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Surface magnetoelastic coupling coefficients of single-crystal fcc Co thin films

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We propose a method of analysis that allows indirect determination of magnetoelastic coupling coefficients for thin magnetic films. The method is based on a general phenomenological equation describing the dependence of the effective magnetic anisotropy energy density on film thickness for a thin cubic ferromagnetic film sandwiched between two nonmagnetic layers of the same material. We show that by fitting the measured effective anisotropy energy density with this equation using independently measured elastic strain in the film and the saturation magnetization M_s, we can extract the Néel surface magnetic anisotropy energy density K^s and either of the surface magnetoelastic coupling coefficients B_1^s or B_2^s depending on the film orientation. The model is applied to published data for fcc Co/Cu(111) superlattices. We find that $B_2^s = -23.5$ erg/cm^2 and K^s(Co/Cu)(111) = +0.47 erg/cm^2. While such large values of B^s relative to K^s are surprising, recent direct measurements of B^s in polycrystalline Ni films show these values to be reasonable.

I. INTRODUCTION

The atomic coordination at the surface of a solid is reduced compared to that in the bulk. This form of broken symmetry is reflected in a dramatic change in the electronic structure of the outermost layers of the solid which, in the case of a ferromagnetic material, can give rise to enhanced magnetic moments at the surface[1–4] and to a uniaxial magnetic surface anisotropy.[5–8] In the case of a thin ferromagnetic film where the surface to volume ratio is relatively large, the magnetic surface anisotropy often plays a crucial, if not the dominant role in determining the magnetization easy axis. Based on the Néel model, one also expects the magnetoelastic (ME) coupling coefficients to differ significantly between the bulk and the surface. These coefficients describe the extent to which externally applied or intrinsic strains contribute to the total magnetic anisotropy energy density. In fact, Sun and O'Handley[9] measured the surface ME coupling coefficient B^s of cobalt-rich and iron-rich amorphous alloys and found that B^s can be approximately three times the bulk value for the cobalt-rich alloy and half the bulk value for the iron-rich alloy. Very recently, Song, Ballentine, and O'Handley[10] have reported giant surface magnetostriction in polycrystalline NiFe/Ag/Si, NiFe/Cu/Si, and Ni/SiO_2/Si thin films where the effective ME coupling coefficients were found to diverge to positive values as the ferromagnetic film thickness was decreased below 4–6 nm. These observations suggest that knowledge of the surface ME coupling coefficients B_1^s and B_2^s may be as important as the knowledge of the magnetic surface anisotropy energy density K^s in order to fully understand the behavior of the magnetic anisotropy in ultrathin films. Unfortunately, these coefficients have not yet been reported in any single-crystal magnetic material and are completely omitted in phenomenological models attempting to explain the behavior of magnetic anisotropy in ultrathin films.

In the present paper, we propose a phenomenological model that describes the behavior with film thickness of the total effective magnetic anisotropy energy density of an epitaxial cubic thin film sandwiched between two identical cubic nonmagnetic metals with a (100) or a (111) orientation. We show that it is possible to obtain an indirect estimate of either $K_1^s(100)$ and B_1^s, or $K_2^s(111)$ and B_2^s for the film by measuring the effective magnetic anisotropy energy density, the saturation magnetization, and the average in-plane elastic strain of the film as a function of film thickness. We apply this model to published data[11,12] on fcc Co/Cu(111) superlattices. The fit allows us to estimate B_2^s and K_2^s(Co/Cu) for fcc single-crystal cobalt.

II. PHENOMENOLOGICAL MODEL

In a thin film, the most important contributions to the magnetic anisotropy energy are the magnetocrystalline (MC), the magnetostatic (MS), the magnetoelastic (ME), and the magnetic surface anisotropy energies. In the present discussion, we will neglect the MC anisotropy (well justified for fcc Co thin films where $f_{MC} \approx 0.1 f_{MS}$) and concentrate on the other three energies which often dominate in the case of a thin film. Let θ be the angle that the magnetization vector makes with the film normal. We use the standard convention for the sign of the magnetic anisotropy energy density: positive energies favor perpendicular magnetization and negative energies favor in-plane magnetization. The total magnetic energy density of a thin ferromagnetic film sandwiched between two identical nonmagnetic layers can be expressed phenomenologically in cgs units as follows:

$$f = f_{MS} + f_S + f_{ME} = \left[-2\pi M_s^2 + \frac{2K^s}{h} \right] \sin^2\theta + f_{ME}, \quad (1)$$

where $-2\pi M_s^2$ and $2K^s/h$ represent the MS and Néel magnetic energy densities, respectively, and h is the thickness of the ferromagnetic film. Strain gives rise to the ME energy density f_{ME} which can be expanded as a function of the strain components e_{ij} and the direction cosines α_1, α_2, and α_3 of the magnetization vector in the crystallographic coordinate system. For cubic materials, f_{ME} can be written to the lowest order in the following form:[13]

$$f_{\rm ME}=B_1(e_{11}\alpha_1^2+e_{22}\alpha_2^2+e_{33}\alpha_3^2)+B_2(e_{12}\alpha_1\alpha_2+e_{23}\alpha_2\alpha_3+e_{31}\alpha_3\alpha_1), \quad (2)$$

where e_{ii} refer to the strain along the crystallographic $\langle 100\rangle$ directions, e_{ij} ($i,j=1,2,3$ and $i\neq j$) refer to shear strains, and B_1 and B_2 are the respective first-order ME coupling coefficients. The strain experienced by an epitaxial film growing in a [100] direction on a (100)-oriented substrate is generally biaxial (e.g., misfit strain) and can be represented approximately by the following tensor in the crystallographic frame:

$$\varepsilon=\varepsilon_0\begin{pmatrix}1&0&0\\0&1&0\\0&0&\dfrac{-2\nu}{1-\nu}\end{pmatrix}\approx\varepsilon_0\begin{pmatrix}1&0&0\\0&1&0\\0&0&-1\end{pmatrix}. \quad (3)$$

Here, we have assumed that the film is elastically isotropic and that Poisson's ratio $\nu=\frac{1}{3}$ which are good approximations for most transition metals. By convention, $\varepsilon_0>0$ and $\varepsilon_0<0$ mean that the film is under tensile and compressive strain, respectively. Equation (3) shows that when the film is under in-plane biaxial tensile strain, it is also compressed along its normal, and vice versa. It is important to emphasize that the strain ε_0 is an average strain and that the local strain $\varepsilon(x,y,z)$ can vary significantly on an atomic scale especially in the film-substrate interface if misfit dislocations are present. Substituting the above strain tensor in Eq. (2) and keeping only angle-dependent terms leads to

$$f_{\rm ME}^{(100)}=2B_1\varepsilon_0\sin^2\theta. \quad (4)$$

For a thin film grown epitaxially with a [111] orientation on a (111)-oriented substrate, it has been shown that[14,15]

$$f_{\rm ME}^{(111)}=-\tfrac{3}{2}\lambda_{111}\sigma_0, \quad (5)$$

where λ_{111} is the saturation magnetostriction along the $\langle 111\rangle$ directions and where σ_0 is the average biaxial in-plane stress in the film. Using the relation $\lambda_{111}=-\frac{1}{3}B_2/c_{44}$ (Ref. 13) and assuming the ferromagnetic film is elastically isotropic, it can be easily shown that

$$f_{\rm ME}^{(111)}=2B_2\varepsilon_0\sin^2\theta. \quad (6)$$

Substituting Eqs. (4) and (6) in Eq. (1) for a [100]- and a [111]-oriented film, respectively, we get

$$f=K_{\rm eff}\sin^2\theta, \quad (7)$$

where

$$K_{\rm eff}^{(100)}=2B_1\varepsilon_0-2\pi M_s^2+\frac{2K_1^s}{h} \quad (8)$$

and

$$K_{\rm eff}^{(111)}=2B_2\varepsilon_0-2\pi M_s^2+\frac{2K_2^s}{h}. \quad (9)$$

Based on the Néel model[7] and as confirmed experimentally by Song, Ballentine, and O'Handley,[10] we can expand the first-order ME coupling coefficients B_1 and B_2 in the following form:

$$B_1=B_1^b+\frac{B_1^s}{h}, \quad (10)$$

$$B_2=B_2^b+\frac{B_2^s}{h}, \quad (11)$$

where the superscripts b and s refer to the bulk and surface coefficients, respectively. Substituting Eqs. (10) and (11) in Eqs. (8) and (9), respectively, we obtain

$$K_{\rm eff}^{(100)}=2\left(B_1^b+\frac{B_1^s}{h}\right)\varepsilon_0(h)-2\pi M_s^2+\frac{2K_1^s}{h}, \quad (12)$$

$$K_{\rm eff}^{(111)}=2\left(B_2^b+\frac{B_2^s}{h}\right)\varepsilon_0(h)-2\pi M_s^2+\frac{2K_2^s}{h}. \quad (13)$$

If we know the dependence of the strain ε_0 on film thickness h, we can deduce which linear combination of powers of h, in Eq. (12) or (13), should be used to fit the measured dependence of $K_{\rm eff}$ on h. From such a fit we can then get an indirect yet accurate value for the saturation magnetization M_s and either pair of surface energies B_1^s and K_1^s or B_2^s and K_2^s. Depending on the thickness dependence of the film strain, it may be possible also to get either of the bulk ME coupling coefficients B_1^b or B_2^b. At this point we would like to emphasize the fact that although it may be possible to fit the magnetic anisotropy data by omitting the B^s/h term, which is particularly true when the strain in the multilayer is independent of thickness as one can see from Eqs. (12) and (13), the physical significance of the magnetic anisotropy energy that one would extract from such a fit may remain questionable. We give an example to illustrate the above method.

III. MAGNETOELASTIC COUPLING IN EPITAXIAL (111) Co/Cu MULTILAYERS

Lee and co-workers have grown a series of Co/Cu superlattices on GaAs (110) substrates by molecular-beam epitaxy.[11,12] Their work is among the most complete and thorough in the literature on ultrathin films because it includes measurements of anisotropy energy density, strain, and saturation magnetization as a function of thickness. The Co thickness was varied from 5 to 40 Å while the Cu thickness was fixed at 25 Å. The total superlattice thickness was 1500 Å in all cases. They showed that the Co layers grow in a (111) orientation with fcc stacking. They have also measured the saturation moments in the various superlattices and shown that the average, 1241 emu/cm^3, is a good value for all thicknesses; therefore we take $f_{\rm MS}=2\pi M_s^2=9.7\times10^6$ erg/cm^3. Finally, they measured the strain in the Co layers and showed that it can be well fit by the following equation:

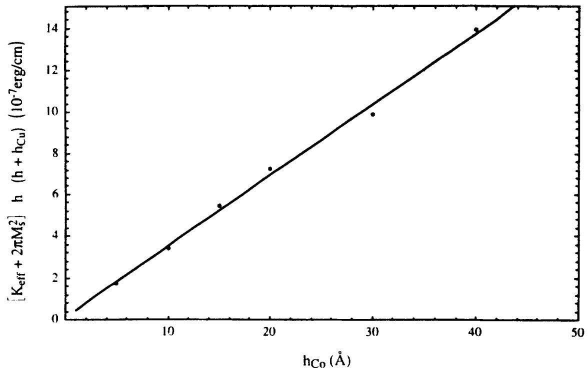

FIG. 1. $[K_{\rm eff}+2\pi M_s^2]h(h+h_{\rm Cu})$ as a function of Co film thickness. $K_{\rm eff}$ is the effective anisotropy energy density and M_s is the saturation magnetization of the Co films. The points represent measured data and were obtained from Ref. 11. The solid line is the linear fit to the data points using Eq. (16).

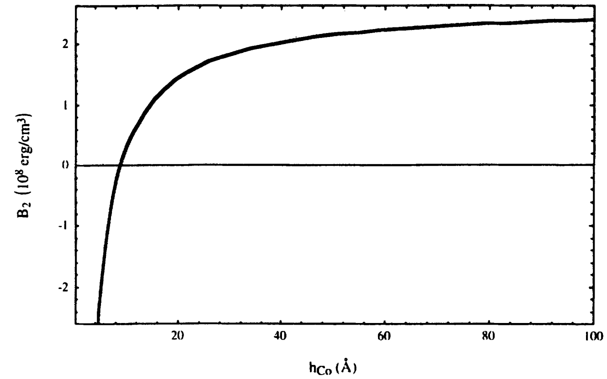

FIG. 2. Effective magnetoelastic coupling coefficient $B_2=B_2^b+B_2^s/h$ as a function of film thickness for fcc Co. B_2^s was obtained from the fit shown in Fig. 1.

$$\varepsilon_0=m\frac{h_{\rm Cu}}{h+h_{\rm Cu}}\,, \qquad (14)$$

where $h_{\rm Cu}=25$ Å and $m=1.9\%$ is the fcc Co-Cu lattice misfit. Substituting Eq. (14) in Eq. (13) and dropping the (111) superscripts, we obtain the thickness dependence of the effective anisotropy energy density for the (111) Co/Cu superlattices:

$$K_{\rm eff}=2\left(B_2^b+\frac{B_2^s}{h}\right)m\frac{h_{\rm Cu}}{h+h_{\rm Cu}}-2\pi M_s^2+\frac{2K_2^s}{h} \qquad (15)$$

which is more conveniently written as follows:

$$[K_{\rm eff}+2\pi M_s^2]h(h+h_{\rm Cu})=2(K_2^s+mh_{\rm Cu}B_2^b)h+2h_{\rm Cu}(K_2^s+mB_2^s). \qquad (16)$$

By plotting $[K_{\rm eff}+2\pi M_s^2]h(h+h_{\rm Cu})$ data vs h and fitting the points with a straight line, we can determine K_2^s(Co/Cu) and B_2^s for single-crystal fcc Co knowing B_2^b. Figure 1 shows the plot of the data points, obtained from Fig. 3 of Ref. 11, and the appropriate fit. We also used the measured value $2\pi M_s^2=9.7\times10^6$ erg/cm^3 in obtaining Fig. 1. The fit gives

$$[K_{\rm eff}+2\pi M_s^2]h(h+h_{\rm Cu})=0.3406h+0.1152 \quad (10^{-7}\ {\rm erg/cm}) \qquad (17)$$

where h is in Å in the right-hand side of the equation. Fujiwara, Kadomatsu, and Tokaunaga[16] extrapolated, from their data on fcc Co-Pd alloys, that for fcc Co, $\lambda_{111}=-6.7\times10^{-5}=-\frac{1}{3}\ B_2/c_{44}$ at $T=0$ K. Taking c_{44}(fcc Co)$=1.28\times10^{12}$ erg/cm^3,[17] one can extrapolate $B_2^b=2.6\times10^8$ erg/cm^3. This value is the best experimental value available for the bulk ME coupling coefficient of fcc Co.[7] Comparing Eqs. (16) and (17), we obtain K_2^s(Co/Cu)$\approx+0.47$ erg/cm^2 and $B_2^s=-23.5$ erg/cm^2. Using Eq. (11) and our estimate of B_2^s, we plot the dependence of B_2 (fcc Co) on Co thickness in Fig. 2. The plot indicates that B_2 decreases below 100 Å due to surface effects and becomes negative for $h\leqslant9$ Å. This result therefore questions the assumption, often encountered in the literature, that bulk Co ME coupling coefficients also apply for ultrathin Co films. The surface ME coupling coefficient was recently measured in polycrystalline Ni/SiO_2/Si thin films by a direct method[10] and was found to be approximately 20 erg/cm^2. Therefore, unlike magnetic surface anisotropy energy densities which are of the order of 0.5 erg/cm^2, surface ME coupling coefficients can be an order of magnitude greater and may strongly affect the value or even the sign of the effective ME coefficient.

Lee *et al.* pointed out that an expression $K_{\rm eff}(h)$ containing the bulk hcp Co MC energy, the bulk hcp Co ME coupling coefficients, and the measured value for $2\pi M_s^2$ does not fit the data of their fcc Co films. They limited themselves to showing that they can fit their anisotropy data by arbitrarily varying the ME coefficients and the MC energy in the expression of $K_{\rm eff}(h)$ of hcp bulk Co; they attributed no physical significance to their fitting parameters. In contrast, we have proposed a more general model appropriate to any epitaxial magnetic sandwich structure or magnetic multilayer that includes a surface magnetoelastic as well as a surface magnetic anisotropy term. When we fit their data with this model, we get physically plausible and meaningful results. This was only possible because they reported a careful measurement of the saturation magnetization and of the thickness dependence of the strain in their films.

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