. Rev. Lett. **73**, 1166 (1994).
[8] CrysTec GmbH, Köpenickerstrasse 325, D-12555 Berlin; email: crystec@aol.com
[9] P. M. Marcus, Surf. Sci. **366**, 219 (1996).
[10] K. Thürmer, R. Koch, M. Weber, and K. H. Rieder, Phys. Rev. Lett. **75**, 1767 (1995).
[11] R. Koch, *The Chemical Physics of Solid Surfaces*, edited by D. A. King and D. P. Woodruff, Growth and Properties of Ultrathin Epitaxial Layers Vol. 8 (Elsevier Science B.V., Amsterdam, 1997), p. 448–489.
[12] R. Koch, Appl. Phys. A (to be published).
[13] J. H. Van der Merwe, J. Appl. Phys. **34**, 123 (1962).
[14] J. W. Matthews and A. E. Blakeslee, J. Cryst. Growth **27**, 118 (1974).
[15] We remark that the strain relief of Cr at a thickness of 2–3 nm is less complete than in the case of Fe (compare Fig. 1). Actually the strain relief of Cr which is a kinetically controlled process could be further optimized by variation of the deposition conditions (growth temperature, deposition rate, etc.).
[16] D. Sander, Rep. Prog. Phys. **62**, 809 (1999).
[17] S. W. Sun and R. C. O'Handley, Phys. Rev. Lett. **66**, 2798 (1991).