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

K.-W. Lin,1,a) T.-C. Lan,1 C. Shueh,1 E. Skoropata,2 and J. van Lierop2,a)
1Department of Materials Science and Engineering, National Chung Hsing University, Taichung 402, Taiwan
2Department of Physics and Astronomy, University of Manitoba, Winnipeg R3T 2N2, Canada

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We have investigated the magnetism of NiFe/CoO/Co trilayers with different CoO spacer thicknesses. The dependence of the coercivity (Hc) and exchange field (Hex) 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 magnetocrystalline anisotropy and exchange 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 Fe IEC have been observed by varying the CoO thicknesses. In particular, the formation of nanodomains that only resulted from electron diffraction patterns (insets in Fig. 1(a)). The microstructures of the NiFe (12 nm)/CoO (5–20 nm)/Co are observed.

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 (VeH = 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 × 10−10 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 ~ 3.55 Å), rock-salt CoO (a ~ 4.32 Å), and f.c.c. Co (a ~ 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,
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. 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. 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. 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)).

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. The larger $H_c$ ($\sim28$ Oe) (cf. $H_c$ $\sim12$ 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.

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 ($\sim200$ Oe) in order to reduce the interfacial energy. 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, (2) coupling either by dipole interactions or an oscillatory interlayer coupling, and (3) the enhancement of the ferromagnetic anisotropy. 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 ($H_c$ $\sim250$ 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
domains near FM interfaces via coupling to the CoO layer or (2) the presence of the coupling either by dipole interactions or an oscillatory interlayer coupling. 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 $t_{CoO} = 20$ 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 $H_{ex} \sim -70$ 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.

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 $t_{CoO} = 20$ 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 $t_{CoO} = 20$ nm could be explained by the domain state model 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) bilayer and Co (10 nm)/CoO (20 nm) bilayer that the interfacial exchange energy, $J_{ex,CoO/CoO} \approx 23.4 \times 10^{-7}$ erg/cm$^2$ is greater than $J_{ex,NiFe/CoO} \approx 1.44 \times 10^{-2}$ erg/cm$^2$ 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. For $t_{CoO} \geq 5$ 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. 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) at a blocking temperature, $T_{B,SP}$. Below $T_{B,SP}$, the difference in magnetization ($\Delta M_{FC,ZFC}(T)$) 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 $\sim 60$ K, $\Delta M_{FC,ZFC}(10 K)$ indicates a change in the blocking behavior that is reflected clearly in $M_{FC}(T)$ (Fig. 4(d)). In trilayers with $t_{CoO} > 5$ nm $\Delta M_{FC,ZFC}(10 K)$ decreases with increasing $t_{CoO}$. This indicates that the thicker CoO layers were able to stabilize the FM magnetization for $T < T_{B,SP}$. 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 $\Delta M_{FC,ZFC}(10 K)$ with $t_{CoO}$. The larger $\Delta M_{FC,ZFC}(10 K)$ 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 $M_{FC}(T)$ measurement for $T < T_{B,SP}$. 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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**References**