Perpendicular Exchange Bias of Co/Pt Multilayers

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Exchange bias measurements of ferromagnetic/antiferromagnetic (F/AF) bilayers are typically performed with the magnetization of the F layer parallel to the AF interface. We describe measurements of Co/Pt multilayers with out-of-plane magnetic easy axis that are exchange biased with CoO. Field-cooling experiments with the applied field perpendicular and parallel to the sample plane exhibit loop shifts and enhanced coercivities. Modeling and comparison to biasing of samples with planar easy axis suggests such measurements provide a way to probe the spin projections at F/AF interfaces.

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When a ferromagnetic (F) thin film in contact with an antiferromagnetic (AF) thin film is cooled through the Néel point of the AF in an applied magnetic field the hysteresis loop of the F develops a loop-shift and enhanced coercivity [1]. These exchange biasing effects arise as the order of the AF is established in the presence of the F through the interfacial F-AF interaction. Since the AF spins are only weakly coupled to the external field through the F-AF interaction, they remain in the state they were frozen into as the F is rotated and are responsible for the spin memory observed macroscopically as a hysteresis loop shift [2]. Microscopically the shift has been explained by random field theory [3], which assumes the formation of AF domains due to interfacial roughness as the F/AF system is field cooled through the Néel point. Various models suggest spin-flop coupling due to frustrated interfacial spins [4–6], hysteretic AF grains [2], and domain wall pinning due to defects [7] at the F/AF interface as responsible mechanisms for the coercivity enhancement. Although exchange biased thin films have found applications in magnetic thin film sensors, a satisfying micromagnetic description that explains all observed phenomena has not, as yet, been developed.

Numerous experimental investigations on exchange biased F/AF bilayers and multilayers have been performed to understand the origin of the effect as summarized in recent reviews [8,9]. However, only exchange biased F/AF systems with a ferromagnetic in-plane easy axis have been reported. Here we present the study of exchange bias experiments on a F/AF system with an out-of-plane or perpendicular easy axis. Co/Pt multilayers are used as the F layer with out-of-plane magnetic anisotropy. Thin Co films are known to exhibit an out-of-plane easy axis due to strong interfacial anisotropy for thickness values $\lesssim$10 Å. However, the interfacial anisotropy can be exploited in Co/Pt multilayers and a strong out-of-plane anisotropy can be maintained for thicker films.

The multilayers were dc magnetron sputtered in a 3-mTorr Ar atmosphere onto Si_3N_x-coated Si(100) wafers which were heated to a temperature of 150 °C. The substrates were rotated during the deposition to ensure uniformity of the films. Typical base pressures prior to deposition were 5×10^{-8} Torr. For each film, a 100 Å thick Pt buffer layer was deposited onto the substrate immediately followed by a multilayer film composed of a number of Co(4 Å)/Pt(5 Å) bilayers. A subset of the multilayers were capped with 15-Å thick Co layers. The films were cooled to ~40 °C and exposed to ambient atmosphere. The 15-Å Co metal films partially oxidize to CoO, which is the natural antiferromagnet of Co, and are used as the AF to exchange bias the Co/Pt multilayers. Since the top Co layer is polycrystalline, the CoO is polycrystalline and will exhibit AF domains with in-plane and out-of-plane spin axis.

The crystallographic structure of the Co/Pt multilayers was determined by x-ray diffraction (XRD) with Cu K_α radiation ($\lambda = 1.541$ Å). The out-of-plane XRD pattern of the multilayers reveal a Pt (111) peak originating from the Pt underlayer and peaks corresponding to the average (111) lattice spacing of Pt and fcc Co with multilayer peaks corresponding to the nominal 9-Å periodicity of the Co/Pt bilayer. Typical full width at half maximum values of the rocking curves, which measures the mosaicity of the sample, are ~9°. No peaks corresponding to other crystallographic orientations are observed. The CoO thickness was determined by SQUID magnetometry of the Co/Pt multilayers with and without the 15-Å Co overlayer. We found that the top 6–7 Å of the Co overlayer oxidize and form ~10 Å of CoO.

Field-cooling experiments from 300 K down to 10 K were performed in a 5-tesla quantum design SQUID. The cooling and measurement fields were applied in both the in-plane and the out-of-plane geometries. To saturate their moment, the applied field during cooling was 40 kOe for samples exhibiting hard-axis behavior and 4 kOe for samples exhibiting easy-axis behavior. Shown in Fig. 1 are the out-of-plane hysteresis loops of $[Co(4\ Å)/Pt(5\ Å)]_5$ multilayers with and without a 15-Å (Co + CoO) cap measured at 10 K after field cooling perpendicular to the sample plane. At 300 K both films have coercive field values $H_C = 460$ Oe. After field cooling both films exhibit a square out-of-plane hysteresis loop. The CoO-capped film displays a hysteresis loop shift $H_S = -0.9$ kOe and a coercivity $H_C = 3.1$ kOe,

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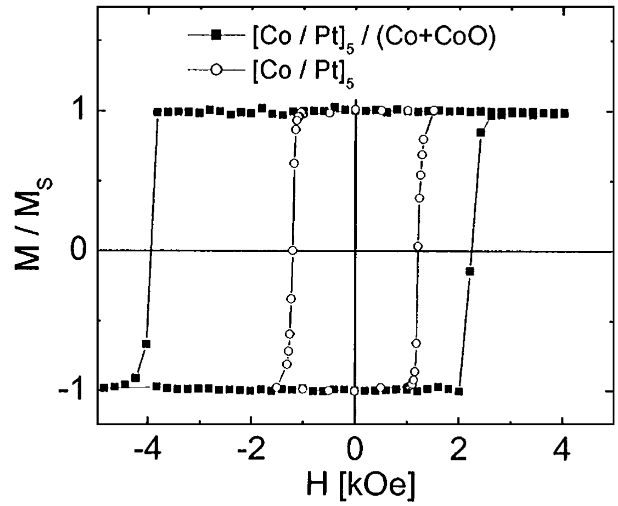

FIG. 1. Out-of-plane hysteresis loops of $[Co(4\ Å)/Pt(5\ Å)]_5$ multilayers with and without a Co + CoO(15 Å) cap after field cooling to 10 K perpendicular to the sample plane.

while the uncapped film exhibits no loop shift and a less enhanced coercivity of 1.2 kOe.

The temperature dependence of the coercivity and the loop shift for both samples are shown in Fig. 2. The blocking temperature T_B of an exchange biased system is defined as the temperature at which the loop shift vanishes. (The term blocking temperature is historic and is not to be confused with the same term describing the temperature at which superparamagnetic particles become magnetically stable.) This is caused by the loss of unidirectional anisotropy, which originates in the AF/F coupling. For some systems T_B equals the Néel temperature, T_N, but in other systems, $T_B < T_N$ and can be explained by thermal activation of the AF grains. From Fig. 2, a blocking temperature of $T_B \sim 220$ K is inferred, which is ~70 K below the bulk Néel temperature of CoO (293 K). The magnitude of the coercivity enhancement for the exchange biased sample is determined by comparing the coercivity dependence on temperature of the biased and unbiased samples. At low temperatures the coercivity of the unbiased sample is well below that of the biased sample and decreases linearly with increasing temperature. The coercivities of the biased and unbiased samples coincide at ~250 K, which is roughly the Néel temperature of a 10-Å CoO film [10]. The interfacial exchange energy at 10 K was calculated as $\sigma = 0.31\ \mathrm{erg/cm^2}$ using

$$\sigma = M_S H_S t\,, \tag{1}$$

where M_S is the magnetic moment density and t is the thickness of the ferromagnet, i.e., Co/Pt multilayer. Because of the interfacial nature of the AF/F exchange coupling, the biasing effect scales with the inverse thickness of the ferromagnetic layer, which is well documented for longitudinal biasing. This dependence also holds true for perpendicular biasing. For example, a single Co(4 Å)/Pt(5 Å)/Co + CoO(15 Å) trilayer exhibited a loop shift of $H_S = -2.4$ kG and a coercivity of $H_C = 9$ kG after field cooling to 10 K perpendicular to the sample plane.

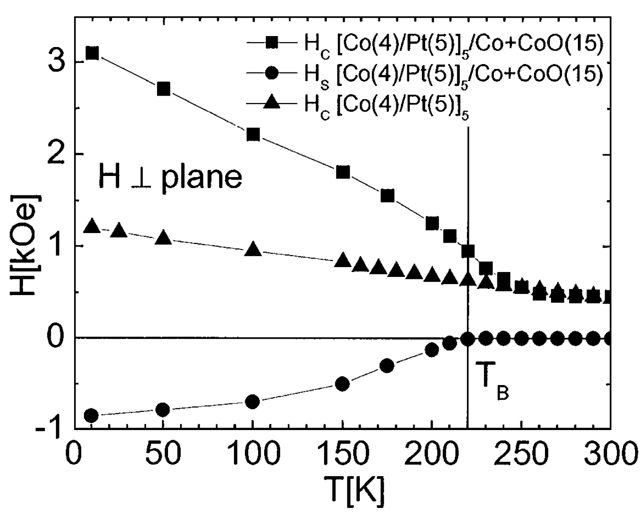

FIG. 2. Temperature dependence of the coercivity H_C (squares) and loop shift H_S (circles) of the out-of-plane hysteresis loops of the $[Co(4\ Å)/Pt(5\ Å)]_5$ multilayers with and without (triangles) Co + CoO(15 Å) cap.

The in-plane hysteresis loop measurements at 300 K revealed a hard-axis loop with $H_C = 160$ Oe. Field cooling to 10 K with the saturating magnetic field applied in the sample plane (i.e., the hard axis of the multilayer) resulted in an enhanced $H_C = 2.5$ kG and a loop shift $H_S = -2$ kG. The exchange energy according to Eq. (1) is $\sigma = 0.61\ \mathrm{erg/cm^2}$, which is roughly twice the value observed for perpendicular field cooling. In agreement with the out-of-plane biasing measurements, the blocking temperature is $T_B \sim 220$ K and the coercivity drops to its room temperature value at $T_N \sim 250$ K. In contrast to the out-of-plane loops, low squareness was observed for the in-plane loops indicating rotation rather than domain nucleation as the reversal mode observed for the perpendicular loop.

To rule out the nature of the reversal mode as the origin for the measured difference in perpendicular versus longitudinal bias values, we compared the relative strengths in similar samples with out-of-plane and planar anisotropy. A $Pt(100\ Å)/[Co(4\ Å)/Pt(5\ Å)]_{10}/Co + CoO(15\ Å)$ multilayer (sample 1) with out-of-plane anisotropy is compared to a Pt(100 Å)/Co(40 Å)/Pt(5 Å)/Co + CoO(15 Å) sample with planar anisotropy (sample 2), but the same total Co thickness. Since we measured a thickness dependent oxidation of the Co cap, we kept a 5-Å Pt spacer layer between the 40 Å Co and the Co + CoO cap to achieve similar CoO biasing layers. Sample 1 has a ~10% higher moment, presumably resulting from polarization of the Pt in the multilayer. The hysteresis loops of these two samples at room temperature and after in-plane and out-of-plane field cooling are shown in Fig. 3.

Upon out-of-plane field cooling of sample 1 the coercivity increased from $H_C = 230$ Oe at 300 K to 1.5 kOe

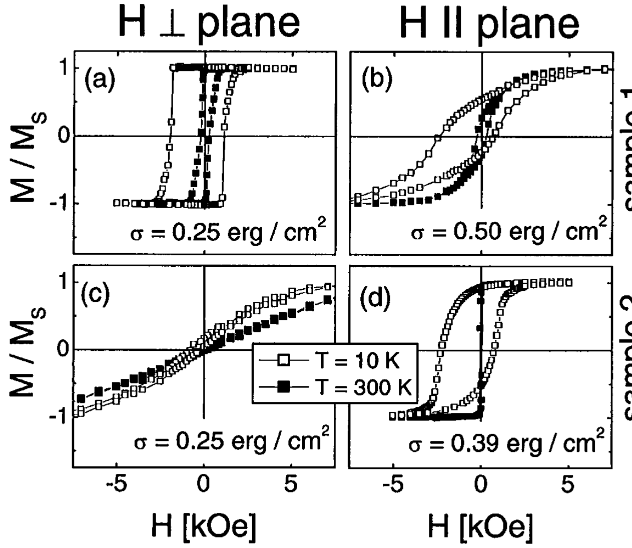

FIG. 3. Hysteresis loops for $[Co(4\ Å)/Pt(5\ Å)]_{10}/Co + CoO(15\ Å)$ (first row) and $Co(40\ Å)/Pt(5\ Å)/Co + CoO(15\ Å)$ (second row) at 300 K and at 10 K after field cooling, measured perpendicular (first column) and parallel (second column) to the plane.

at 10 K with a loop shift of $H_S = -430$ Oe ($\sigma = 0.25$ erg/cm^2). In-plane field cooling resulted in a coercivity increase from $H_C = 270$ Oe to 1.5 kOe and a loop shift of $H_S = -890$ Oe ($\sigma = 0.5$ erg/cm^2). Out-of-plane field cooling of sample 2 yields a coercivity increase from 80 to 280 Oe and $H_S = -490$ Oe ($\sigma = 0.25$ erg/cm^2), in-plane field cooling yielded an increase from 60 Oe to 1.5 kOe and a shift of $H_S = -770$ Oe ($\sigma = 0.39$ erg/cm^2). Again σ was found to be lower for field cooling perpendicular to the sample plane than for field cooling in the sample plane independent of the anisotropy direction of the F layer.

For sample 1 the easy-axis switching fields are much lower than the saturation fields of the hard-axis loops, which suggests nucleation followed by domain wall motion as a reversal mechanism for the easy axis and rotation for the hard axis. Both the easy- and hard-axis loop shapes also appear symmetric for the increasing and decreasing fields. In contrast, sample 2 shows an asymmetric reversal after in-plane field cooling. This asymmetry is often observed for longitudinal biasing experiments [1,11] and is attributed to a different reversal mechanism for the increasing and decreasing field part of the loop [11]. The observed difference in loop symmetry may be linked to the uniaxial anisotropy of sample 1 limiting the reversal modes as compared to the planar anisotropy of sample 2.

To describe the observed behavior qualitatively, we posit the following model: Upon field cooling the interfacial AF spins are "frozen-in" to the spin anisotropy axes that are closest to the applied field. No AF domain wall is assumed, since only ~4 monolayers of CoO are formed, and thus the concept of a domain wall seems inadequate [16,17]. This model is particularly applicable for CoO because of its high magnetocrystalline anisotropy. If the coupling between F and AF is bilinear then the measured bias will be proportional to the projection of the AF spins to the field-cooling direction. Thus out-of-plane AF spin components are necessary to obtain perpendicular exchange bias, while in-plane AF spin components are necessary to obtain longitudinal exchange bias. Accordingly an out-of-plane loop shift would vanish, if the AF spins would completely lie in the plane. In that sense the AF layer in our system must be arranged differently from systems, such as CoO/Fe_3O_4, where CoO spins were found to completely lie in the growth plane rather than in their bulk directions [12].

As determined from XRD, the Co/Pt multilayers are (111) textured. CoO is formed as oxygen atoms are placed in the interatomic sites of the Co lattice, so that Co and CoO have the same texture. For our analysis we assume the bulk spin structure of CoO, where the spins are aligned in ferromagnetic (111) sheets and point in ⟨117⟩ directions [13–15]. In the case of field cooling and measuring the multilayer perpendicular to the sample plane, the CoO spins of various AF domains will be distributed on a cone surface with half-apex angle 43.3° if a perfect (111) texture is assumed. The angle of 43.3° is given by the [111] surface normal and the [117], [171], and [711] spin easy axes of CoO, which are closest to the [111] normal, as shown in Fig. 4a. For this case, the calculated loop shift is simply given by

$$H_S = -\frac{\sigma_0 \cos\Theta_{AF}}{M_S t} = -0.72\,\frac{\sigma_0}{M_S t}\,, \qquad (2)$$

where $\Theta_{AF} = 43.3°$ is the angle of the AF spins with respect to the plane normal.

In the case of field cooling and measuring the multilayer parallel to the sample plane, the CoO spins freeze into a direction closest to the cooling field direction. For a (111) texture, these are the $[\bar{1}\bar{1}7]$, $[\bar{1}7\bar{1}]$, $[7\bar{1}\bar{1}]$ and the $[11\bar{7}]$, $[1\bar{7}1]$, $[\bar{7}11]$ directions. Because of the texture, these directions for the various AF domains lie on two cone surfaces with a half-apex angle of 66.2° as depicted in Fig. 4b with only 1/6th of the cone surfaces populated. In this case the average AF spin projection is given by

$$H_S = -\frac{\sigma_0 \sin\Theta_{AF}\langle\cos\varphi_{AF}\rangle}{M_S t} = -0.87\,\frac{\sigma_0}{M_S t}\,, \qquad (3)$$

where

$$\langle\cos\varphi_{AF}\rangle = \frac{1}{\pi/3}\int_{-\pi/6}^{\pi/6}\cos\varphi_{AF}\,d\varphi_{AF} = 0.95 \qquad (4)$$

is the average azimuthal AF spin component and $\Theta_{AF} = 66.2°$ is used. The calculated loop shift after in-plane cooling is ~20% higher than that after out-of-plane cooling. Such a model, although phenomenological in character, is able to explain the higher value for the longitudinal bias compared to the perpendicular bias for anisotropic samples. It also suggests perpendicular together with longitudinal biasing experiments could be a general approach

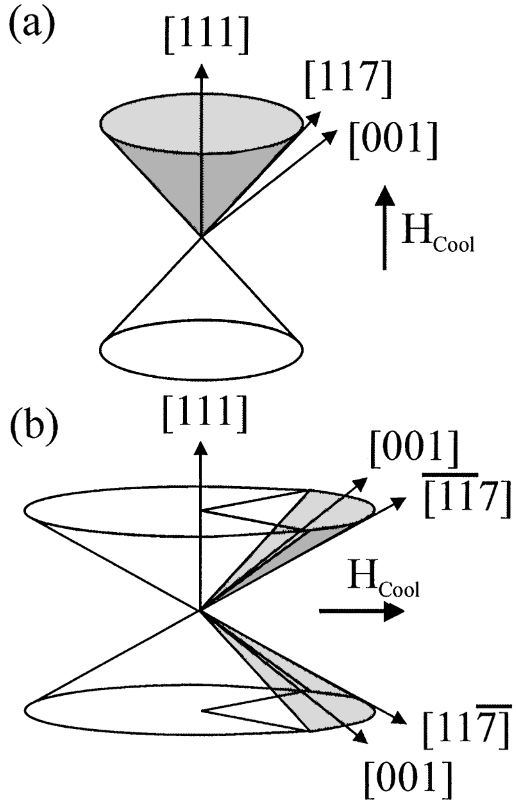

FIG. 4. AF CoO spins are frozen in to their easy axis, which is closest to the direction of the applied cooling field. (a) For out-of-plane field cooling the CoO spins will lie on a cone defined by the ⟨117⟩ type directions; (b) for in-plane field cooling, the spins lie on 1/6 of the two cone surfaces defined by the $[\bar{1}\bar{1}7]$ and $[11\bar{7}]$ directions.

to study the projections of the AF spins out of and into the plane. It is understood that spin flop coupling between the F and AF can contribute to the coercivity enhancement, but not to the loop shift [6]. Although CoO has a very high anisotropy [18] (29.2×10^7 erg/cm^3), spin-flop tendencies were observed in Co/CoO [19] and Fe_3O_4/CoO [12] systems. Thus, a spin flop may not be excluded in the mechanism to provide for additional uniaxial anisotropy of perpendicular exchange biased multilayers.

In summary, we observed the existence of perpendicular exchange biasing for AF/F multilayers with an out-of-plane easy axis, which was demonstrated using Co/Pt multilayers exchange coupled to a thin CoO layer. The interfacial exchange energies between AF and F were determined to be about twice as big for the in-plane compared to the out-of-plane field-cooling geometry. We suggest that the quantitative difference between in-plane and out-of-plane field biasing has its origin in anisotropy in the CoO induced by the textured growth of the films shown by XRD. This holds true for both films with in-plane and out-of-plane easy axis. The coercivity enhancement of the perpendicular biasing effect is comparable to the longitudinal biasing effect, but the interfacial exchange energy is different. Since the difference reflects the spin direction of the AF after field cooling, perpendicular together with longitudinal exchange bias experiments are a simple technique to probe the AF spin distribution of thin F/AF bilayers.

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