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Symmetry influence on interlayer coupling in epitaxial Co/Cr trilayers grown on MgO $(1 0 0)$ and $(1 1 0)$ substrates

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Abstract

Trilayers of Co(15 Å)/Cr(x)/Co(17 Å) have been epitaxially sputtered onto MgO (1 0 0) and (1 1 0) substrates coated with Cr(1 0 0) and (2 1 1) buffer layers, respectively. The Cr thickness x is varied from 6 to 50 Å. Both sample sets have the Co *c*-axis lying in the plane of the film, however, due to the four-fold symmetry of MgO (100), the Co layers form bicrystals with perpendicularly oriented *c*-axis. Ferromagnetic resonance measurements clearly show large two-fold and four-fold magnetic in-plane anisotropy for MgO (1 1 0) and MgO (1 0 0) samples, respectively. Magnetization measurements reveal interlayer coupling strengths peaked at a Cr thickness of approximately 10 Å for both symmetries, but the strength decreases more slowly with increasing t_{Cr} in the MgO(1 0 0) system. \odot 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Recently, investigators have utilized epitaxial growth techniques to deposit coherent HCP Co layers with inplane *c*-axis orientation $\lceil 1-5 \rceil$. A series of investigations have been reported $[6-8]$ on the anisotropy and exchange coupling in Co/Cr superlattices, in which the in-plane symmetry was primarily two-fold, although for some samples with lower Co thicknesses four-fold symmetry was observed. An investigation of Co/Cr multilayer samples grown on MgO $(1 1 0)$ and MgO $(1 0 0)$ was reported [9] in which the sample symmetry was determined by the substrate symmetry. In particular, the four-fold symmetry was realized for Co thickness ranging from 15 to 100 Å. This present study on structurally simpler trilayers has been undertaken to address the influence of the sample symmetry on the anisotropy and interlayer exchange coupling.

2. Experimental details

Two series of $Co(15 \text{ Å})/Cr(x)/Co(17 \text{ Å})$ trilayers with $(6 \leq t_{\text{Cr}} \leq 50 \text{ Å})$ were epitaxially sputtered onto single crystal MgO (1 0 0) and (1 1 0) substrates. The substrates were mounted side by side onto the sample holder and simultaneously deposited. A 100 \AA Cr layer was intially deposited at a substrate temperature of 600° C resulting in epitaxial Cr $(2 1 1)$ and $(1 0 0)$ buffer layers on MgO $(1 1 0)$ and $(1 0 0)$, respectively [10]. The substrate was then cooled to 150° C and the trilayers were grown by sequential deposition of the Co and Cr layers. This growth procedure has been successfully used to grow Fe/Cr and Co/Cr superlattices [2,10]. Due to the limited material in these trilayer samples, X-ray measurements

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are not feasible. However the trilayers were grown under identical conditions used to prepare \lceil Co (20 Å)/ $Cr(7 \le x \le 22 \text{ Å})$]₁₀ multilayer samples [9]. Therefore it is assumed that the trilayers possess structure similar to that found for the multilayers. Those results indicate that BCC-Cr (2 1 1) and coherent HCP-Co $(1 1 0 0)$ are formed on MgO $(1 1 0)$ substrates and that BCC-Cr (100) and epitaxial, strained HCP-Co (1120 are formed on MgO (1 0 0). In both cases, the Co grows with the c -axis in plane of the film, but the MgO $(1 0 0)$ series has two equivalent perpendicular directions in which the *c*-axis may be oriented [11].

In-plane anisotropy symmetry was determined using 35GHz ferromagnetic resonance (FMR) with the sample placed film side down on the bottom of TE102 mode cavity and the magnet rotated about the sample. Hysteresis loops were measured on a vibrating sample magnetometer (VSM) built in our lab primarily by the two undergraduate authors (JJP and JZH). Absolute magnetization values were obtained on a SQUID magnetometer.

3. Results and discussion

Saturation magnetization values for the Co in the samples reported vary in the range of 1100 to 1200 emu/cm3. While less than the bulk value of 1400 emu/cm^3 , these results are consistent with values reported by others [6] and are consistent with a small amount of alloying at the Co/Cr interface.

FMR spectra for all samples reported are symmetric, single mode with linewidth of approximately 1000 G and signal-to-noise ratio exceeding 100. The two-fold symmetry of the MgO (1 1 0) series of samples and the fourfold of the (1 0 0) series is readily apparent in the FMR resonance positions as exemplifed in Fig. 1. These resonance positions fit very well a model that assumes the standard form for Co uniaxial anisotropy

$$
E(\theta) = K_1 \sin^2 \theta + K_2 \sin^4 \theta, \tag{1}
$$

where θ is the angle between the magnetization *M* and the Co c -axis, and K_1 and K_2 are first-and second-order anisotropy energy densities. The model also includes an out-of-plane anisotropy term which is cast in the form of a shape anisotropy $4\pi M_{\text{eff}}$. The two-fold sample shown in Fig. 1 is characterized by $K_1 = 1.8(\pm 0.4)$ $\times 10^6$ erg/cm³, $K_2 = 0.55(\pm 0.12) \times 10^6$ erg/cm³. These *K* values are approximately 40% of the bulk Co values, but have the same 3 to 1 ratio as is observed in the bulk. The out-of-plane anisotropy for the two-fold sample shown in Fig. 1 is characterized by $M_{\text{eff}} = \frac{1}{2} M_{\text{sat}}$, which implies an easy-axis out-of-plane anisotropy that counters the easy-plane demagnetization. This rather large additional $(4\pi M_{\text{eff}} - 4\pi M_{\text{sat}})$ anisotropy could

Fig. 1. Ferromagnetic resonance fields as a function of in-plane angle. Symbols are data, solid line is fit to model described in the text.

arise from surface anisotropy or stress-induced crystal fields normal to the film surface. All $MgO(110)$ samples reported here have anisotropy results similar to the above.

As mentioned above, the MgO (1 0 0) substrate promotes bicrystalline Co growth with mutually orthogonal *c*-axis leading to four-fold in-plane anisotropy. As has been pointed out previously [9], in the limit of strong inter-crystallite coupling, the Co layer behaves as a single moment with anisotropy given by the sum of $E(\theta)$ and $E(\theta + \pi/2)$ in Eq. (1), resulting in an average in-plane anisotropy determined by

$$
E(\theta) = \frac{1}{2}K_2 \cos^2(2\theta). \tag{2}
$$

Again very good agreement between experiment and theory is observed as the model yields a value of $K_2 = 9.9(\pm 0.9) \times 10^5 \text{ erg/cm}^3$ for the $t_{\text{Cr}} = 10 \text{ Å}$ MgO $(1 0 0)$ sample. This is significantly larger than for the two-fold samples, being still only slightly less than 70% the K_2 for bulk Co. All MgO (1 0 0) trilayer samples reported here have anisotropy results similar to the above.

In none of the FMR spectra observed is there any evidence for modes other than a single uniform resonance mode. This supports, in the case of the four-fold samples, the assumption of strong inter crystalline coupling within a Co layer (if the crystallites were weakly coupled, their magnetizations would precess independently and two resonances, one from each *c*-axis orientation, would be observed at each in-plane angle). Such weak intercrystalline coupling was reported in one Co/Cr multilayer sample [8]. The lack of additional modes also means that we were not observing the anticipated outof-phase resonance modes associated with interlayer coupling effects. These out-of-phase modes may be suppressed somewhat by the large in-plane anisotropies present in the samples.

Although we did not observe the coupling effects in FMR spectra, anitiferromagnetic (AF) interlayer coupling is clearly manifest as symmetrically offset hysteresis loops shown in Figs. 2 and 3, where, in all cases, the field is applied along the in-plane easy axis. The interlayer

Fig. 2. Hysteresis loops for MgO (1 1 0) sample series. Magnetic field as applied in-plane along the easy axis.

Fig. 3. Hysteresis loops for MgO (1 0 0) sample series. Magnetic field is applied in-plane along one of the easy axes.

coupling energy per unit area may be simply expressed as (ignoring biquadratic coupling)

$$
Et_{\text{Co}} = -J\cos(\theta_1 - \theta_2),\tag{3}
$$

where $J > 0$ ($<$ 0) implies ferromagnetic (antiferromagnetic) coupling, t_{Co} is the Co layer thickness and θ_i represent the angle between M_i (one for each Co layer) and the field direction which is also the in-plane easy direction.

In the MgO (1 1 0) system, samples with $t_{\text{Cr}} = 8, 10,$ 11.5 and 13 \AA show an abrupt transition from antiparallel to parallel alignment. Given the strong in-plane anisotropy in these samples, the antiparallel magnetizations are aligned along the easy axis ($\theta_1 = 0$, $\theta_2 = \pi$). As θ switches from π to 0 (via domain wall propagation) the anisotropy energy remains unchanged and therefore the H_s is a direct measure of the effective coupling field. Since this transition occurs in a metamagnetic or non-equilibrium fashion, the Zeeman energy at the switching field is equal to the change in coupling energy as θ_2 switches form π to 0. Therefore, the magnitude of *J* is given by

$$
J = M_{\rm s} t_{\rm Co} H_{\rm s} \tag{4}
$$

and the results are plotted in Fig. 4. The *J* values are somewhat smaller over the same Cr thickness range than the Fe/Cr system [10]. However, the present *J* values are comparable to those observed in Co/Cr multilayers with Co *c*-axis in-plane [7]. It is interesting to note that the

Fig. 4. Exchange energy densities as a function of t_{Cr} . Diamonds and squares are MgO (1 1 0) and (1 0 0), respectively.

coupling peak occurs in the $8-10$ A range whereas the peak occurred at 13 Å in similarly prepared superlattice samples [9].

As seen in Fig. 3, where again the field is applied along one of the in-plane easy axes, the MgO (1 0 0) system also exhibits offset hysteresis loops indicative of antiferromagnetic interlayer coupling at $t_{\text{Cr}} = 8, 10, 11.5$ and 13 Å. The approach to saturation for the four-fold system is in principle more complicated than for the two-fold, often requiring fitting via energy minimization to reliably extract coupling energies. However, in the case where the anisotropy is considerably larger than the coupling energy one can demonstrate that the centroid of the offset hysteresis is the effective coupling field. For example, minimization simulations were done utilizing Zeeman, coupling and fourfold anisotropy energies (as per Eqs. (2) and (3)) with the field along an easy in-plane axis. With anisotropy fields 2, 4 and 10 times the exchange field, switching fields were 10, 5 and 0% greater, respectively, than the exchange field. In our MgO $(1 0 0)$ AF coupled samples, the anisotropy fields (approximately 3500 G) are 3–5 times greater than the switching fields. Given that our accuracy in determining the switching centroid is no better than \pm 5%, we conclude that *J* may be obtained by Eq. (4) above. The results are plotted in Fig. 4. Even though both occur via domain wall motion, the switching transitions are not as sharp in the (1 0 0) as in the (1 1 0). This is likely due to impeded wall motion in the (1 0 0) due to domain wall pinning arising from the bicrystal nature of a Co layer.

Biquadratic coupling would not affect the $(1 1 0)$ loops seen in Fig. 2, however, if present, it should manifest itself as additional, low field, abrupt transitions in the $(1 0 0)$ loops of Fig. 3. Since no such transitions are apparent, it must be concluded that any biquadratic coupling energy is negligible in these samples.

The long-period oscillatory coupling (the period, phase, and strength) in Fe/Cr has been shown to be independent of crystallographic orientation and, therefore, is believed to arise from a common feature of the Cr Fermi surface [10]. This isotropic behavior has been attributed to spanning vectors across a d-derived lens [12] or the N-centered ellipse of the Cr Fermi surface [13]. As seen in Fig. 4, the Co/Cr trilayer coupling energies do depend on the Cr spacer-layer symmetries. The (1 0 0) coupling is somewhat greater than that for the (2 1 1) spacers and persists through greater Cr thicknesses. These differences can be explained as a simple phase shift in the peak coupling strength as a function of Cr thickness. Although the period of the exchange coupling depends predominantly on the details of the Cr Fermi surface, the phase and strength of the coupling also depend on the overlap of the Co and Cr wavefunctions [13], and on the structure of the interface. Even though the Co/Cr and Fe/Cr systems have the same Cr epitaxy, the magnetic layers have very different symmetries and, in addition, there is significant dependence of the interfacial strains on crystallographic orientation in the Co/Cr system that is not common to the Fe/Cr system.

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