Surface Magnetoelastic Coupling

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The surface magnetoelastic coupling coefficient B^s is determined for cobalt-rich and iron-rich amorphous alloys by measuring the field and strain dependence of the spin polarization of secondary electrons emitted from their surfaces. B^s is found to be approximately 3 times the bulk value for the cobalt-rich alloy and half the bulk value for the iron-rich alloy.

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The surface of a material is expected to have an electronic structure that reflects lower symmetry and reduced coordination relative to the bulk. Thus, all properties that derive from electronic structure should be different at the surface. The outermost layer of atoms in a transition metal generally relaxes to a position closer to the first subsurface layer because of lower coordination and surface charge redistributions. This surface relaxation can be as great as 9% (e.g., for V).[1] There is some evidence for enhanced magnetic moments[1-4] and much support for different magnetic anisotropy[5-7] at surfaces. It is expected, therefore, that the surface magnetoelastic coupling constants also would differ from those of the bulk.

The magnetoelastic (ME) coupling coefficients B_{ij} describe the extent to which strains contribute to the magnetic anisotropy energy density. They are defined from an expansion of the anisotropy energy density in powers of strain:

$$fa = K_{ij}\alpha_i\alpha_j + B_{ij}\alpha_i\alpha_j\varepsilon_{ij} + \cdots , \tag{1}$$

where K_{ij} are the magnetocrystalline anisotropy constants and α_i are the direction cosines of the magnetization. We define B_{ij} measured at the surface as B^s; it reflects the unique electronic structure there. Because $|B_{ij}| \approx E\lambda_s$, where E is Young's modulus ($E \approx 10^{11}$ N/m^2 for amorphous alloys) and λ_s is the saturation magnetostriction constant ($\lambda_s \approx 10^{-5}$), it is clear that strains of even 1% would result in very large anisotropy energy densities (10^4 J/m^3) *if bulk coupling parameters were to apply at the surface.* It is important, therefore, to be able to characterize the strength of the magnetoelastic coupling at a surface.

We report the first direct measurements of the surface magnetoelastic coupling coefficient B^s in two representative amorphous alloys. We have chosen amorphous alloys for these measurements because only one ME coefficient will be required. The strain derivative of the spin polarization of secondary electrons coming from the outermost few atomic layers of the sample is measured as a function of applied field. For amorphous $Co_{76}Cr_4B_{20}$ (with bulk magnetostriction $\lambda_s = -3.8\times10^{-6}$ and ME coupling coefficient $B = +1.8\times10^5$ J/m^3) we determine $B^s = +(6.0\pm0.2)\times10^5$ J/m^3. For the amorphous $Fe_{77}Cr_6B_{17}$ (characterized by bulk parameters $\lambda_s = +7\times10^{-6}$ and $B = -3.0\times10^5$ J/m^3) we find $B^s = -(1.6\pm0.2)\times10^5$ J/m^3.

Zuberek *et al.*[8] have used strain-modulated ferromagnetic resonance on Ni-C and Ni-Ag multilayers to show that the effective magnetostriction $\lambda_{\rm eff}$ due to *interfacial strain* varies strongly with the Ni layer thickness. They find $\lambda_{\rm eff}$ becomes less negative ($\lambda_s^{\rm bulk} \approx -36\times10^{-6}$) as the Ni thickness decreases. Mazumdar and Juretschke[9] have used the metallic field effect in 90%-Ni Permalloy thin films to measure the fractional change in anisotropy K with strain: $\eta = \partial(\ln K)/\partial\varepsilon \approx \lambda^s E^s/K$. They determined that the ME coupling at the surface is an order of magnitude less than in the bulk.

The present paper describes our measurements of *surface* magnetoelastic coupling B^s in amorphous $Co_{76}Cr_4B_{20}$ and $Fe_{77}Cr_6B_{17}$ using secondary-electron spin polarization. The secondary electrons, whose spin polarization (at E_K, above the low-energy, polarization-enhanced region) is measured as a probe of sample magnetism, come from a depth of approximately 1 nm. The magnetoelastic measurements described below derive from bending a thin ribbon of a magnetic sample so that its front surface is under pure uniaxial tension or compression.

In principle, we should treat the surface magnetoelastic problem in uniaxial or lower symmetry. In the present case, the single measurement to be made does not distinguish different magnetoelastic parameters, so we assume, as a first approximation, isotropic magnetoelastic behavior. Appropriate to this geometry we minimize the magnetoelastic plus elastic free energy density[10] and solve for the strain ε_{xx}, neglecting shear strains in the thin surface layer. With the definitions of Young's modulus $E = (C_{11}-C_{12})(C_{11}+2C_{12})/(C_{11}+C_{12})$ and the Poisson ratio $\nu = C_{12}/(C_{11}+C_{12})$, ε_{xx} can be expressed as

$$\varepsilon_{xx} = -(B/E)[\alpha_1^2 - \nu(\alpha_2^2+\alpha_3^2)] . \tag{2}$$

The α_i are the direction cosines of the magnetization relative to the axes defined in Fig. 1. Using $\alpha_1^2 = (M/M_s)^2 = m^2$ and, therefore, $\alpha_2^2+\alpha_3^2 = 1-m^2$, and taking $\nu = \frac{1}{3}$

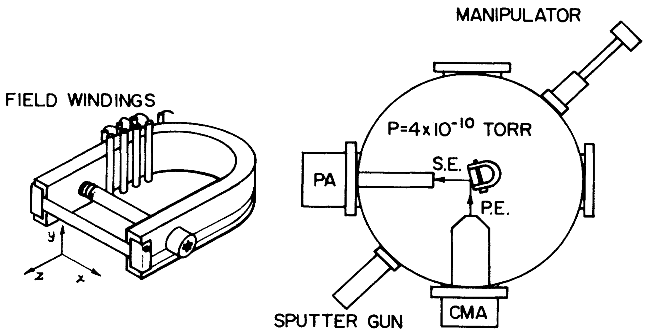

FIG. 1. Left: Flexible, externally adjustable yoke holding ribbon sample. Some field windings are shown; heater is not shown. Right: Schematic of UHV chamber showing primary electron (PE) source in Auger analyzer (CMA), secondary electrons (SE), and polarization analyzer (PA).

which is appropriate for the Co-rich amorphous materials studied here ($\nu=0.32$ is reported by Chou, Davis, and Narasimhan[11]) and is reasonable for the Fe-base alloy ($\nu=0.30$ is reported for $Fe_{80}B_{20}$),[12] we have

$$\varepsilon_{xx}=-(B/3E)(4m^2-1)\,. \tag{3}$$

We relate this imposed strain to the strain dependence of m by the thermodynamic relations which define the magnetostrictivity d

$$d=\mu_0\left(\frac{\partial M}{\partial\sigma}\right)_H=\left(\frac{\partial\varepsilon}{\partial H}\right)_\sigma \tag{4}$$

[Eq. (4) comes from the linear responses $dB=\mu^\sigma dH+d\times d\sigma$ and $d\varepsilon=s^H d\sigma+d\,dH$, with $s^H=(E^H)^{-1}$]. Integration of (4) gives

$$\varepsilon(H)-\varepsilon(0)=\int_0^H d\,dH=\int_0^H\frac{\mu_0}{E^H}\left(\frac{\partial M}{\partial\varepsilon}\right)_H dH\,. \tag{5}$$

It is $(\partial M/\partial\varepsilon)_H$ that can be measured in the bending experiment. For $\varepsilon_{xx}\gg\lambda_s$, $E(H)\approx E(0)$, i.e., there is no ΔE effect. In this case we can safely remove E^H from the integral in Eq. (5). From Eqs. (3) and (5), we have

$$-\frac{4B}{3}[m^2(H)-m^2(0)]=\mu_0\int_0^H\left(\frac{\partial M}{\partial\varepsilon}\right)_H dH\,. \tag{6}$$

Equation (6) allows a determination of the magnetoelastic coupling coefficient B even if the measuring field H is less than saturation. The method of imposed stress or strain is a common one for measuring ME parameters.[13]

Surface magnetization is probed to a depth of order 1 nm by measuring the spin polarization of secondary electrons emitted from the sample surface. It has been shown[14] that the spin polarization $P=(N^+-N^-)/(N^++N^-)$ of the true secondary electrons[2] at energies above about 10 eV is equal to the polarization of the valence electrons at the surface of the material, n_B/n_v. Here N^+ is the number of secondary electrons with positive spin component along a chosen axis, n_B is the valence-band magneton number n^+-n^-, $n_v=n^++n^-$ is the total number of valence electrons, and n^+ is the number of spin-up valence electrons. For kinetic energies below about 10 eV, the secondary-electron polarization is enhanced, probably due to a spin-dependent inelastic mean free path. We can measure or reject these electrons by the voltage we apply to a grid located between our extraction lens and drift tube. The spin polarization of the secondary electrons can be detected from the asymmetric spin-orbit scattering[15] $A=(N_L-N_R)/(N_L+N_R)$. The polarization of the electron beam is related to A by the asymmetric scattering efficiency S of the detector $S=A/P$. Thus we have for the surface magnetization (rejecting the low-energy, polarization-enhanced secondaries):

$$M^s=(N/V)n_B\mu_B=(N/V)\mu_B n_v A/S\,, \tag{7}$$

where $N/V=N_A\rho f/w$ is the number of magnetic atoms per unit volume, each with moment $n_B\mu_B$, N_A is Avogadro's number, ρ is the mass density, f is the fraction of the atoms present that are magnetic, and w is the molar mass. For our diffuse scattering polarimeter, $S=0.14\pm0.01$. Equation (7) has been used to determine surface magnetization in a variety of systems.[2,14,16]

We may now combine Eqs. (6) and (7) to give the magnetoelastic coupling coefficient at the surface, B^s:

$$B^s=-\frac{3D/4}{m^2(H)-m^2(0)}\int_0^H\left(\frac{\partial A}{\partial\varepsilon}\right)_H dH\,, \tag{8}$$

with $D=\mu_0 n_v\mu_B\rho f N_A/Sw$.

The surface of the sample to be probed by measuring the strain derivative of its secondary-electron polarization is bent *in situ* on its magnetizing yoke (Fig. 1). The ribbon is clamped at the ends of each tine so that it forms a flat surface across the open end of the yoke. Electrical windings are applied to the yoke so the sample can be magnetized. The sample can be heated to 300 °C by a separate resistance heater. A specially machined screw joins the tines of the yoke a few mm behind the flat face of the sample. Before being placed in the UHV chamber the curvature and displacement of the sample were optically calibrated. When the tines are drawn together by the screw, the ribbon is bent so that its front surface is either convex or concave. Surface strains up to 6×10^{-4} in increments of approximately 3×10^{-5} are achievable. The primary electron energy was 2 keV which is well above the knee of our secondary-electron yield versus primary energy curve.[2]

For the cobalt-based amorphous alloy Auger spectra showed the oxygen and carbon contents of the sputter-cleaned surface to be less than 10% of the boron content and the overall composition to be essentially equal to that of the bulk. The solid lines in Fig. 2(a) show typical $A(H)$ loops at zero strain. Here the energy-selecting grid was set at -18 V to reject all the polarization-enhanced secondaries. Extrapolation of the surface polarization to saturation using a $1/H$ approach law gives $P=13.6\%$ ($\mu_0 M_s^s=0.89$ T) for the Co-based amorphous alloy (92% of the *bulk* saturation magnetization). The dotted curve in Fig. 2(a) shows $A(H)$ at one value of compressive strain. (Several different values of strain were applied to each sample and the magnetoelastic coefficients were calculated from the slopes $\partial A/\partial \varepsilon$.) The increasing polarization in Fig. 2(a) indicates that λ^s is negative. The surface ME constant B^s is proportional to the change in area between two such curves with respect to strain [Eq. (8)]. B^s is found to be $(6.0\pm0.2)\times10^5$ J/m^3, more than 3 times the bulk value for the cobalt-based alloy (Table I).

For the iron-based amorphous alloy the Auger spectra showed the oxygen and carbon concentrations of the sputter-cleaned surface to be less than 3% of the boron content. However, the surface composition was enriched in boron relative to the bulk. (The surface composition of the fully annealed sample was calculated from Auger spectra, including relative sensitivity factors, to be $Fe_{51}Cr_5B_{42}$ with 1 at.% each of oxygen and carbon.) The decreasing polarization with compressive stress [Fig. 2(b)] is compatible with positive magnetostriction. We find a surface spin polarization $P=9.1\%$ ($\mu_0 M_s^s=0.71$ T or 63% of the bulk value) and $B^s=(1.6\pm0.2)\times10^5$ J/m^3, approximately half of the bulk value. Our results on this clean surface are valid for these conditions but the comparison with bulk values must be considered in light of the surface segregation which may be characteristic of amorphous iron-boron alloys.[2]

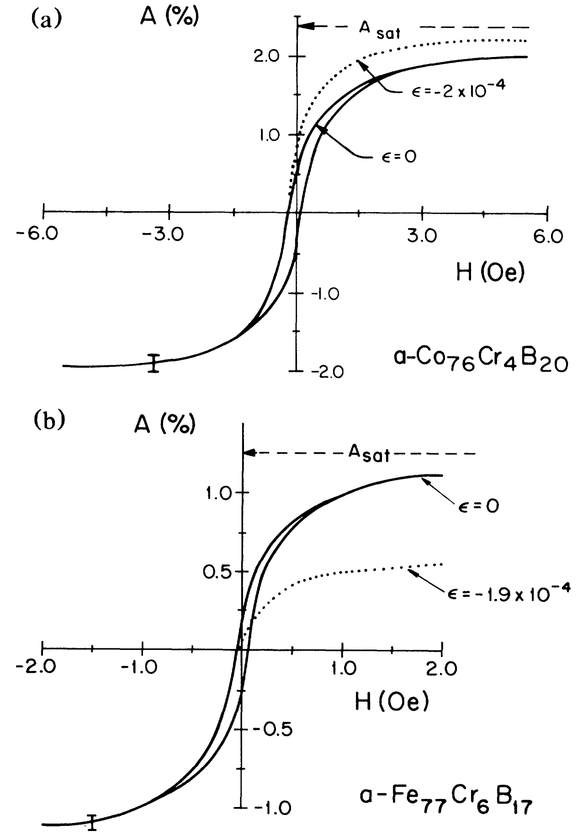

FIG. 2. Asymmetry vs applied field for zero strain (solid lines) and for modest strain (dashed lines) at front surface of ribbon (a) $Co_{76}Cr_4B_{20}$ and (b) $Fe_{77}Cr_6B_{17}$. Saturation asymmetry based on extrapolation of surface polarization data is indicated.

TABLE I. Properties of amorphous $Co_{76}Cr_4B_{20}$ and $Fe_{77}Cr_6B_{17}$.

	$Co_{76}Cr_4B_{20}$	$Fe_{77}Cr_6B_{17}$
Valence electrons per formula unit: n_v	7.68	7.03
Molar weight: w (g/mole)	49	48
Mass density: ρ (g/cm^3)	8.2 [a]	7.5
Saturation moment: n_B (μ_B/T atom)	1.1 [c,d]	1.01 [b]
Saturation magnetization: σ (emu/g)	93 [c,d]	118 [b]
$\mu_0 M_s$ (T)	0.955	1.11
Surface saturation magnetization: $\mu_0 M_s^s$ (T)	0.89	0.71
Young's modulus: $E^H=1/s^H$ (10^{10} N/m^2)	6.2 [e]	5.8 [e]
Bulk saturation magnetostriction: λ_s (10^{-6})	-3.8 [c]	$+7.0$ [b]
Bulk ME coefficient: B^b (10^5 J/m^3)	1.8	-3.0
Surface ME coefficient: B^s (10^5 J/m^3)	6.0 ± 0.2	-1.6 ± 0.2

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[b] A. Collins, Ph.D. thesis, MIT, 1987 (unpublished).

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[e] Values given are for Metglas 2605_s-3A ($Fe_{77}Cr_2B_{16}Si_5$) and for 2714A ($Co_{66}Fe_4B_{14}Si_{15}$) from Metglas Products catalog.

The depth probed by the secondary electrons in our experiment is difficult to specify accurately. Measurements[17] place it from 3 to 5 monolayers, so in all likelihood, in addition to measuring surface ME response, we are also measuring part of the bulk response with this method. Our measured values of B^s are, therefore, an average of B^s and the bulk ME coefficient B^b weighted by the effective depths probed. The true surface ME coefficients may differ more from the respective bulk values than our measurements indicate.

The surface magnetoelastic coupling coefficients of two different amorphous alloys have been measured to be markedly different from the corresponding bulk values. Thus, the calculated effects of strain on magnetic surface anisotropy should be viewed with skepticism unless the appropriate surface magnetoelastic coupling constant(s) have been explicitly measured. These measurements need to be extended to other materials and particularly to single crystals where their significance and implications for surface anisotropy may be more readily assessed and more widely applied. Such measurements are critical to the proper understanding of magnetism at surfaces and interfaces, and in ultrathin films.

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