



Modification of the ferromagnetic anisotropy and exchange bias field of NiFe/CoO/Co trilayers through the CoO spacer thicknesses

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We have investigated the magnetism of NiFe/CoO/Co trilayers with different CoO spacer thicknesses. The dependence of the coercivity (H_c) and exchange bias field (H_{ex}) on the CoO thicknesses indicated that different pinning strengths from the CoO were acting on the top NiFe and bottom Co layers, respectively. DC susceptibility indicated the different interlayer coupling energies and showed that the anisotropy of CoO layer strongly affected the temperature dependence of the magnetization. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4861216>]

Interlayer exchange coupling (IEC) has been of great interest due to its potential application in magnetoelectronic devices. Essentially IEC results from interfacial exchange and propagates through a spacer layer by RKKY interactions.¹ In a Fe/CoO/Co trilayer system,² changes in both magnetic domain structure (resulting from the competition between magnetocrystalline anisotropy and exchange) and Fe IEC have been observed by varying the CoO thicknesses. In particular, the formation of nanodomains that only occurred within a specific range of CoO thicknesses is connected with an instability generated by magnetic frustration at the FM/AF interfaces. Recently, Liu *et al.*³ have shown in Co/Cr₂O₃/Fe trilayers that the competition between IEC and interfacial coupling depends on the temperature, where for $T < T_N$ the interfacial exchange coupling dominates the magnetism, whereas the interlayer exchange coupling prevails at $T < T_N$. The strength of the exchange is usually characterized by the interfacial exchange coupling energy,⁴ $J_{ex} = H_{ex} M_s t_{FM}$, where the H_{ex} , M_s , and t_{FM} represent the exchange bias field, saturation magnetization and thickness of the FM layer, where H_{ex} and M_s have their intrinsic temperature dependencies (typically mean-field like). The interlayer coupling strength (usually expressed by^{3,5} $J(T)/(T/T_0)/\sinh(T/T_0)$ where $T_0 = \hbar v_F/2\pi\kappa_B d$ represents the characteristic temperature, and v_F , κ_B , and d are the Fermi velocity, Boltzmann constant, and spacer thickness, respectively) is known to increase with increasing temperature.⁶ IEC is dependent on the mechanism acting on the individual interfaces, and on the whole AF spin structure.

We studied the magnetic properties of FM NiFe and Co separated by different thicknesses of AFM CoO (with $T_N \sim 289$ K and anisotropy constant⁸ $K \sim 2 \times 10^5$ erg/cm³ (Ref. 8)). The evolution of H_{ex} and H_c as a function CoO thicknesses suggested changes in CoO domain structure. A two-step (or “kinked”) hysteresis loop or a shifted (large exchange bias, H_{ex}), but symmetric loop indicated either a step-wise or simultaneous reversal of the NiFe and Co layer magnetizations.

The trilayers were prepared on amorphous SiO₂ substrates by using a dual ion-beam sputtering deposition technique.⁹ A Kaufman ion source (800 V, 7.5 mA) was used to focus an Argon ion-beam onto a commercial Ni₈₀Fe₂₀ (at%) or Co target surface in order to fabricate the top NiFe or bottom Co layer. An End-Hall ion source ($V_{EH} = 50$ V, 500 mA) was used to *in-situ* bombard the substrate during deposition with a fixed 15% O₂/Ar mixture in order to fabricate the spacer CoO layer. The base pressure and working pressure during deposition were 3×10^{-7} Torr and 5×10^{-4} Torr, respectively. A JEOL (JEM-2010) transmission electron microscope (TEM) operating at 200 kV was used for the microstructural analysis. Magnetic hysteresis loop measurements were performed in a commercial ADE-DMS 1660 vibrating sample magnetometer (VSM) and a Quantum Design Magnetic Property Measurement System (QD MPMS) under a 12 kOe field-cooled (FC) process from 350 down to 10 K. The temperature dependence of the zero-field-cooled (ZFC) and FC (100 Oe) DC susceptibility ($M(T)$) of the NiFe/CoO/Co trilayers was measured with a QD MPMS.

The microstructures of the NiFe (12 nm)/CoO (4–20 nm)/Co (12 nm) trilayer thin films have been characterized by TEM, as shown in Fig. 1. In the NiFe/CoO (4 nm)/Co trilayer, polycrystalline f.c.c. NiFe ($a \sim 3.55$ Å), rock-salt CoO ($a \sim 4.32$ Å), and f.c.c. Co ($a \sim 3.54$ Å) were identified from electron diffraction patterns (insets in Fig. 1(a)). The grain sizes ranged from 3 to 15 nm, as shown in Fig. 1(a). Increasing the spacer (CoO) layer thickness between the top NiFe and bottom Co layer from 4 nm to 6 nm and 20 nm resulted in a small increase in the average layer grain size (5–20 nm) but did not change the structures. A representative cross-sectional TEM image of the NiFe/CoO (6 nm)/Co trilayer is shown in Fig. 1(b); interfaces between NiFe/CoO and CoO/Co are observed.

The temperature dependence of the hysteresis loops of NiFe/CoO (4 nm)/Co is shown in Fig. 2. At 10 K a typical signature of interfacial coupling from the two interfaces of NiFe/CoO and Co/CoO is revealed by the loop shift, as shown in Fig. 2(a). The two FM layers reverse together via coupling with the thin CoO layer (4 nm). However, at 160 K,

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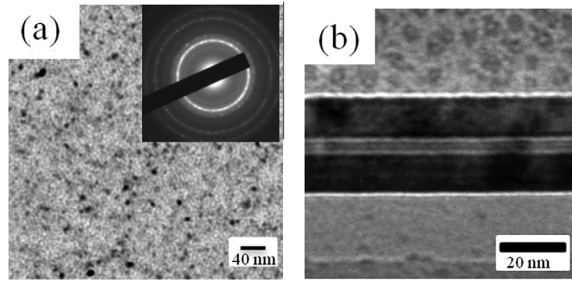


FIG. 1. Transmission electron microscopy (TEM) images of NiFe (12 nm)/CoO (6 nm)/Co (12 nm) showing (a) the planar-view, with the corresponding electron diffraction pattern shown in the inset, and (b) the cross-sectional view.

a rounded hysteresis loop with $H_{ex} = 0$ is seen (Fig. 2(b)). This indicates a decrease in the interfacial coupling, so that interlayer coupling is no longer overwhelmed with increasing temperature.¹¹ The blocking temperature $T_{B,ex}$ (above which $H_{ex} = 0$, < 160 K) is much lower than those found for similar thickness NiFe/CoO (Ref. 6) or Co/CoO bilayers.¹⁰ This reduced $T_{B,ex}$ in the trilayer is likely attributable to changes in the CoO layer domain structure since the AF CoO layer coupling is a competition between the two different FM layers (NiFe and Co) with different anisotropies; an equilibrium AF domain structure should be obtained after the field-cooling. In addition, the changes in the shape of the hysteresis loop at different temperatures suggests a transition in the magnetization reversal process from domain wall movement to domain rotation.¹² At 298 K, where the CoO is no longer antiferromagnetic (no longer pins the FM layer), and only IEC is present, the reversal process between the two FM layers results in a symmetric, yet rounded, hysteresis loop (Fig. 2(c)).

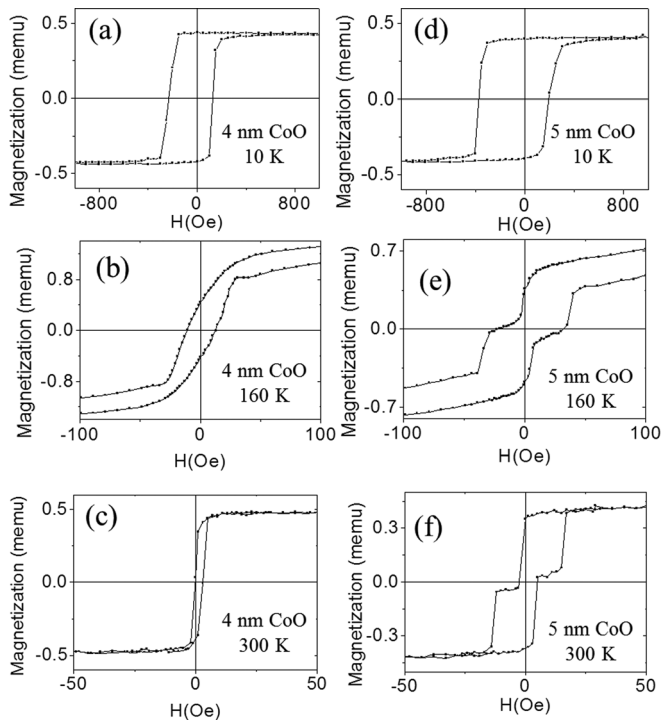


FIG. 2. Hysteresis loops for NiFe/CoO (t nm)/Co collected at 10 K ((a) and (d)), 160 K ((b) and (e)), and 300 K ((c) and (f)) after field cooling in 12 kOe in the film plane from 350 K. Results are shown for $t_{CoO} = 4$ nm ((a)–(c)), and $t_{CoO} = 5$ nm ((d)–(f)).

The role of a greater AF CoO thickness creating more pinning sites and stronger exchange (bias) coupling to the two FM layers is shown by the NiFe/CoO (5 nm)/Co trilayer in which both stronger H_{ex} and the largest coercivity H_c at 10 K were measured (Fig. 2(d)). However, at 160 K a decrease in the interfacial coupling and an increase in the IEC relative NiFe/CoO/Co at 160 K is observed, as evidenced by a clear two-phase hysteresis loop (Fig. 2(e)), typical of IEC.¹⁵ The larger H_c (~ 28 Oe) (cf. $H_c \sim 12$ Oe in a trilayer with $t_{CoO} = 4$ nm) but no H_{ex} is attributed to the larger switching field required to reverse the Co layer (set by AF CoO pinning layer) after the soft NiFe layer has been reversed at a smaller field. The double hysteresis loop is maintained even at 298 K (Fig. 2(f)). The transition from single loop (two FM layers reverse together) to two-phase loops at different temperatures for the trilayer films reflects the competition between interfacial exchange coupling (that dominates at much lower temperatures) and IEC.¹⁴

The hysteresis loops of the trilayers with thicker CoO layers ($t_{CoO} = 6, 12,$ and 20 nm) at 10 K are shown in Fig. 3. The NiFe/CoO (6 nm)/Co trilayer (Fig. 3(a)) exhibited a kink in the third quadrant with an increase in H_{ex} and a decrease in H_c . In the increasing field branch (first quadrant), no step or kink is observed and the magnetization of the FM layers reverse in a smaller field (~ 200 Oe) in order to reduce the interfacial energy.¹³ Further, no kink or step in the hysteresis loops is found in this trilayer either at 160 K or at 298 K. The different reversal processes (either step-wise or simultaneous for different CoO thicknesses [e.g., $t_{CoO} = 5$ nm (Fig. 2(d)) and 6 nm (Fig. 3(a))] are likely due to (1) changes in the FM domain structures via coupling to the CoO layer,¹⁵ (2) coupling either by dipole interactions or an oscillatory interlayer coupling,¹⁶ and (3) the enhancement of the ferromagnetic anisotropy.¹⁷ Further, doubling the CoO thickness from 6 to 12 nm (Fig. 3(b)) resulted in the largest H_{ex} at 10 K amongst all trilayers, while the coercivity ($H_c \sim 250$ Oe) was similar to the other films. This indicated that while the thicker CoO layer can pin the FM layer (resulting in an enhanced H_{ex}), the asymmetry of the hysteresis loop steps from different t_{CoO} (6 nm and 12 nm) implies (1) a possible formation of spiral

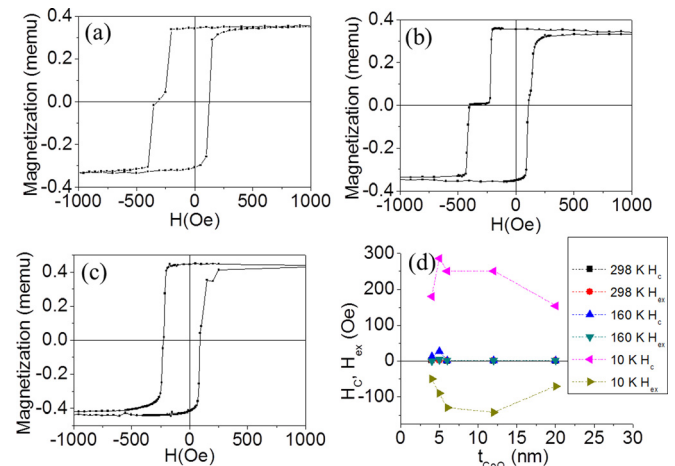


FIG. 3. The hysteresis loops of NiFe/CoO (t nm)/Co trilayers measured at 10 K: (a) $t_{CoO} = 6$ nm, (b) $t_{CoO} = 12$ nm, and (c) $t_{CoO} = 20$ nm. The variation of H_c and H_{ex} vs. CoO thicknesses is shown in Fig. 4(d).

domains near FM interfaces via coupling to the CoO layer¹⁸ or (2) the presence of the coupling either by dipole interactions or an oscillatory interlayer coupling.¹⁶ However, when increasing the temperature to 160 K or 298 K, the trilayers exhibited similar magnetic properties (single phased hysteresis loops with $H_c \sim 1-2$ Oe). In contrast, for the trilayer with the thickest $t_{\text{CoO}} = 20$ nm, the step in the third quadrant of the hysteresis loop disappeared at 10 K, accompanied by a decrease in $H_c \sim 155$ Oe and $H_{\text{ex}} \sim -70$ Oe (Fig. 3(c)), while in the ascending field branch the step in the first quadrant indicated there might be incoherent domain rotation upon saturation.¹¹

The CoO thickness dependence of H_c and H_{ex} for all trilayer films is shown in Fig. 3(d). The magnitude of H_{ex} increases with increasing CoO thicknesses up to 12 nm, and decreases for the trilayer with the thickest $t_{\text{CoO}} = 20$ nm. The H_{ex} increase can be attributed to the pinning of the AF CoO spins to the NiFe and Co interfaces, whereas the decrease in H_{ex} for $t_{\text{CoO}} = 20$ nm could be explained by the domain state model¹⁹ in which the H_{ex} is predicted to be inversely proportional to the AF domain size (closely related to the AF thicknesses). Accordingly, a decreased number of AF domain walls is expected for thicker films, which results in weaker exchange coupling and reduction of H_{ex} . We have shown in a reference NiFe (10 nm)/CoO (20 nm) bilayer²⁰ and Co (10 nm)/CoO (20 nm) bilayer¹⁰ that the interfacial coupling energy, $J_{\text{ex,Co/CoO}} \sim 23.4 \times 10^{-2}$ erg/cm² is greater than $J_{\text{ex,NiFe/CoO}} \sim 1.44 \times 10^{-2}$ erg/cm² at 200 K after FC. However, in the present case of the NiFe/CoO/Co trilayers, since the AF CoO layer is shared by the FM layers with different anisotropies and exchange coupling strengths, different equilibrium AF domain structures would be obtained for the interfaces between both NiFe and Co layers. Thus, the final interfacial exchange energy could either increase or decrease, depending on the relative strengths of J_{ex} at the two interfaces and the micromagnetic energy of the AF.¹¹ For $t_{\text{CoO}} \geq 5$ nm H_c increases with decreasing t_{CoO} , consistent with H_c enhancement from irreversible transitions of the AF domain states resulting from thermal activation.¹⁵ Thus, it is expected that

the trilayers with thinner CoO layers are more prone to the thermal fluctuation effects and thus possess larger H_c . The further decrease in H_c for the trilayer with thinnest t_{CoO} (4 nm) is likely due to the weakening of the CoO anisotropy from finite size effects such that the coupling strength of CoO to the two FM layers is decreased and H_c becomes close to that of a plain NiFe/Co bilayer.

For the NiFe/CoO (20 nm)/Co trilayer, a divergence between ZFC and FC curves was observed, shown in Fig. 4. The maximum of the ZFC magnetization is related to the thermal demagnetization of the layer as the crystallites become superparamagnetic (thermal fluctuation effects described above)⁷ at a blocking temperature, $T_{\text{B,SP}}$. Below $T_{\text{B,SP}}$, the difference in magnetization ($\Delta M_{\text{FC-ZFC}}(T)$) between FC and ZFC curves increases with decreasing t_{CoO} at the lowest temperature (10 K), as shown with Figs. 4(a)–4(c). This behavior qualitatively indicates that the exchange coupling that sets the energy where thermal fluctuations of domains can occur between two interfaces of NiFe/CoO and Co/CoO becomes significant at lower temperatures, an interpretation consistent with our previous work. Since T_{B} for all trilayers was ~ 60 K, $\Delta M_{\text{FC-ZFC}}$ (10 K) indicates a change in the blocking behavior that is reflected clearly in $M_{\text{FC}}(T)$ (Fig. 4(d)). In trilayers with $t_{\text{CoO}} > 5$ nm $\Delta M_{\text{FC-ZFC}}$ (10 K) decreases with increasing t_{CoO} . This indicated that the thicker CoO layers were able to stabilize the FM magnetization for $T < T_{\text{B,SP}}$. It is also possible that the weakening in FM-FM coupling (either dipolar or IEC) due to larger FM layer separation with increasing t_{CoO} contributed to the decrease in $\Delta M_{\text{FC-ZFC}}$ (10 K) with t_{CoO} . The larger $\Delta M_{\text{FC-ZFC}}$ (10 K) at thin t_{CoO} implies that the changes in domain structures of the CoO layer resulted in a weakening of the anisotropy, and the layer could not stabilize the FM magnetization during the $M_{\text{FC}}(T)$ measurement for $T < T_{\text{B,SP}}$. This was in contrast to the trilayers with thicker t_{CoO} , which had a more bulk-like CoO anisotropy. Our results indicate that the magnetic properties (in particular H_c and H_{ex}) can be mediated by CoO thicknesses.

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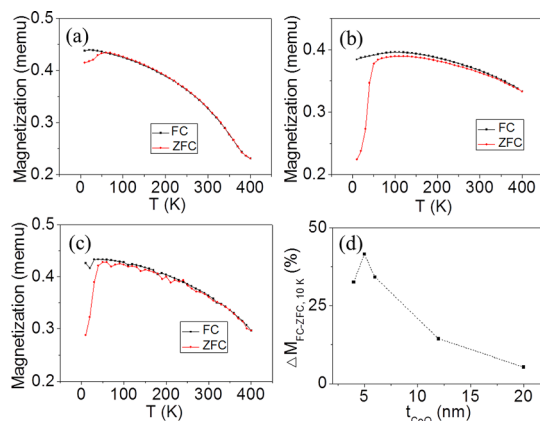


FIG. 4. The temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetization of NiFe/CoO (t nm)/Co trilayers ((a) 20 nm, (b) 5 nm, and (c) 4 nm) and (d) the difference in magnetization ($\Delta M_{\text{FC-ZFC}}$) at 10 K for each film for comparison of the exchange coupling and interlayer coupling strengths.

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