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Sources of interface magnetization and interface anisotropy in Fe/Cu multilayers as revealed by thermal behavior

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Abstract

Sources on interface magnetization and anisotropy in Fe/Cu multilayers have been investigated via thermal behavior using DC magnetization and ferromagnetic resonance techniques. The samples were prepared by DC sputtering resulting in equal thickness elemental layers ranging in modulation wavelength from 1 to 30 nm. The interface anisotropy remains nearly constant at 0.32 erg/cm^2 through the temperature range 4–300 K, which is in stark contrast to large thermal effects observed in Ni-based systems, but is consistent with weak thermal variations of the crystal field in bulk Fe. Both the interface and volume Fe magnetizations follow Bloch $T^{3/2}$ behavior, but whereas the volume Fe coefficient matches the bulk value of $3.5 \times 10^{-6} K^{-3/2}$, the interface coefficient is 17 times greater. This indicates an interface Fe–Fe exchange which is much weaker than would be expected at a free Fe surface. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

In recent years there has been considerable interest in the properties at the interface between dissimilar magnetic materials. Preeminent among these have been the interfacial magnetization [1,2] and anisotropy [3,4]. Owing to reduced dimensionality and interfacial interactions in multilayer systems, magnetizations and anisotropies have been obtained which differ significantly from those observed in bulk Co, Ni and Fe. In order to understand and control these interfacial properties, one must investigate their sources. Much insight has been gained in this area by probing the material's thermal properties. The role of finite size and the interface on the Curie temperature and spin wave behavior have been studied both theoretically [5,6] and experimentally [7]. Co/Pt and Co/Au

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multilayers have been prepared to study the effects of miscibility on the interface magnetization of Co (Pt is miscible in Co, Au is not) [8]. A much stronger magnetization temperature dependence was observed in the Co/Pt than in the Co/Au. This was attributed to the higher degree of interdiffusion giving rise to a decreased Co–Co exchange. Mossbauer investigations [7] on variable thickness single Fe films revealed Bloch law ($1 - bT^{3/2}$) behavior in the magnetization temperature dependence for even the thinnest (3.4 Å) Fe layers, however, the spin wave parameter, b, scaled with $1/d_{Fe}$ and had a value more than six times larger than bulk for the thinnest Fe layer. The magnetization temperature dependence in Fe/Au multilayers [9] was interpreted in terms of a generalized Bloch law where both the temperature coefficient and exponent are fit to the data. The exponent was observed to increase from 1.1 to 1.5 (2D to 3D) with increasing number of bilayers and the coefficient in the 3D limit was observed to be approximately six times larger than expected for bulk Fe. The results were interpreted in terms of a simplified spin wave model with the introduction of an interlayer Fe exchange. Fe (10 Å)/Cr multilayers were grown [10] with Cr thickness of 10, 20 and 100 Å to provide AF, ferro and uncoupled Fe layers, respectively. The temperature dependence of the magnetization was then examined and found to exhibit temperature exponents of approximately 2, 1.5 and 1, for the three cases. A Heisenberg model consisting of 2D planes with interlayer coupling in the third dimension was sucessfully employed in analyzing the data. Comparison with magnetotransport results indicated that within an Fe layer, the interface had a different magnon temperature dependence from that of the interior of the layer. In a polarized neutron reflectometry study of ultrathin, epitaxial Fe layers in proximity to Ag, Au, Cu and Pd layers, an enhanced interface moment was observed in all cases [11]. A similar observation was reported employing a novel FMR technique [12]. This is in contrast to the present results, presumably owing to the greater level of interfacial intermixing in our polycrystalline samples. In addition, the above mentioned neutron results found only weak temperature dependence in the Fe magnetization. This also is in contrast with the present results, again presumably owing to interfacial structural differences.

Although the interface anisotropy has been studied extensively [3], relatively few investigations have addressed its thermal behavior. Strong temperature dependence of the anisotropy in Ni/Mo multilayers indicates Ni crystal field modified by interfacial strain as the source of the interfacial contribution [13]. Epitaxial Fe/Ag multilayers have been characterized by an interface anisotropy which varies with temperature as M^n, with n between 2 and 3 [14]. Studies of Co/Ag and Fe/Ag [15] and of Fe/Au [16] multilayers have revealed little or no temperature dependence in the interface anisotropy.

A previous report [17] on these present Fe/Cu samples at room temperature indicates anisotropy well characterized by first-order terms as was evidenced by equivalent values obtained from static and dynamic techniques. In addition, the magnetization was found to fit a model wherein only a single Fe monolayer at an interface experienced a reduced magnetization (30%) relative to that Fe in the interior of the layer. The present investigation addresses the sources of the interface magnetization and anisotropy by examining their thermal behavior.

2. Experimental details

Fe/Cu superlattices ($d_{Fe} = d_{Cu}$ = 5, 7, 10, 13, 20, 37, 75 and 150 Å) were prepared by DC magnetron sputtering onto ambient temperature sapphire and Si substrates. A Cu layer of 500 Å was grown as a base layer on which the superlattice, of total thickness $\sim$ 2700 Å was deposited. The base pressure was $2–4 \times 10^{-7}$ Torr, the Ar pressure during deposition was 2 mTorr and the sputtering rates were close to 5 Å/s.

The crystal structure as determined by high- and low-angle X-ray diffraction on these samples has been reported in detail elsewhere [18]. The samples are textured polycrystalline with the FCC Cu(1 1 1) and BCC Fe(1 1 0) oriented perpendicular to the film surface. For the samples used in this investigation (equal Cu and Fe layer thicknesses), neither the Cu nor the Fe deviate appreciably from the bulk

lattice parameters down to the lowest modulation wavelengths. Well resolved low-angle diffraction peaks indicate a well defined layer structure for all modulation wavelengths. Thermal scans of saturation magnetizations were taken with a SQUID magnetometer. Anisotropy values were obtained from FMR measurements at 9.2 GHz with the applied field in the plane of the film using a standard TE102 mode cavity-based spectrometer. The temperature was varied using an Oxford Instruments flow-through cryostat.

3. Results and discussion

3.1. Magnetization

Square in-plane M versus H hysteresis loops were observed and coercivity values were less than 10 Oe for samples with $d_{Fe} > 10$ Å [17]. For samples with smaller modulation wavelengths there was an increase in both the coercivity and the field required to saturate the in-plane moment. The Fe volume used in the magnetization calculation is determined utilizing the magnetic layer thickness obtained by X-ray analysis and careful measurements of the sample area.

Fig. 1 shows the magnetization temperature dependence as represented by four of the eight samples investigated. The increasingly strong temperature dependence with decreasing Fe layer thickness, d_{Fe}, is particularly striking. One should note that the absolute not reduced magnetization is plotted and that even the thinnest layer magnetizations reach values of approximately 1600 emu/cm^3 at low temperatures. (This low-temperature magnetization is lower than that of bulk Fe as has been observed in other studies of sputtered Fe/Cu superlattices [19–21] and is consistent with Mössbauer studies [18].) Furthermore, the Mössbauer results showed that the monolayers closest to the interface had a reduced hyperfine field relative to the center of the layer. Motivated by this and by the observed (Fig. 1) magnetization dependence upon d_{Fe} at a given temperature, the data is analyzed using a model which assumes a reduced magnetization M_i in the monolayer of Fe adjacent to each Cu interface and a bulk-like magnetization M_b in

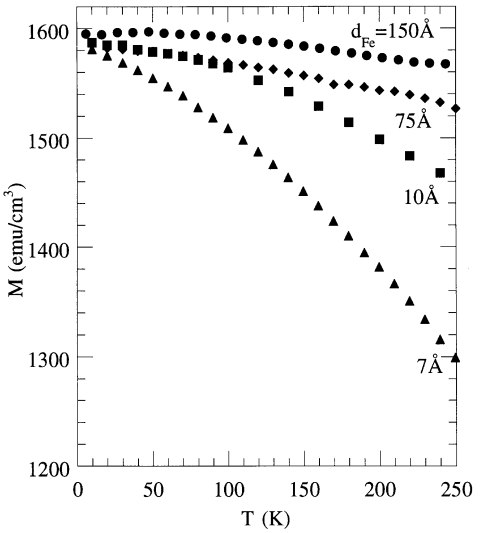

Fig. 1. Magnetization versus temperature for $d_{Fe} = 150$ Å (circles), 75 Å (diamonds), 10 Å (squares) and 7 Å (triangles) samples.

the interior of an Fe layer. Therefore, the measured magnetization of a given sample at a given temperature will be given by

$$M_{meas} = \frac{M_b(d_{Fe} - 2d_{ML}) + 2M_i d_{ML}}{d_{Fe}}, \quad (1)$$

where d_{ML} is the thickness of a monolayer of Fe (approx. 2 Å). One then does a linear fit of the measured magnetization as a function of the inverse Fe layer thickness at each temperature. The intercept yields M_b and the

$$\text{slope} = (M_i - M_b)2d_{ML} \quad (2a)$$

from which one can obtain M_i. The temperature dependence of M_b and M_i thus extracted is shown in Fig. 2. The interior magnetization M_b is observed to follow the low-temperature Bloch model

$$\frac{M(T)}{M(0)} = 1 - bT^{3/2} \quad (2b)$$

with the spin wave parameter $b_b = 3.5 \times 10^{-6}K^{-1.5}$, which is in excellent agreement with the value of $3.4 \times 10^{-6}K^{-1.5}$ obtained for macroscopic sized samples of bulk Fe [22]. Therefore, one observes that the interior of an Fe layer is described

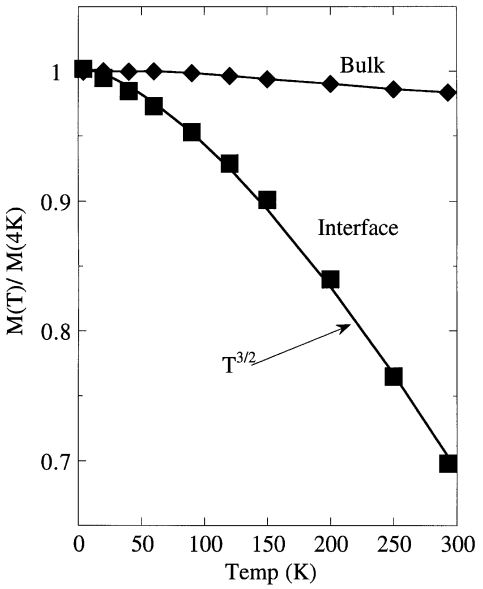

Fig. 2. Reduced interface (squares) and bulk (diamonds) magnetization versus temperature. Error bars are comparable to the size of the markers and are estimated from uncertainties in the SQUID signal. The solid line is a fit to the Bloch Law model.

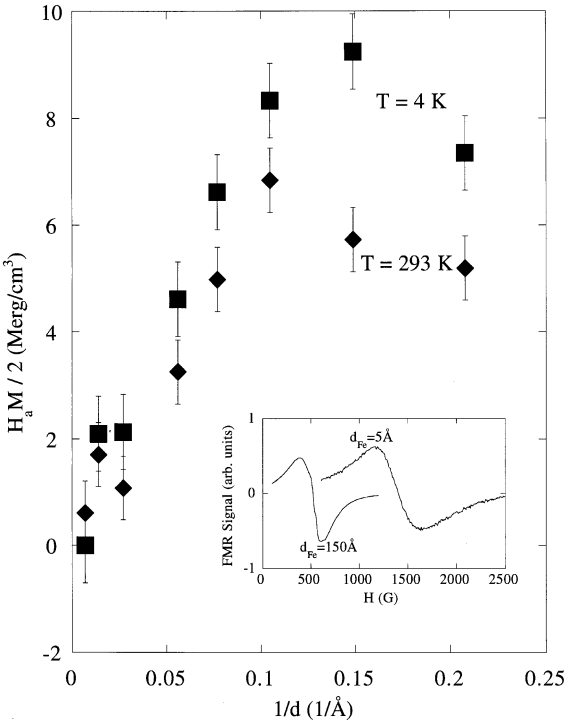

Fig. 3. Intrinsic anisotropy energy as a function of inverse Fe layer thickness. (See model in text.) Error bars are estimates obtained from uncertainties in magnetization and resonance position. Note deviation from linearity for the thinnest samples.

by both the expected temperature dependence and the expected spin wave prefactor. Building upon the understanding obtained from analysis of the film interior, we turn our attention to the interface. The interface magnetization is also observed to follow a Bloch model, but with a spin wave prefactor $b_i = 60 \times 10^{-6} K^{-1.5}$, which significantly exceeds the factor of two, increase relative to bulk expected for the free surface of a semi-infinite ferromagnetic solid [23]. It is important to note that, since the interface magnetization approaches the bulk value as the temperature is decreased, one can conclude that the Fe atoms at the interface are not magnetically 'dead', but rather possess approximately bulk Fe moments. It should be pointed out that our approach is equivalent to that employed by Korecki et al. [7], wherein a 'b' is obtained for each sample and b_i and b_b are extracted from a plot of b versus $1/d_{Fe}$.

3.2. Anisotropy

In the absence of in-plane anisotropy and with the external field applied in the film plane, the FMR resonance condition is given by

$$\omega = \gamma[H(H + D)]^{1/2}, \tag{3}$$

where ω is the experimental microwave frequency, H is the applied field at resonance and D is the total effective uniaxial anisotropy field. This anisotropy can be written as a linear combination of the shape anisotropy and the intrinsic anisotropy H_a according to

$$D = 4\pi M_s - H_a. \tag{4}$$

$H_a > 0$ ($H_a < 0$) implies an easy axis normal to (coincident with) the film plane.

FMR spectra were obtained with the field parallel to the film plane for each sample as a function of temperature. Large absorption signals were observed, which systematically shift to higher fields for decreasing modulation (Fig. 3, inset). D values are extracted using Eq. (3) via the measured value

of H (applied field where the spectrum crosses background), and the microwave frequency. In a fashion similar to the magnetization treated above, contributions to the intrinsic anisotropy are expected to arise from both volume (crystalline) and interface effects averaged over the magnetic layer. The anisotropy energy density is then commonly written as:

$$\frac{H_a M_s}{2} = K_v + \frac{2K_s}{d_{Fe}}, \tag{5}$$

where K_v is the crystalline anisotropy and $2K_s/d_{Fe}$ is the interface anisotropy treated in the homogeneous magnetization approximation [24]. Fig. 3 contains this energy density plotted as a function of inverse Fe layer thickness for the temperature extremes measured. The M_s values used are those for the specific sample at the stated temperature. Note that positive energy densities indicate the intrinsic anisotropy has its easy axis normal to the plane surface. The data in Fig. 3 display linear behavior in the thicker Fe layer regime. Fitting this linear region to Eq. (5) gives K_v approximately equal to zero and $K_s = 0.32 \pm 0.04\ \mathrm{erg/cm^2}$ at room temperature. The negligible K_v is consistent with the small crystal fields in bulk Fe [25] and K_s is in excellent agreement, in both sign and magnitude ($0.29\ \mathrm{erg/cm^2}$), with previous measurements of Fe(1 1 0)/Cu multilayers [26]. Since values as high as $0.62\ \mathrm{erg/cm^2}$ have been reported in epitaxial Fe(0 0 1) on Cu [4], it is quite clear that the interface anisotropy is dependent upon crystalline orientation and interfacial structure. The value for K_v remains zero (within error bars) with decreasing temperature and K_s exhibits only weak temperature dependence as is shown in Fig. 4. This is in contrast with the strong temperature dependence of the interface anisotropy observed in Ni multilayers [13], but is consistent with the rather weak temperature dependence (below room temperature) of the crystal field in bulk Fe [25].

Eq. (5) assumes a purely two-dimensional magnetic interface anisotropy in which the magnetization changes abruptly at the interface. However, as can be seen in Fig. 3, this model breaks down for thinner Fe layers, which most likely results from the finite width of the interface [13,18,27–29]. A more realistic model would characterize the interface as having an effective interface width which penetrates a distance L into the film. H_a would then scale inversely with the Fe thickness down to an Fe thickness of $2L$ and would then be a constant. At room temperature, this crossover occurs for the Fe thicknesses of ≈ 10 Å which corresponds to approximately two Fe monolayers at each interface. This result is consistent with the room temperature Mössbauer results on similarly prepared Fe/Cu multilayers [18]. It is interesting to note that this crossover decreases to ≈ 7 Å as the temperature is lowered to 4 K.

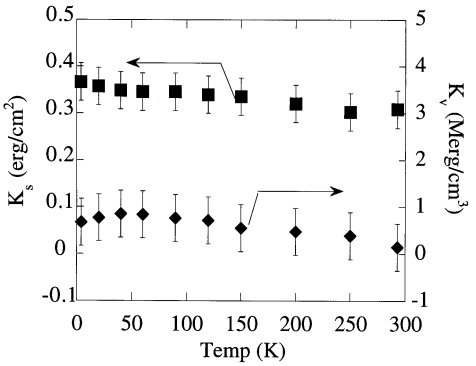

Fig. 4. Interface (squares) and bulk (diamonds) anisotropies as a function of temperature.

3.3. Comparison between magnetization and anisotropy

How can one understand the weak temperature dependence of the interface anisotropy, and the strong temperature dependence in the interface magnetization? Since the interface anisotropy likely has its source in the crystal field, it is primarily determined by the interface chemical structure, which one does not expect to change dramatically below room temperature. This combined with the fact that the crystal field of Fe depends weakly upon temperature, leads one to expect the interface anisotropy to also exhibit weak temperature dependence. On the other hand, the interfacial chemical structure is undoubtedly characterized by intermixing of Fe and Cu atoms at the monolayer level. This intermixing has the effect of decreasing both the Fe–Fe coordination number and the

exchange integral, each of which contributes to a decrease in the interfacial spin wave stiffness constant and consequently an increase in the interface spin wave parameter (b). As mentioned earlier, similar thermal effects arising from interfacial intermixing have been reported in a comparison between Co/Pt and Co/Au multilayers [8].

4. Summary

In conclusion, sources of interfacial of magnetization and anisotropy in a series of Fe/Cu multilayers have been characterized according to their thermal properties. The strongly temperature-dependent interface Fe magnetization is characterized by a full Fe atomic moment, but with a significantly reduced interatomic exchange relative to the interior of the layer. The weakly temperature-dependent interface anisotropy is consistent with the thermal behavior of bulk Fe crystal fields.

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