Correlations Among Sputter Pressure, Thickness, and Coercivity in Al/Co/Cu Magnetic Thin Films Sputter-Deposited on Si(001)

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***Abstract*—We demonstrate the effect of sputter gas pressure and film thickness, d, on the coercivity of Co in Al/Co/Cu sputter-deposited on Si(001). Increased sputter gas pressure produces increased rms roughness and increased O content in our films. The deposited Co thickness at which the onset of ferromagnetism is first observed, d_c, increases with sputter gas pressure. Above this thickness, $d > d_c$, the coercivity increases with increasing Co thickness. For film thickness $d \gg d_c$, the coercivity is thickness independent. We show that coercivity is directly controllable at technologically relevant thicknesses by regulating sputter-deposition parameters.**

***Index Terms*—Cobalt, coercive force, rough surfaces, sputter deposition.**

I. INTRODUCTION

T ULTRATHIN ($d < 5$ nm) magnetic film thicknesses, surface roughness or interface roughness significantly influence the coercivity, making the characterization of these films particularly relevant. Though others have examined coercivity, thickness, and roughness at the same time in evaporated films [1], [2], our emphasis in this paper is studying correlations among deposited Co thickness, d; Ar sputter pressure, P_{Ar}; and coercivity, H_c; in Al/Co/Cu sputter-deposited thin films on Si(001) as sputter deposition is preferred in commercial applications [3]. In addition, the capability to control P_{Ar} during deposition also implies an ability to control the surface roughness. Increases in P_{Ar} lead to more numerous collisions between atoms on the film and ambient gas atoms [4], which in addition to self-shadowing effects [5] should result in a greater roughness for the resulting surface. Specifically, we examine the role of sputter pressure on the onset of ferromagnetism at $d = d_c$, on the increase of H_c with d for $d > d_c$, and on the thickness independence of H_c for $d \gg d_c$. We shall further address difficulties inherent in the sputter deposition of ultrathin films while presenting the opportunities this method provides for tailoring the coercivity of the magnetic layer via deposition parameters.

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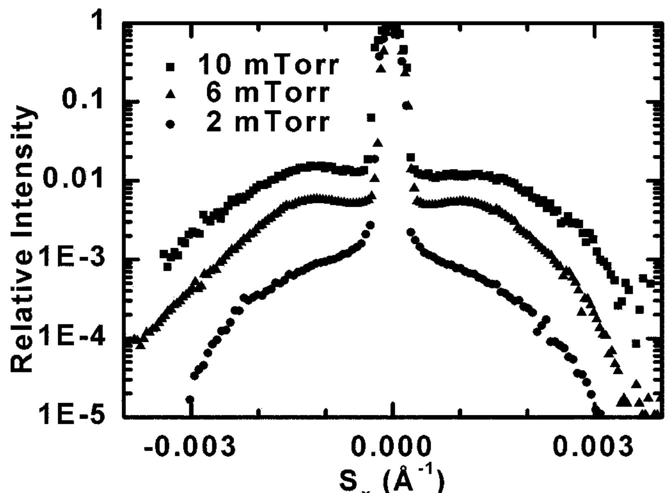

Fig. 1. Rocking curves for three sputter-deposited films grown at 2, 6, and 10 mTorr. The sharp break between the specular peak and the diffuse intensity implies that there is a long-wavelength cutoff to the roughness and allows the determination of unique triplets of the rms roughness σ, lateral correlation length ξ, and roughness parameter h. The magnitude of the diffuse component determines σ, while the width of that component and its detailed shape yield ξ and h. The increasing amount of diffuse component from 2 mTorr to 10 mTorr immediately shows an increase in rms roughness with sputter pressure.

II. EXPERIMENTAL

Film wedges were grown by d.c.-magnetron sputter deposition in a chamber with a base pressure of $<1 \times 10^{-6}$ Torr at three different Ar sputter gas pressures, $P_{\mathrm{Ar}} = 2$, 6, and 10 mTorr. We first grew a thick polycrystalline Cu buffer layer onto Si(001). The Cu thickness differs for each sample but each is at least $>$40 nm. We then deposited a continuous wedge of Co to have $d = 0$ to $5 \sim 6$ nm (assuming the density of pure, bulk Co) over a lateral distance of 15 mm, using a moving knife edge, and finally a $\sim$3 nm Al capping layer to protect the Co from oxidation. Deposition rates were calibrated for 2 mTorr through x-ray scattering experiments on similarly deposited multilayer films. Rates for the 6 and 10 mTorr Co films are discussed below.

Surface roughness measurements were performed using synchrotron-radiation-generated soft-x-ray diffuse scattering [6]. A scattering angle of $2\theta = 5 \sim 6°$ and a wavelength of $\lambda = 0.93$ nm ($E = 1.33$ keV) were chosen to maximize surface sensitivity and radiant flux; the x-rays at this grazing angle do not penetrate through the Co layer, thus only the Co–Al interface and the uppermost Al layer contribute to the detected x-ray scattering. As seen in Fig. 1, all wedges yield a two-component profile: an instrument limited peak in the specular direction and a broad diffuse component. Such a profile implies a roughness

TABLE I
GROWTH DETAILS AND CHARACTERIZATION RESULTS

P_{Ar} (mTorr)	2.18	6.00	9.78
Co deposition rate (nm/min)	5.0 ± 0.5	5.5 ± 0.6	6.0 ± 0.6
σ (nm)	0.24 ± 0.01	0.45 ± 0.01	1.45 ± 0.01
ξ (nm)	85 ± 13	50 ± 8	10 ± 2
h	0.5 ± 0.1	0.5 ± 0.1	0.5 ± 0.1
$H_{c,max}$ (Oe)	50	97	117
d_c (nm)	0.7 ± 0.1	1.6 ± 0.2	2.4 ± 0.2

with a long-wavelength cutoff. We may then treat the film as a self-affine fractal [7], [8] on a short length scale and as smooth on a long length scale, and using the model of [8], fit this profile to determine three morphological parameters: σ, the rms amount of vertically correlated roughness; ξ, the lateral correlation length of the vertically correlated roughness; and h, the roughness exponent which yields a measurement of the "jaggedness" of these features. The height-height correlation function, written in terms of σ, ξ, and h, is

$$C(x) = \sigma^2 \exp[-(|x|/\xi)^{2h}]. \quad (1)$$

Values for σ, ξ, and h for these growth conditions are given in Table I. The rms roughness increases for higher sputter gas pressures while the correlation length decreases. Although we do not directly measure the roughness of the magnetic layer, we expect this roughness relationship to reflect that of the internal interfaces as well as in particular that of the Co film.

Coercivity measurements were performed *ex situ* by reflecting polarized laser light ($\lambda = 632.8$ nm) off these wedges and detecting changes due to an applied magnetic field situated in the film plane and the plane of incidence [longitudinal magneto-optical Kerr effect (MOKE)]. Hysteresis loops were measured at several points along the Co wedge. Fig. 2(b) shows the coercivity, H_c, versus thickness, d, for the samples, with a representative hysteresis loop shown in the inset.

Auger electron spectroscopy (AES), performed on the $P_{\rm Ar} =$ 2 and 10 mTorr samples, reveals a small dependence of the deposition rate of the Co layer on sputter pressure, also given in Table I. More significant, however, is the presence of appreciable amounts of oxygen alloying uniformly throughout the Co layer for the 10 mTorr sample. A similar preferential alloying between oxygen and an Co-containing layer was seen previously using AES for CoCrTa films [9]. In contrast, the distribution of the oxygen in the 2 mTorr film was shifted toward the surface, indicating that oxygen alloyed with the Al layer as well as the Co layer.

III. RESULTS AND DISCUSSION

We must first address the mechanisms responsible for oxygen in our films. Possible sources are the background pressure before and during growth, the Ar pressure during growth, and exposure to atmosphere after growth. The oxygen content is likely

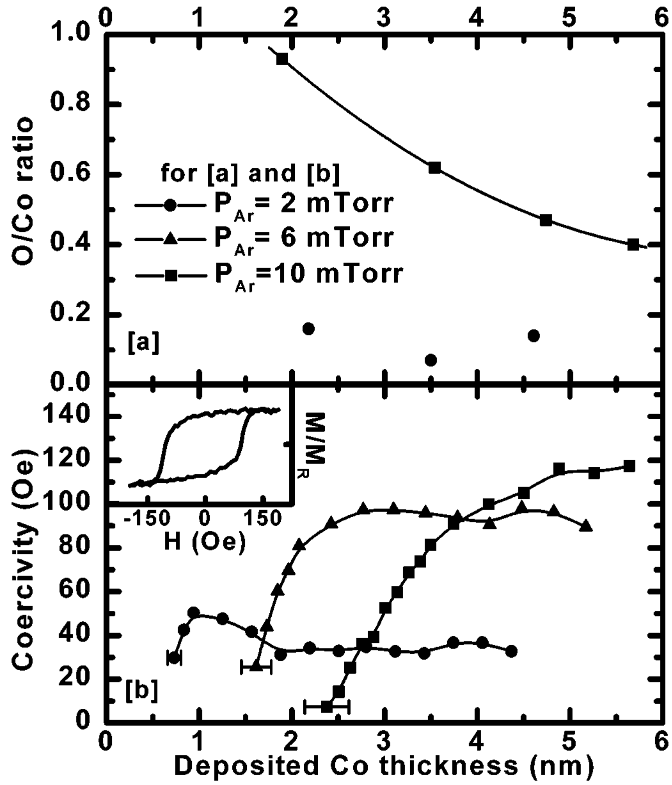

Fig. 2. (a) Ratio of O to Co atomic concentration, as determined using Auger electron spectroscopy, as a function of deposited Co thickness (O incorporation increases geometrical thickness) for Co wedges sputter-deposited at pressures of 2 mTorr and 10 mTorr (shown with polynomial fit). (b) Coercive field as a function of deposited Co thickness for film wedges grown separately at Ar pressures of 2, 6, and 10 mTorr. Inset: Representative hysteresis curve for the wedge grown at a pressure of 6 mTorr at $d = 4.5$ nm. Data show magnetism normalized to the remanent magnetization as a function of externally applied magnetic field strength, H. The onset of ferromagnetism for the 10 mTorr sample does not occur until the O–Co ratio is <0.80. To first order, increases in coercivity for the 10 mTorr sample seem to scale inversely with O–Co ratio while the 2 mTorr sample exhibits practically no change in either O–Co ratio or coercivity for $d > d_c$. This description does not account for any effects of roughness.

due to higher sputter pressure and atmospheric exposure. First, it has been reported previously that an increase in $P_{\rm Ar}$ from 2 to 4 Pa led to an incorporation of oxygen without any intentional addition of O to the sputter gas [10]. Second, we must take into account that while we have $\sim$3 nm Al capping layer on all these films, the Al cap on the 10 mTorr sample must cover a surface for which $\sigma = 1.45$ nm. It is quite possible that atmospheric oxygen would be able to reach the Co layer for this film. The O–Co ratio for this film is not <1 until $d > \sigma$.

We discuss three distinct thickness regions: $d = d_c$, $d > d_c$, and $d \gg d_c$. Previous reports of the value $d_{c(Co)}$ range from 0.2 nm to 2.2 nm [2], [11], [12]. The Cu surface roughness in [12] was ~ 250 nm, much larger than that of our roughest sample. The values of d_c appear to first order to follow with roughness. Our smoothest sample (2 mTorr) sample exhibited $d_c = 0.7$ nm, falling at the low end of the range of observed values of d_c. Our roughest sample, much smoother than the one in [12], nevertheless has similar d_c. The presence of O in our Co film appears to hinder the onset of ferromagnetism. As Co–O alloys are in general antiferromagnetic, no ferromagnetic ordering can occur when all the Co in the film is interacting with O. Ferromagnetism, first seen at $d = d_c$, does not occur until there is an O/Co ratio <0.80 for the film grown at $P_{\mathrm{Ar}} = 10$ mTorr.

We previously found a sudden increase in coercivity at a thickness greater than that for the onset of ferromagnetism in an evaporated Co/Cu(001) wedged film [13]. The in-plane coercivity of an evaporated film along the $\langle 100 \rangle$ direction increased suddenly at $d \sim 2.136$ nm, which in [13] is attributed to the thickness of the wedge increasing past that range where Co films grow in a metastable face-centered tetragonal phase. While such a transition may be important here also, we did not test for the phase of our films as oxygen plays such a dominant role and would make comparison impossible to our previously evaporated film. However, the transition to a thickness-independent value of H_c lower than its maximum value at $d = 1.0$ nm might indicate the increasing predominance of bulk effects in Co with increasing d for the $P_{\mathrm{Ar}} = 2$ mTorr sample. The bump in coercivity for this 2 mTorr sample is perhaps due to surface (e.g., morphology) influences on the coercivity, such as domain wall pinning.

For samples prepared at higher pressures (and thus having higher roughness and O content), the rate of increase in coercivity is more gradual. The relationship between O/Co ratio and H_c is illustrated for the roughest sample by comparing Fig. 2(a) and (b). The coercivity increases with decreasing O–Co ratio. It is possible that the ferromagnetic Co regions of the film gradually grow more numerous and coalesce with thickness causing the coercivity to increase gradually. The role of roughness in this thickness region for the 10 mTorr sample is unclear given the incorporation of oxygen.

For $d \gg d_c$, our data suggest that both roughness and O content determine the final, thickness-independent, value of H_c. We would expect an increased coercivity with increasing roughness, due to domain wall pinning. In addition, AES suggests that for this thickness region that the O/Co ratio is constant for our films. Therefore, while we cannot quantify whether the roughness or the oxygen content leads to these particular values for the coercivity, we can assert that some combination of controlling roughness and O/Co ratio would allow the tailoring of magnetic properties at commercially relevant thicknesses ($d \sim 5$ nm). Further work with lower base pressures, commercially-used alloys, and in situ MOKE will help further define the mechanisms responsible for adjusting coercivities in ultrathin films.

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