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Interface magnetism and superparamagnetism of Co/Cu multilayers

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The magnetic properties of Co/Cu multilayers were investigated from 5 to 380 K and analyzed in terms of the interface magnetism and mean-field model. The interface, which is about 1.2 Å thick, is nonmagnetic at room temperature and becomes magnetic at 5 K with the average magnetization of 40% of the pure Co magnetization. The samples of X Å Co/10 Å Cu behaved superparamagnetically as X ranged from 4 to 6.5 Å and did not show superparamagnetic behavior for thinner or thicker Co layer thickness.

I. INTRODUCTION

Many magnetic phenomena in Co/Cu multilayers, such as the reduction of magnetization at interfaces,[1] the magnetic anisotropy,[2] and interlayer coupling[3,4] have been of great interest in recent years. For the multilayered structure, the interfaces play a crucial role in determining the magnetic behavior. In order to understand better the effect of interface magnetism, we deal with structural and magnetic properties, in particular the temperature dependence of interface magnetism which is analyzed in terms of a magnetic two-phase model involving Co and a mean-field Co-Cu alloy interface.

II. EXPERIMENT

Co/Cu multilayers with the form of X Å Co/10 Å Cu ($X=2.2,3,4,5,6.5,10,20,40,80,160$) were fabricated by the dc magnetron sputtering with the sputtering rate of ~ 1 Å/s. The structure was characterized by x-ray diffraction and the magnetic properties were studied with the vibrating sample and SQUID magnetometers from 5 to 380 K.

III. STRUCTURE

The small-angle x-ray diffraction revealed that the peaks corresponding to layered structure appeared at the right positions [see Fig. 1(a)]. Only the first order peak was observed for $X \leqslant 5$ in our samples and a small second order peak was found for $X=10$, and the third and fifth order peaks were observed for $X=40$ and 80, respectively. The large-angle x-ray diffraction [see Fig. 1(b)] showed that both Cu and Co have the fcc structure, and it was found that the diffraction peak positions of the multilayer are between the positions of pure fcc Co and Cu materials.

IV. MAGNETIC PROPERTIES

A. Temperature character of interface magnetism

One of the interesting problems is to understand the interface magnetism of such multilayers. Since Co and Cu are immiscible, the interface may be expected to have a rather sharp boundary. Assuming the interface thickness is Δd and its average magnetization is $\langle\sigma_{\text{int}}\rangle$, the interface magnetism can be analyzed as follows.

The Co layer magnetization in the presence of the interface, whose thickness is Δd, can be expressed as

$$\begin{aligned}\tilde{\sigma}'_{\text{Co}} &= \text{moment of Co layer/mass of Co layer}\\ &= [\tilde{\sigma}_{\text{Co}}(d_{\text{Co}}-2\Delta d)+2\Delta d\langle\sigma_{\text{int}}\rangle]/d_{\text{Co}}\\ &= \tilde{\sigma}_{\text{Co}}\left(1-k\frac{2\Delta d}{d_{\text{Co}}}\right),\end{aligned} \tag{1}$$

where d_{Co} is the nominal thickness of Co layer, σ_{Co} is the magnetization of pure Co film, and

$$k=\frac{\sigma_{\text{Co}}-\langle\sigma_{\text{int}}\rangle}{\sigma_{\text{Co}}}. \tag{2}$$

If the interface layer is nonmagnetic, then $\langle\sigma_{\text{int}}\rangle=0$ and Eq. (1) reduces to[1,5]

$$\sigma'_{\text{Co}}(\Delta d)=\sigma_{\text{Co}}\left(1-\frac{2\Delta d}{d_{\text{Co}}}\right). \tag{3}$$

Equation (1) indicates that the plot of $\sigma'_{\text{Co}}(\Delta d)$ vs $1/d_{\text{Co}}$ should be a straight line if this model is reasonable.

When $T=300$ K, the experimental data and the fitted curve based on Eq. (3), i.e., $\langle\sigma_{\text{int}}\rangle=0$, is shown in Fig. 2.

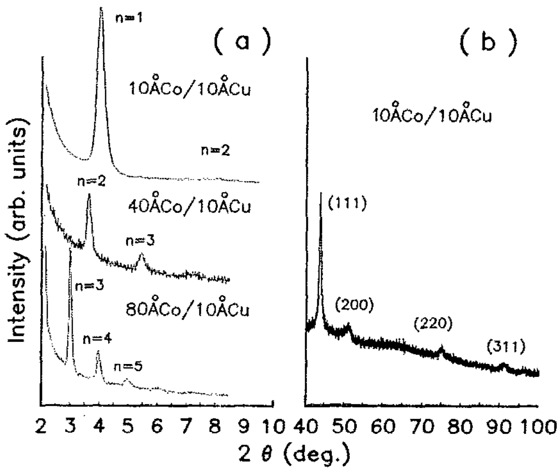

FIG. 1. Co $K\alpha$ small-angle (a) and large-angle (b) diffraction intensity as a function of 2θ.

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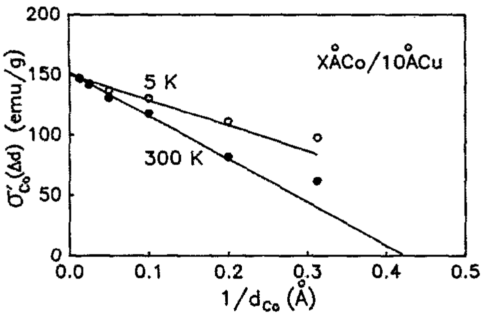

FIG. 2. Spontaneous magnetization σ_{Co} (emu per Co mass) as a function of ($1/d_{Co}$) for X Å Co/10 Å Cu at 300 and 5 K. d_{Co} is the nominal Co layer thickness.

The interface thickness Δd determined from the intercept with the x axis is 1.2 Å. The agreement between the experimental data and fitting curve is very good until $1/d_{Co} = 0.2$ ($d_{Co} = 5$ Å).

When $T = 5$ K, the interface is magnetized which will be discussed in more detail in Sec. IV B. The experimental data and the fitted curve based on Eq. (1) with the parameters of $k = 0.6$, $2\Delta d = 2.4$ Å and $\sigma_{Co} = 152(\text{emu}/g_{Co})$ is also shown in Fig. 2. Then Eq. (2) can be rewritten as

$$\langle\sigma_{\text{int}}\rangle = \sigma_{Co}(1-k) = \sigma_{Co}(1-0.6) = 0.4\sigma_{Co}, \qquad (4)$$

i.e., the average of the interface magnetization at 5 K is 40% of the pure Co magnetization.

B. Origin of the temperature dependence of magnetization

The magnetic moment of Co/Cu multilayers originates from the Co layers and their interfaces. The inner part of Co layer can be regarded as the pure Co region and the interface may be treated as a Co_XCu_{1-X} alloys with $X \simeq 0.5$. Since the magnetization of pure Co is almost independent of temperature, the temperature dependence of multilayers comes from the interface magnetism. Using the mean-field model,[7,8] the magnetization $\sigma(X,T)$ of Co_XCu_{1-X} can be calculated as follows:

$$\sigma(X,T) = N\mu_B X g \bar{S}(X,T), \qquad (5)$$

$$\bar{S}(X,T) = S B_S(g\mu_B S H(X,T)/k_B T), \qquad (6)$$

$$H(X,T) = 2(JZX\bar{S}(X,T)/g\mu_B), \qquad (7)$$

where N is the total number of atoms per unit volume, μ_B is the Bohr magneton, g is the gyromagnetic factor of Co, $\bar{S}(X,T)$ and S are the effective spin at room and zero temperatures, respectively. J is the exchange constant between Co-Co pairs. $H(X,T)$ is the effective internal field. K_B is the Boltzmann constant. B_S denotes the Brillouin function. Parameters S and J are adjusted by optimally fitting the calculated $\sigma(X,T)$ to the experimental data over a wide range of composition X and temperature T. Z is the maximum number of nearest Co neighbors. In the thick Co_XCu_{1-X} films, Z is equal to 12 and in the monolayer case, Z is equal to 6 which is close to the Z value in the interfaces. Figure 3(a) shows the calculated $\sigma(X,T)$ curves for $T = 5$ and 300 K with $Z = 12$ and the experimental data, and they agree with each other reasonably well. Figure 3(b) shows the calculated magnetization curves with $Z = 6$ simulating the Co configuration in the interfaces.

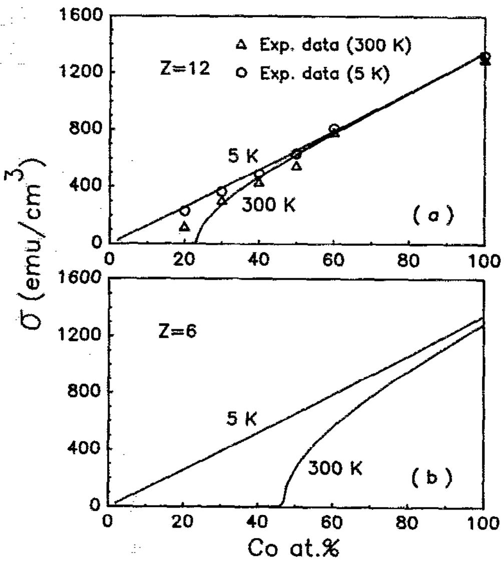

FIG. 3. A comparison between the calculated and experimental magnetization curves for thick Co_XCu_{1-X} films (a) and the simulating magnetization curves for the interfaces (b).

It is seen (i) when $X \simeq 0.4$–0.5, the Co_XCu_{1-X} alloy is magnetically disordered at 300 K, and its magnetization is ~40% of pure Co at 5 K [Fig. 3(b)]. (ii) The magnetization of pure Co, i.e., $X = 1$, almost does not change its value as T decreases from 300 to 5 K for the thick Co film [Fig. 3(a)], and there is only little change for the Co monolayer or interface [Fig. 3(b)].

C. Anisotropy

The anisotropy of Co/Cu multilayers can be investigated using the following equation[8]

$$\lambda K'_u = 2K_i + [K_v - 2\pi\sigma_{Co}^2]d_{Co}, \qquad (8)$$

where λ is the bilayer thickness, d_{Co} is the Co layer thickness, σ_{Co} is the Co layer magnetization in the presence of the interface, and K'_u, K_i, K_v are the measured, interface, and volume anisotropy, respectively.

Figure 4 shows the experimental results for X Å Co/10 Å Cu at 300 and 5 K. The following features are worth mentioning. (i) The $\lambda K'_u$ curve at 5 K is only shifted down slightly from that at 300 K. The temperature effect is very weak. (ii) The "downshift effect" is larger in thinner Co layer region than in thicker Co layer region, because the interface magnetism, which shows strong temperature dependence, plays a more important role in multilayers with thinner Co layer. (iii) $K_i \simeq 0.12$ (erg/cm^2) for both $T = 300$ and 5 K. Assuming that the interface contains only a single atomic layer, the interface anisotropy per Co ion is equal to $K_i/n^{2/3} = 0.12/(8.5\times10^{22})^{2/3} = 6\times10^{-17}$ (erg/Co ion). (iv) The volume anisotropy determined

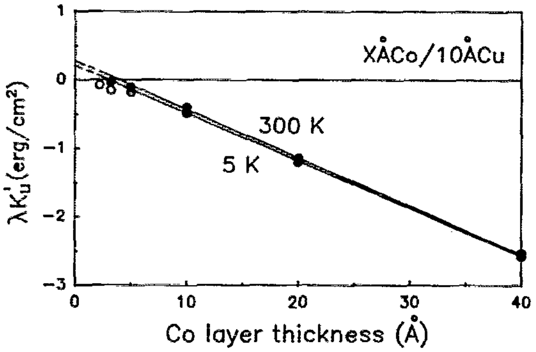

FIG. 4. Dependence of λK_u on Co layer thickness for X Å Co/10 Å Cu at 300 and 5 K.

from the curve slope is $K_v=4.4\times10^6$ (erg/cm^3). Then the volume anisotropy per Co ion is equal to $K_v/n=4.4\times10^{22}/8.5\times10^{22}=5\times10^{-17}$ (erg/Co ion). (v) It is well known that the anisotropy of a magnetic atom at the interface is usually larger than that at the inner layer, because the structural symmetry is broken at the interface. However in the Co/Cu multilayers, the volume anisotropy per Co ion (5×10^{-17} erg/Co ion) is roughly equal to the interface anisotropy per Co ion (6×10^{-17} erg/Co ion). Therefore, the symmetry breaking effect is very weak in such multilayers. (vi) We notice that the thermal expansion is 12×10^{-6} for Co and 16.6×10^{-6} for Cu.[9] Their difference reaches ∼30% which should make larger thermal stress between the Co and Cu layers as the temperature goes down from 300 to 5 K. However, Fig. 4 shows there is no remarkable change in the anisotropy and this implies that the stress-induced anisotropy is not an important source of the anisotropy.

D. Superparamagnetism

We have reported in our previous paper[10] that the sample 4 Å Co/11.5 Å Cu behaved superparamagnetically. Recent work reveals the samples of X Å Co/10 Å Cu show superparamagnetic properties for X between 4 and 6.5 Å and did not show superparamagnetic properties for thinner or thicker Co layers. It is seen in Fig. 5 that for 5 Å Co/10 Å Cu, all the datum points at 200, 250, 300, and 350 K are superimposed on one curve which is expected for superparamagnetic particles. By contrast for 3 Å Co/10 Å Cu and 10 Å Co/10 Å Cu samples, the datum points deviate distinctly from one curve. A possible reason is that in the 10 Å Co/10 Å Cu sample, the continuous Co layer shows ferromagnetic properties; and in the 3 Å Co/10 Å Cu sample, the Co cluster size is very small and the surface magnetization or oxidation gives the appreciable effect on magnetic properties which does not lead to showing the classical superparamagnetic behavior.

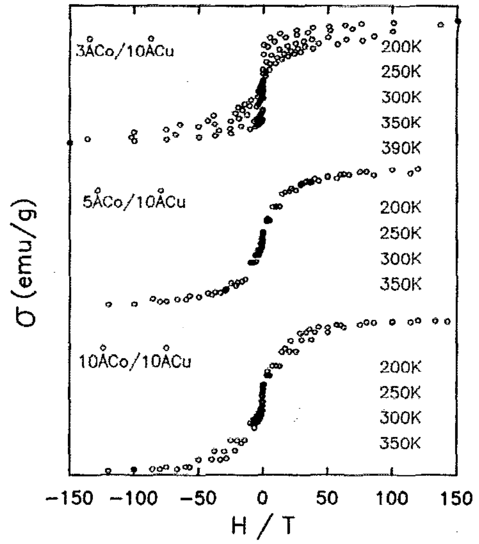

FIG. 5. Magnetization for X Å Co/10 Å Cu ($X=3$, 5, 10) as a function of (H/T).

V. SUMMARY

The magnetic properties can be analyzed in terms of the interface magnetism with the mean-field model. The interface, which is about 1.2 Å thick, is nonmagnetic at room temperature and becomes magnetic at 5 K with the average magnetization of 40% value of the pure Co magnetization. The stress-induced anisotropy appears not to be important and the magneto-crystalline anisotropy is the major source of the anisotropy.

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