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Origin of anomalously high exchange field in antiferromagnetically coupled magnetic structures: Spin reorientation versus interface anisotropy

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Magnetization reorientation from in-plane to perpendicular direction, observed in Co thin film coupled antiferromagnetically to high perpendicular magnetic anisotropy (Co/Pd) multilayers, is studied systematically for Co thickness ranging from 0 to 2.4 nm. The sample with 0.75 nm thick Co showed an exchange coupling field (H_{ex}) exceeding 15 kOe at room temperature and 17.2 kOe at 5 K. With an increase of Co thickness, H_{ex} decreased as expected and beyond certain thickness, magnetization reorientation was not observed. Indeed, three regions were observed in the thickness dependence of magnetization of the thin layer; one in which the thin layer (in the thickness range up to 0.8 nm) had a perpendicular magnetic anisotropy due to interface effects and antiferromagnetic coupling, another in which the thin layer (0.9–1.2 nm) magnetization had no interface or crystallographic anisotropy but was reoriented in the perpendicular direction due to antiferromagnetic coupling, and the third (above 1.2 nm) in which the magnetization was in-plane. In addition, Hall effect measurements were carried out to observe the anomalous and planar Hall voltages and to quantify the perpendicular and in-plane components of magnetization. The sample with thicker Co layer (2.4 nm) showed an in-plane component of magnetization, whereas the sample with 0.75 nm Co showed no in-plane component. The high value of H_{ex} observed in 0.75 nm Co samples can have important implications in spintronics and bit patterned media. © 2011 American Institute of Physics. [doi:10.1063/1.3658843]

I. INTRODUCTION

Materials such as Co/Pd multilayers and FePt with high perpendicular magnetic anisotropy (PMA) are intensively studied for bit patterned media (BPM) (Refs. 1–5) and heat assisted magnetic recording (HAMR).^{6–8} These two alternative technologies are considered as potential techniques to overcome the superparamagnetic effect of the current granular media based magnetic recording technology beyond 1 Tbit/in². These materials are also being investigated for magnetic random access memory (MRAM) applications.^{9,10} In BPM technology, besides write-synchronization and manufacturing issues, wide switching field distribution (SFD) of media is a crucial problem that needs to be addressed. One of the main factors that can exacerbate the SFD is the dipolar interaction between magnetic bits.^{1–5} Antiferromagnetically coupled (AFC) bit patterned structure was proposed to study the effect of dipolar interactions and as a potential candidate to minimize the SFD.^{11–13} In AFC structure, when two ferromagnetic layers are separated by a very thin nonmagnetic layer such as Ru, magnetizations of the recording layer (thicker film) and stabilizing layer (thinner film) are aligned anti-parallel to each other.¹⁴ To achieve AFC after patterning, it is important to have higher antiferromagnetic (AF) exchange coupling field (H_{ex}) than the coercivity of stabiliz-

ing layer.^{12–15} Therefore, materials which show a high H_{ex} are desired. A thin Co layer antiferromagnetically coupled to a magnetic layer with perpendicular anisotropy provides such a possibility.¹⁶

Moreover, magnetization reorientation is an interesting phenomenon for academic curiosity. Our preliminary study on magnetization reorientation looked at a few samples only at discrete set of thickness values, whereas a detailed study would be needed to understand this observation further. Therefore, in this paper, we have carried out a systematic study on spin-reorientation at room temperature and 5 K. We report a very high H_{ex} of 15 kOe at room temperature and 17.2 kOe at 5 K between a 7.5 Å-thick Co thin film and (Co/Pd) multilayer. In addition to conventional magnetometric techniques, we have used anomalous Hall effect (AHE) technique to observe the components of magnetization and to further understand the magnetization reorientation in such complex structures.

II. EXPERIMENTAL DETAILS

Thin film samples were deposited using dc magnetron sputtering system at 1.5 mTorr Ar pressure on thermally oxidized Si (100) substrates. The base pressure prior to the deposition was below 5×10^{-9} Torr. The samples had a structure of Ta (5 nm)/Cu(5 nm)/Pd(3 nm)/Co(*t*)/Ru(0.8 nm)/Co(0.5 nm)/[Co(0.3 nm)/Pd(0.8 nm)]_{×15}/Pd(3 nm)/Ta (3 nm), and a schematic of the film structures is shown in Figure 1.

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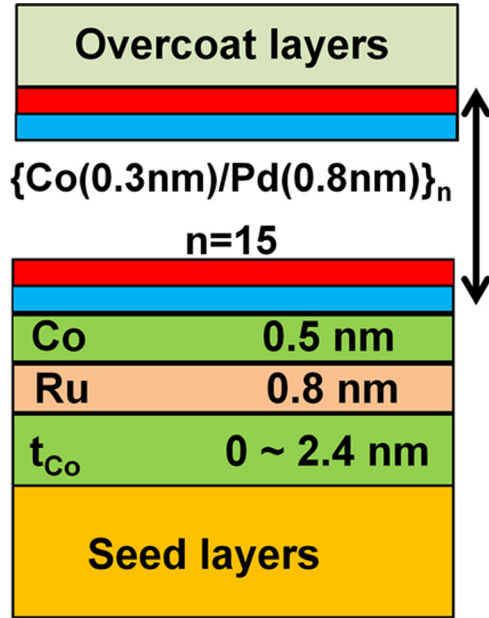


FIG. 1. (Color online) Schematic illustration of complex multilayer structures with different thickness of Co thin film (0–2.4 nm).

The thickness, t , of bottom Co layer was varied from 0.4 nm to 2.4 nm. The structural and magnetic properties were investigated using x-ray diffractometer (XRD), alternating gradient magnetometer (AGM), and superconducting quantum interference device (SQUID) magnetometer.

For the AHE measurements, the Hall cross-bar was first lithographically patterned onto thermally oxidized Si substrates using the Karl-Suss mask aligner and a positive-tone photoresist. Materials consisting of the film stack were then deposited into the exposed region followed by a lift-off process. Contact pads were then fabricated in the same manner onto each arm of the Hall bar and finally wire-bonded to a chip carrier for measurement. Figure 2 shows a typical scanning electron microcopy (SEM) image of a fabricated device ready for AHE measurements. A vibrating-sample magnetometer (VSM) with magnetoresistance (MR) controller was used to perform the AHE measurements. The magnet

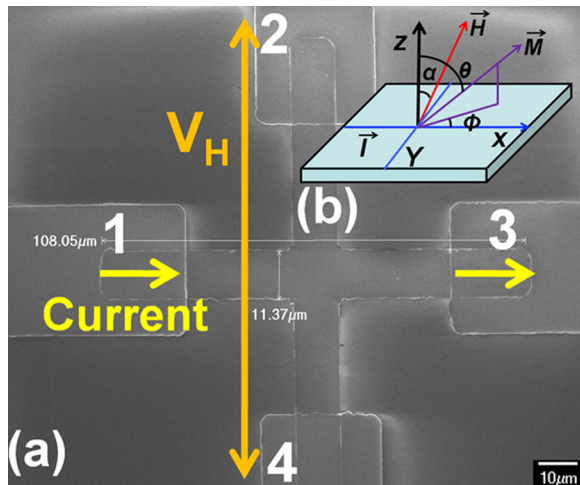


FIG. 2. (Color online) (a) SEM image from Hall bar and (b) schematic diagram showing the relative angles between the external magnetic field H , magnetization M , and the current I .

in the VSM provides a field up to 20 kOe at various angles with respect to the film normal. The MR controller provides a constant current of 2 mA through the Hall bar and measures the resistance in a direction perpendicular to the current flow. The Hall voltage is obtained by multiplying the measured resistance with the applied current as shown in following equation:^{17–21}

$$V_H = \left(\frac{R_H I}{t} \right) H \cos(\alpha) + \left(\frac{\mu_0 R_{SH} I}{t} \right) M \cos(\theta) + \left(\frac{K I}{t} \right) M^2 (\sin \theta)^2 \sin(2\phi) + V_{offset} \quad (1)$$

where t is the layer thickness, R_H the ordinary Hall coefficient, R_S is the anomalous Hall coefficient, and K is the planar-Hall coefficient.

III. RESULTS AND DISCUSSION

Figure 3 shows the perpendicular hysteresis loops of exchange coupled structures with different thickness of Co bottom layer as measured by AGM. The inset in Figure 3 shows the enlarged images of the magnetization reversals of the Co bottom layer. A very high exchange coupling field of 15 kOe can be observed for 0.75 nm thick Co. With the increase of Co thickness to 1 nm, the exchange field reduces to a value of about 10 kOe. For relatively high values of Co thickness (e.g., 2.4 nm), no exchange coupling was observed. These results are, to some extent, very similar to our earlier observations where—at Co thickness of 1 nm—we reported magnetization reorientation of Co magnetization from in plane to out of plane direction. For a Co layer thickness of about 2.4 nm, no magnetization reorientation could be observed. While the reversal characteristics of the 1 nm Co layer is very similar to that in our earlier work, the obvious difference in the present study is the sharp magnetization reversal characteristics of the 0.75 nm Co layer. Table I summarizes the switching field distribution, as measured from the width of the peaks in differentiated M-H loops. It can be noticed that the SFD increases from 0.6 kOe for t of 0.75 nm to a value of 1.6 kOe for t_{Co} larger than 0.9 nm. While the SFD in our earlier work is as high as 1.6 kOe, the relatively

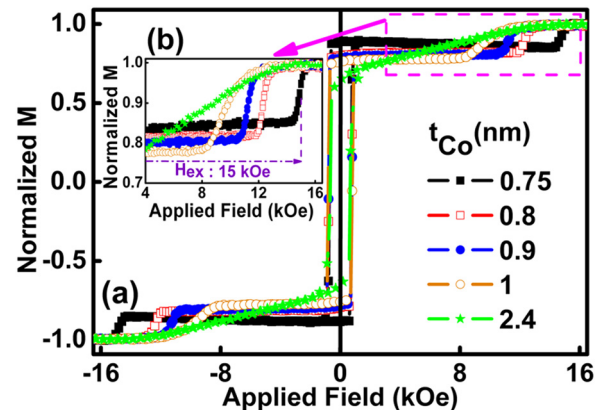


FIG. 3. (Color online) (a) Hysteresis loop complex multilayer system (CS) and (b) Zoomed in view of the exchange coupling.

TABLE I. Switching field distribution, as measured by the full width at half-maximum of the derivatives of the M-H loops Co layers in perpendicular and in-plane magnetization direction, in single-layer and complex multi-layer system.

Switching field distribution (kOe)			
Co thickness (nm)	Complex system	Thin film (perpendicular)	Thin film (in-plane)
0.75	0.6	0.35	2
0.9	0.6	1	0.37
1	1.6	2.2	0.37

sharp switching and a large exchange coupling in 0.75 nm was not observed.

In order to understand this effect further, thin film samples of the type Ta (5 nm)/Cu(5 nm)/Pd(3 nm)/Co(*t*)/Ru(0.8 nm)/Pd(3 nm)/Ta (3 nm) were made. This sample structure, without the complexity of the multilayers on the top, helps to understand the anisotropy of the thin Co layer per se. Figure 4 shows the hysteresis loops of the single-layered Co films, measured when field was applied in perpendicular and in-plane directions. It can be noticed that the sample with $t=0.75$ nm shows a PMA with a narrow SFD (as seen from Table I), possibly induced by a contribution of surface and magneto-crystalline anisotropy arising from the Pd(111) seedlayer below.²² The PMA vanishes for thickness greater than 0.9 nm, and the films show in-plane magnetization orientation. The SFD of the films greater than 0.9 nm is larger in the perpendicular direction and narrower in the in-plane direction (Table I).

The following discussion can be made from the results shown in Figures 3 and 4. In the complex multilayers, the Co layer at the bottom has a PMA for a maximum thickness of about 0.85 nm. Therefore, for samples with a Co layer

thickness of 0.7–0.85 nm, there is no magnetization reorientation. The bottom layer is antiferromagnetically coupled to the top Co/Pd multilayer through the Ru layer, as shown in several other studies.^{11–13} For the films with a Co layer thickness of 0.9 and 1 nm, magnetization reorientation arises due to the antiferromagnetic coupling and hence is the observation of an exchange field. For too large values of Co layer thickness (~ 2.4 nm), the antiferromagnetic coupling is not strong enough to cause reorientation, and hence, the magnetization simply lies in the film-plane in the absence of an external magnetic field.

Figure 5 shows the values of H_{ex} as a function of thickness of the bottom Co layer. The H_{ex} shows a steady decrease until a thickness of about 1 nm, beyond which the exchange coupling cannot be observed. Three regions corresponding to the different behavior of bottom Co layer are marked. In region (I), Co thin layer (in the thickness range up to 0.8 nm) has a PMA due to interface effects and is antiferromagnetically coupled to (Co/Pd) multilayer. In region (II), where the bottom Co layer thickness is between 0.9 nm and 1.2 nm, the magnetization of the film as such had no PMA but was reoriented due to the antiferromagnetic coupling with the Co/Pd multilayers on the top. In the third region, for large values (> 1.2 nm) of thickness, H_{ex} vanishes due to the in-plane orientation of the Co layer. This result is in contrast with the case of conventional AFC system, where the value of H_{ex} decreases monotonically as a function of thickness.^{11,12,23} Another interesting point to notice from this study is that the value of H_{ex} in the complex multilayers is much larger than that observed in our earlier AFC systems.^{16,23}

As the value of H_{ex} increases inversely proportional to the thickness, it is meaningful to look at the value of exchange coupling energy J , as calculated from the formula $J = H_{ex} \cdot M_S \cdot t$, where $M_S \times t$ is the magnetic areal moment.

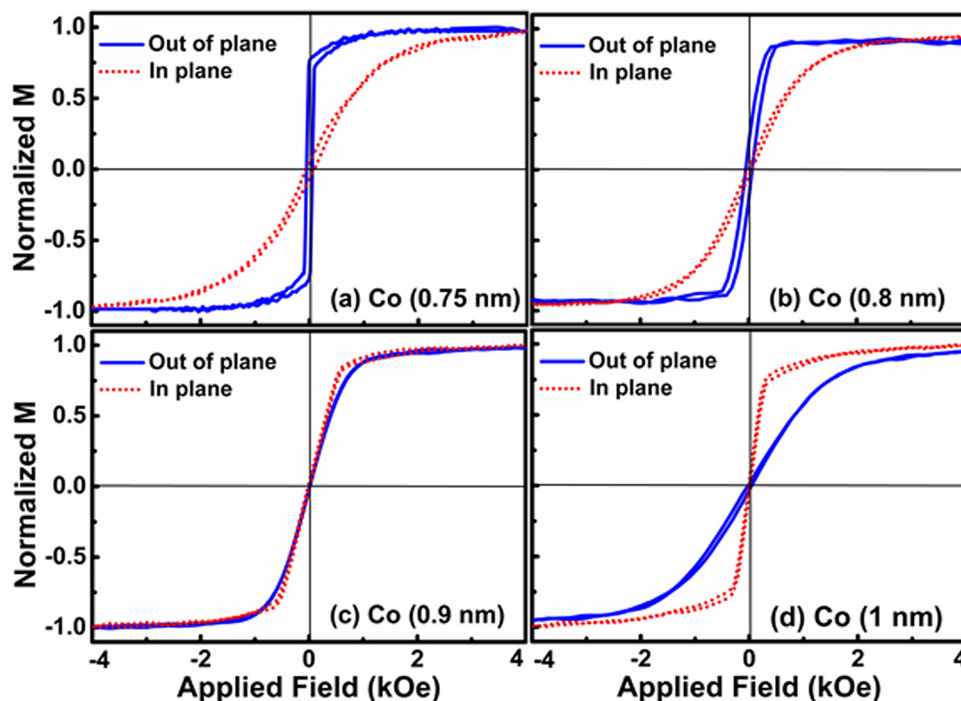


FIG. 4. (Color online) Out of plane and in-plane magnetizations of Co thin film with thickness of (a) 0.75 nm, (b) 0.8 nm, (c) 0.9 nm, and (d) 1 nm, respectively.

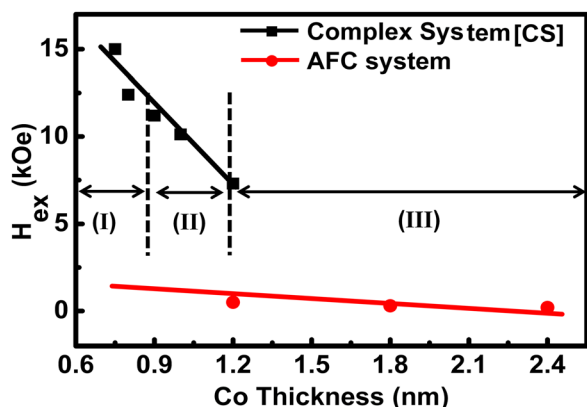


FIG. 5. (Color online) Exchange coupling field (H_{ex}) versus Co thickness for complex system in this study and a typical AFC system.

In our samples, J was found to be close to 1.24 erg/cm^2 for a sample with 0.75 nm thick Co layer. In the case of longitudinal recording media, an increase of J from 0.08 to 0.32 erg/cm^2 has been reported by the introduction of thin Co layers at the Ru interface.^{23–25} In the case of perpendicular media with antiferromagnetically coupled configuration, J was found to be in the range of 0.2 to 0.44 erg/cm^2 .^{12,24} In the pioneering work on oscillatory coupling in Co/Ru/Co, Parkin *et al.* have reported a value of J of about 5 erg/cm^2 .²⁶ From the hysteresis loops plotted in Fig. 3, a saturation field of about 14 kOe for 2.4 nm thick Co layer can be observed. Using the method of Parkin *et al.*, this yields a J value of 4.7 erg/cm^2 , which is very close to the reported value.^{14,26}

In order to confirm if this higher value of saturation field is due to AF coupling, hysteresis loop of 2.4 nm Co layer without Co/Pd multilayers was measured by AGM for comparison. Fig. 6 shows the hysteresis loops for the two cases. A saturation field of 9.7 kOe was observed for single Co layer which is smaller compared to the case (14 kOe) when it is exchange coupled antiferromagnetically to (Co/Pd) multilayer. This result is in contrast to what was observed in case of two ferromagnetically coupled layers.^{27,28} In the case of two magnetic layers that were coupled ferromagnetically, the saturation field was lower as compared to a single layer. These results indicate that the difference of 4.3 kOe is the

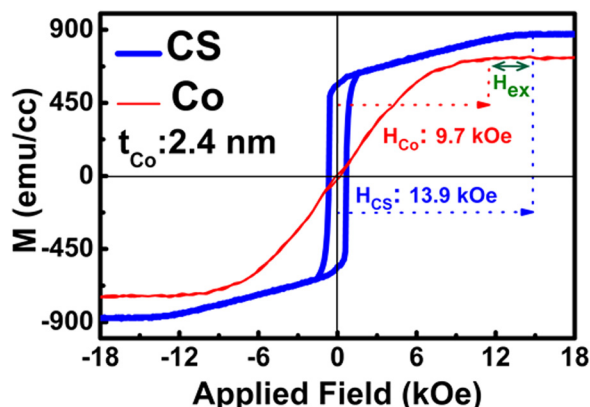


FIG. 6. (Color online) M-H loop of complete CS with 2.4 nm thick Co layer and only 2.4 nm Co layer without Co/Pd.

magnitude of field needed to overcome the antiferromagnetic coupling.

In order to further confirm that the differences in the crystallographic structure for different thickness of Co layers are not the cause of the observed phenomenon, rocking curves were measured by XRD. The full width at half maximum (FWHM) was found to be in the range of 4° to 5° (shown in Fig. 7(b)) indicating that the crystallographic growth of the (Co/Pd) multilayer was not affected. The absence of any significant trend with Co thickness (0.9 – 1.2 nm) indicates that the reorientation is essentially magnetic in origin.

AHE provides the in-plane and perpendicular components of magnetization. As the signal from AHE is inversely proportional to the film thickness, it is a useful technique to study magnetization components of these samples in detail.^{17,29} This is especially important, as the thickness of the Co-film is in the range of 0.7 nm to 2.4 nm and the VSM and AGM measurements do not separate out the different components. Therefore, we carried out a systematic study of these samples using AHE at various angles and applied fields.

We followed the method of extracting the in-plane and out of plane magnetization signal as reported elsewhere.^{17–20,29} Fig. 8(a) shows anomalous Hall voltage (AHV) and planar Hall voltage (PHV) for the case of 0.75 nm thick Co layer exchange coupled to 17 nm (Co/Pd) multilayer. Each curve was measured for different angles α between the film-normal and applied magnetic field. It can be noticed from the AHV loops that the kink representing the magnetization reversal for Co layer with thickness up to 1.2 nm decreased in magnitude as the angle α varied from film-normal toward in-plane direction. Consequently, an increase of the in-plane magnetization component was observed by changing the angle of applied magnetic field from 0° to 85° as shown in Fig. 8(b). Since ordinary Hall voltage which is proportional to the magnetic field has been subtracted from AHV, the tilt in hysteresis curves at large angles shown in Fig. 8(a) is due to genuine reduction in the perpendicular component of magnetization. For the case of 0.75 nm -thick Co, it was observed that the magnetization of Co layer is strongly antiferromagnetically

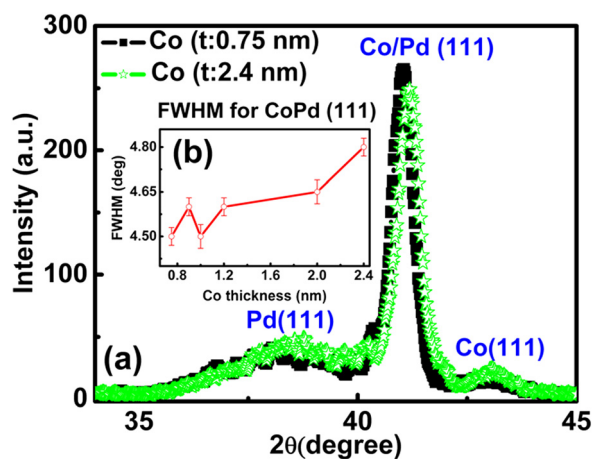


FIG. 7. (Color online) (a) X-ray θ - 2θ scan of complete CS with thickness of Co thin film 0.75 nm and 2.4 nm , (b) FWHM of Co (FCC:111) peak for complete complex multilayer.

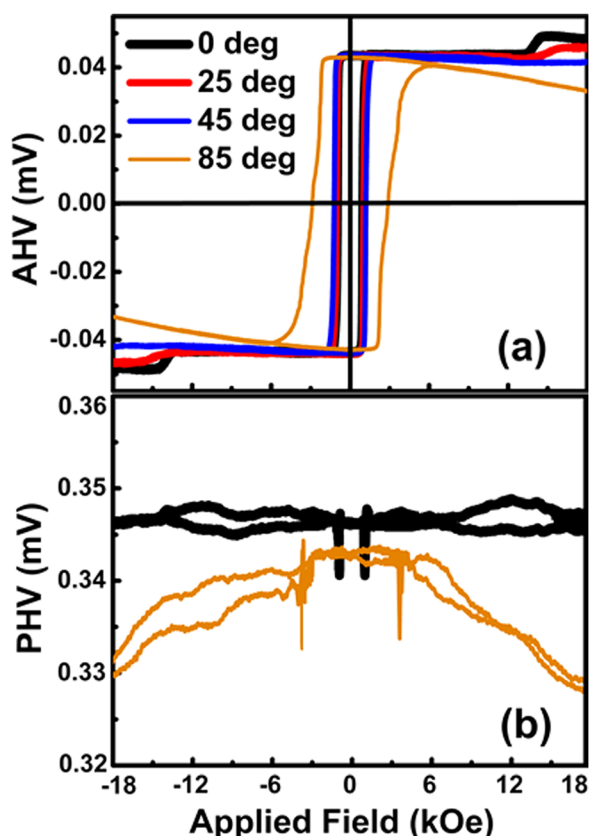


FIG. 8. (Color online) (a) AHV and (b) PHV at different angles for Co layer thickness of 0.75 nm.

coupled with the (Co/Pd) multilayer. The AHE measurements, shown in Fig. 9(a), also revealed that the amount of in-plane magnetization component becomes higher when the Co layer thickness was increased. As a result, spin reorientation could not be achieved for thicker Co layer. Different magnetization states for the case of 2.4 nm Co layer have been identified in Fig. 9(a) as indicated by the numbers 1 to 4. Until state 1, the magnetization of Co layer is in the perpendicular direction due to Zeeman energy. From state 1 to 2, the magnetization starts to rotate towards the in-plane direction. From state 2 to 3, the switching of the (Co/Pd) multilayer with perpendicular anisotropy occurs. The rotation of the magnetization of 2.4 nm-thick Co layer due to Zeeman energy continues until state 4, where saturation occurs.

The corresponding in-plane magnetization curves for film with 2.4 nm Co thickness are shown in Fig. 9(b). The in-plane magnetization curves show M-shape reversal loops which is typical of films with both in-plane and perpendicular anisotropy. However, the M-shape loops are only present for exchange coupled structures with bottom Co layer of 2.4 nm, indicating that there was no reorientation in this sample. The in-plane magnetization reaches peak value twice; this is due to the magnetization rotation in the x-y plane (change in the angle ϕ). As magnetization switches from $-z$ to $+z$ direction, ϕ changes by 180° , but the planar Hall voltage changes by 2ϕ , hence the magnitude of the in-plane magnetization is the same. In addition, angle dependence of perpendicular coercivity was measured for different Co thickness, and it was observed that the coercivity increases

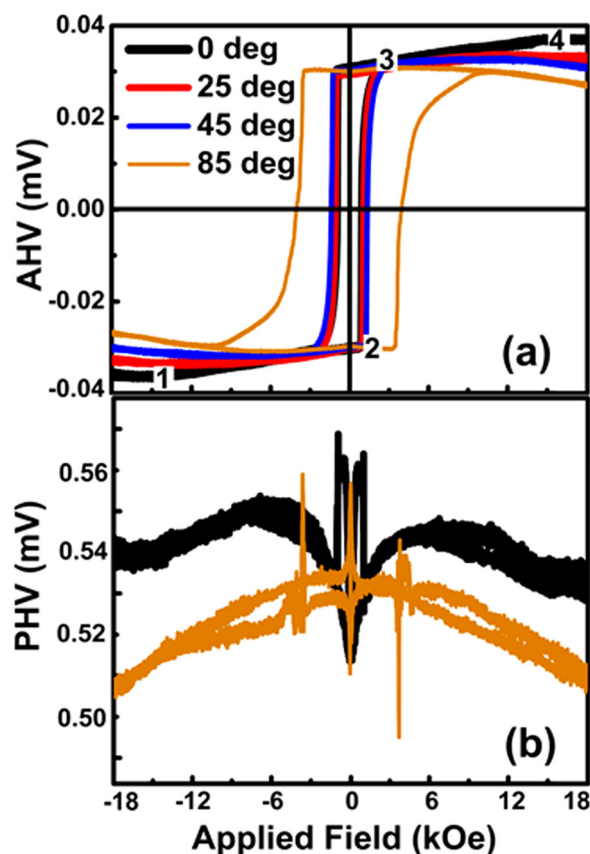


FIG. 9. (Color online) (a) AHV and (b) PHV at different angles for Co layer thickness of 2.4 nm.

with the angle α (shown in Figure 10). This behavior indicated that the magnetization reversal in these films is due to domain wall motions.³⁰

In order to understand the interlayer exchange coupling further, the temperature dependency of magnetization in complex structures with different Co thickness film thicknesses (0.75 nm and 2.4 nm) was studied using SQUID. The minor loops of these samples were measured with field increment of 150 Oe at different temperatures ranging from 5 K to

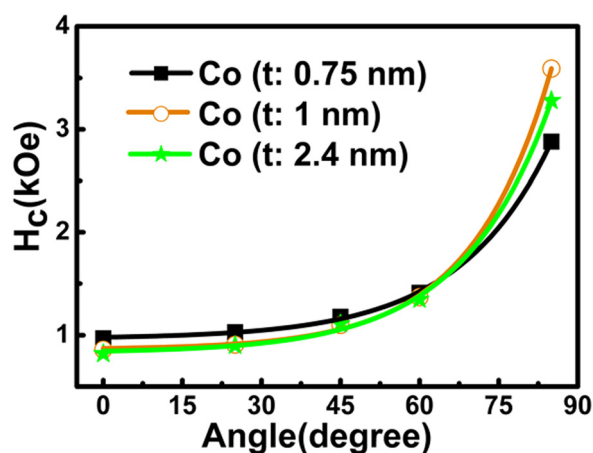


FIG. 10. (Color online) Variation of coercivity as a function of applied magnetic field direction in complex structures for selected Co thin film thicknesses (0.75, 1, and 2.4 nm).

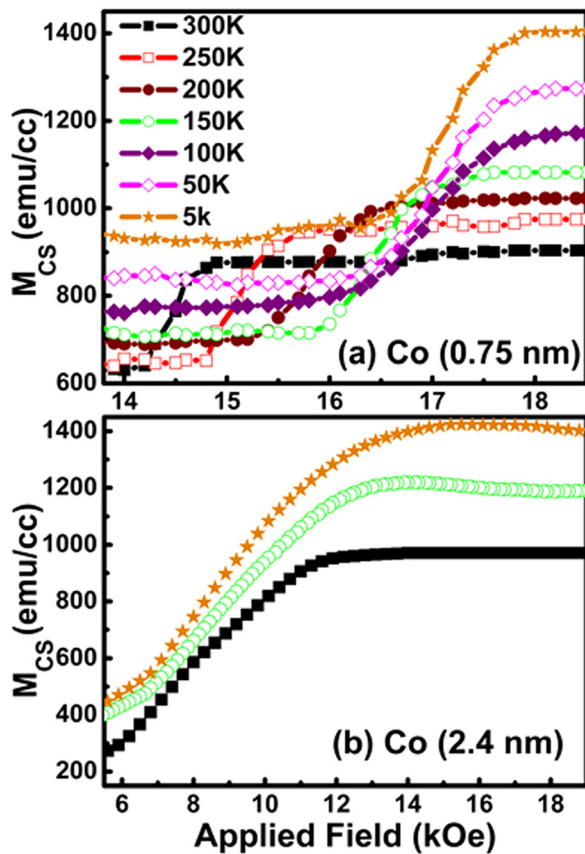


FIG. 11. (Color online) Magnetization of complex multilayer system (M_{CS}) with (a) Co film of 0.75 nm in thickness and (b) Co film with 2.4 nm in thickness measured at different temperatures ranging from 5 K to 300 K.

300 K as shown in Figures 11(a) and 11(b). It can be noticed that the sample with 0.75 nm Co, in general, switches in a narrower range of field, indicating spin reorientation. On the other hand, the sample with 2.4 nm thick Co does not show a reversal at all temperatures, indicating the absence of spin-reorientation even at 5 K. For sample with 0.75 nm thick Co layer, the measured FWHM of the derivative of M-H loop (SFD) is 0.51 kOe at 300 K and 0.89 kOe at 5 K. This is attributed to the increase of saturation magnetization at reduced temperatures.³¹ It can also be seen from Fig. 11(a) that the exchange coupling field of a complex structure with 0.75 nm Co thin film increases to 17.2 kOe at 5 K. The enhancement in H_{ex} (as temperature decreases) may arise from an increase in J or a decrease in M_s . However, our measurements indicated an increase of saturation magnetization of Co, as reported by Gong *et al.*,³² from about 800 emu/cc at 300 K to about 1300 emu/cc at 5 K. Therefore, the increase in H_{ex} is expected to arise due to an increase in J . The value of exchange coupling energy J , calculated using the experimentally measured values of M_s , H_{ex} , and t shows an increase from 1.22 erg/cm² at room temperature to 2 erg/cm² at 5 K. Moreover, it was observed that the saturation field increases from 13.6 kOe and 15.11 kOe at room temperature to 18.1 kOe and 18.25 kOe at 5 K for a multilayer system with 2.4 nm and 0.75 nm Co thin film, respectively. These enhancements of saturation field for the films indicate that a large interlayer coupling is induced between ferromagnetic layers.^{32,33}

IV. CONCLUSIONS

In summary, spin-reorientation in antiferromagnetically coupled structures was studied using AGM and AHE. A very high exchange coupling field of 15 kOe at room temperature was observed due to a large antiferromagnetic coupling constant J for Co with thickness of 0.75 nm. AHE was used to investigate magnetization reversal of Co thin films with in-plane anisotropy exchange coupled antiferromagnetically to 17 nm (Co/Pd) multilayers with a perpendicular magnetic anisotropy. For thin Co film with a thickness of 2.4 nm, the exchange coupling strength is not high enough to induce a spin reorientation of its magnetization. The observed high exchange coupling field of 15 kOe obtained in samples with 0.75 nm of Co is significant for spintronics and bit patterned media applications.

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