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Abstract

A series of ferromagnetic Ni₈₀Fe₂₀(55 nm)/antiferromagnetic CoO (25 to 200 nm)/ferromagnetic Co (55 nm)/SiO₂(substrate) trilayer thin films were fabricated by ion-beam assisted deposition in order to understand the role of ion beam modification on the interfacial and interlayer coupling. The microstructural study using transmission electron microscopy, X-ray reflectometry, and polarised neutron reflectometry showed that ion-beam modification during the deposition process led to an oxygen-rich Co/CoO nanocomposite interface region at the bottom layer. This interface caused a high exchange bias field for the ferromagnetic cobalt. However, the exchange bias for top permalloy ferromagnet remained low, in line with expectations from the literature for the typical interfacial energy. This suggests that the ion-beam enhancement of the magnetic exchange bias is localized to the Co/CoO interface where local microstructural effects provide the dominant mechanism.

Keywords

modification, beam, ion, via, films, magnetic, thin, enhancement, trilayer, co, coo, nife, interface, specific, energy, exchange, interfacial

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Enhancement of the magnetic interfacial exchange energy at a specific interface in NiFe/CoO/Co trilayer thin films via ion-beam modification

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A series of ferromagnetic Ni₈₀Fe₂₀(55 nm)/antiferromagnetic CoO (25 to 200 nm)/ferromagnetic Co (55 nm)/SiO₂(substrate) trilayer thin films were fabricated by ion-beam assisted deposition in order to understand the role of ion beam modification on the interfacial and interlayer coupling. The microstructural study using transmission electron microscopy, X-ray reflectometry, and polarised neutron reflectometry showed that ion-beam modification during the deposition process led to an oxygen-rich Co/CoO nanocomposite interface region at the bottom layer. This interface caused a high exchange bias field for the ferromagnetic cobalt. However, the exchange bias for top permalloy ferromagnet remained low, in line with expectations from the literature for the typical interfacial energy. This suggest that the ion-beam enhancement of the magnetic exchange bias is localized to the Co/CoO interface where local microstructural effects provide the dominant mechanism. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4865569>]

I. INTRODUCTION

The unusual magnetic anisotropy known as exchange bias was first discovered in Co/CoO nanoparticles, and results in the unidirectional shift of the magnetic hysteresis.^{1,2} More recently, the deliberate implantation of oxygen atoms using ion-beam techniques has been shown to lead to an anomalously high interfacial exchange energy in ion-beam modified Co.^{3,4} The origin of this exchange field enhancement in disordered Co/CoO nanocomposites remains unclear, but the relative strength compared to high quality Co/CoO thin films is both noteworthy and interesting. Moreover, our past work showed that the deposition order of the two layers, which may be either Co/CoO/Substrate or CoO/Co/Substrate, affected the penetration and diffusion of ions in the thin film interfaces. However, it is a topical question whether the bulk of the anti-ferromagnet, away from the ferromagnetic and interfacial regions, is an important component in this enhanced field. It is not clear, for instance, whether, as proposed for FeF₂, a long-range spin structure in the (antiferromagnet) AF could lead to a different exchange bias field sensed at the opposite interface from the ion-beam modified interface.^{5,6} In previous work, we showed that exposure of the Co layer to moderate energy oxygen ion-beams during the deposition of the CoO led to the implantation and diffusion of oxygen into the underlying Co, leading to a pillar-like microstructure with an anomalously high exchange bias.^{2,3} A natural question is whether similar effects can be realized in a trilayer structure, and whether the third layer on the opposite side of the AF structure is affected by the modified exchange coupling at the nanocomposite interface. Previous work on (ferromagnet) FM/AF/FM

layers in the 20–200 nm thickness regime reported a strong coupling between the two layers ascribed to the important role of bulk AF spins in such a structure. In this work, we fabricated Ni₈₀Fe₂₀/CoO/Co spins valves using ion-assisted deposition to test whether, upon modification of the cobalt layer, the permalloy layer also experienced an altered exchange field or whether the effect was localised to the bottom interface.

II. THEORY

By selecting two ferromagnetic materials with different coercive fields, it is possible to create a well-resolved double-step magnetic hysteresis loop that is the superposition of the two independent magnetic layers as shown in Figure 1.^{6,7} In this work, we chose Ni₈₀Fe₂₀ as the second ferromagnet, due to its soft magnetic character, and used CoO as the intermediary layer to form a Ni₈₀Fe₂₀/CoO/Co trilayer structure. This allows for the direct calculation of the exchange bias fields of each interface independently. For the scenario illustrated in Figure 1, one can then calculate the coercivity and exchange bias for each ferromagnetic layer labelled FM1 and FM2 using the formulae

$$H_{EB}^{FM1} = \frac{(A + B) + (C + D)}{4}, \quad (1)$$

$$H_{EB}^{FM2} = \frac{(E + F) + (G + H)}{4}, \quad (2)$$

$$H_c^{FM1} = \frac{(C + D) - (A + B)}{4}, \quad (3)$$

$$H_c^{FM2} = \frac{(G + H) - (E + F)}{4}. \quad (4)$$

If the ferromagnetic layers are truly independent, then this method may be used to reliably deconvolute the resulting

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FIG. 1. Schematic illustration of double hysteresis loop resulting from volume-average measurement of two uncoupled ferromagnetic phases. (a) Softer magnetic phase. (b) Harder magnetic phase. (c) Superposition of hard and soft magnetic phase.

double hysteresis to obtain the collinear configurations of each decoupled ferromagnet.⁷ A depth-resolved probe, however, such as polarized neutron reflectometry can provide a direct check of the magnetic depth profile. Once the behavior of such a trilayer is understood, the judicious selection of a specific field selected at points A-F can set the nanoscale spin alignment of each ferromagnetic layer independently during the field-cool process. For the typical case of negative exchange bias, this allows to one to safely field cool parallel or antiparallel alignments through the temperature range of the field cool process. Past work has shown that the formation of partial or anti-phase domains in the antiferromagnet can mediate magnetic frustration through the antiferromagnetic layer, with two possible mechanisms illustrated in Figure 2.^{5,7} In one case, it was shown that antiparallel alignment during the field cool process resulted in a lowering of exchange bias in one layer,⁵ whereas in other cases there is a change in the overall loop shape associated with a new anisotropy axis.⁷ Using similar techniques, we investigated whether ion-beam modification of the Co/CoO nanocomposite interface had a detectable effect on the bias at the $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}$ interface across the CoO spacer layer in a trilayer structure.

III. EXPERIMENTAL

A dual ion-beam deposition technique was used to prepare the $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ trilayers on a Si(100) substrate that had previously been annealed to provide a thick SiO_2 surface layer.^{8,9} A Kaufman ion source (800 V, 7.5 mA) was used to focus an argon ion-beam onto a commercial Co or $\text{Ni}_{80}\text{Fe}_{20}$ target surface. An End-Hall source ($V_{EH} = 100$ V, 500 mA) was used to *in-situ* bombard the growing film during CoO layer deposition with a mixture of 15% $\text{O}_2/(\text{Ar} + \text{O}_2)$, which was previously found to form the rock-salt phase.^{9,10} Five samples were deposited with the layer thicknesses summarised in Table I.

The thicknesses were chosen in the same length scale regime as Ref. 5 and the two ferromagnet layers were deposited to give a constant thickness across the series, whereas the thickness of the oxide spacer layer (x) was systematically varied. The antiferromagnet CoO was selected because of its similarity to FeF_2 in terms of antiferromagnetic domain state formation.^{11,12} The deposition was done at room temperature, and no external magnetic field was applied during deposition. Cross sectional transmission electron microscope (TEM) was performed on a JEOL [JEM-2010] TEM operating at 200 kV. X-ray reflectometry and diffraction was conducted on a Panalytical X'Pert Pro lab source using $\text{Cu-K}\alpha$ radiation ($\lambda = 0.154$ nm). Polarised neutron reflectometry was conducted on the PLATYPUS reflectometer at the OPAL Research Reactor, Sydney.^{13,14} Variable temperature measurements were performed on a commercial Quantum Design Physical Property Measurement System vibrating sample magnetometer. Except where explicitly stated, the samples were field-cooled by applying a +10 kOe field at 350 K to saturate the sample, before applying a smaller field (0–1000 Oe) while cooling to 200 K where the exchange bias was measured.

IV. DATA AND ANALYSIS

A. Film structure

Figure 3 shows the X-ray diffraction patterns for each of five samples in the trilayer series collected using $\text{CuK}\alpha$ radiation, where the data for each sample has been offset for clarity. From the appearance of the broad diffraction peaks, the top two layers are polycrystalline, consisting of the face-centered cubic $\text{Ni}_{80}\text{Fe}_{20}$ ($a = 3.55$ Å) and the CoO ($a = 4.25$ Å) rock-salt phase. The cobalt layer is highly polycrystalline with nanoscale crystallites leading to the lack of clear X-ray diffraction features. Figure 4(a) is a high resolution cross-sectional TEM of sample 1. It shows a clear 3

FIG. 2. Two possible mechanisms for long-range frustration mediated through an antiferromagnetic spacer in a FM/AF/FM structure for the situation that the top ferromagnet is dominant, and both interfacial exchanges are positive ($J_{int1} > J_{int2} > 0$). (Left) Partial spiral domain wall parallel to the surface causing a non-collinear arrangement at the bottom interface. (Right) Different populations of antiferromagnetic phase domains with walls perpendicular to the surface. In both figures, only one antiferromagnetic sub-lattice has been drawn for clarity.

TABLE I. Measured layer thickness for the five $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ trilayer samples using X-ray reflectometry (see Figure 5). The root-mean-squared interface roughness for the NiFe/CoO interface σ_1 and the Co/CoO interface σ_2 are also given.

Sample	$\text{Ni}_{80}\text{Fe}_{20}$ (nm)	CoO (nm)	Co (nm)	σ_1 (nm)	σ_2 (nm)
1	56	23	58	0.8	7.5
2	56	44	53	1	5
3	58	89	59	1.4	7
4	56	130	56	1	10
5	57	184	55	1	14

layer structure with a reasonably sharp interface between the $\text{Ni}_{80}\text{Fe}_{20}$ and the CoO layers, corresponding to low interface roughness (≈ 2 nm). However, the boundary between the cobalt layer and the CoO layer is less distinct, with a diffuse interface showing evidence of layer-intermixing on a longer length scale (> 5 nm). Figure 4(b) is the selected area electron diffraction (SAED) pattern of the Co/CoO region. It confirms that three nanocrystalline phases co-exist in this vicinity: hcp metallic Co, rock-salt phase CoO, and a smaller component of the Co_3O_4 spinel phase. Figures 4(c) and 4(d) are the bright field and dark field image of the Co layer showing small grains with sizes in the range of 3–18 nm. Thick grain boundaries are evident separating grains of the same phase. The presence of some oxygen ($\approx 4\%$ – 11% per atom) within the cobalt layer was qualitatively confirmed using energy dispersive spectroscopy mapping. Figure 5 shows the X-ray reflectometry (XRR) pattern for each sample in the series, where the data and fit for each sample has been offset horizontally by $0.05 \text{ \AA}^{-1}$ for clarity. The X-ray reflectometry was fitted with a 3-layer model using the genetic fitting and least-squares algorithm in the Motofit software package.¹⁵ Table I shows the fitting results for the thickness of each of the three layers for samples 1–5. The spacer-layer thickness ranged from 25 to 150 nm showing a linear dependence on the deposition time. For all samples, from the XRR fits, the roughness of the $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}$ boundary was found to be significantly lower (0.8–1.8 nm) than the CoO/Co boundary (6–23 nm).

FIG. 3. X-ray diffraction patterns for the $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ samples 1–5 in this work. Intensities have been offset for each sample for the sake of clarity. Peaks have been indexed using the cubic $\text{Ni}_{80}\text{Fe}_{20}$ and CoO crystal structures.

FIG. 4. (a) TEM cross-section, (b) electron diffraction pattern, (c) bright field, and (d) darkfield images for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ sample 1.

Figure 6(a) shows a high resolution TEM image for sample 5, compared with the structural model derived from the XRR fit in Fig. 6(b). Both techniques gave complementary information regarding the film chemical profile. A low roughness (< 1.3 nm) is evident at the top permalloy surface. However, the Co/CoO interface displays a high degree of phase intermixing. The highly diffuse interface at the CoO/Co interface may result from local heating and implantation of oxygen ions due to the use of moderate energy ions. At the ion energies used (0.1 keV), the penetration of oxygen ions through the first CoO monolayer as it grows should only be 4–7 Å, according to Monte Carlo simulations using the TRIM software package¹⁶ for oxygen implantation into cobalt with mass density of 8.8 g/cm^3 . However, from the TEM and XRR result it is clear that oxygen is present on a far longer length scale (> 5 nm), suggesting that local ion-beam effects promote grain-boundary assisted transport of oxygen deeper into the Co layer.

FIG. 5. X-ray reflectometry fits and data for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}(x)/\text{Co}$ (samples 1–5). Intensities have been systematically offset in Q_z for samples 2–5 for the sake of clarity. Thick black lines are the experimental data and thin colored lines are the fit to the data. Solid lines are fits to the data for each sample. Table I summarises the fitted layer thicknesses for the three layers in each of the five samples.

FIG. 6. Comparison of the cross-sectional TEM image (a) for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ (sample 5) with the 1D X-ray scattering length density (b) used to fit the data in Figure 5. The two techniques give a similar chemical profile normal to the film surface. A gradient interface is observed at the Co/CoO boundary indicating intermixing between the two layers due to implantation and diffusion of oxygen into the cobalt.

B. Magnetic properties

Figure 7 presents the room temperature magnetometry data for samples 1–5. The data have been over-plotted to emphasize the overall similarity in the magnetic properties of the films. A clear double step hysteresis is seen for all samples, reminiscent of the schematic diagram in Fig. 1. This is interpreted as the independent switching of each ferromagnetic layer separated by the paramagnetic CoO spacer layer. All samples were found to be saturated at fields above 2 kOe, as evident in the flat magnetic response for increasing fields up to 50 kOe. At room temperature, using Eqs. (3) and (4), the coercivity for the two phases is calculated as $H_c^{FM1} \approx 10$ Oe and $H_c^{FM2} \approx 250$ Oe. The inner loop coercivity is essentially identical for all samples, whereas small differences occur for the various films at the outer-loop switching point, which shows a distribution of coercive fields in a relatively narrow range (200–350 Oe). The exchange bias values for H_{EB}^{FM1} and H_{EB}^{FM2} are both zero within experimental uncertainty (0 ± 5 Oe), which is to be expected since the Néel temperature of CoO is usually below room temperature (293 K for bulk).¹⁷ Figure 8 summarises the angular dependent in-plane magnetic properties for sample 5 obtained at room temperature by taking multiple hysteresis loops at different

in-plane angles. The lack of any strong angular dependence for the quantities H_c^{FM1} and H_c^{FM2} confirms the polycrystalline nature of the ferromagnetic grains, which, on average, lack a uniaxial or biaxial in-plane easy-axis. It should be noted that, from the room temperature magnetometry presented in Fig. 7, it is ambiguous which of the quantities (H_c^{FM1} and H_{EB}^{FM1} or H_c^{FM2} and H_{EB}^{FM2}) belongs to the cobalt layer because neither of the dual loops gives the properties normally expected of cobalt. For instance, one could assume the typical case, which is that the cobalt layer has a higher coercivity (25–130 Oe)¹⁸ than the permalloy (2–10 Oe), in which case the outer loops belong to cobalt. However, in that scenario, the outer loop should have a step-size that is $\approx 2/3$ of the overall magnetic saturation, since bulk cobalt has 1400 emu/cm^3 versus bulk permalloy which has 780 emu/cm^3 ,¹⁹ and the layers are of almost the same thickness. Therefore, from the room-temperature magnetometry, there are two possibilities: either the cobalt has a reduced magnetic moment or the $\text{Ni}_{80}\text{Fe}_{20}$ has an enhanced coercive field. To resolve this anomaly, and correctly understand the magnetic depth profile of the sample in the saturated state, we conducted polarised neutron reflectometry at 4000 Oe and room temperature (i.e., in the saturated state).

FIG. 7. In-plane room temperature magnetometry data for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}(x)/\text{Co}$ samples 1–5, over-plotted to emphasize the similarity in all films. The $\text{Ni}_{80}\text{Fe}_{20}$ and Co ferromagnetic phases have different coercive fields, and are separated by a paramagnetic CoO spacer, resulting in the observed double step hysteresis.

C. Magnetic depth profile

Figure 9 reveals the polarised neutron reflectometry pattern obtained for sample 1. No spin-flip scattering was observed in the saturated state, implying a collinear arrangement of moments with the field.²⁰ Fitting of the two non-spin-flip (NSF) channels resulted in the magnetic depth profile illustrated on the right of Fig. 9. From the high-quality fit, it is obvious that there are missing magnetic moments located in the cobalt layer. The permalloy layer obtains a value close to its bulk magnetization ($0.9 \mu_B$ per formula unit $\text{Ni}_{80}\text{Fe}_{20}$) whereas the average magnetic moment in the cobalt layer is reduced to $0.4 \mu_B$ per Co, which is only 25% of the bulk value ($1.7 \mu_B$ (Ref. 21)). To prove that this magnetic depth-profile is the more feasible fit between the two possibilities elicited from the magnetometry data, Fig. 10 shows a low-quality fit resulting from the magnetic depth model where the cobalt retains a higher average magnetic moment ($0.9 \mu_B$ per formula unit), but the

FIG. 8. Polar plots of the angle-dependent in-plane magnetic quantities for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}/\text{Co}$ sample 5 obtained using Eqs. (1)–(4). The absence of strong angular dependence is a result of a random in-plane crystallite arrangement in both the ferromagnet and the antiferromagnet, manifested in the lack of an in-plane easy axis.

permalloy is oxidized ($0.4 \mu_B$ per formula unit). It is obvious that the model depicted in Fig. 9 is the only one of the two possibilities which accurately describes the data. Although in some magnetic multilayers, a non-collinear relationship of one ferromagnet with respect to the other has been found, leading to a similar reduction in the aligned magnetic moment, in that case, neutron spin-flip scattering would be expected,^{20,22} and it should not be possible to fit the NSF data with a collinear model.¹⁸ The best-fit nuclear scattering length density is also slightly higher for the cobalt layer, consistent with an increased oxygen-content. Recently, we studied CoO/Co bilayers fabricated in the same way and found that during the deposition of the CoO , oxygen ions penetrated into the underlying Co layer, forming pillar-like CoO features embedded in the Co layer.²³ It is believed that a similar mechanism is at work in these trilayer films, explaining the diffuse interface and the nanocomposite diffraction peaks found in the cobalt region. Both the coercivity enhancement and the lower magnetic saturation in the film series are attributed to the penetration of oxygen into the cobalt, and the formation of CoO nanoscale structures embedded within the Co layer. The interpenetrating CoO and Co_3O_4 regions are nominally antiferromagnetic and lower the average magnetization in the bottom layer, whilst also acting as defects and altering the dipolar interactions between the remaining “columns” of metallic cobalt. This agrees well with previous work showing that there is a direct correlation in partially oxidized cobalt thin films between the oxygen content and the coercivity enhancement/perpendicular anisotropy.²⁴

D. Exchange bias and layer-resolved interfacial energy

Sample 1 was field-cooled in one of two cooling fields (± 1000) Oe to 200 K, which is below the blocking

temperature of CoO . Figure 11 is the in-plane magnetic hysteresis measured for sample 1 under each of these two different field-cooling conditions, where only the first (untrained) loop is shown. In both cases, it is apparent that inner and outer loops shift either left or right, giving an exchange bias with the opposite sign to the cooling field. This is typical behaviour for most exchange bias systems. Table II summarises the quantities H_c^{FM1} , H_c^{FM2} , H_{EB}^{FM1} and H_{EB}^{FM2} for samples 1–5 under the +100 Oe cooling condition after first saturating at 10 kOe at 300 K.

The magnitude of the loop-shifts, H_{EB}^{FM1} , is found to be similar for all films within experimental uncertainty, whereas a maximum in H_{EB}^{FM2} is apparent for sample 3, which had a 89 nm CoO layer. The exchange bias shift is direct evidence for magnetic coupling between ferromagnetic ($\text{Ni}_{80}\text{Fe}_{20}$ or Co) and antiferromagnetic (CoO) spins. The lack of a strong spacer layer thickness implies that, in this case, the interfacial regions for the $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}$ and CoO/Co regions dominate the magnetic effects. We note that the loop-shift (H_{EB}^{FM1}) for the permalloy inner-loop is an order of magnitude smaller than that of Co at 200 K (H_{EB}^{FM2}). Moreover, the coercive field of the permalloy H_c^{FM1} is seen to be < 10 Oe for both the 300 K and 200 K measurements, but the cobalt layer shows a five-fold enhancement in coercive field at low temperature ($H_c^{FM2} \approx 1000$ Oe). Taking account of the saturation magnetization for the nanocomposite layer measured directly by polarized neutron reflectometry in the saturated state, along with thicknesses and exchange biases measured separately for the cobalt layers and permalloy layers, the interfacial energy $E_{int} = t_{FM} H_{EB} M_{sat}$ for the cobalt and permalloy layers can be calculated for each layer. The magnitude of the exchange bias for the cobalt layers in the five samples ($t_{FM} = 55$ nm, $M_{sat} = 400$ emu/m²) is anomalously high, leading to an estimate of the interfacial energy E_{int} in the range of 0.45–0.6 ergs/cm² at 200 K, which is higher than values

FIG. 9. Room temperature polarised reflectometry data fitted for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}(x)/\text{Co}$ (sample 1) using Model 1 (left) and fitting model for Model 1 (right). Circles are experimental data. Solid lines are fits to the data. The cobalt layer has a reduced magnetic moment of only $0.4 \mu_B$ per formula unit, suggesting oxygen implantation from ion-beam modification during growth.

FIG. 10. Room temperature polarised reflectometry data for $\text{Ni}_{80}\text{Fe}_{20}/\text{CoO}(x)/\text{Co}$ (sample 1) fitted using Model 2 (left) and fitting model for Model 2 (right). Circles are experimental data. Solid lines are fit to the data. The low quality of fit between the model and experimental data proves that the model with near-bulk Co magnetic moment cannot describe the data.

reported for typical thin films that use CoO ($0.16\text{--}0.28\text{ ergs/cm}^2$) measured at $>100\text{ K}$ with similar blocking temperatures.¹⁷ On the other hand, the interfacial energy for the permalloy layers ($M_{\text{sat}} = 790\text{ emu/cm}^2$, $t_{\text{fm}} = 55\text{ nm}$, and $H_{\text{EB}} = 10\text{ Oe}$) is $0.04\text{--}0.05\text{ ergs/cm}^2$, which is a more typical value for polycrystalline CoO pinning layers in thin films ($0.03\text{--}0.12\text{ ergs/cm}^2$). The vast difference in interfacial energies for the two ferromagnets coupled to nominally the same antiferromagnet implies that the modified nanocomposite interface structure at the Co/CoO has a strong, local effect on the exchange bias.

Along with the microstructural investigations, the increased low temperature exchange bias and coercivity in the Co/CoO layer provides indirect evidence for the implantation of oxygen into the cobalt layer, which alters the magnetic spin structure of the resulting nanocomposite, leading to stronger apparent coupling between the Co and CoO regions. One way to understand this is to remember that for two perfect thin film layers, there is only a single planar interface, whereas for a nanocomposite there can be numerous interfaces formed between Co/CoO, which may resemble an array of core-shell nanoparticles. In such a system, there is a natural tendency towards higher effective “surface” area for the interfacial magnetic coupling to occur. Indeed, the exchange bias effect was first discovered in core-shell Co/CoO nanoparticles,¹ and the loop shift of disordered magnetic particles is often higher than in the film equivalents. In general, for films with high quality interfaces, the exchange bias loop-shift usually decrease with a $1/t_{\text{FM}}$ relationship, where t_{FM} is the thickness of the ferromagnet layer.²⁵ The data, however, shows that the nanocomposite

region breaks this trend allowing for a strong exchange bias to exist even for a comparatively thick ferromagnetic layer (55 nm). This agrees with the recent finding that strong exchange bias could be realized in 100 nm epitaxial Co thin films ion-implanted with oxygen, although in that work, a high energy implantation energy was used (60 keV), presumably resulting in a different microstructure.³ It should be noted, that while increased, the interfacial energy is somewhat lower than the previous report of interfacial energies up to $2.5\text{--}3.5\text{ ergs/cm}^2$ in ion-beam modified Co/CoO bilayers, suggesting that the unique morphology in that case played an important role.

The vast difference in interfacial energies also implies a second important point: The two ferromagnets are probably decoupled across the spacer layer, since upon parallel and antiparallel field cooling; we find the exchange bias has the same magnitude for the individual ferromagnets, and the relative magnitude is unchanged. A previous study found that antiparallel field cooling led to a lower exchange bias in one of the two ferromagnets. It is also conceivable that, in the case of a negative interfacial exchange constant at one of the interfaces, one may expect the inverse result, therefore necessitating that all configurations are considered. To test this directly, we explored the different field-cooling configurations. To this end, sample 1 was saturated in $+1000\text{ Oe}$ at room temperature. It was then field-cooled to 200 K in one of 4 fields ($\pm 25\text{ Oe}$ or $\pm 1000\text{ Oe}$). In principle, the positive applied fields should not alter the exchange bias magnitude since they preserve the parallel alignment during cooling. A negative -25 Oe field, however, should reverse the permalloy layer during cooling, but preserve the positive direction in the cobalt, while a -1000 Oe field should reverse both the cobalt and permalloy causing their alignment to cool in a collinear-negative direction. Figure 12 shows that only the inner hysteresis is affected by the change in field-cooling

FIG. 11. In-plane magnetic properties of sample 1 at 200 K after field-cooling from 300 K to 200 K in either $+1000\text{ Oe}$ or -1000 Oe . Inset shows an enlarged region near the origin.

TABLE II. Exchange bias and coercivity at 200 K for $\text{Ni}_{80}/\text{Fe}_{20}/\text{CoO}/\text{Co}$ samples 1–5 after field-cooling in $+100\text{ Oe}$ (after initial saturation).

Sample	H_c^{FM1} (Oe)	H_{EB}^{FM1} (Oe)	H_c^{FM2} (Oe)	H_{EB}^{FM2} (Oe)
1	11	−12	1235	−265
2	5	−14	1285	−245
3	4	−9	1113	−293
4	4	−12	1134	−216
5	11	−12	1169	−221

FIG. 12. $\text{Ni}_{80}\text{Fe}_{20}$ (56 nm)/CoO(23 nm)/Co(58 nm) trilayer after saturating at room temperature, and field-cooling to 200 K in ± 25 Oe. The inset shows an enlargement of the low field region.

from +25 Oe to -25 Oe. This is consistent with the permalloy layer being reversed during cooling whereas the magnetic moments of cobalt remains in the same initial direction. Moreover, the fact that the absolute magnitude of the exchange bias is nearly equal in both cases suggests that we do not detect the type of effects found in Ref. 5. Using larger fields of 1000 Oe aligns both the cobalt and permalloy layer in the positive or negative direction. Figure 11 shows that in this case both the inner and outer loops are shifted in the same direction. The symmetry in both cases suggests that, to a large extent, the system behaves as two sets of independent ferromagnetic/antiferromagnet bilayers should, and there is no detectable coupling occurring across the CoO. This may well be due to the presence of grain boundaries in the polycrystalline CoO layer, as well as the role of competing interfaces in the Co nanocomposite. Although it neither proves nor disproves the particular result of Ref. 5, it certainly shows that a long length scale spin structure in the antiferromagnet is not a universal prerequisite for exchange bias in nanocrystalline systems, and it is quite possible to design spin-valve-like structures where this does not occur. In this case, the ion beam modification only affects one interface.

V. CONCLUSION

Fabrication of nanocrystalline thin films by low energy ion-assisted deposition leads to magnetic nanocomposites that show unusual magnetic properties that deviate from typical bulk materials and epitaxial thin films. Clear double step hysteresis loops were seen for all samples, with both ferromagnetic layers showing a low temperature exchange bias that was dependent on field-cooling conditions. A detailed microstructural study demonstrates the existence of multiple crystalline phases near the Co/CoO interface, along with oxygen in the underlying cobalt region due ion-beam

modification during deposition. This resulted in a larger coercivity and exchange bias for the bottom cobalt layer. However, the top $\text{Ni}_{80}\text{Fe}_{20}$ /CoO interface was nominally more ideal, and did not sense the enhanced exchange field.

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